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**Resource Constraints for the Diffusion of Fuel Cell Vehicles: Assessing the Role of Recycling in Meeting Future Platinum Demand**

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## **Abstract**

Classified as 'zero emission vehicles', fuel cell vehicles do not emit any greenhouse gases while driving, but still offer the familiar convenience of conventional vehicles. For this reason, they are expected to play a major role in combating climate change under the European Union's quest for reducing greenhouse gas emissions.

However, the widespread deployment of fuel cell vehicles entails trade-offs and unintended rebound effects, especially concerning the depletion of non-renewable resources. The excessive platinum requirement is of major concern, as this metal is used as the predominant catalyst material in automotive proton exchange membrane fuel cells. Platinum recycling is viewed as a vital strategy for mitigating environmental and resource-related impacts of extraction, as well as enhancing security of supply.

This thesis thus aims to assess to what extent the recycling of fuel cell vehicles can contribute to meeting this industry's future platinum demand. Following a theoretical basis that provides the fundamental principles of current proton exchange membrane fuel cell technology, the market status of fuel cell vehicles and the global platinum market, a literature review examines the potential recycling chain of fuel cell vehicles in qualitative terms. Analysing possible deficits in the recycling chain that could lead to sub-optimal platinum recovery rates in the future, this part concludes that the recycling of fuel cell vehicles is still in its early stages and many aspects require further research.

Using the software STAN 2.5, a dynamic material flow analysis is prepared to determine in quantitative terms the expected platinum requirements resulting from the diffusion of fuel cell vehicles in Europe, as well as the platinum flows resulting from the recycling of end-of-life fuel cell vehicles.

In the course of this research it is shown that the diffusion of fuel cell vehicles in Europe is unlikely to cause a depletion of primary platinum deposits, but could have severe impacts on the global platinum market and exacerbate structural and temporal scarcities. These effects could not only impede the market adoption of fuel cell vehicles, but also impact on other platinum-dependant technologies. The co-development of recycling technologies and frameworks is hence considered a prerequisite, and suggestions for the mitigation of deficits in the recycling chain of end-of-life vehicles are given. Further research on all steps of the recycling chain of fuel cell vehicles is deemed essential.

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## List of Acronyms and Abbreviations

°C	degree Celsius
°K	degree Kelvin
µm	micrometre
a	annum
Ba	barium
BEV	battery electric vehicle
BoP	balance of plant
Ca	calcium
CaCO <sub>3</sub>	calcium carbonate
CCM	catalyst coated membrane
CCS	catalyst coated substrate
cf	confer
Cl	chlorine
cm	centimetre
Co	cobalt
CO	carbon monoxide
CO <sub>2</sub>	carbon dioxide
COS	carbonyl sulfide
DERA	Deutsche Rohstoffagentur ('German resource agency')
ELV	end-of-life vehicle
EoL	end-of-life
EPR	Extended Producer Responsibility
EU	European Union
EU-15	all member states of the European Union before the Eastern extension in 2004
EU-27	all member states of the European Union up until the extension in 2007
F	fluorine
FC	fuel cell
FCV	fuel cell vehicle
Fe	iron
g	gram
G8	Group of 8 (the 8 leading industrialised nations)
GDL	gas diffusion layer
GHG	greenhouse gas
H	atomic hydrogen
H <sub>2</sub>	hydrogen
H <sub>2</sub> O	water
H <sub>2</sub> O <sub>2</sub>	hydrogen peroxide
H <sub>2</sub> PtCl <sub>6</sub>	hexachloroplatinic acid
H <sub>2</sub> S	hydrogen sulfide
HCl	hydrochloric acid
HF	hydrogen fluoride
HHI	Hirschman-Herfindahl-Index
HNO <sub>3</sub>	nitric acid
i.e.	id est ('that is')
ibid	ibidem ('in the same place')

ICE	internal combustion engine
k oz	kilo ounces
kg	kilogram
kHz	kilohertz
kW	kilowatt
l	litre
LCA	life cycle assessment
MEA	membrane electrode assembly/ies
Mg	manganese
mg	milligram
mm	millimetre
Ni	nickel
nm	nanometer
NO <sub>x</sub>	nitrogen oxide
NSTF	nano-structured thin film
O	atomic oxygen
O <sub>2</sub>	oxygen
oz	ounce
p	page
Pa	Pascal
Pd	palladium
PEM	proton exchange membrane
PEMFC	proton exchange membrane fuel cell/s
PFA	perfluoroalkyl sulfonyl fluoride
PFSA	perfluoro sulfonic acid
PGM	platinum group metals
pp	pages
ppm	parts per million
PS	horsepower
Pt	platinum
PTFE	polytetrafluorethylene
Rh	rhodium
Sr	strontium
t	metric ton
TFE	tetrafluorethylene
Ti	titanium
UK	United Kingdom
US	United States of America
US DOE	United States Department of Energy
WGI	World Governance Indicators
WIPO	World Intellectual Property Organization
wt. %	percentage by weight

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# 1 Introduction

## 1.1 Background and context

Climate change has become “*the major, overriding environmental issue of our time, and the single greatest challenge facing environmental regulators*” (UNEP 2015). Despite international agreements on climate change mitigation, which have, amongst others, delivered the long-term goal of limiting global warming to 2 degrees Celsius compared to pre-industrial times, climate change remains a growing concern. As global temperatures continue to climb, ice loss in the Arctic and Antarctic is accelerating, sea levels are rising and weather patterns are changing, it is increasingly impacting on human interests such as food production, health and safety, availability of ecosystem services and the functioning of the global economy (cf. WMO 2013).

While individual media statements may portray it differently, there is a general scientific consensus that the global climate is indeed changing, and that humanity’s emissions of greenhouse gases<sup>1</sup> (GHG) are contributing to this. Although there is already alarming evidence that certain tipping points in the Earth’s climate are being approached, UNEP (2015) asserts that the worst impacts may still be avoided by installing specialised adaptation programmes and, above all, by facilitating the transition towards a low carbon economy<sup>2</sup>.

In 2011, the European Council reconfirmed its determination to meet the 2 degree-target through EU-wide reductions of GHG emissions by 80% to 95% (compared to 1990 levels) by 2050 (European Commission 2011, p. 3). As part of its ‘Roadmap for moving to a competitive low carbon economy in 2050’, the European Commission (2011) has identified key sectors and development pathways to enable the EU to reduce GHG emissions according to the targets agreed. Among these, transport plays a major role, as it represents Europe’s second-largest source of GHG emissions today, according to the European Climate Foundation (2015). Road transport alone makes up one-fifth of the EU’s total emissions of carbon dioxide (European Commission 2015b). In order to meet

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<sup>1</sup> in particular carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) (UNEP 2013, p. 3).

<sup>2</sup> No common definition for this term exists; however, it generally signifies an economy that produces only minimal GHG emissions. The reduction targets agreed by EU and G8 leaders in 2009 may be seen as a reference level for a ‘low carbon economy’ (RSC 2015).

the overall reduction target of 80% to 95% by 2050, transport-related GHG emissions in the EU must therefore be cut by 54% to 67% by 2050 (European Commission 2011, p. 6).

Despite ongoing improvements to the emission standards of internal combustion engine vehicles, under the current vehicle portfolio (i.e. the mix of small, medium and large vehicles) it will not be possible to meet these ambitious reduction targets without the integration of so-called ‘zero-emission vehicles’, which include battery electric and fuel cell vehicles (Mohr dieck et al. 2014, p. 60). In light of these developments, the concept of electromobility is gaining momentum and promises additional benefits. As the European Commission (2011) states:

*“The synergies with other sustainability objectives such as the reduction of oil dependence, the competitiveness of Europe's automotive industry as well as health benefits, especially improved air quality in cities, make a compelling case for the EU to step up its efforts to accelerate the development and early deployment of electrification”* (p. 7).

Although battery electric vehicles have been at the forefront of public attention, due to their short range, limited by battery capacities, they are best-suited for urban driving<sup>3</sup>. Fuel cell vehicles, on the other hand, combine the emission benefits of battery electric vehicles with the range and refuelling time provided by conventional internal combustion engine vehicles. With these characteristics, they are ideally suited to personal transportation in medium and larger cars, as well as longer trips (cf. BMVBS 2011). According to McKinsey & Company (2010, p. 32), this market segment alone accounts for around half of all passenger cars in the EU and is responsible for 75% of transport-related CO<sub>2</sub> emissions. Hence, substitution of conventional vehicles with fuel cell vehicles within this segment offers the potential of relatively higher reductions in CO<sub>2</sub> emissions (ibid. p. 4). This powertrain technology will hence be at the focus of this thesis.

However, potential trade-offs and unintended rebound effects resulting from the endorsement of novel powertrain technologies have to be considered carefully. As highlighted by a number of research publications, electromobility not only entails environmental benefits, but also has substantial impacts on the demand for non-renewable re-

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<sup>3</sup> Note that the advantages and disadvantages of the respective powertrain technologies will be discussed in more detail in Chapter 3.

sources, in particular for so-called special or technology metals (cf. Råde 2001; Gordon et al. 2006; Elshkaki 2007; Angerer et al. 2009; Buchert et al. 2009; Yang 2009; Buchert et al. 2011).

With regards to fuel cell vehicles, platinum is of particular concern, as it represents the main catalyst material of the proton exchange membrane fuel cell that is used in automotive applications. The extraction of platinum group metals from primary deposits involves high environmental impacts, including emissions of sulphur dioxide, of CO<sub>2</sub> equivalents in the range of 13,000 tons per ton of platinum group metals (Saurat & Bringezu 2008, pp. 760-761), excessive water and energy consumption (Mudd 2010, pp. 108-110), as well as habitat destruction, air and water pollution and generation of dust, particulate matter and solid waste (Cairncross 2014). In addition, platinum is considered a critical metal in the EU due to its geological scarcity, its use in a variety of technologies and its highly concentrated supply base (EC 2014b). Because of the limited annual supply and unfavourable mining conditions, platinum prices are expected to remain high, which has been identified as a major barrier to the diffusion of fuel cell vehicles by raising the cost of production (Bernhart et al 2014, p. 15). Taken altogether, these aspects cast doubts on the endorsement of fuel cell vehicles for sustainable development.

As UNEP (2012) points out, “*Metal recycling is an important strategy to increase the economic benefit of extracted metals and to improve pressure on primary metals and the environment*” (p. 22). However, while various publications discuss the diffusion of fuel cell vehicles and the resulting platinum demand (cf. Appendix 1), few examine the role of recycling in satisfying the industry’s platinum requirements. This is especially astonishing considering the fact that recycling rates vary significantly between applications. According to UNEP (2012), valuable precious metals, including platinum, typically have high recycling rates, “*except in some applications*” (p. 23). Passenger cars represent one such application with relatively low platinum recycling rates, as the recycling quotas for exhaust gas catalysts reach a mere 50% to 60% (Hagelüken 2012, p. 5). This makes the automotive industry the largest net consumer of platinum today, even when growth of vehicle sales is ignored (Bernhart et al. 2014, p. 15).

*“These rates signal a large amount of wasted metal and point to the need for strengthening institutional frameworks, and the logistics and technologies for metal recycling in many countries in the world. [...] Enhancing recycling for consumer applications needs to be a priority in developing policy, and practical solutions are required.”* (UNEP 2012, p. 23).

It is therefore in the interests of both fuel cell and car manufacturers to close these loops and focus on the contribution of recycling for meeting their future platinum demand.

## **1.2 Aims and research objectives**

Accordingly, this thesis aims to assess in what way the recycling of fuel cell vehicles could play a role in mitigating or compensating for the resource-related issues explained above.

Hence, the central research problem of this thesis is as follows: To what extent can the recycling of platinum from fuel cell vehicles contribute to satisfying the industry’s future platinum demand?

Research objectives arising from this central research problem are:

1. How will fuel cell vehicles diffuse over time and what impact will this have on the global platinum demand?
2. To what degree can this demand be satisfied by primary production considering the competition with other future-oriented technologies and the classification of platinum as a critical resource?
3. What is the current status of platinum recycling from automotive fuel cells, which recovery rates can be achieved and what are barriers to effective recycling?
4. What lessons can be learned from the recycling of internal combustion engine vehicles in order to design efficient recycling frameworks for fuel cell vehicles?
5. What platinum flows can be expected from the recycling of fuel cell vehicles at a future point in time?

Due to data availability, but especially with a view to the EU’s paradigm of sustainable development and its objective of reducing GHG emissions by 2050 as described above, this thesis focuses on the European market until 2050 in particular. This restriction applies both to the qualitative assessment of expected levels of market penetration and the recycling potential as well as to the quantitative assessment of platinum demand and recycling flows arising from a diffusion of fuel cell vehicles in Europe.

### 1.3 Methodical approach and thesis structure

The methodical approach employed in this thesis serves the purpose of answering the central research problem and subordinate objectives outlined above. It consists of a qualitative and a quantitative part, the general structure of which is shown in Figure 1.

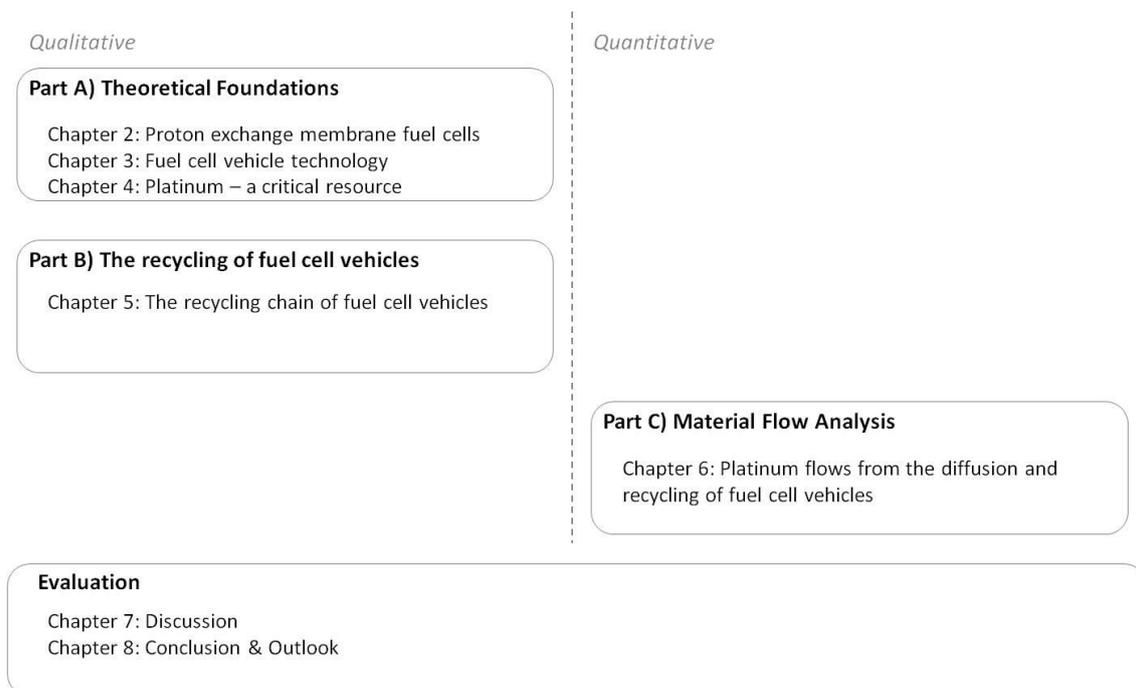


Figure 1: Structure and outline of thesis (own source)

In order to lay down the theoretical foundations for the subsequent analyses, the thesis begins with a literature-based part (A) that addresses research objectives (1) and (2). This part discusses current proton exchange membrane fuel cell technology, the basic functioning of fuel cell vehicles and their market potential, as well as providing insight into the various factors that influence the global platinum market and the classification of platinum as a critical resource.

The following part (B) addresses research objectives (3) and (4) and forms the qualitative basis for the later quantitative assessment of the recycling potential of fuel cell vehicles. Based on a review of technical and non-technical literature, this part examines the current status of fuel cell vehicle recycling as well as options for the design of a future recycling system. Drawing on analogies to the recycling of end-of-life internal combustion engine vehicles, potential deficits that could lead to suboptimal rates of platinum recovery are identified.

At the core of this thesis is a dynamic material flow analysis<sup>4</sup> (part C), modelling the flows of platinum throughout a fuel cell vehicle's life cycle, in order to determine the potential for meeting platinum demand through the recycling of end-of-life fuel cell vehicles. This part hence forms the quantitative assessment for attending to research objective (5). It is based on the information gained in the preceding parts, as data concerning, for example, the manufacturing process of fuel cells, the expected development of platinum loads and the market penetration of fuel cell vehicles, as well as possible recycling rates, govern the mass flows, stocks and processes established within the material flow analysis.

Based on the information presented in Part A, B and C, the discussion then focuses on evaluating both the qualitative and quantitative data to answer the central research question. A conclusion is given concerning the role of recycling in meeting the future platinum demand arising from a diffusion of fuel cell vehicles in Europe.

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<sup>4</sup> Note that the concept of material flow analysis will be explained in more detail in Chapter 6.1.

## 2 Proton exchange membrane fuel cells

*'Fuel Cells [...] are electrochemical devices that directly convert chemical energy stored in fuels such as hydrogen to electrical energy'* (Wang et al. 2010, p. 982). Different types of fuel cells, which are classified by their respective electrolyte, have distinguishing features and characteristics. Due to its favourable attributes, such as low operating temperature, fast start-up and fast response to varying loads, the type of fuel cell used in current automotive applications is the Proton Exchange Membrane fuel cell, which is also known as Polymer Electrolyte Membrane fuel cell (PEMFC)

(Nel 2004, pp. 290-291). In the following, a brief overview of the function and design of a PEMFC is given.

### 2.1 Function and design of proton exchange membrane fuel cells

PEMFC consist of a membrane electrode assembly (MEA) and bipolar plates (cf. Figure 2). The MEA comprises a compound structure of polymer electrolyte membrane (PEM), gas diffusion layer and catalyst. Serving as the fuel cell's electrolyte, the PEM divides the cell into two "chambers", i.e. a positive electrode (cathode) and a negative electrode (anode) (Mohr dieck et al. 2014, p. 78). Gas-proof but proton-conductive, the PEM consists of a 50 to 150 µm thick foil, which is known mainly by its registered trade name Nafion® (Kurzweil 2013, p. 79). The PEM is sandwiched between the electrodes, which consist of a gas diffusion layer (GDL) and a catalyst layer that is usually composed of carbon-supported platinum or platinum-alloys (ibid., pp. 87-88).

While the GDL serves several purposes, including gas dispersion and removal of by-produced water (Mohr dieck et al. 2014, p. 79), the catalyst layer is incorporated into the MEA in order to speed up the required reactions by lowering the energy thresholds that must be overcome, i.e. lowering activation energy (Lehmann & Luschtinetz 2014, p. 31). Flow field plates, made of graphite, specially coated steel or carbon-containing synthetics, frame the MEA and serve the purpose of electrical contact, gas supply, water removal and cooling (Kurzweil 2013, p. 94). The term 'bipolar plates' is derived from the fact that PEMFC are usually connected in series (i.e. fuel cell 'stacks') to increase voltage. Electrical contacting between individual cells takes place at the flow field plates, which hence must be bipolar (Lehmann & Luschtinetz 2014, p. 32).

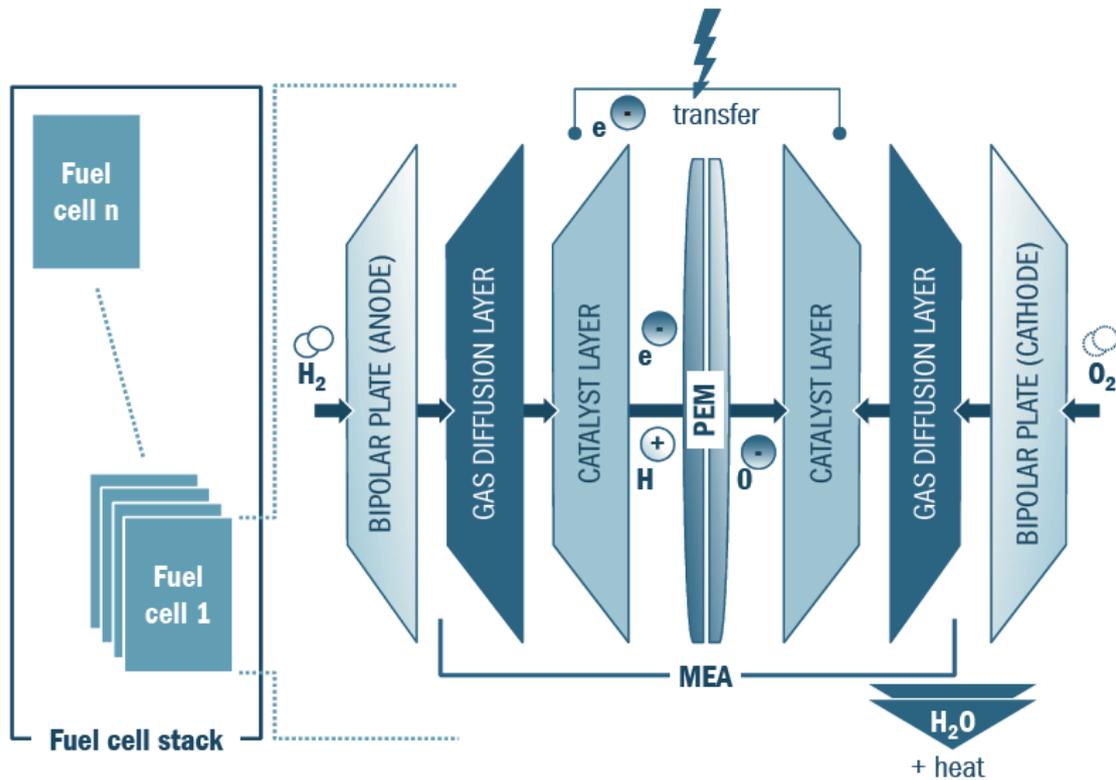
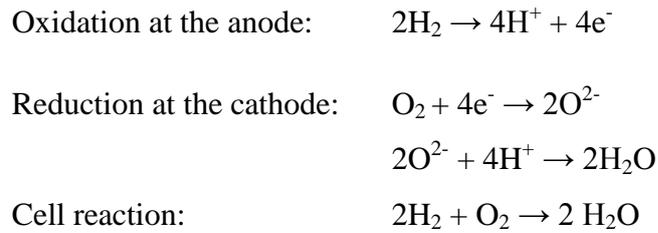


Figure 2: Schematic view of a PEMFC (Bernhart et al. 2014, p. 6)

PEMFC generate electricity as follows (cf. Figure 2): Hydrogen is supplied to the anode and oxygen to the cathode. At the anode, hydrogen molecules ( $H_2$ ) adsorb onto the platinum catalyst, where the hydrogen-hydrogen bond is broken. The resulting atomic hydrogen ( $H$ ) releases an electron, thus leaving the catalyst surface as protons (Holton & Stevenson 2013, p. 260). The hydrated PEM lets protons pass through, while forcing electrons to flow towards the cathode through the external circuit, thus creating a current (Bernhart et al. 2014, p. 6). At the cathode, oxygen molecules ( $O_2$ ) are separated into atomic oxygen ( $O$ ) by means of the catalyst. The atomic oxygen reacts with the protons that have passed through the PEM and the electrons supplied through the external circuit to form water ( $H_2O$ ) (Lehmann & Luschtinetz 2014, p. 31). This redox reaction takes place at a maximum of  $80^\circ C^5$  and involves the following reaction steps (Kurzweil 2013, p.78):

<sup>5</sup> This is true for the low-temperature PEMFC used in automotive applications, while high-temperature PEMFC operate at temperature of up to  $180^\circ C$  (Lehmann & Luschtinetz 2014, p. 30).



Similar to a battery, a single PEMFC produces only a limited electrical potential below 1 Volt. As mentioned above, fuel cells are therefore connected in series, in so-called ‘fuel cell stacks’, in order to provide enough voltage to power the desired application (Lehmann & Luschtinetz 2014, p. 32). In this case, seals between individual cells are required to separate reactants and cooling mediums, as are metallic or synthetic end plates to ensure the stack’s solid bond (Mohr dieck et al. 2014, pp. 79-80).

In order to integrate fuel cell stacks into the respective application, fuel cell systems that contain additional parts, such as balance of plants and other supporting components, are necessary. The definitions of fuel cell systems, and hence the components included, differ between publications. However, they may comprise pumps for fuel circulation, air blowers and compressors, power conditioning units (i.e. voltage converters), electric motor, fuel tank and control valves (Nel 2004, p. 292).

## 2.2 Membrane electrode assembly fabrication

According to Bladergroen et al. (2012), “*the true potential of the MEA is not only determined by the properties of the individual components but mainly by the MEA production method*” (p. 47). While different methods for the design of the electrode and configuration of the MEA exist, all are aimed at achieving a MEA architecture with optimal utilisation of the ‘three-phase-boundaries’, a term which denotes the three-dimensional interface where reactant gases, catalyst and membrane meet and the reactions described in Chapter 2.1 take place (ibid.). In the following, designs and production methods common at the time of writing will be described briefly. It must, however, be noted that various other MEA configurations are available and data concerning weight proportions, sizes and densities may vary accordingly.

At present, the most widely used membrane material remains to be Nafion®, which is made of a polytetrafluorethylene (PTFE, also known as Teflon®) layer that is bonded to sulphonic acid ions by either dispersion or extrusion casting. Nafion® has a density of 1.97g/cm<sup>3</sup> and is 0.0508 mm thick when used as a PEMFC membrane.

The ratio of tetrafluoroethylene (TFE) to perfluoroalkyl sulfonyl fluoride (PFS) is approximately 57.4 wt.% TFE to 42.6 wt.% PFS (Simons & Bauer 2015, p. 3). Nafion® provides a light-weight and chemically stable membrane. It must, however, be kept hydrated at all times and is susceptible to freezing at low temperatures as well as local overheating, which may cause tears (Kurzweil 2013, p. 79). Current research therefore focuses on increasing thermal stability, for example via the use of hydrophilic additives (Simons & Bauer 2015, p. 4). The membrane is typically purchased in a ready-to-use roll form and only needs to be cleaned and protonated before further processing (Koraishy et al. 2009, p. 5).

The gas diffusion layer (GDL) commonly consists of carbon cloth or carbon paper, as this provides the desired characteristics of porosity, electrical conductivity, high stability and corrosion resistance. This base material is hydrophobically treated with PTFE in order to prevent water from accumulating in the pores, a process called ‘flooding’ which reduces the ability of gases to contact the catalyst sites. Since the GDL pores are much larger in size than the catalyst particles, an additional micro-porous layer consisting of carbon or graphite particles mixed with PTFE is adhered to the GDL to improve both electrical contact and water transport. A GDL including a micro-porous layer has a density of 0.55g/cm<sup>3</sup> and a thickness of 0.41 mm, respectively, and contains on average 15% PTFE (Simons & Bauer 2015, p. 4). Development efforts focus on porosity, water management and reduction of area-specific resistance through an increased use of nano-materials (ibid., p. 5). Figure 3 summarises the GDL production process.

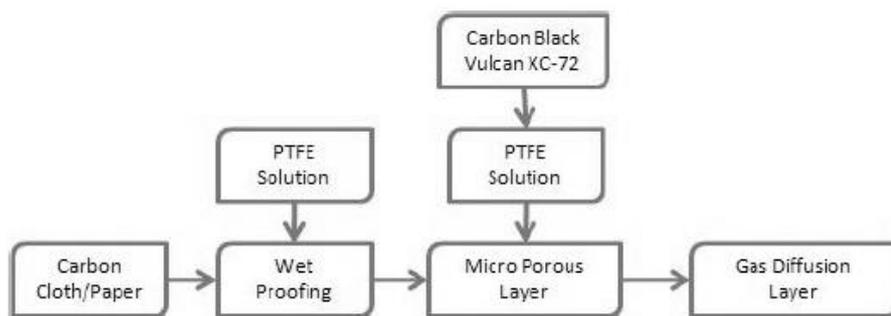


Figure 3: Gas diffusion layer fabrication process (Koraishy et al. 2009, p. 6)

Simply put, the catalyst layer enables the desired reactions within the fuel cell by providing the surface area on which these reactions take place. As Simons and Bauer (2015) state “*a high surface area is therefore far more important for high performance than high mass of catalyst material*” (p. 5). Due to its preferable characteristics, platinum is used as the main catalyst material in PEMFC (cf. Chapter 2.3). Small particles of the catalyst material (10 to 100 nm in size) are finely dispersed on a porous substrate, which usually consists of carbon powders. The resulting catalyst layer is between 10 and 30  $\mu\text{m}$  thick (ibid).

According to Litster and McLean, the catalyst layer can be applied to either the PEM or the GDL, with the objective of bringing the catalyst particles as close to the membrane as possible (Litster & McLean 2004, p. 62). In addition, an intermediate procedure (the so-called ‘Decal-method’) can be employed, in which the electrode is cast onto a substrate, such as Teflon film, hot-pressed to the membrane and the film then peeled off (Koraishy et al. 2009, p. 4).

Figure 4 provides an overview of available techniques for application of the catalyst layer, which can be used in both catalyst coated membrane (i.e. the catalyst layer is applied to the membrane) and catalyst coated substrate (i.e. the catalyst is applied to the GDL) processes. With a view to the limited scope of this thesis, the various catalyst application methods listed below cannot be reviewed in detail, but may be examined further in Litster & McLean (2004), Kaz (2008), Koraishy et al. (2009) and Bladergroen et al. (2012).

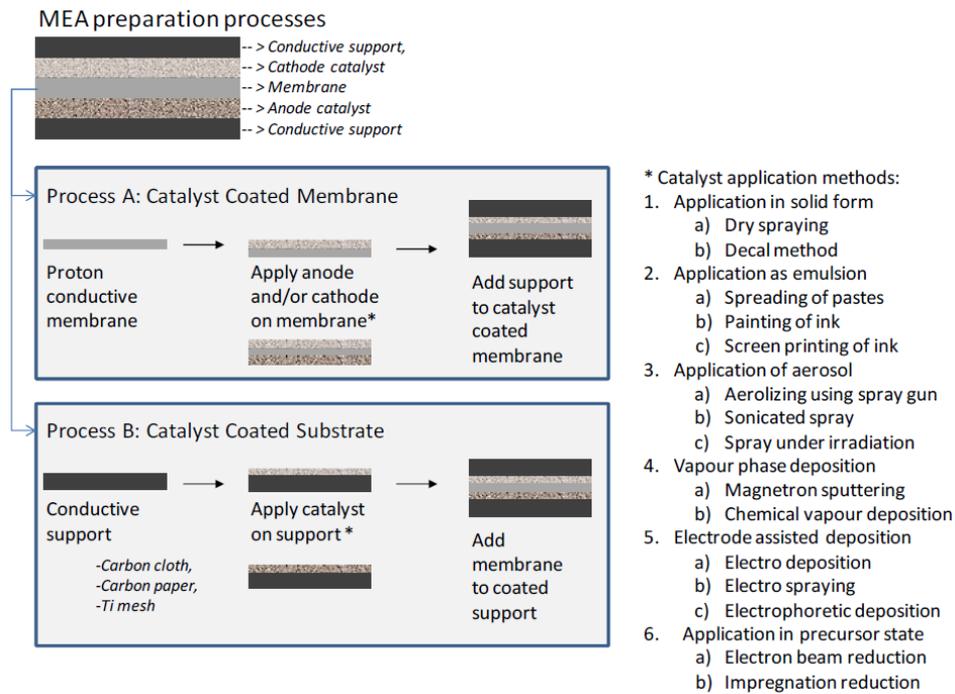


Figure 4: Overview of catalyst application methods (Bladergroen et al 2012, p. 50)

Many of the procedures listed above are used for low-volume, manual production in academia or for specific applications only and do not lend themselves to higher production volumes. Commercial producers today typically use procedures similar to those of the printing industry, in which the supported or unsupported catalyst material is mixed with solvents, binder and other additives to form a ‘catalyst ink’ and then applied in wet form (Kaz 2008, p. 29; Koraisly et al. 2009, p. 6). For future higher-volume production, however, dry coating methods, in which the electrode material is deposited onto the substrate in the form of a powder, are considered a viable route of development (Kaz 2008, p. 29).

The most commonly used catalyst application methods today involve a spray-coating or doctor blade-coating process, in which a thin film of catalyst ink is applied directly to the membrane, resulting in a catalyst coated membrane (CCM) (cf. Kurzweil 2013; Bernhart et al. 2014; Simons & Bauer 2015). Various catalyst ink formulations have been developed, the exact recipes of which depend not only on the MEA’s desired performance and application, but also on the fabrication method to be used, as the viscosity and solid content must be tuned to fit the procedure.

In general, however, the catalyst ink consists of the following ingredients in varying proportions (Koraishy et al. 2009, p. 6):

- solvent (deionised water, isopropyl alcohol, glycerol etc.),
- binder (perfluorosulfonic acid (PFSA) or other ionomer in protonated form),
- catalyst (platinum, unsupported or supported on carbon powder),
- additives (gelling agents, release agents, pore formers etc.).

Although the development of such thin-film electrodes has resulted in a significant reduction of the required platinum loadings by increasing the surface area of the catalyst, research on improved MEA production methods, aimed at reducing the platinum load further, continues to be a major focus of fuel cell technology research (cf. Chapter 2.3).

One possible development pathway is presented by current research on nanostructured thin-film (NSTF) catalysts, a technology developed and pursued by the US-based company 3M that features significantly lower platinum loadings, while also addressing performance and durability issues. For the production of these catalysts, platinum alloys (platinum alloyed with nickel (Pt-Ni), cobalt (Pt-Co), manganese (Pt-Mn), iron (Pt-Fe) or titanium (Pt-Ti)) in a 3 to 1 ratio are sputtered consecutively, i.e. finely deposited in the form of a vapour, onto a polymer support. This produces polymer ‘whiskers’ or tubes finely covered with a thin film of the platinum alloy, thus increasing the active area of the catalyst (van der Vliet 2010, p. 105; Simons & Bauer 2015, p. 5). A further differentiation of this technology from other electrocatalysts is beyond the scope of this thesis, but can be reviewed in, for example, Debe (2012), Debe et al. (2012), Debe (2013), Steinbach et al. (2014) and US DOE (2014).

In a final step, the catalyst coated membrane is hot-pressed to the GDL at around 130°C and 80 bar pressure in order to form the MEA (Simons & Bauer 2015, p. 5).

### **2.3 The role of platinum in proton exchange membrane fuel cells**

As described in the preceding chapters, a platinum catalyst is generally used for both the oxidation reaction taking place at the anode and the reduction reaction occurring at the cathode. While the oxidation of hydrogen proceeds very fast and incurs negligible voltage losses even at very low platinum loadings, the reduction of oxygen involves a much slower and more complicated mechanism.

Due to the slower kinetics, the oxygen reduction is responsible for a large portion of the voltage loss of a PEMFC and the cathode thus requires a several times higher platinum loading than the anode (cf. Holton & Stevenson 2013). Platinum content, however, is considered one of the main cost drivers of PEMFC, significantly limiting their market potential (cf. Chapters 2.3.2 and 3.3). Reduction of the platinum load is therefore a major focus area of fuel cell technology research. Although platinum loadings at the anode do of course contribute to PEMFC's overall platinum content, research on catalyst improvement generally focuses on the cathode rather than the anode process (Holton & Stevenson 2013, pp. 260-261). In the following, the characteristics of platinum which make it the material of choice for PEMFC catalysts, as well as the required platinum loadings at current and future technology levels, will be reviewed.

### 2.3.1 Characteristics of platinum

According to Holton and Stevenson (2013, p. 269), pure platinum outperforms all currently known substitutes in the characteristics *Activity*, *Selectivity* and *Stability*, which are crucial for an effective PEMFC catalyst.

Adsorbing reactants strongly enough to break chemical bonds, but weakly enough to discharge the resulting products once the reaction has been completed, is the balancing act that effective catalysts must achieve. This ideal relationship between the catalyst material and the reactant is known as the 'Sabatier Principle' and can be visualised by a volcano diagram, which plots the catalyst activity against the binding energy for a given reaction (Holton & Stevenson 2013, p. 262). Figure 5 shows a volcano plot for binding of atomic oxygen with a number of metals, illustrating the fact that platinum is closest to the optimum activity level. Note that the optimal catalyst according to the Sabatier principle would sit at the peak of the diagram.

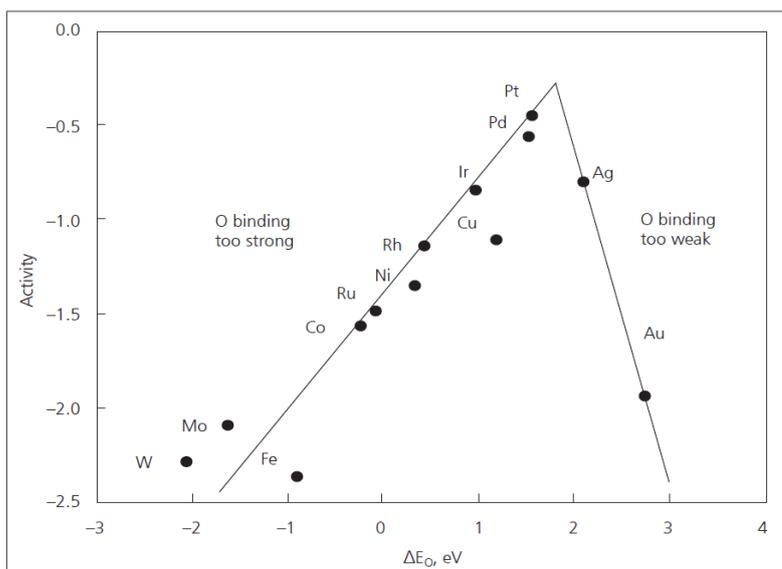


Figure 5: Oxygen reduction activity plotted as a function of oxygen binding energy (Norskov et al. 2004 in Holton & Stevenson 2013, p. 263)

At the same time, catalyst materials must ensure that the oxygen reduction reaction follows the desired pathway leading to the formation of water ( $H_2O$ ) rather than the corrosive hydrogen peroxide ( $H_2O_2$ ), which happens when oxygen is adsorbed onto a metal surface without the bond between the two oxygen atoms being broken. Platinum has been shown to demonstrate the highest selectivity for the desired  $H_2O$ -producing mechanism among various metal catalysts, proceeding almost exclusively according to the desired pathway, as the oxygen bond is broken immediately upon adsorption (Holton & Stevenson 2013, p. 264).

Since conditions inside the fuel cell include the presence of oxidants, reactive radicals, low pH value, high temperatures and potential fluctuations, stability within such a chemical environment is another prerequisite for an effective catalyst. The few other metals, including for example gold and iridium, which like platinum are noble enough to not dissolve in the PEMFC's harsh conditions, perform worse in terms of activity and selectivity, making platinum the only pure metal suitable as a PEMFC catalyst material (Holton & Stevenson 2013, p. 265).

Despite these favourable attributes, however, durability is an issue for platinum-based catalysts. For example, poisoning (i.e. chemical deactivation) by impurities contained in the fuel cell or the supplied reactants affects all catalysts and here, platinum is no exception. Poisoning by carbon monoxide (CO), sulphur ( $H_2S$ , COS) and organic compounds is most problematic for platinum-based catalysts (Kurzweil 2013, p. 92).

While quality standards for hydrogen aimed at minimising impurity levels have been agreed, alloying of platinum with other metals to improve poisoning resistance would be beneficial (Holton & Stevenson 2013, p. 267). Additionally, sintering of platinum particles (i.e. accumulation of particles due to movement on the carbon support or dissolution and subsequent precipitation onto larger metal particles), dissolution of platinum from the carbon surface and corrosion of the carbon support may all negatively affect the stability and hence the durability of platinum catalysts (He et al. 2005, p. 43-44).

### 2.3.2 The platinum content of fuel cell vehicles

The superior characteristics of platinum in terms of activity, selectivity and stability combined with its high cost (cf. Chapter 4.4.2), as well as the above mentioned issues concerning durability, have sparked research aimed at reducing or replacing platinum loadings while maintaining or even improving PEMFC performance. Current research activities focus on the following approaches (cf. He et al. 2005; Holton & Stevenson 2013; US DOE 2014):

- alloying with other transition metals to bring the catalyst's adsorption properties even closer to the Sabatier optimum,
- layering platinum on or below the surface of another metal in order to create a structure binding oxygen atoms somewhat weaker than pure platinum,
- core-shell approaches, in which a cheaper metal core is coated with platinum,
- de-alloying approaches, in which the base metal content is leached from nanoparticles of platinum-alloy catalysts,
- novel technologies aimed at replacing platinum by alternative materials entirely.

Dedicated research programs have resulted in significant technology advances, allowing a considerable reduction of PEMFC platinum loadings without compromising performance in recent years. For example, He et al. (2005, p. 44) report a 50-fold reduction of catalysts' platinum content from around 25 mg/cm<sup>2</sup> several decades ago to below 0.5 mg/cm<sup>2</sup> in 2005. Although this presents a significant improvement, such platinum loadings still amounted to a total platinum content of almost 1 gram/kW<sub>net</sub> (Spendelow & Marcinkoski 2014, p. 4) or around 80 gram per FCV in 2005, when a 100 kW<sub>gross</sub> fuel cell stack is assumed (NRC 2013, p. 32).

Research efforts have, however, continued over the past decade and succeeded in further reducing the required platinum content. Table 1 lists the reduction in platinum loadings achieved from 2007 to 2013 through research programs governed by the US Department of Energy (US DOE), as well as the corresponding stack performance. Note that the increases in platinum content per kW in 2012 and 2013 are due to newly introduced mandates by the US DOE concerning minimisation of waste heat generation that required changes to system design parameters.

Table 1: PEMFC stack power and corresponding platinum content (adapted from Spendelow & Marcinkoski 2014, p. 3)<sup>6</sup>

Characteristics	Unit	2007	2008	2009	2010	2011	2012	2013
Stack Power	kW <sub>gross</sub>	90	90	88	88	89	88	89
System Power	kW <sub>net</sub>	80	80	80	80	80	80	80
Platinum loading	mg/cm <sup>2</sup>	0.35	0.25	0.15	0.15	0.19	0.20	0.15
Platinum, total content	g/kW <sub>gross</sub>	0.6	0.35	0.18	0.18	0.17	0.20	0.23
Platinum, total content	g/kW <sub>net</sub>	0.68	0.39	0.20	0.20	0.19	0.22	0.25

Assuming a fuel cell stack of 80 kW<sub>net</sub>, the platinum loading reported for 2013 still translates into a total platinum requirement of around 20 gram per FCV. Moreover, as these are fuel cell stacks produced and tested under laboratory conditions, the platinum load of fuel cell stacks installed in FCV technology currently ‘on the road’ is likely still higher. Due to the proprietary nature of such data, exact and reliable numbers for the platinum content of FCV models manufactured today cannot be obtained; instead, assessment of the current platinum load relies on estimates and proposals published by automobile manufacturers and researchers.

General Motors, for example, propose a platinum content of 30 gram per automotive fuel cell system in 2015 (Lehman & Lushtinetz 2014, p. 118). Bernhart et al. (2014, p. 13), on the other hand, deem a platinum load of 0.4 mg/cm<sup>2</sup> feasible at current technology levels. Based on their assumption of an average active area of 300 cm<sup>2</sup> per MEA and 350 MEA within one vehicle’s fuel cell stack (ibid. p.15), this adds up to 42 grams of platinum per FCV. According to estimates by Sui (P.-C. Sui 2015, pers. comm., 05 June 2015), the 114 kW fuel cell stack of the current Toyota Mirai (cf. Chapter 3),

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<sup>6</sup> Note that Spendelow & Marcinkoski (2014) use the term ‘PGM’ rather than ‘platinum’ loading and content in the cited table, but base cost calculations on platinum prices only and also mention only platinum throughout the accompanying text. It is therefore assumed that ‘PGM’ in this table does not denote a range of platinum group metals but is used synonymously for platinum.

which uses a Pt-Co alloy as catalyst material, is likely to contain around 40 grams of platinum, although no official figures have been published.

While these estimates vary significantly, either amount is incommensurate with the 1 to 8 grams of PGM used in current catalytic converters of ICE vehicles (Lucas et al. 2011, p. 33). This not only adds significantly to the cost of FCV, but also raises concerns regarding resource availability and sustainability (cf. Chapters 1 and 0).

While research aimed at eliminating the use of PGM as catalyst materials altogether and replacing it with alternatives is ongoing, it is unlikely to produce any viable options at least in the coming decades (cf. Holton & Stevenson 2013; Bernhart et al. 2014). Hence, a further reduction of platinum load remains a vital prerequisite for commercialisation of FCV. Table 2 shows the platinum content target for PEMFC electrocatalysts in 2020, as applied throughout the US Department of Energy’s combined research programs on fuel cell technology (cf. US DOE 2015).

Table 2: Technical targets for electrocatalysts in 80 kW automotive fuel cell systems (adapted from US Drive FCTT 2013, p. 8)

Characteristics	Unit	Status 2013	Target 2020
Total platinum content	g/kW <sub>rated</sub>	0.14	0.125
Total platinum loading	mg/cm <sup>2</sup> electrode area	0.15	0.125

The figures given for the current status within Table 2 refer to platinum loadings achievable for so-called nanostructured thin-film (NSTF) electrocatalysts (cf. Chapter 2.2).

It must be noted that such ultralow-platinum NSTF catalysts at this point in time remain at a laboratory phase and are unlikely to represent the technology employed in current FCV. However, since NSTF catalysts demonstrate promising results in terms of performance and durability and are cited as the technology to be pursued in order to achieve the technical targets set by the US Department of Energy’s hydrogen and fuel cell research program (cf. US Drive FCTT 2013; Spendelow & Marcinkoski 2014; US DOE 2014; Simons & Bauer 2015), the development of such catalysts on a larger scale appears feasible.

The total content of platinum within FCV, however, will depend not only on technological developments concerning the required platinum loading, but also on the vehicle power demanded by consumers. Developments in the sector of conventional combustion engine cars have shown that advances in fuel economy, enabling a reduction in CO<sub>2</sub> emissions in theory, were in practice counteracted at least in parts by increases in vehicle weight, size and power, which in turn diminished the reduction of CO<sub>2</sub> emissions (cf. US EPA 2014). In 2014, for example, BMW's i3 electric vehicle was sold 2233 times in Germany. In the same year, however, more than five times as many exemplars of BMW's X5, a sport utility vehicle with a horsepower of 306 PS and CO<sub>2</sub> emissions of around 199 g/km, were ordered despite a higher purchasing price (Brauck et al. 2015, p. 67). This underlines the fact that "*consumer desires for large, powerful vehicles have been the major stumbling block in achieving a "green car"*" (MacLean & Lave 2003, p. 5445).

The same effect could present an issue in the further development of FCV. For example, the 80 kW<sub>net</sub> fuel cell stack proposed by the US DOE, as mentioned above, carries a total platinum content of 20 grams per vehicle assuming a platinum load of 0.25 g/kW in 2013. Considering the fact that the 2014 Toyota Mirai offers a maximum power output of 114 kW (cf. Chapter 3), a future FCV with a fuel cell stack of 150 kW may well be imagined. Based on the targeted platinum loading of 0.125 g/kW in 2020, such a FCV would still require a total of 18.75 grams of platinum, rendering any technological advances largely irrelevant.

Similarly, governmental and other regulations may impact on the required platinum loading of future FCV. This can be observed in Table 1, showing that more stringent heat rejection regulations of the US DOE led to increased platinum loadings from 2012 onwards due to necessary system changes. While a more detailed analysis of such trends in consumer wants as well as governmental and other regulations and the resulting impact on vehicle specifications would be highly useful, due to constraints of space and time these aspects cannot be considered further as part of this thesis.

### **3 Fuel cell vehicle technology**

#### **3.1 Classification of fuel cell vehicles**

Converting the chemical energy of a fuel to electric energy and waste heat, fuel cells alone cannot propel a vehicle but require an electric motor to do so (Lehmann & Lutschinetz 2014, p. 52). Fuel cell vehicles therefore fall into the category of electric vehicles, a term which is used to describe a wide variety of vehicle concepts and propulsion systems. Electric vehicles can be further divided into hybrid electric vehicles, using both a conventional internal combustion engine (ICE) and an electric motor in varying ratios as power sources, and all-electric vehicles, which include battery electric vehicles (BEV) and fuel cell vehicles (FCV) (Tie & Tan 2013, p. 5). While both battery electric vehicles and fuel cell vehicles have zero tail-pipe emissions while driving and are hence classified as ‘zero-emission vehicles’ (CEPA 2012, p.1), they differ in inherent attributes such as driving range, efficiency, cost and recharging speed (Offer et al. 2010, p. 25).

Although superior to ICE vehicles in terms of power and acceleration, with a maximum range of approximately 150 kilometres per battery charge and a battery charging time of up to 8 hours, the current BEV technology cannot compete with typical ICE vehicles’ performance (Thielmann et al. 2012, p. 7). Due to the comparably low energy density of the power source, BEV are therefore best suited for ‘urban driving’, which typically constitutes small cars and short trips, as well as car sharing concepts (McKinsey & Company 2010, p. 4). FCV, on the other hand, combine the advantages of BEV with the convenience provided by conventional ICE vehicles. Able to deliver average power at a much higher efficiency than an ICE (Offer et al. 2010, p. 25), a FCV’s driving range is determined by its fuel tank size, which can be refuelled in approximately 3 minutes and achieves a range of around 400 kilometres (Thielmann et al. 2012, p. 7). FCV are therefore ideally suited to personal transportation in medium and larger cars and longer trips. According to McKinsey & Company (2010, p. 32), this market segment alone accounts for around half of all cars in the EU and is responsible for 75% of transport-related CO<sub>2</sub> emissions. Hence, the diffusion of FCV within this segment offers the potential of relatively higher reductions in CO<sub>2</sub> emissions (ibid. p. 4) and will therefore be at the focus of this thesis.

Though FCV are often praised for their comparably higher efficiency, realised through the electric motor's above 90% efficiency (Lehmann & Luschtinetz 2014, p. 54) and the fuel cell's efficiency of around 60% (Kurzweil 2013, p. 79), their well-to-wheels<sup>7</sup> efficiency compared to BEV and conventional ICE vehicles remains disputable. This is due to the fact that the hydrogen supplied to the fuel cell can be derived from a variety of sources, including renewable and fossil resources, and through different production methods. Hence, the required energy input and greenhouse gas emissions resulting from hydrogen production vary widely (Heywood 2010, p. 8). In addition, any vehicle's fuel economy depends not only on the efficiency of energy conversion, but also on size, weight, road conditions and driving style (CAFCEP 2014, p. 7).

Assuming hydrogen production from a range of renewable and non-renewable feedstocks, McKinsey & Company (2010, p. 54) conclude that FCV achieve a well-to-wheels efficiency competitive with ICE vehicles, while BEV remain the most efficient vehicle option at the current state of technology. As several studies point out, a portfolio of different vehicle concepts is therefore required to incorporate the various transport requirements as well as economic and environmental concerns in the move towards a low-carbon economy in Europe (cf. McKinsey & Company 2010; BMVBS 2011; European Commission 2013).

In addition to personal transportation, fuel cell-powered vehicles are already deployed in public transport showcase projects in order to improve pollution levels in areas suffering from high traffic loads and congestion (cf. Hydrogen London 2013). Other application areas include heavy-duty vehicles used in freight transportation and material-handling equipment, such as forklifts (cf. IEA/OECD 2009; NREL 2015). However, since not all of these applications are expected to have significant impacts on platinum demand (Nel 2004, p. 293), and not least with a view to the limited scope of this thesis, FCV other than light-duty passenger cars used for personal transportation are not going to be discussed further.

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<sup>7</sup> The term „well-to-wheels“ was coined by TIAX LLC (2007) to assess the net material impact of different vehicle/fuel combinations considering both the fuel cycle (i.e. fuel production, processing and delivery) and the vehicle cycle (i.e. vehicle energy use).

### 3.2 Technology overview

Although the distinction between hybrid electric and all-electric vehicles described above is common, it may in fact be misleading, as the fuel cell vehicles developed and manufactured today are actually hybrids using both fuel cells and batteries as power sources for propulsion (Fuel Cell Today 2013, p. 3). Pure fuel cell vehicles as well as configurations with onboard reforming of methanol or petrol to generate hydrogen are continuously discussed in literature (cf. Nel 2004; Offer et al. 2010; Franzen 2013; Kurzweil 2013), but such technologies have, at least to this date, not been pursued by car manufacturers on a larger scale and will therefore not be included in the further analysis.

From a structural perspective, FCV can be considered a type of hybrid vehicle, in which the fuel cell replaces the ICE (Chan et al. 2010, p. 591) (cf. Figure 6).

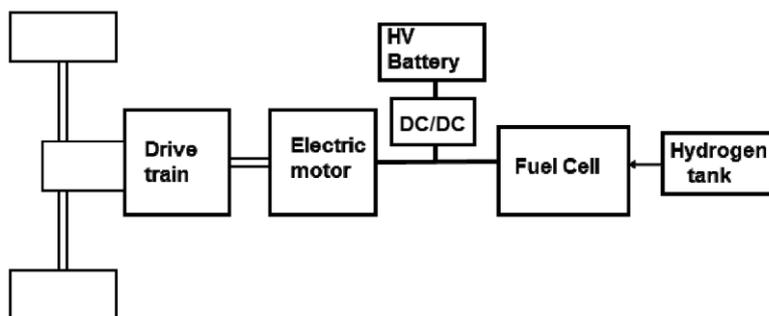


Figure 6: Schematic illustration of FCV power-train (Edwards et al. 2014, p. 75)

Using atmospheric oxygen and compressed gaseous hydrogen supplied from the on-board tank, the fuel cell generates electricity, which powers the electric motor that propels the vehicle (Daimler AG 2015). In order to supply the necessary peak power during acceleration and warm-up, fuel cell vehicles not only contain a battery as an auxiliary power source for peripherals, such as lights, radio etc., but also a traction battery. The traction battery recovers energy from the electric motor during deceleration – a process called regenerative braking – and adds power to the fuel cell’s output during acceleration, thus increasing efficiency (Lehmann & Luschtinetz 2014, pp. 54-55). A power control unit (not included in Figure 6) is required to effectively manage the energy flows between the electric motor and the two power sources (Kong et al. 2010, p. 1), as is a DC/AC and DC/DC converter to level voltages between fuel cell, electric motor and battery (Toyota Motor Corporation 2015).

This hybridization of the power-train offers certain advantages. Supported by battery power, the fuel cell does not need to be able to supply the only briefly required peak power, but can be configured for delivering average power only. This allows the use of smaller fuel cell stacks without reducing performance, as well as protecting the fuel cell from early degradation (Lehmann & Luschtinetz 2014, p. 55). Recharging the battery through regenerative braking, as well as allowing the fuel cell to operate at its optimum range by directing peak power demand to the battery, may lead to efficiency gains of up to 15%, depending on the level of hybridisation (Suh & Stefanopoulou 2005, p. 2).

Fast transient response of the power source is critical in automotive applications but difficult to achieve with fuel cells alone, a problem which is negated by utilising battery power for start-up and fast power responses while driving (ibid. p. 1). Since the above-mentioned drawbacks of BEV, including limited range and extensive recharging time, are counteracted by the higher energy density of the FCV's energy storage system (i.e. the hydrogen carried onboard), hybrid FCV may be regarded as combining the benefits of both technologies (cf. Offer et al. 2010; Franzen 2013).

### **3.3 Market penetration**

*'FCVs have largely overcome the technical barriers to commercialization'* (Fuels Institute 2013, p. 21), as for the first time several automobile manufacturers have recently moved beyond the production of mere demonstration models and are preparing for sales of market-ready FCV. While FCV, such as the Daimler F-Cell or Honda FCX Clarity, have been leased to select customers (predominantly in California) for several years in order to test the technology under real driving conditions and integrate consumer feedback (Fuel Cell Today 2013, p.5), the launch of the Toyota Mirai in Japan in December 2014 marks their commercial debut. Sales of the Toyota Mirai, priced at around € 66.000 before tax, are expected to launch in Europe in autumn 2015, but will be limited to several hundred vehicles (Webster 2015). Other brands, including Daimler, Honda and Hyundai, have announced plans to introduce series-produced versions of their FCV by 2017 (Fuel Cell Today 2013, pp. 14-20).

### 3.3.1 Hydrogen infrastructure as a barrier to market penetration

Market introduction and subsequent diffusion of FCV within the abovementioned time scale faces two major barriers: infrastructure and cost. The development of a hydrogen supply infrastructure has to be driven alongside vehicle development, since the hydrogen-fuelled FCVs require a sufficient network of hydrogen stations, while operators of hydrogen stations rely on a sufficient number of customers for economic viability, a dilemma which is often referred to as the ‘chicken-and-egg problem’ (Fraunhofer ISE 2013, p. 11). The construction of a suitable hydrogen infrastructure requires sophisticated investment plans as well as political support, with a theoretical future FCV market share of 25% translating into investment costs of € 3 billion for Europe in the first decade in order to provide the required critical mass (McKinsey & Company 2010, p. 7).

A number of automobile manufacturers, energy providers, public transport companies and oil companies are collaborating to develop hydrogen infrastructure solutions in Europe and several member states have dedicated development plans in place, aimed at achieving the required critical mass in hydrogen stations over the coming years. Nonetheless, the options for hydrogen refuelling to this date remain limited. Germany, which is considered a leading market for FCV, for example, possessed only 30 hydrogen stations in 2011, though a further 20 are to be constructed by 2015 (Fraunhofer ISE 2013, p. 12).

Hydrogen infrastructure developments in other European countries are lacking even further. Spain and France, for example, which can be considered large European countries in terms of surface area, currently operate only three and six hydrogen stations, respectively, while most Eastern European countries at this point in time offer no hydrogen infrastructure at all (TÜV Süd 2015). The United Kingdom and to some degree the Scandinavian countries present an exception and provide a more extensive hydrogen network due to political support for FCV development (Fraunhofer ISE 2013, p.12). Throughout the world, 594 hydrogen stations were operating at the time of writing, with many more currently at the planning or construction stage, especially in leading markets such as Germany, Japan and California (TÜV Süd 2015).

As mentioned above, fuel availability is a crucial prerequisite for consumer acceptance of fuel cell vehicles. However, it remains uncertain how many hydrogen stations must be available and in what proximities for successful market integration (Greene & Duleep 2013, p. 34). Early market introduction will therefore likely focus on *'government fleet and other return-to-base fleet operations and the high-end consumer car market in areas with an appropriate level of infrastructure'* (Fuel Cell Today 2013, p. 5).

### 3.3.2 Cost as a barrier to market penetration

A more widespread and accelerated adoption of FCV by other consumer segments can be expected with further hydrogen infrastructure developments, but will ultimately depend on how future FCV compare to alternatives in terms of cost and performance (Fuel Cell Today 2013, p. 5). Researchers of the *US Hydrogen and Fuel Cells Program* determined that FCV costs need to be reduced to US\$ 30 to 45 per kW in order to be competitive with ICE vehicles (Ahluwalia et al. 2011, p. 4619) and strive to achieve a fuel cell system cost of US\$ 40 per kW and a fuel cell stack cost of US\$ 20 per kW by 2020, respectively (US Drive FCTT 2013, p. 3).

As FCV share a number of common components and architecture with both BEV and ICE vehicles (cf. Offer et al. 2010; Greene & Duleep 2013), the following discussion will focus on the differentiating aspects of FCV, i.e. the fuel cell system. Note that the required hydrogen storage tank represents another major cost driver (Greene & Duleep 2013, pp. 18-19), however, due to the limited scope of this thesis it will not be examined further.

While cost is frequently cited as a major barrier to FCV market penetration (cf. Offer et al. 2010; Fuel Cell Today 2013; Fuels Institute 2013; Greene & Duleep 2013; US Drive FCTT 2013; Bernhart et al. 2014; James et al. 2014), detailed cost breakdowns are hard to obtain, as such information is highly confidential. The limited information on production costs that is published by manufacturers or otherwise estimated is also difficult to analyse and compare, since different sources use varying levels of detail, define the term 'cost' in different ways and in some cases do not disclose the underlying calculations (Greene & Duleep 2013, p. 13).

While some include only costs that are directly production-related, such as labour, material and energy inputs, others also count research and development, engineering, amortisation of tools and machinery, as well as overhead costs towards the total production costs. FCV cost estimates also vary significantly depending on assumptions made concerning the starting point of FCV development (i.e. whether fuel cell technology may be integrated into existing ICE vehicle architectures or structural modifications are required) and the applied learning curves<sup>8</sup> (i.e. the rate of technological progress). Further important factors influencing suggested cost levels are the assumed production volumes, as higher volumes generally result in lower unit costs due to economies of scale, and assumptions regarding the FCV's technical specifications, since these have a direct impact on material requirements (Greene & Duleep, p. 13). The estimates given in Appendix 2 exemplify cost breakdowns for the production of crucial FCV components and demonstrate the impact of economies of scale on unit costs.

While the studies in Appendix 2 were selected based on their relative comparability and their extensive data basis, as mentioned above, these estimates rest upon differing assumptions and calculations and hence this compilation does not raise claim to completeness. Rather, it serves the purpose of illustrating the fact that the MEA – and here predominantly the MEA's catalyst – takes up a large share of FCV costs, ranging from 51 to 33% of total fuel cell system costs, depending on the underlying assumptions. While other specialist components, such as the PEM, obviously contribute to the MEA's high proportion of total fuel cell system costs (cf. Bernhart et al. 2014), the catalyst layer is responsible for a major share of the MEA's production cost, of the order of 10 to 20%. Moreover, catalyst costs benefit less by economies of scale than other components, leading to a higher share of costs at increased production volumes (cf. Appendix 2).

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<sup>8</sup> Also known as 'experience curve', it describes the idea that proficiency with any process increases as it is repeated over time (cf. Yelle 1979; Stump 1987).

As can be derived from the cost breakdown pictured in Figure 7, Bernhart et al. (2014, p. 13) come to a similar assessment, concluding that material costs are the driving factor and account for around 90% of MEA production costs and virtually all of the catalyst costs.

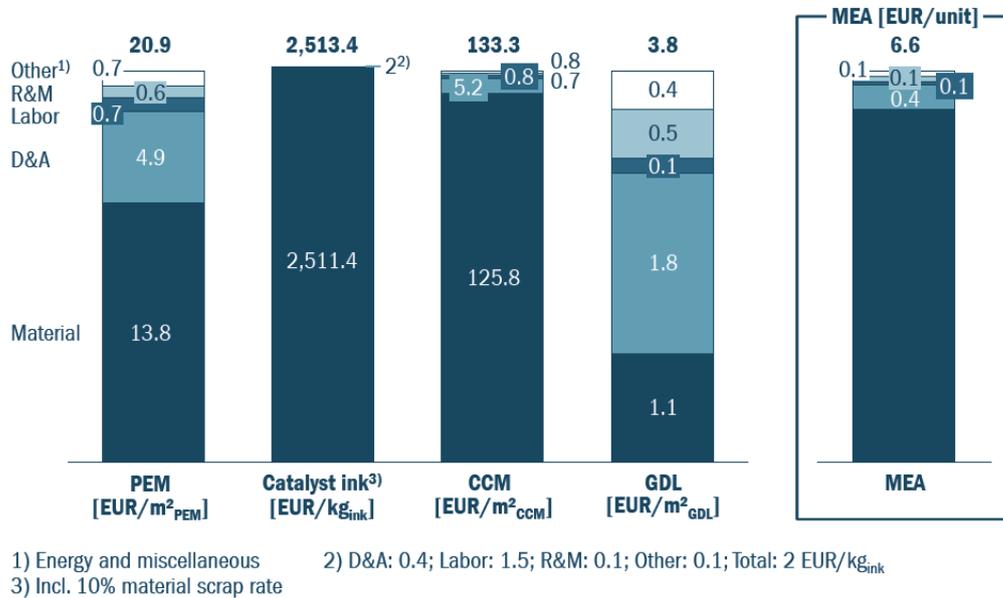


Figure 7: Cost structure by MEA component (Bernhart et al. 2014, p. 13)

As discussed in Chapter 2.3, due to its favourable characteristics platinum serves as the main catalyst material in PEM fuel cells and is likely to continue to do so over the coming decade. However, at a current market price<sup>9</sup> of around € 37 per gram, the platinum content is also responsible for a large proportion of catalyst and thus MEA costs. Assuming, as examined in Chapter 2.3, current FCVs' MEA contain 20 to 42 grams of platinum, at current market prices the platinum costs alone would amount to € 740 to € 1554.

Adding to this not only the remaining MEA costs, but also the costs of other vehicle components, such as electric motor, hydrogen tank etc., and assuming manufacturers apply the usual mark-up to recover overheads and generate a profit, Greene & Duleep (2013, p. 21) propose a retail price equivalent of US\$ 77,000 (€ 70,900) at low production volumes, which is consistent with the Toyota Mirai's expected launch price of € 66,000 before tax (Webster 2015). At such prices, FCV compare unfavourably with ICE vehicles and therefore face an economic barrier, limiting their potential for market

<sup>9</sup> As quoted by Johnson Matthey (2015a) on May 11, 2015.

adoption. This is likely to remain the case at least for the coming decade, unless technological breakthroughs (Greene & Duleep 2013, p. 21) or political measures, such as tax incentives (McKinsey & Company 2010, p. 5), are able to counteract this limitation.

Since the high price of platinum contributes significantly to the material cost of FCV, the drivers of platinum prices, the supply-demand relationship and the assessment of platinum as a critical metal will be considered in the following chapter.

## 4 Platinum – a critical resource

As analysed in the previous chapter, the current FCV technology requires platinum-based catalysts and will likely continue to do so over the coming decade; however, platinum also occupies a major share of material costs, which ultimately act as a barrier to the diffusion of FCV. With a view to this thesis' research focus, this chapter investigates the global production capacity of platinum and the competing demands of its applications, considering the impact of price developments and the contribution of recycling to satisfying global demand.

### 4.1 Physical properties and occurrence

The six precious metals referred to as the platinum group metals (PGM) are ruthenium, rhodium, palladium, osmium, iridium and platinum. All PGM are extremely rare in the Earth's crust, with platinum present at about 0.0005 parts per million (ppm) (Zientek & Loferski 2014, p. 1). Although all six PGM are chemically similar, platinum and palladium are the most commercially important, followed by rhodium (Gunn 2014, p. 285).

The physical properties of the PGM vary markedly, though “*all display properties typical of metals, including the ability to form alloys, to conduct heat and electricity, and some degree of malleability and ductility*” (Gunn 2014, p. 285). Platinum and palladium are soft and ductile, but at the same time highly resistant to heat and corrosion, while platinum, iridium and osmium present the densest known metals on Earth (ibid.). The important properties that make PGM attractive for many industrial applications include their catalytic properties as well as high strength, high melting point and high resistance to corrosion (ibid. p. 297).

The PGM preferentially form bonds with iron, nickel, copper and sulphur, rather than with oxygen, and their distribution is thus governed by metallic phases (Gunn 2014, p. 285). Rarely occurring as native metals, they commonly form alloys with one another or with other metals, particularly with iron and less often with tin, copper, lead, mercury and silver (Polinares Consortium 2012, p. 2). Most of the world's PGM resources are magmatic ore deposits, which are formed through the cooling and crystallisation of magma (Gunn 2014, p. 286).

At concentrations of 1 to 10 ppm of PGM, ore deposits are considered viable for exploitation (European Commission 2014a, p. 124). Such enrichment of PGM can be found in several types of deposits, mainly associated with nickel and copper. PGM resources can be split into PGM-dominant resources (i.e. PGM are the main products with nickel and copper as by-products) and nickel-copper-based deposits (i.e. PGM are a by-product of nickel and copper production) (ibid.).

## **4.2 World resources and primary production**

Estimating the global resources and reserves<sup>10</sup> appears difficult and cited amounts differ between publications (cf. Cawthorn (2010) for a summary of publications on South African PGM deposits and Mudd (2010) for detailed estimates of global resources and reserves). A figure cited throughout the relevant literature (cf. Mudd 2012, p. 3; Polinares Consortium 2012, p. 3; European Commission 2014a, p. 125; USGS 2015, p. 121) is the estimate by USGS (2011, p. 121) quoting global PGM reserves of 66,000 metric tons and resources of more than 100,000 metric tons. According to Cawthorn (2010, p. 213), platinum resources in South Africa's Bushveld Complex alone exceed 10,000 metric tons (approx. 350 million troy ounces) per kilometre depths. Due to the high cost of exploration and strict regulations concerning the publication of 'proven' or 'inferred' resources and reserves, mining companies generally only report resources they are confident can be extracted within the next 20 years. PGM-containing geological deposits for which extraction may become feasible in the future, are therefore likely much greater than estimates published today suggest (ibid., p. 210).

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<sup>10</sup> Resources = a concentration of naturally occurring material in such a form that economic extraction is currently or potentially feasible (includes reserves). Reserves = the share of total resources that can be economically extracted with technology available at the time of determination. (cf. Polinares Consortium 2012, p. 15)

In any case, South Africa dominates in both world resources and production. Figure 8 shows the distribution of the 2014 global platinum supply by country, illustrating the fact that with 3,070,000 troy ounces (approx. 95,000 kg) South Africa produced nearly 65% of global mining supply, not counting changes in producer inventory. Although this represents an impressive market share, due to issues with labour rights and energy supply blocking the South African mining industry in 2014 (cf. WPIC 2015), this figure is actually far below the market shares achieved in previous years.

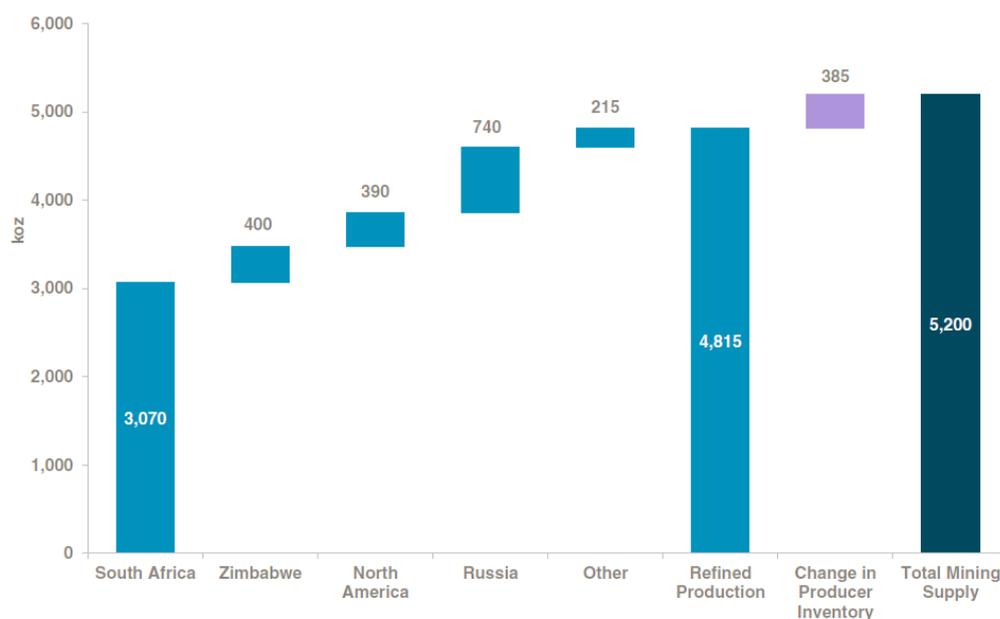


Figure 8: Global platinum mining supply 2014 (WPIC 2015, p. 3)

In 2010, for example, South African production accounted for 76% of global platinum supply (Gunn 2014, p. 302) and according to European Commission (2014a, p. 125), the country holds 95% of global PGM reserves. In addition, South Africa and to a limited extent Zimbabwe and the USA, are the only countries with PGM-dominant resources, whereas in other locations PGM are produced as by-products of, for example, nickel and copper extraction only (Gunn 2014, pp. 286-293). While other countries do play an important role in the diversification of platinum supply sources, as can be seen in Figure 8, their production capacity is insignificant compared to that of South Africa as the dominant supplier.

The production of platinum and PGM in general involves highly complex, costly and labour-, water-, and energy-intensive processing steps. Concerns regarding environmental impacts and workers' safety are an issue. While such aspects not only affect supply but also make PGM production highly controversial and should not be neglected

when assessing the sustainability of a continued reliance on platinum for ‘clean’ transportation, discussing these issues is beyond the scope of this thesis. For a further examination of environmental impacts as well as health and safety and workers’ rights issues, confer Steinweg & de Haan (2007), Saurat & Bringezu (2008), Mudd (2010), Buchert et al. (2011), Mudd (2012), Yager et al. (2013) and Gunn (2014).

Simply put, “*The mining of pgm ores is through conventional underground or open cut techniques. The next stage is grinding and gravity-based separation, followed by flotation to produce a pgm-rich concentrate. [The] concentrate is then smelted to produce a pgm-rich nickel-copper matte, with the pgms extracted and purified at a precious metals refinery*” (Mudd 2012, p. 3).

A more detailed description of the production processes is not part of this thesis, but can be reviewed in, for example, Buchert et al. (2011) and especially Gunn (2014).

### **4.3 Platinum areas of application**

Demand for PGM, especially platinum and palladium, has experienced rapid growth in the past decades. Of the 14,200 metric tons of PGM produced between 1900 and 2011, 95% were produced since 1960 (Zientek & Loferski 2014, p. 2), as demand for platinum and palladium has increased by 180% since 1980 (SATW 2010, p. 23).

#### **4.3.1 Current demand**

Today, platinum is used in a wide range of applications. In 2014, global demand amounted to 7,925,000 troy ounces (approx. 218,500 kg), according to WPIC (2015, p. 2). Since the contribution of recycling to platinum supply varies between sectors, a distinction between net and gross demand is useful, where net demand signifies gross demand minus the amount of platinum supplied by recycling (ibid.). Table 3 shows the main uses of platinum by sector in 2011.

Table 3: Main uses of platinum and net demand by sector in 2011 (Gunn 2014, p. 298)

Sector	Main Uses	Consumption (thousand troy ounces)
Autocatalysts	Catalysts for vehicle exhaust emission control	3,105
Investment	Exchange Traded Funds, ingots, bars, coins	460
Jewellery	Fabrication of platinum jewellery	2,480
Chemical	Catalysts for production of nitric acids and other bulk and speciality chemicals	470
Electrical	Hard disk coatings, thermocouples	230
Petroleum	Catalysts for petroleum refining and production of petrochemicals	210
Glass	Vessels for glass manufacture, brushings for fibre glass production	
Dental, medical and biomedical	Anti-cancer drugs, implants such as heart pace-makers	230
Other	Spark-plug tips, oxygen sensors for engine management, fuel cells.	355
Total global consumption		8,095 ( $\approx$ 251.783 t)

As evident from Table 3, the largest net use of platinum today is in autocatalysts. Demand from this sector has grown significantly since the 1970s, when emission control legislation was introduced in a number of countries, necessitating the use of catalytic converters to eliminate toxic emissions. In Europe, where half of all newly registered vehicles are diesel-powered, catalysts and particulate-filters for diesel-engines are the main source of platinum demand (Gunn 2014, p. 297).

Due to its white colour, strength and tarnish resistance, platinum is also a popular material for jewellery (Zientek & Loferski 2014, p. 1). Traditionally used in Japan, demand for platinum-based jewellery has lately also been growing in China and Europe and in 2011, this sector accounted for 31% of global platinum demand (Gunn 2014, p. 297).

While in the past, platinum coins, ingots and bars have been sold to safely store wealth, in the last few years investors have increasingly purchased Exchange Traded Funds (ETF)<sup>11</sup>. Although interest in platinum ETF was relatively low when they were first launched between 2007 and 2010, they create opportunities for speculative buying,

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<sup>11</sup> ETF = A marketable security, i.e. a type of fund which owns the underlying assets and divides ownership of these into shares. ETFs combine the liquidity of shares with the risk diversification of a fund or index (cf. Finanzen.net GmbH 2015).

which could potentially lead to artificial platinum shortages in the world market (Yang 2009, p. 1807).

Another important application of platinum is its use in industrial catalysts for the production of chemicals, pharmaceuticals and in petroleum refining. Although gross demand from these applications is significantly higher than the net demand given in Table 3, the catalysts are recycled with a high efficiency and thus additional platinum is only required to create additional catalysts or replace them at the end of their lifetime (Gunn 2014, p. 298).

Platinum is also a vital component of computer hard disk drives and multilayer ceramic capacitors (Zientek & Loferski 2014, p. 1), with demand from this sector growing rapidly in recent years. Finally, platinum is used in a number of drugs and medical equipment, including anti-cancer drugs, pacemakers and dental alloys (Gunn 2014, p. 298).

#### 4.3.2 Future demand

Although fuel cells represent only a minor use of PGM today, demand from this and other future-oriented applications<sup>12</sup> is expected to rise. While demand for palladium may be affected, for example, by the need for desalinisation plants and hydrogen storage technology, future demand for ruthenium could be driven by production of enhanced photovoltaic cells, in which ruthenium functions as the photoelectrical element. The major drivers for platinum demand, however, are expected to be increasing needs for catalysts in chemical and petrochemical industries and, particularly, a wide-spread deployment of fuel cells (Angerer et al. 2009, p. 295).

The impact of FCV diffusion on platinum demand has been examined in various projections over the past 15 years. These have led to differing conclusions, depending on the time of publication, development status of technology at the time, forecast horizons, as well as assumptions concerning the future number of registered vehicles, market share of FCV, development of platinum loadings and recycling rates. Appendix 1 gives an overview of publications assessing the impact of FCV market penetration on global platinum demand, listing select assumptions and results, respectively..

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<sup>12</sup> This term has no official definition but rather describes an assumption of technologies with the potential for positive impacts on economic, environmental and social sustainability (cf. Angerer et al. (2009); Buchert et al. (2009)).

As indicated by the wide range of platinum demand forecasts in Appendix 1, neither the mode nor the impact of FCV market penetration are straightforward, but remain subject to uncertainty and speculation. Not only do estimates concerning the platinum demand of a future FCV fleet vary significantly between sources depending on the applied assumptions, it is also highly uncertain to what degree this demand will be *additional* to the current and future demand of other industries. Total primary platinum requirements will depend not only on technology and market developments in other industries, such as chemical, petrochemical or electronic industries, and supply from secondary sources, but also on the degree to which FCV will replace ICE vehicles fitted with catalytic converters, thus ‘liberating’ the contained platinum for use in other applications. While examining such issues would be highly useful, it is beyond the scope of this thesis.

It must also be stressed that projections, by definition, only forecast *possible* developments and must not be misunderstood as precise predictions of the future. Based on a set of specific assumptions, different scenarios may provide a fan of projections that, hopefully, capture the actual development if any set of assumptions comes true. Despite these limitations, projections provide a valuable tool by indicating the drivers and influences of future developments (cf. Angerer et al. 2009, p. 7).

#### **4.4 The classification of platinum as a critical resource**

*“In most of its industrial applications, platinum is either irreplaceable or can only be substituted with significant compromises in performance”* (Yang 2009, p. 1805). This quote by Yang once again stresses the fact that platinum plays a vital role in a number of current and especially future-oriented technologies, which was already alluded to in Chapter 4.3. Due to this importance for the competitiveness of entire industries, platinum and PGM in general are deemed strategically important resources by a number of governments. Combining this strategic importance with their relative scarcity, PGM are typically considered ‘critical metals’ (cf. Buchert et al. 2009; US DOE 2011; BMBF 2012; European Commission 2014b).

Although there is no common definition for the term ‘criticality’ of metals, the concept can be understood as a kind of risk assessment based on a range of determinants including geological availability, concentration of supply, political conditions in the supplying countries, possibilities of substitution and recycling, and environmental implications of production and processing, combined with high demand (cf. Graedel et al. 2012; Albach 2014). Consequently, “*the denotation ‘critical’ could be considered a label, which is based upon various factors and circumstances that may change over time, and which may be differently weighted by actors e.g. nations or industries*” (Albach 2014, p. 35). Several methodologies for assessing the criticality of resources exist. In the following, factors resulting in the classification of platinum as a critical metal will be addressed.

As mentioned above, platinum can be considered a scarce metal and concerns regarding the availability of enough platinum for current and future demand have persisted for many years. This can not only be deduced from the number of studies analysing the impact of, for example, FCV deployment on platinum demand (cf. Appendix 1), but also from publications stressing the fact that platinum is a finite resource whose deposits will eventually be depleted if extraction rates continue to exceed geological processes of enrichment (cf. Gordon et al. 2006; Yang 2009).

Although “*unlike oil, which is irremediably consumed when used, metals have the potential for almost infinite recovery and reuse*” (Gordon et al. 2006, p. 1209), in practice the usability of metals depends on the thermodynamic form and concentrations at which they are available (Albach 2014, p. 25). As metal-containing goods are produced, metal stocks are transferred from the lithosphere to the anthroposphere<sup>13</sup>, with some amounts ‘lost’ in mining and refining wastes at concentrations too low to be recovered. Over time, the metals in use are reused and recycled and again, some amounts are irrecoverably lost for human use through corrosion and wear, ineffective recycling processes and disposal in those end-of-life products that are not recycled (Gordon et al. 2006, p. 1209). Considering platinum as a non-renewable and finite resource therefore makes sense and the irrevocable depletion of its resources may indeed occur at some point in time.

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<sup>13</sup> Anthroposphere = that part of the environment which is influenced by human activity, encompassing our technology, infrastructure, culture and activities connected to them (cf. Baccini & Brunner 2012, p. 1).

However, although some sources propose this to be a matter of only decades and even warn against an “*impending platinum crisis*” (Yang 2009, p. 1805; also cf. Gordon et al. 2006; Halada et al. 2008; Bardi & Corporali 2014), the majority assume that enough platinum is available in the Earth’s lithosphere for meeting demand for the foreseeable future (cf. Sealy 2008; SATW 2010; Zientek et al. 2010; Gunn 2014), or as Gunn (2014) states, “*The physical availability of PGM is not likely to be a constraint on economic growth in any sector in the long term. New supplies from both primary (mining) and secondary (recycling) sources are likely to be available with existing technology*” (p. 309). ‘Scarcity’ in the context of PGM supply therefore does not necessarily refer to geological scarcity, but rather to structural scarcity created by the highly concentrated supply base and unstable conditions in the supplier countries, as well as temporal scarcity caused by price volatility and inelastic supply.

#### 4.4.1 Criticality assessment of Germany and the European Union

As mentioned in Chapter 4.2, South Africa holds 95% of platinum resources and has produced up to 76% of annual supply in recent years, with the remainder supplied by only four other countries (Zimbabwe, USA, Canada and Russia)<sup>14</sup>. This supply concentration is represented by the Herfindahl-Hirschman-Index (HHI), a measure used in the criticality assessments of both the European Commission and the Deutsche Rohstoffagentur (DERA), as well as in several research publications (cf. Angerer et al. 2009; DERA 2012; Polinares Consortium 2012; Silberglitt et al. 2013; European Commission 2014a; European Commission 2014b).

The HHI is the sum of the squared market shares of all market participants, with the value range limited as follows:  $1/[\text{number of market participants}] < \text{HHI} < 1$ . The HHI reaches its lowest value if all market participants have equal shares, and its highest value (i.e. 1) if there is only one market participant claiming a monopoly. Since the HHI reaches very low numbers for markets with many participants, it is generally multiplied by 10,000 for ease of accounting. A value above 2,500 denotes a highly concentrated market (DERA 2012, p. 9).

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<sup>14</sup> Note that the column ‘others’ in Figure 8 has been excluded from this recital.

Since countries exhibiting poor governance pose further supply risks, because political instability and unrest, but also infrastructure concerns may lead to interruptions in supply, the World Governance Indicators (WGI) published annually by the World Bank are a common measure of political stability used by both DERA (2012) and European Commission (2014a; 2014b). The WGI is a weighted aggregate between +2.5 and -2.5, composed of the following indicators (cf. DERA 2012, p. 10; World Bank 2014):

- Voice and accountability,
- Political stability and absence of violence,
- Government effectiveness,
- Regulatory quality,
- Rule of law,
- Control of corruption.

The derived country risk is weighted with mining and refining shares of the respective country by DERA (2012) to arrive at an overall country risk between +1.5 and -1.5, with values below +0.5 considered a moderate risk and values below -0.5 considered critical.

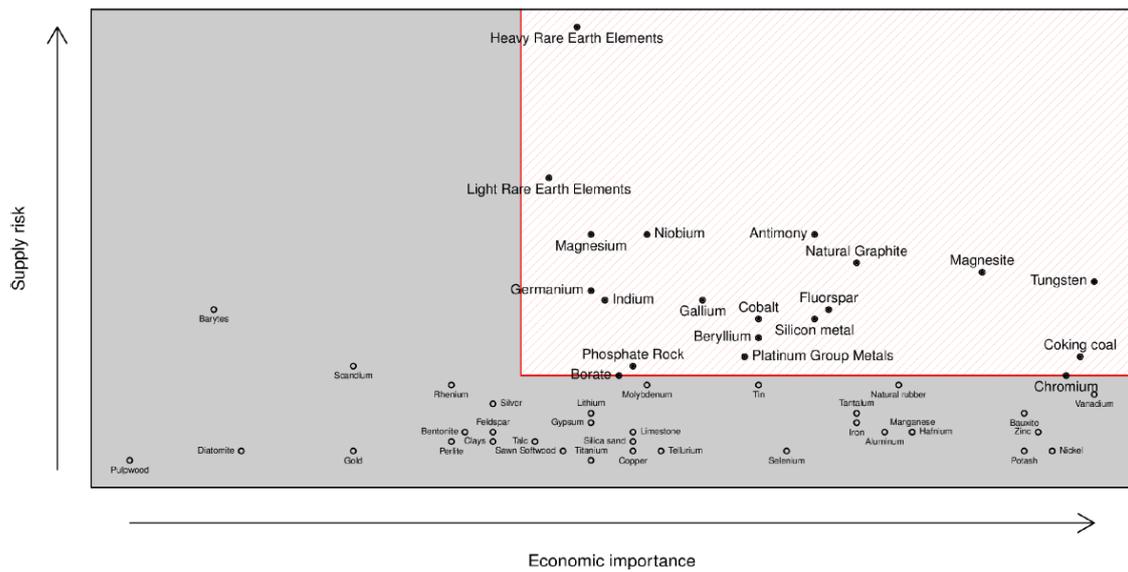


Figure 9: Criticality assessment of select elements for the EU in 2013 (European Commission 2014b, p. 24)

Figure 9 shows the result of the European Commission’s criticality assessment, plotting the identified supply risk (measured by HHI and WGI) against economic importance. All elements within the lightly-shaded box are considered critical for the EU at the time of publication. Details for HHI and WGI as calculated by DERA (2012) are given in Table 4.

Table 4: Herfindahl-Hirschman Index and World Governance Indicators as calculated by DERA (2012, p. 14)

	HHI	WGI
Platinum	6,226	0.09

Colouration denotes criticality assessment as follows: red = high risk, yellow = moderate risk

#### 4.4.2 Platinum price developments

As can be derived from the above criticality assessments, the accessibility and vulnerability of platinum supply raises concerns at national and international levels, a fact which is also reflected in the volatility of platinum price developments. Metal prices in general are driven by a number of factors, including structural costs (e.g. energy), declining ore grades, long lead times between exploration and exploitation of deposits, rising labour costs due to labour shortages and currency-related cost issues. In addition, the market for platinum in recent years was governed by both price-inelastic (automobile catalysts) and price-elastic demand (jewellery) as well as an increased presence of financial players, coupled with restrained responses to the increasing platinum price due to disruptions and expenditure cuts on the supply side (Perrot-Humphrey 2006, p. 40).

Hence, market prices of platinum have oscillated considerably over the past decades and demonstrated a worrying inelasticity of supply, with supply driven by world events rather than platinum price (cf. Yang 2009, p. 1806; Zientek et al. 2010, p.80). Figure 10 shows the platinum price developments in US\$ per troy ounce from 1968 to 2011, illustrating the impact of world events.

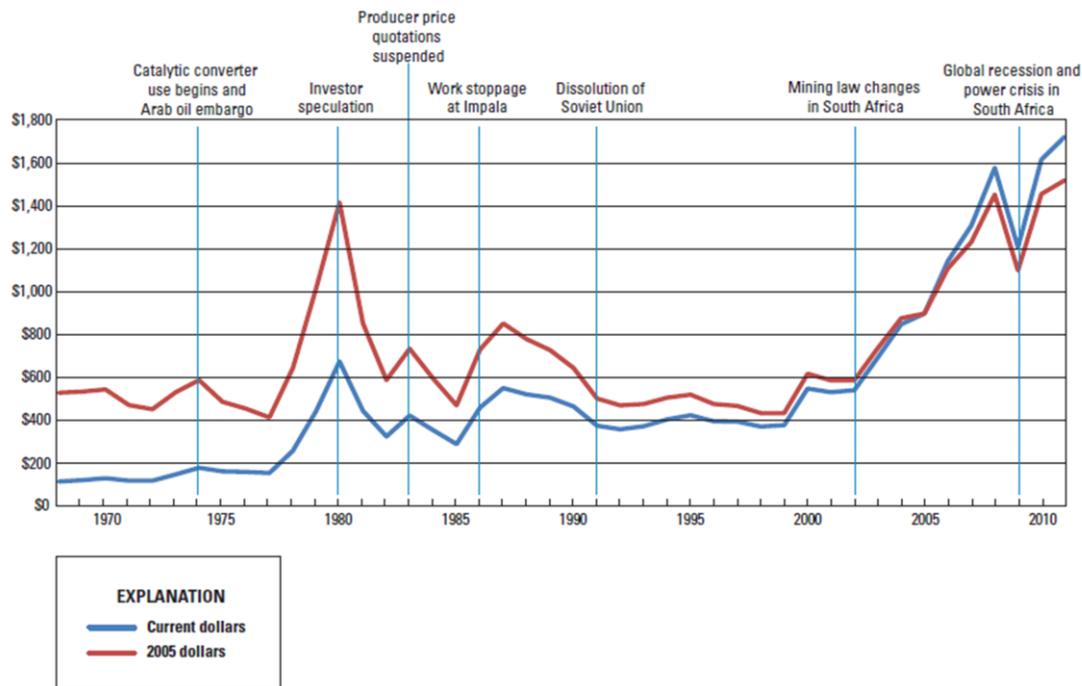


Figure 10: Real and nominal (2005) platinum price in US\$ per troy ounce from 1968-2011 and major events affecting prices (Zientek et al. 2010, p. 80)

As outlined in Figure 10, the price of platinum has experienced times of strong volatility. Prohibitively high material costs can present a market barrier for new technologies, such as fuel cell vehicles (cf. Chapter 3.3), but may also have serious consequences for the profitability of established industries. Analysing the impact of PGM price fluctuations on the automobile industry, Hagelüken (2005, p. 126), for example, concludes that PGM-related purchasing costs alone increased by US\$ 2.4 billion for the whole industry in just one year (1999 to 2000). While this price pressure eased considerably in the following years, such time scales are too small for businesses to react and develop technologies with lower precious metal contents (cf. Alonso et al. 2007).

Not only may unexpected price surges and a lack of raw materials seriously harm businesses, price volatility can also encourage inventory build-up, as companies or national governments form buffer stocks in order to prepare for times of high prices or low supply (Pindyck 2004, p. 1030). For example, national stockpiles of platinum are held by a number of governments, including the US Strategic Materials National Defense Stockpile (USGS 2015, p. 120). Inventory building, however, can lead to unexpected increases in demand and raise prices in the short term, thus adding to price volatility (Pindyck 2004, p. 1030). Since a reliable availability of resources and planning security

are vital prerequisites for production and investment, price volatility can directly affect the competitiveness of both industries and entire economies, making the minimisation of supply risks a priority for national governments and international organisations (cf. Buchert et al. 2009; US DOE 2011; BMBF 2012; DERA 2012; European Commission 2014a; European Commission 2014b).

#### **4.5 Secondary supply from recycling**

In its more comprehensive meaning<sup>15</sup>, recycling is defined as the recovery of raw materials from waste, their return to the economic cycle and their processing into new products (Umweltdatenbank 2015), and represents the most desirable waste management option after reduction and reuse (cf. Gertsakis & Lewis 2003).

The benefits of recovering platinum through recycling compared to its primary production have been commented on in a number of studies. Due to the low ore grades and complex mining processes, primary production of PGM involves high environmental impacts as a matter of principle. These include emissions of sulphur dioxide, of CO<sub>2</sub> equivalents in the range of 13,000 tons per ton PGM (Saurat & Bringezu 2008, pp. 760-761), excessive water and energy consumption (Mudd 2010, pp. 108-110) as well as habitat destruction, air and water pollution and generation of dust, particulate matter and solid waste (Cairncross 2014). Altogether, each ton of platinum from primary production comes with an ecological rucksack of 680,000 tons (Lucas et al. 2010, p. 22).

The environmental impacts resulting from state-of-the-art secondary production using specialised equipment and processes are significantly lower, especially with regards to sulphur dioxide emissions and material requirements (Saurat & Bringezu 2008, p. 765). The ecological rucksack of platinum from secondary production is thus reduced by a factor of 78 (Lucas et al. 2010, p. 22), sulphur dioxide emissions are reduced by a factor of 100 (Handley et al. 2002, p. 346) and CO<sub>2</sub> emissions lowered by a factor of 5, respectively (Hagelüken 2009, p. 16). In addition, secondary production leads to slowing resource depletion, consumes up to 90% less energy, avoids solid waste from mining and reduces space requirements for landfills (Crundwell et al. 2011, p. 537).

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<sup>15</sup> compared with the more restrictive definition of e.g. the Directive 2000/53/EC (cf. the following footnote)

In combination with the high supply risk examined in Chapter 4.4, it can thus be concluded that “*recycling the platinum in a fuel cell is an enabling technology for the fuel cell industry as it stabilises the supply of this metal*” (Shore 2009, p. 4).

Recovery of platinum from secondary sources not only bears environmental benefits, but most importantly, it contributes to an improved security of supply. Several studies emphasize the role of recycling in meeting future platinum requirements. Buchert et al. (2009), for example, state “*A successful recycling of [...] critical metals is very important regarding increase of resource efficiency, avoidance of possible scarcities, and reduction of the overall environmental impacts*” (p. V). Tiax LLC (2003), assessing platinum demand from FCV production, stress that “*Recycled platinum from the transportation sector will be an increasingly critical source of supply*” (p. 7). Boudreau et al. (2008, p. 41) conclude that assuming a longer FCV lifetime in the economic model applied for their study results in a higher platinum price, as less platinum is available through recycling. Also, as more and more FCV enter the end-of-life phase, the modelled platinum price becomes increasingly sensitive to assumed recycling efficiencies (ibid.). Similarly, Saurat and Bringezu (2009) state “*the requirement for primary PGM will remain constantly high [...] as long as recycling rates are not further increased. [...] This pinpoints again the need to arrange for improved international recycling.*” (p. 419).

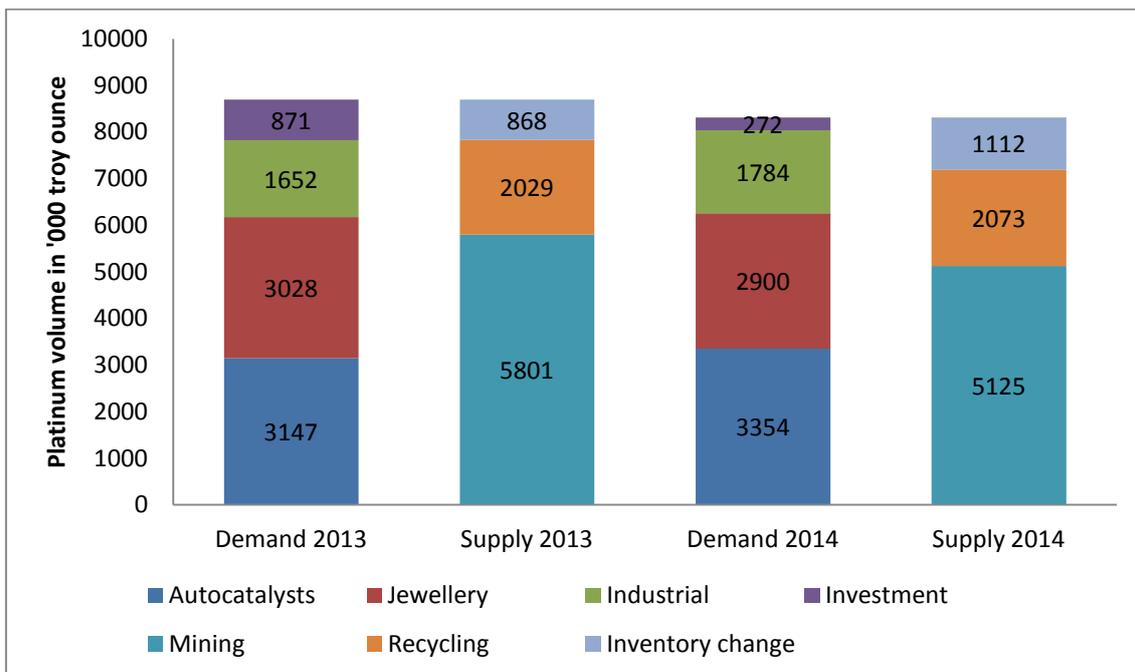


Figure 11: Platinum supply and demand in 2013/14 (adapted from Johnson Matthey 2015b)

Secondary production already contributes significantly to meeting gross demand for platinum today. Figure 11 compares global platinum demand by sector with the global supply from mining and recycling for the years 2013 and 2014. In 2014, for example, 2,073,000 troy ounces (approx. 64.477 t) of platinum were recovered through recycling, which equates to around 25% of gross demand for that year. However, despite this considerable contribution from secondary sources, in 2014 the platinum market ended in deficit, with global supplies falling short of demand by around 700 troy ounces (approx. 0.022 t). For the second consecutive year, this forced companies and national governments to release part of their buffer stocks (WPIC 2015, p. 2). Again, this points to the importance of increasing recycling rates.

## 5 The recycling chain of fuel cell vehicles

While it is technologically possible to recover virtually all of the platinum contained in end-of-life scrap without any quality losses (Lucas et al. 2011, p. 23), realised recycling rates differ highly between sectors, with industrial applications achieving recovery rates of 80 to 90% over the whole life cycle compared to only 5 to 60% for consumer goods (Hagelüken 2012, p. 33). This peculiarity can be explained by the fact that the recycling chain for any product consists of several consecutive steps, which are pictured in Figure 12.

According to Schluep et al. (2009), “*the efficiency of the entire recycling chain depends on the efficiency of each step and on how well the interfaces between these interdependent steps are managed*” (p. 12). If, for example, the collection rate for any product (or a specific material) is 50%, the efficiency of the dismantling and pre-processing procedures 70% and the efficiency of material recovery 95%, this results in an efficiency of the overall recycling chain of only 23%. It follows thus that “*all [...] steps should operate and interact in a holistic manner in order to achieve the overall recycling objectives*” (ibid, p. VI).

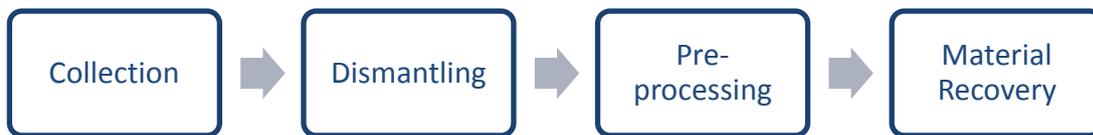


Figure 12: Recycling chain (adapted from Umicore 2015)

Substantial deficits in some of the steps illustrated in Figure 12 cause certain PGM applications to exhibit much lower recycling rates than others. As can be derived from Figure 13, which shows the dynamic recycling rate<sup>16</sup> for a range of PGM-dependant applications, this is especially true for consumer goods, such as automotive catalysts or electronic equipment.

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<sup>16</sup> The dynamic recycling rate states what proportion of the PGM amount that was used in a certain application at the time  $x$  is recovered at the end of its life cycle ( $x+n$ ). It is therefore more meaningful in assessing the recycling efficiency than the static recycling rate, which states the ratio of secondary material supply to gross demand at a given point in time, neglecting the product life time and any market shifts (cf. Hagelüken et al. 2005a, pp. 228 + 232)

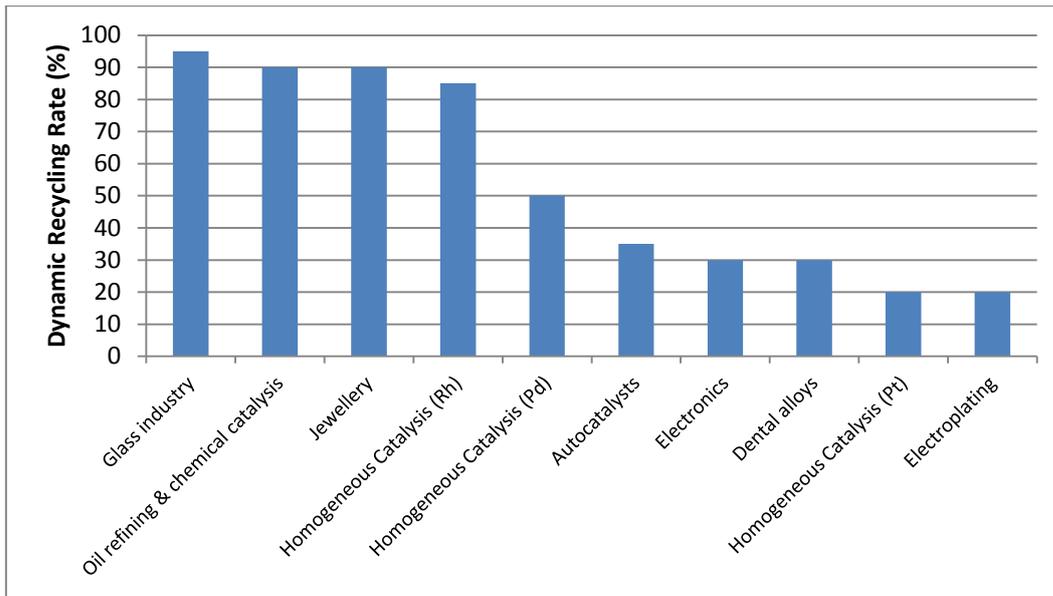


Figure 13: Dynamic recycling rate of PGM from selected applications (adapted from Hagelüken et al. 2005a, p. 169)

These significant deviations may be explained by the fact that consumer products pass through fundamentally different life cycles involving a number of different actors. Analysing the PGM material flows of Europe, Hagelüken et al. (2005a, pp. 169-173) conclude that unlike industrial goods, the life cycles of consumer applications often feature so-called ‘open loops’, which are exemplified in Figure 14 and characterised by the following (ibid.):

- Lack of direct business relations between industrial actors throughout the life cycle,
- Multiple change of ownership,
- Lack of transparency of life cycle stages and actors involved,
- Lack of actors’ integrity and reliability,
- Difficulty of identifying and quantifying PGM losses over the life cycle.

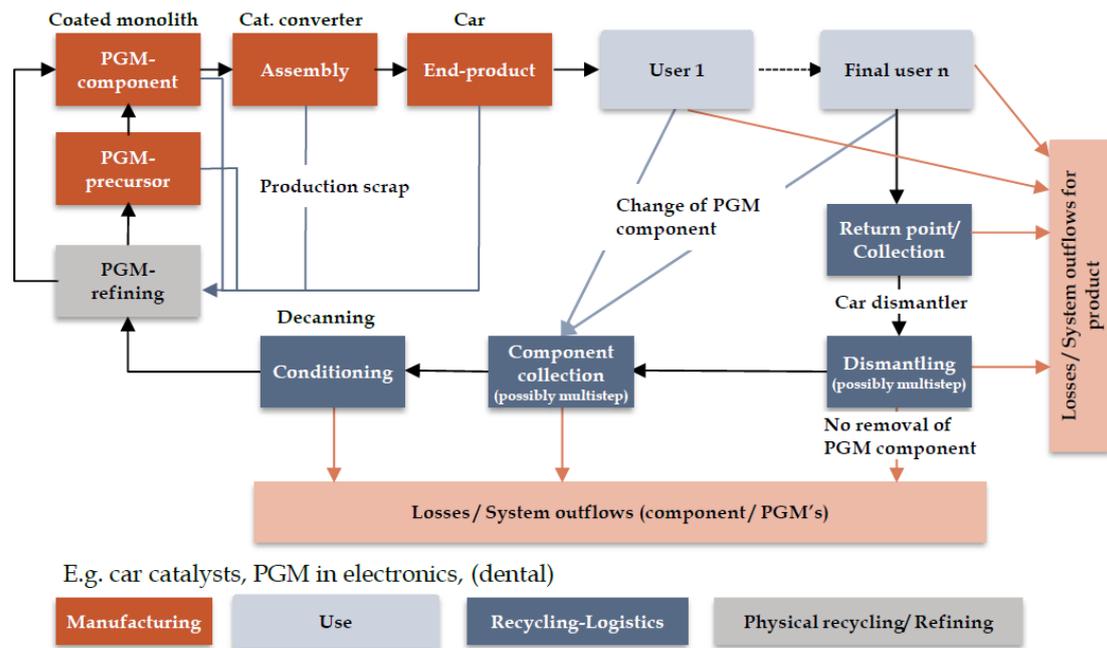


Figure 14: Open loop life cycle of PGM applications (Hagelüken et al. 2005a in Molotova et al. 2013, p. 60)

Although high recovery rates similar to those of industrial applications should be achievable in principle, these attributes lead to deficits in collection, dismantling, pre-processing and material recovery, causing considerable PGM losses at each step of the recycling chain and thus resulting in suboptimal recycling efficiencies (Hagelüken et al. 2005a, pp. 169-173). In order to assess in how far the preceding considerations may prove problematic for the future recovery of platinum from FCV, this chapter addresses each of the steps in the recycling chain of automotive fuel cells, examining how a future recycling system may be designed and highlighting potential ‘hot spots’ for PGM losses.

## 5.1 Collection and Dismantling

### 5.1.1 End-of-life vehicle treatment in the European Union

In the European Union, the end-of-life treatment of vehicles is governed by the Directive 2000/53/EC on end-of-life vehicles, although other directives, such as Directive 2012/19/EU on waste electrical and electronic equipment, Directive 2011/65/EU on the restriction of the use of certain hazardous substances in electrical and electronic equipment and the REACH (registration, evaluation, authorisation and restriction of chemicals) regulation No. 1907/2006, play an important role for the manufacturing and disposal of certain vehicle components, too.

The end-of-life vehicle (ELV) Directive prescribes that 85% of the weight of vehicles per year is to be reused or recycled and 95% reused or recovered<sup>17</sup> by January 1, 2015, respectively. It consists of 13 Articles which determine the Directive's scope, set requirements for waste prevention, collection, treatment standards and reporting procedures, establish reuse and recovery targets and list exemptions from the restrictions on hazardous substances. Automobile producers are liable for take-back, treatment and recycling of ELV and incur the costs involved with these requirements; the end-of-life treatment is thus free of charge to consumers (Heiskanen et al. 2013, p. 2). While the precise organisation of collection and dismantling systems differs between the EU's member states and typically involves a number of different business entities, the disposal of ELV follows the general steps outlined in Figure 15.

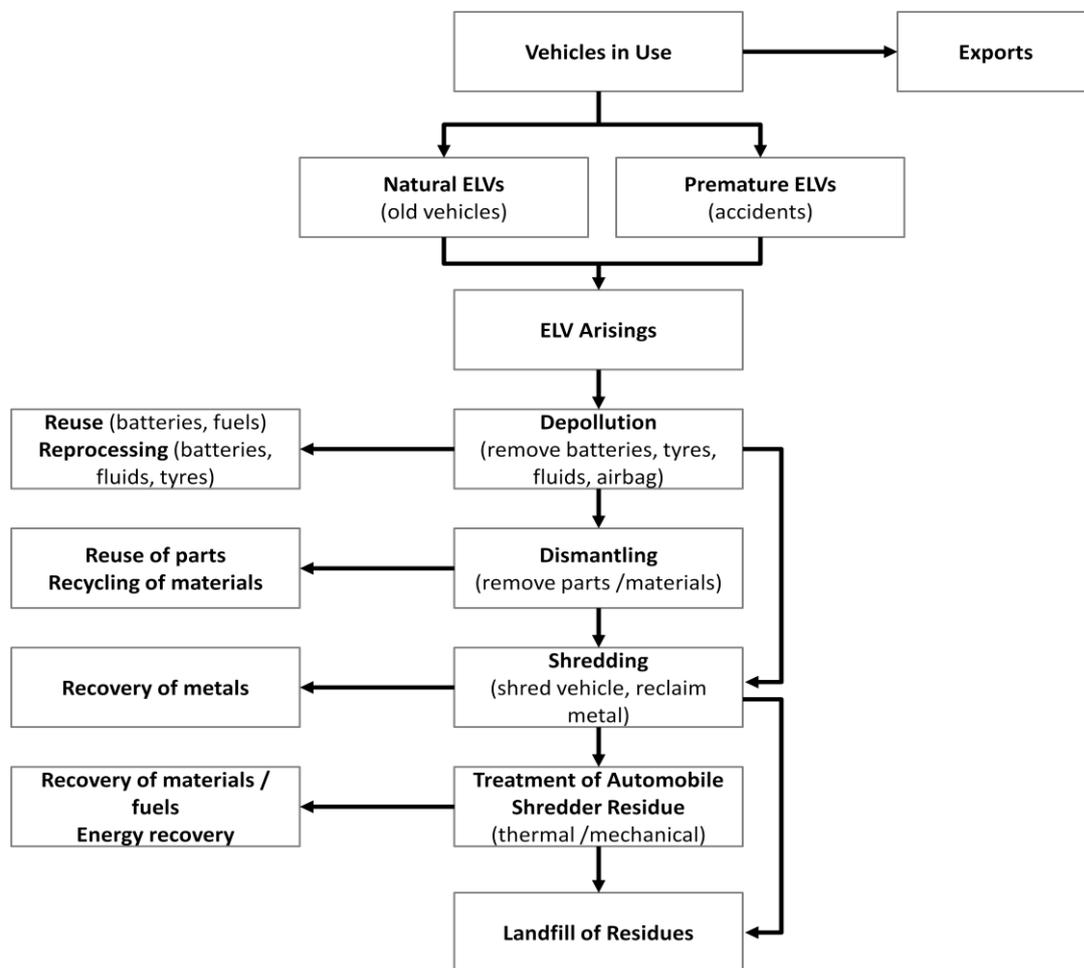


Figure 15: Overview of ELV treatment process (GHK 2006, p. 2)

<sup>17</sup> Reuse = reuse of components for their original purpose. Recycling = reprocessing of waste material for original or other purpose. Recovery = any operation provided by Annex IIB to Directive 75/442/EEC, which includes recovery of metals and other organic and inorganic materials, as well as use as fuel for energy recovery (cf. Directive 2000/53/EC).

The composition of the vehicle fleet and hence also the composition of ELV entering the recycling chain differs very much between member states, as automobile ownership in the original member states (EU-15) is high and the vehicles in use are relatively new (typically less than 10 years old), while ownership in the newer member states tends to be much lower and the fleet significantly older (Fergusson 2007, p. 57; Konz 2009, p. 435). The scrapping age, however, determines the dismantling and material recovery processes applied to ELV, as the material composition and thus value of recyclable components of cars is constantly evolving, which has a strong impact on the profitability of recycling businesses (Heiskanen et al. 2013, p. 5).

As Fergusson (2007) notes, “*the quality of data on car scrapping is extremely variable across Europe*” (p. 57); however, it is estimated that around 12.6 million vehicles are deregistered in the EU-25 each year, 75% of which arise in just five member states (i.e. Germany, United Kingdom, France, Spain, Italy). Approximately 3 million of these deregistered vehicles are exported per year, with the majority (2 million) going to other, mainly Eastern European countries. Considering the rising weight of new vehicles as well as increased vehicle usage, it is estimated that by 2030, the total mass of ELV generated per year in the EU-25 will reach 14 to 17 million tons (Heiskanen et al. 2013, p. 2).

### 5.1.2 End-of-life treatment of electric vehicles

In force since 2000, the ELV Directive, however, does not make any separate provisions for the treatment of EoL *electric* vehicles, which contain a number of components that differ very much from those installed in ICE vehicles. Depending on the type of powertrain technology, these may include, for example, power electronics, power control systems, traction batteries and fuel cell systems. Although significant volumes of EoL electric vehicles will not arise at least for several years to come, they will undoubtedly provide challenges to the current ELV recycling and disposal framework eventually, as questions concerning the waste classification of components, adaption needs of collection and dismantling systems, safety issues and recycling technologies remain unsolved (cf. NAFTC 2013; UBA 2014; European Commission 2015).

According to Simons and Bauer (2015, p. 6), the balance of plant of a current fuel cell system accounts for 54% of its overall weight and consists of components that are relatively similar to those used in ICE vehicles. Due to ongoing efforts aimed at producing

lighter stacks, this weight proportion is likely to increase further in future years. Given the weight-based recycling quota of the ELV Directive it is therefore assumed that FCV, once established, will follow the same EoL treatment processes as conventional ICE vehicles, a view which is shared by Saurat and Bringezu (2009, p. 409). Schiemann et al. (2007, p. 73), on the other hand, assume that EoL fuel cells, irrespective of their application, will be treated as waste electric and electronic equipment according to Directive 2012/19/EU.

Since BEV represent a more mature technology than FCV and their disposal may hence be considered a more immediate ‘problem’, the above-mentioned issues concerning this powertrain technology are currently the subject of ongoing research efforts (cf. TU Braunschweig 2009; Kwade & Bärwaldt 2012; UBA 2014; TU Braunschweig 2015). Kwade and Bärwaldt (2012, p. 35) assume that BEV-specific components will follow the EoL processes laid out by the ELV Directive up until the dismantling step, in which the battery is removed. The authors further suggest that as long as only limited numbers of EoL BEV arise, only few, branded workshops will have the specialist knowledge, personnel and equipment required for removal of the batteries. This is likely to change with increasing volumes of EoL BEV, which will force also independent workshops and ELV dismantlers to handle the contained batteries. Following the removal, the battery is expected to enter a dedicated recycling process consisting of the steps outlined in Figure 16.



Figure 16: End-of-life battery processing (own preparation based on Kwade & Bärwaldt 2012, pp. 35-38)

Drawing on structural similarities between the batteries of BEV and fuel cells of FCV (i.e. a powertrain-specific component installed in common vehicle architecture), the author of this thesis assumes that future EoL treatment of automotive fuel cells may be organised in a similar way, meaning the FCV will be treated as ELV according to Figure 15 and upon removal in the dismantling step, the fuel cell will be directed to a separate waste stream and enter a dedicated recycling process. In this regard, fuel cells hence also resemble PGM-loaded exhaust gas catalytic converters, whose removal prior to shredding is compulsory under the ELV Directive.

### 5.1.3 Potential deficits in collection and dismantling

#### 5.1.3.1 Collection

According to Hagelüken et al. (2005a, p. 78) collection deficits represent the single largest cause for PGM losses in the recycling chain of automotive catalysts. Although, as mentioned above, the organisation of ELV collection and treatment frameworks differs between member states in detail, all states certify ‘Authorised Treatment Facilities’ that issue a ‘Certificate of Destruction’ to consumers when their deregistered vehicle enters the ELV process (cf. Fergusson 2007; Heiskanen et al. 2013). In many member states, however, the number of ELV registered this way represents less than 50% of the deregistered vehicles, making it clear that by no means all ELV in Europe are disposed of as prescribed by the ELV Directive (Heiskanen et al. 2013, p. 2).

Figure 17 shows the destinations of the 3 million vehicles that were deregistered<sup>18</sup> in Germany in 2008, demonstrating that less than 15% of these vehicles entered the ELV treatment process in Germany. This phenomenon can also be observed in other EU countries (cf. Heiskanen et al. 2013), although to a lesser extent. In 2000, for example, only 6.9 million vehicles were scrapped EU-wide while 10.9 million were deregistered (Hagelüken et al. 2005a, p. 68).

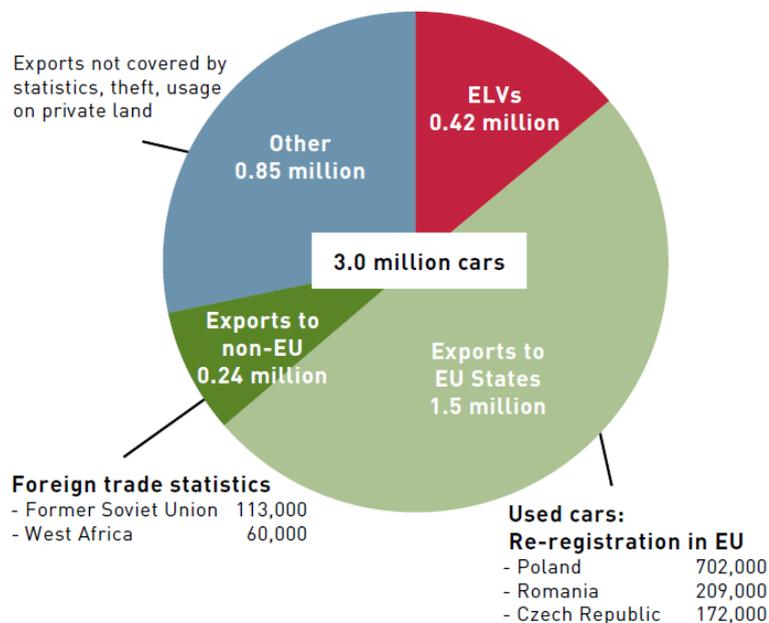


Figure 17: Destinations of vehicles deregistered in Germany in 2008 (Wilts & Bleischwitz 2012, p. 3)

<sup>18</sup> i.e. finally deregistered; vehicles re-registered (as possible in e.g. Germany) have been deducted

While the majority of deregistered vehicles are exported to other (mainly Eastern European) EU countries and could therefore be available for recycling in the receiving countries after a second use phase, in practice a major share of these vehicles is resold to countries even further East or to developing countries, where a suitable recycling infrastructure is lacking (Wilts & Bleischwitz 2012, p. 4). Only a small number of vehicles are exported directly from Germany to non-EU countries, the majority of these vehicles, however, end up in countries lacking the infrastructure required for environmentally sound recycling. In addition, the used vehicle market is characterised by numerous small traders, however, the intra-EU trade statistics cover only merchants whose transaction values exceed € 300,000 per year (Lucas et al. 2010, p. 39), thus explaining at least part of the 0.85 million vehicles with unknown whereabouts in Figure 17. Issues such as illegal exports of ELV falsely classified as used vehicles, lack of legal rigorosity, unlicensed scrap operators, abandoned and garaged ELV represent further significant barriers to maximising collection rates (Fergusson 2007, pp. 57-58).

While the issues concerning ELV collection rates described above persist despite ongoing research on possible causes and counter-measures, it is disputable in how far such issues will also prove problematic to a future FCV fleet. Saurat and Bringezu (2009, p. 409) argue that not only is the efficiency of ELV collection schemes likely to improve in future years as a result of corrective actions taken today, but more importantly, the use of pure hydrogen as a fuel will provide a significant barrier for exports to non-European countries. Requiring a suitable hydrogen supply infrastructure, FCV will be unattractive in countries where such an infrastructure does not exist, which at the time of significant market diffusion in Europe is likely to still be the case in the poorer non-EU countries that provide the typical final destination of European ELV today. Exports of deregistered FCV to countries without a suitable recycling infrastructure should therefore only be a minor issue in the coming decades, meaning a large proportion of the platinum in use by this application will remain in Europe.

In contrast, Cerri et al. (2012) proclaim that “*recycling of PGM from Fuel Cells is not a technical issue but may be a challenge with regard to re-collection of stacks from used cars*” (p. 19) and stipulate the development of highly efficient logistic chains to assure that EoL fuel cells actually reach specialised recycling facilities where recovery of the contained PGM is possible.

Similarly, Schittl (2012, p. 56) assumes that once FCV have achieved a relevant level of market penetration, deficits in the collection of EoL FCV could indeed lead to significant losses of the contained precious metals, although detailed reasons for this assessment are not given. The author highlights the need for economic incentives to increase collection rates and proposes the development of producer-led schemes, such as a reclaimable deposit on fuel cells or a lease programme, in order to close the life cycle loop.

Similarly, Kromer et al. (2009) discuss the option of a platinum lease program for FCV, in which ownership of the contained platinum remains with the (possibly governmental) lessor throughout the life cycle. Although the study's main objective is not to analyse opportunities for increasing recycling rates but rather to assess such a programme's impact on FCV costs, the authors emphasize that "*by concentrating ownership of the metal, a leasing program could greatly enhance the efficacy of vehicle or stack recycling programs*" (Kromer et al. 2009, p. 8287). Furthermore, it is assumed that the high residual value of platinum in FCV could encourage vertical integration of collection, dismantling and recovery businesses, which would lead to an increase in recycling efficiency by reducing the number of actors involved (ibid., p. 8284).

Having already acknowledged the need for closing the loop in automobile recycling (Morton 2014), the UK-based firm Axion Recycling is currently investigating the viability of a take-back system for EoL fuel cells from a number of applications, including fuel cell vehicles. This forms part of a more comprehensive research project that involves also UK-based Johnson Matthey Fuel Cells Limited and Total Fibre Products, and focuses on material recovery. However, more detailed information on the possible design of such a take-back scheme is lacking at the time of writing (Axion Recycling 2015).

The actual platinum losses that could result from ineffective collection of EoL FCV at a future point in time remain subject to speculation. Nonetheless, both public and private stakeholders involved with fuel cell or fuel cell vehicle production, as well as automobile recycling or platinum refining would do well to devise strategies aimed at minimising losses at the collection stage prior to a widespread diffusion of FCV.

### 5.1.3.2 *Dismantling*

In the context of this paragraph, dismantling refers to the removal of the fuel cell system from the FCV and its subsequent partitioning into fuel cell stack, balance of plant and additional supporting components (cf. Chapter 2). The further disassembly of the fuel cell stack into its respective constituents will be covered in the subsequent Chapter 5.2.

As mentioned previously, it is assumed that EoL FCV will be treated as conventional ELV, with the provision that the fuel cell system must be removed prior to shredding. However, since the ELV Directive prescribes that as of January 2015, 95% of an ELV's weight must be reused or recovered and automobile manufacturers incur the costs involved, dismantlers are under increased pressure to streamline and automate the dismantling process. Dismantling plants must hence be set up to efficiently handle a highly heterogeneous vehicle fleet while meeting the weight-based targets and minimising costs. For manual separation of certain parts to be economically feasible, a significant value must be retained in the respective component. In addition, the exact location of such components as well as safe removal techniques must be known in advance or be immediately detectable (Great Lakes Institute for Recycling Markets 1998, pp. 67/68).

Due to the higher number of 'alternative fuel vehicles' in use, safety instructions for the salvaging and dismantling of FCV have been devised in the US, which not only emphasize the safety hazards involved with inadvertently damaging high-voltage lines, fuel cell stacks, batteries and hydrogen tanks but also provide advice on how to distinguish FCV from conventional vehicles and on where to locate such high-risk components (NAFTC 2013, p. 55). Figure 18 shows the typical locations of those components (shaded in red and orange) that must be disconnected and/or removed with special care in order to avoid serious safety hazards. These components generally necessitate manual removal, discharge or other treatment before the vehicle can be further dismantled, which is likely to add significant costs. While the instructions mentioned above apply to a diverse range of FCV models and additional information may be given in the 'Emergency Responder Information' of vehicle manufacturers (cf. Toyota Motor Sales Inc. 2015), such guidelines are not available for Europe, specifically.

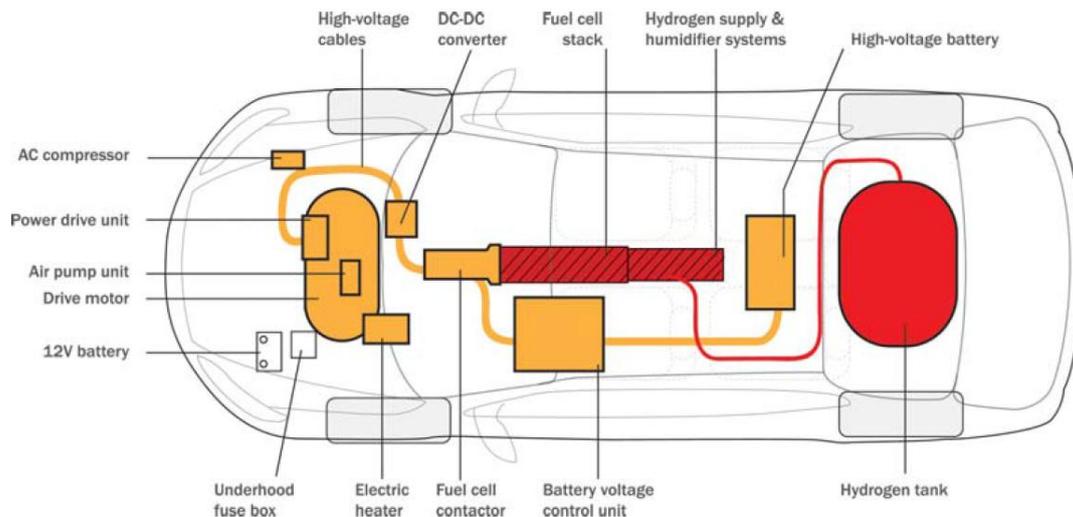


Figure 18: Cutaway view of FCV showing locations of high-risk components (NAFTC 2013, p. 55)

While removal of exhaust gas catalytic converters is mandatory under Annex I of the ELV Directive, according to Hagelüken et al. (2005a, p. 69), around 4% of those catalyst converters that reach an automobile recycling or dismantling facility inadvertently fail to be removed, meaning these precious metal-loaded components enter the shredding process in which they are irrecoverably lost. In addition, catalysts may be stolen from vehicles prior to removal, as well as from recycling facilities (Westfälischer Anzeiger 2012, Europol 2014, ka-news GmbH 2014).. Another issue is the possible damage upon removal, which leads to the loss of monolith dusts (Hagelüken et al. 2005a, p. 83).

It is difficult to assess to what extent incorrect or incomplete removal of fuel cell stacks may lead to PGM losses in future recycling activities, as this will depend on a number of factors, including the degree of automation in dismantling businesses, the variety of FCV brands and models handled by a dismantler and the design and location of the fuel cell stack in different FCV models. However, based on the following deliberations one may assume that platinum losses in the FCV dismantling stage will be considerably lower than in dismantling of ICE vehicles.

Due to the need for series connection, fuel cells are generally concentrated in a relatively compact stack (cf. Figure 18). Therefore, only one component needs to be located and removed, rather than a number of small units dispersed throughout the entire vehicle, as is the case with, for example, electrical equipment or certain types of magnets. Provided the vehicle is recognised as a FCV, incomplete removal of the fuel cell stack is therefore unlikely, even though stack design and shape may differ between brands.

As mentioned above, failure to remove the fuel cell stack and other components manually prior to further dismantling involves serious safety hazards. In addition, due to the stack's high platinum concentration and the associated monetary value, manual removal is not only economically feasible but also less labour-intensive than the decanning of catalytic converters. Since the stack can be removed and transported intact, dust losses as described for catalytic converters are also considered insignificant (Kromer et al. 2009, p. 8284). Whether this fact may pave the way for theft of stored fuel cell stacks from recycling facilities is, at this point in time, idle speculation and will hence not be considered further.

It is, however, uncertain in how far fuel cell stacks of future FCV will be accessible and removable by independent workshops and ELV dismantlers. As mentioned in Chapter 5.1, it is assumed that for some time following the wider market penetration of BEV, only few, branded workshops will have the specialist knowledge, personnel and equipment required for removal of the batteries, which could well be the case for FCV, as well. As Papasavva et al. (2003) note, "*all components of the PEM FC stack could be recycled in principle*" (p.2), but "*recyclability should be considered early in the product engineering design/development process in order to enhance its potential for reuse or recycling at the end of the vehicle's useful life*" (ibid., p. 1). Based on this conviction, a 'Recommended Practice to Design for Recycling of PEM Fuel Cell Systems' was devised in 2011, which can be obtained from the Society of Automotive Engineers (cf. SAE 2011).

## **5.2 Disassembly and pre-processing**

While according to the ELV Directive, the reuse of components is to be favoured over recycling and material recovery, simply reusing a fuel cell vehicle's MEAs at the end of its lifetime is not feasible, as fuel cell failure is often caused by degradation of the MEA and attempting to replace or repair any of its components would risk irrevocably damaging the others (Simons & Bauer 2015, p. 6). Recycling of automotive fuel cells is hence aimed at recovering the valuable raw materials. The required disassembly and pre-processing steps, as well as the fractions produced in this stage, hence depend on the objectives for material recovery and the applied recovery processes.

Similar to the value-oriented disassembly of waste electric and electronic goods, the recycling of fuel cells requires a manual disassembly procedure. Although automated

processing of small fuel cells with a low power range, such as those used as battery replacement, may be conceivable in the future, such processes require high volumes of EoL fuel cells and have hence not been developed to date (Schiemann et al. 2007, p. 77). It must be emphasized that recycling of fuel cells does not take place on a larger scale yet and the disassembly, pre-processing and recovery processes described in the following refer to procedures trialled in laboratory conditions or at very low input volumes.

Schiemann et al. (2007, pp. 28-31) provide exemplary information on the disassembly procedure of a PEM fuel cell stack into its respective fractions, an overview of which is given in Table 5. Note that the exact composition of the fractions produced will depend on the respective PEM technology and may differ between brand and age of the stack (cf. Chapter 2.2).

Table 5: Disassembly process for PEM fuel cells and fractions produced (adapted from Schiemann et al. 2007, pp. 28-31)

Disassembly step	Action taken	Fractions produced
Step 1	Removal of tie rods and casing	<ul style="list-style-type: none"> <li>• Connecting elements: high-grade steel</li> </ul>
Step 2	Removal of end plates	<ul style="list-style-type: none"> <li>• Connecting elements: high-grade steel</li> <li>• Insulation: plastics</li> <li>• End plates: high-grade steel or aluminium alloy</li> </ul>
Step 3	Withdrawal of the stack's individual layers, each consisting of bipolar plates, seal, gas diffusion layer and membrane electrode assembly.	<ul style="list-style-type: none"> <li>• Seal: plastics</li> <li>• Bipolar plates: graphite, polymeric binders, additives, separating agents</li> <li>• GDL: non-woven fabric, carbon paper</li> <li>• MEA: polymeric membrane coated with graphite and platinum</li> </ul>

Nonetheless, the procedure described above produces fractions that are well-known from the disassembly of electric and electronic equipment and thus facilitates the further processing and valuing in the secondary materials market; however, economic feasibility was not considered in this approach (Schiemann et al. 2007, p. 23). It must be noted that this procedure assumes that the GDL is entirely separate from the MEA or can at least be separated with minor effort.

While this may be true for certain brands, age of technology or application of the fuel cell, in practice, the GDL is usually hot-pressed and laminated to the catalyst coated membrane in order to form the MEA (which is then labelled a 5-layer rather than a 3-layer MEA), making separation of GDL and catalyst coated membrane difficult (cf. Chapter 2.2). Attempting to strip the GDL off a 5-layer MEA in order to expose the catalyst-coated membrane is not only labour-intensive but could also result in platinum losses and should thus be avoided (Shore 2009, p. 6).

In addition, the fuel cell stacks of some manufacturers are moulded rather than screwed shut and can therefore not be disassembled by simply removing the casing and individual layers (Wegner et al. 2012, p. 433). In order to separate the MEA from other components, the end adapters need to be cut off and the casing removed. The stack can then be broken into individual units of MEA and bipolar plate, from which the catalyst-coated membrane may be separated. Although judging from the photographic process documentation supplied for this study it appears as though fragments of the catalyst-coated membrane remain attached to the bipolar plates or are otherwise damaged (cf. Figure 19), the authors state that a complete capture of all materials can be assumed (ibid., p. 434).



From top left: End adapter, contact plate, connecting element, unit of MEA and bipolar plate (shown from both sides), separated CCM

Figure 19: Disassembled moulded-type fuel cell stack (Wegner et al. 2012, p. 434)

While other fractions also contain valuable materials and, as pointed out previously, in principle all components could be recycled (Papasavva 2003, p. 2), further processing of the MEA only will be considered in the following.

Once the precious metal-loaded MEA has been separated from other fuel cell components, the need for further processing prior to the material recovery step depends on the ensuing recovery methods as well as the targeted materials. Due to the high value of the contained platinum, it is likely that platinum recovery will constitute the major driver for fuel cell recycling, with other materials, such as the polymeric membrane, being of minor interest. Of the nine platinum recovery processes analysed in Chapter 5.3.2, the majority require a further comminution of the MEA following its separation from other fuel cell stack materials, in order to disintegrate the MEA's laminate structure and expose the catalyst layer.

While in most publications, no further information is given on exactly *how* the MEAs are to be comminuted (cf. Koehler et al. 2006) or the process is simply referred to as 'shredding' (cf. Handley et al. 2002), Shore (2009) gives a more detailed description of the applied procedures and also mentions potential drawbacks of the employed technologies. However, as the different platinum recovery processes analysed in Chapter 5.3.2 require different feed materials, ranging in size from intact MEAs over 1 cm-sized fragments to finely ground powder, it is uncertain in how far these technologies can be employed in the pre-processing for other recovery methods. Nonetheless, the issues experienced by Shore (2009), such as matting together of MEA fragments and subsequent obstruction of milling equipment (ibid., p. 20), overheating of equipment (ibid., p. 39) and failure to successfully comminute MEAs due to the elastic properties of the membrane (ibid., p. 13) are likely to apply to other comminution requirements as well.

In order to counteract these problems, Shore (2009) reports two possible approaches, cryogenic milling and granulation at room temperature followed by milling. Cryogenic milling involves the milling of previously shredded MEAs by an impact mill operating in a chamber filled with liquid nitrogen (i.e. at a temperature of  $-197^{\circ}\text{C}$ ). While this approach successfully produces a fine, free-flowing and homogeneous powder and circumvents the above-mentioned issues (ibid. p. 13), the required equipment adds significant costs. Instead, the shredded MEAs may also be granulated at room temperature using a special cutting mill that can handle flexible feed materials, which sufficiently

disintegrates the laminated MEA structure but requires a subsequent wetting of the material prior the final recovery process (ibid. p. 21).

### 5.2.1 Potential deficits in pre-processing

As recycling of fuel cells is still in its very early stages and no commercial scale recycling plants exist at the time of writing, the research focus appears to be on the development of technically feasible material recovery processes rather than process efficiency. Thus, little information is available on potential losses of platinum in the disassembly and pre-processing stage of the recycling chain, let alone the magnitude of such losses. However, some conclusions may be drawn from the following deliberations.

As mentioned previously, the disassembly of fuel cells is likely to remain a manual process unless significant volumes of EoL fuel cells become available that justify investment in automated disassembly processes. While this fact may decrease the likelihood of false or incomplete disassembly of fuel cell stacks, the possible prevalence of moulded rather than bolted stacks could cause significant platinum losses, should fragments of the CCM remain attached to the bipolar plates after disassembly and thus fail to enter the material recovery process dedicated to MEA. While Wegner et al. (2012, p. 434) state that full material capture in the disassembly process was confirmed as part of their study and an assessment of this statement exceeds the expertise of this thesis' author, it is conceivable that minimal dust losses occur when the stack is forcefully broken into individual units. When applied to high volumes of EoL fuel cells, the platinum losses resulting from such processes could be significant.

As mentioned previously, the forceful removal of GDL from 5-layer MEA should be avoided as platinum-rich fragments may remain attached to the GDL. However, even careful delamination and subsequent removal of the GDL could result in significant platinum losses. This is due to the fact that over a fuel cell's lifetime, platinum particles tend to migrate from the catalyst layer into both the membrane and the GDL. Thus, *"Removal of the gas diffusion layer results in immediate loss of a portion of the precious metals originally present in the fuel cell that is otherwise available for recycling"* (Shore & Matlin 2009).

According to tests performed by Wegner et al. (2012, p. 436), precious metal losses of up to 0.017 mg/cm<sup>2</sup> (equal to 3.4% of the original load) could be experienced. While

these amounts obviously depend on the originally applied platinum load and recovery may still be possible if the catalyst particles accumulate elsewhere in the stack, it is important to take note of this fact in order to avoid significant losses.

Shore (2009, p. 20) also acknowledges the fact that if cryogenic milling forms part of the pre-processing stage, special attention must be paid to prevent loss of platinum-rich dusts, as the finely dispersed platinum may be carried off by the nitrogen gas liberated during gasification of liquid nitrogen. This effect may lower the platinum concentration of the output material from 1.5% when no cryogenic milling is employed to 1.3% with cryogenic milling.

Further difficulties in disassembly and pre-processing arise when it is assumed that in the future, specialised recycling facilities will handle a range of different types of fuel cells from different fields of application, as proposed by Handley et al. (2002, p. 350), Schiemann et al. (2007, p. 54) and Kromer et al. (2009, p. 8284).

In a detailed analysis of the different types and brands of fuel cells available, Schiemann et al. (2007, p. 54) stress that it will likely be tremendously difficult for disposal firms to recognise the exact type of fuel cell system they are dealing with. With a view to recycling efficiency, but especially health and safety issues and environmental protection, the authors emphasise that each type of fuel cell requires unique disassembly and processing steps and propose the use of flow charts, photographic guides and manufacturer catalogues to avoid misjudgement.

Once again it must be acknowledged that fuel cell recycling does not take place on a larger scale yet and thus any assessment of the order of magnitude of process losses can be considered idle speculation. As Kromer et al. (2009) state, “*without real-world experience with a fuel cell recovery supply chain the extent to which the process losses may be mitigated is unknown*” (p. 8284). However, based on the fact that fuel cell stacks may be removed, transported and disassembled largely without damaging the CCM the authors assume that total process losses of platinum recovery from automotive fuel cells will be in the order of 8 to 10%, which is around half of that estimated for automotive catalysts (ibid.).

Nonetheless, the issues portrayed above once more point to the importance of considering fuel cells' end-of-life already in the design phase, as design for disassembly and recycling could avoid significant precious metal losses and thus have not only environmental, but also economic benefits.

### 5.3 Material recovery

#### 5.3.1 Platinum group metal recovery through pyrometallurgical processes

As mentioned in Chapter 4.5, recycling already represents a major source of supply for a number of industries relying on platinum. Depending on the specific application and the concomitant material composition of the EoL product streams, a number of processes for PGM recovery are available, as shown in Figure 20. As evident from Figure 20, pyrometallurgical processes prevail. Since the processing of automotive catalysts is representative of this route and both ICE and fuel cell ELV are expected to pass similar collection and dismantling processes, the recovery of platinum from automotive catalysts will be considered in more detail in the following.

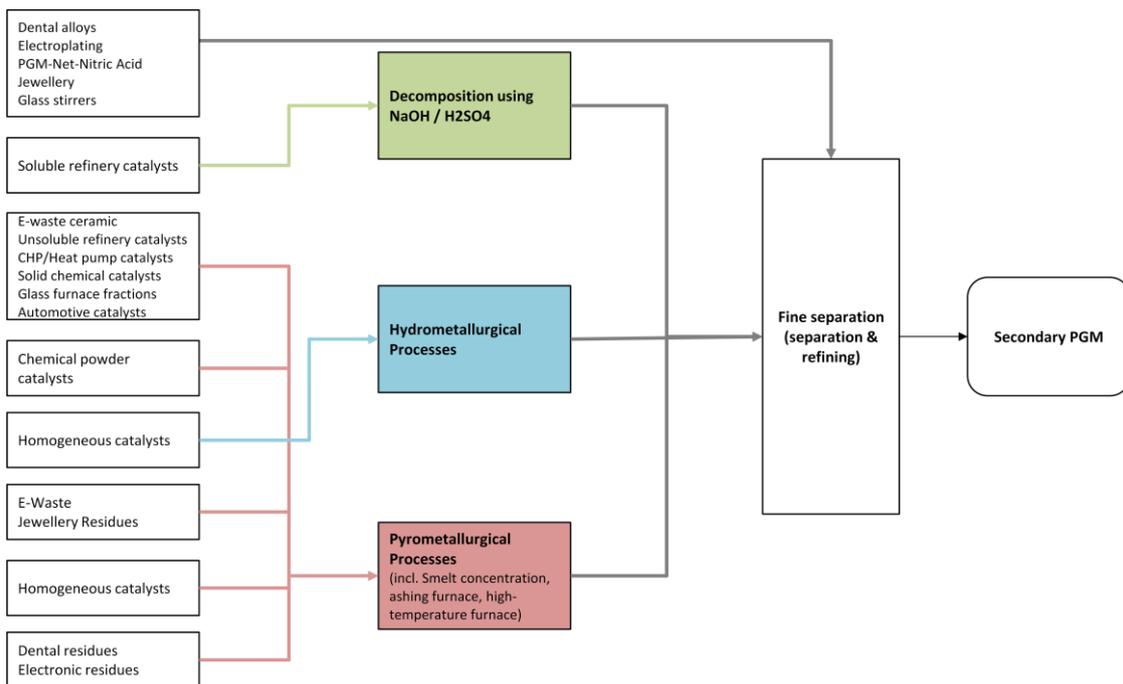


Figure 20: PGM recovery processes of different applications (adapted from Hagelüken et al. 2005a, p. 192)

### *5.3.1.1 Platinum recovery from automotive catalysts*

Due to its high value, the recovery of platinum from automotive catalysts is well-established and represents a major economic driver for the recycling of ELV. After dismantling, catalytic converters are collected, decanned and the catalyst ceramic transported to specialised precious metal refineries, where the PGM recovery can be achieved by hydrometallurgical and pyrometallurgical procedures (Buchert et al. 2009, p. VII). Since hydrometallurgical procedures involve the use of strong acids or leaches to separate the PGM-containing washcoat from its ceramic carrier and thus produce toxic wastewater, as well as achieving only moderate recovery rates, pyrometallurgical processes are commonly employed at copper smelters or specialised PGM refineries in state-of-the-art facilities, where recovery rates of around 98% can be achieved (Hagelüken 2005, p. 289; Lucas et al. 2011, p. 35).

In pyrometallurgical procedures the PGM are separated by smelting washcoat and ceramic together in one process. Due to their high specific density and high melting point (density of 21450 at 25°C and a melting point of 1769°C, respectively, for platinum), the PGM in liquid or solid form fall through this slag into a collector metal bath consisting typically of copper, or less commonly also of nickel, lead or iron (Rumpold & Antrekowitsch 2012, p. 697-698).

This PGM-enriched collector metal is processed further by electrowinning or the more time-consuming electrorefining. During either process, a current is applied to the PGM-rich solution, leading to the chemical reduction of the precious metal ions and their depositing at the cathode (Gottstein et al. 2006, p. 57). The resulting residue is then treated further by hydrometallurgical processes, including solvent extraction, precipitation and ion exchange (for further detail on these processes cf. Gottstein et al. 2006), in order to separate the individual elements with high purity levels (Rumpold & Antrekowitsch 2012, p. 697-698).

In state-of-the art facilities of specialised precious metal refineries, the ceramic slag resulting from the pyrometallurgical treatment as well as any arising side streams are fed into a second blast furnace process to recover any remaining precious metals, thus achieving a recovery rate of 98% and above. The processes described above require the use of sophisticated filter, scrubber and afterburning systems in order to prevent harmful emissions and waste streams (Hagelüken 2005, p. 291).

### 5.3.1.2 *Applicability of pyrometallurgical procedures for PEMFC recycling*

Although FCV, as proposed above, are expected to undergo the same EoL treatment processes as ICE vehicles from collection to dismantling, recovering the platinum contained in the MEA's catalyst layer by the pyrometallurgical processes used for recycling of catalyst converters is, although technically possible, not feasible.

This is due to the fact that incineration of the fluoropolymers contained in Nafion® and in the hydrophobically treated GDLs (cf. Chapter 2.2) leads to the formation of hydrogen fluoride (HF) (Wegner et al. 2012, p. 431). Although hydrogen fluoride is used in a number of industries, for example in glass etching or for the production of fluoride, it is considered a highly toxic compound that may be harmful to humans, animals and plants as well as having a highly corrosive effect on processing equipment (US EPA 2000). Boiling at room temperatures, hydrogen fluoride is typically found in a gaseous phase. If skin contact or inhalation and consequently dissolution in water occur, it is transformed into hydrofluoric acid, which enters and continuously destroys deeper tissue layers, thus causing severe and often fatal injuries (CDC 2013).

Employing the usual pyrometallurgical process route would therefore allow a high platinum recovery comparable to catalyst recycling, but require the refurbishment of existing plants with special protective linings for the furnaces as well as the installation of elaborate filter and scrubbing systems to eliminate HF from the off-gas. Such measures, however, are time-consuming and add significant costs, thus presenting a considerable economic barrier that prevents refineries from investing in such technologies (Stolten 2010, p. 54). In addition, through incineration the valuable membrane material Nafion® is essentially destroyed in the process and cannot be recycled (Shore 2009, p. 4) and any fluorine constituents remaining in the PGM-containing slag would hamper the later separation (Koehler et al. 2006).

### 5.3.2 State of the art in fuel cell recycling

In order to determine in how far the issues described above have been acknowledged by precious metal refiners, researchers and other stakeholders and have sparked the development of alternative recovery processes, a patent search was performed. This search included patents registered under the European Patent Office, the US Patent & Trademark Office and the World Intellectual Property Organization (WIPO) and yielded the results summarised in Table 6.

Note that with a view to the research focus of this thesis, the search focused solely on the recovery of PGM (as well as the more broadly phrased recovery of ‘noble metals’ or ‘precious metals’, which may include additional elements, such as gold or silver) from PEM fuel cells. Procedures aimed exclusively at recycling another type of fuel cell, different constituents, such as the membrane material, or other kinds of fluorine-containing devices, as well as any patents published before the year 2000 or in a language other than English were neglected.

The WIPO database also lists patents registered by national patent offices only and while further methods developed, for example, by Toyota Motor Corporation in Japan, Hyundai in Korea and by research institutes in China and Canada could be found, these were excluded from further analysis due to the limited scope of this thesis. In cases where process descriptions available via the WIPO database obviously represented a repetition of entries published with the regional patent offices, only the international publication is given in Table 6.

Table 6: List of patents for PGM recovery from MEAs avoiding hydrogen fluoride emissions

Patent Number	Inventor/s	Proprietor	Year of Publication	Materials recovered	Input material
WO/2004/102711	Hagelüken Kayser Romero-Ojeda Kleinwächter	Umicore AG & Co KG	2005	PGM	CCM, MEA (possibly shredded); electrolysis cells, batteries
WO/2006/024507	Koehler Zuber Binder Baenisch Lopez	Umicore AG & Co KG	2006	PGM and/or fluo- rine- containing constituents	CCM, MEA, whole fuel cell stacks, electrolysis cells, batteries, sensors; commin- uted to pieces of 1-3 cm edge length or pulver- ised
WO/2006/115684	Shore Robertson Shulman Fall	BASF Cata- lysts LLC	2006	PGM, ionomer	5- and 3-layer MEA
WO/2007/149904	Shore	BASF Cata- lysts LLC	2007	PGM, ionomer	5-layer MEA
WO/2009/029463	Shore Matlin	BASF Cata- lysts LLC	2009	PGM, ionomer	CCM-type MEA (shredded and pulverized)
WO/2009/149241	Shore Matlin Heinz	BASF Cata- lysts LLC	2009	PGM	MEA (shredded and pulverized)
WO/2010/132156	Shore Matlin Heinz	BASF Cata- lysts LLC	2010	PGM	MEA (shredded and granulated / pulverized)
EP2700726A1	Romero Meyer Voss	Heraeus Precious Metals GmbH	2014	PGM	any material with a fluorine content < 5 wt.%
WO/2015/010793	Paepke	Daimler AG	2015	PGM, ionomer	CCM, MEA; seals removed and shredded to pieces of 0.1 mm <sup>2</sup> - 9 cm <sup>2</sup>

In the following, a brief overview of the different approaches outlined in the patents listed above will be given.

#### 5.3.2.1 Procedures developed by Umicore AG & Co KG

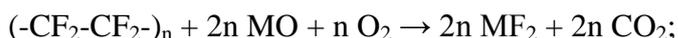
- a) Process for the concentration of noble metals from fluorine-containing fuel cell components (Hagelüken et al. 2005b)

As part of this process, the MEAs are mixed with an inorganic additive, such as calcium carbonate ( $\text{CaCO}_3$ ), which chemically binds hydrogen fluoride and other fluorine compounds during the subsequent thermal treatment. The fluorine-containing reaction product is concentrated in the combustion residue or slag, rather than released into the exhaust gas, and can be separated from the precious-metal containing products using established hydrometallurgical or melting procedures.

For the concentration of platinum from fluorine-containing catalyst residues the electrode layers are detached from CCMs to obtain a fluorocarbon polymer-containing catalyst mixture with a fluorocarbon polymer content of 19 wt.%, a fluorine content of 15 wt.% and a platinum content of 25 wt.%. In the patented sample process, the fluorine content amounts to 0.08 moles. Adding 0.8 moles (i.e. the tenfold amount) of anhydrous calcium carbonate ( $\text{CaCO}_3$ ), the material is mixed vigorously. This mixture is then heated to  $1000^\circ\text{C}$  and held at this temperature for a period of two hours, during which no fluorine-containing combustion gases are detectable.

Analysis of the combustion residue at the end of this process shows a calcium fluoride ( $\text{CaF}_2$ ) content of 7.2 wt.%, while the wash water maintains a neutral pH value and has a fluorine content below 3 parts per million ( $< 3 \text{ mg/l}$ ). Using established hydrometallurgical or melting processes, the fluorine-containing reaction product is separated and the precious metals refined further.

This process can be represented by the following equation:



where:  $(-\text{CF}_2-\text{CF}_2-)_{\text{n}}$  is a perfluoroalkyl compound (that may be substituted by perfluorinated compounds of the form  $(-\text{CFR}_1-\text{CFR}_2-)_{\text{n}}$ )

and:  $\text{M} = \text{M}^{2+} = \text{Mg}, \text{Ca}, \text{Sr}, \text{Ba}$  and others.

According to Schiemann et al. (2007, p. 78) this procedure is not only inexpensive but also able to recover 95% of the contained platinum. Umicore plans to build an industrial size recycling plant once significant volumes of EoL fuel cells arise.

- b) Process for recycling fuel cell components containing precious metals (Koehler et al. 2006)

This process involves dissolving the fluorine-containing components by treatment with a medium in its supercritical state, thus separating the fluorine-containing compounds from the precious metal-containing residues. According to the inventors,

*“Substances such as water or carbon dioxide become highly effective solvents and reaction media when they are heated under pressure to far above the boiling point. At a particular point, the density of the vapour becomes virtually as high as that of a liquid. In this "supercritical state" (i.e. liquid and gaseous at the same time), completely new physical properties are displayed. Thus, above 374°C (647°K) and a pressure of 220.6 bar, water suddenly dissolves oils and other organic compounds”* (Koehler et al. 2006).

The materials to be recycled are shredded using jaw crushers and / or hammer mills to create a homogeneous mix of fragments with 1 to 3 cm in edge length or, if using mills, a fine powder. This mixture is then placed in a pressure reactor and mixed with the reaction medium. In the patented sample process, a CCM, weighing 10 g and consisting of 1 g catalyst coating (platinum supported on carbon black), perfluorinated ionomer (Nafion®) and a 25 µm perfluorinated ionomer membrane (Nafion®), is placed in a heatable stainless steel autoclave equipped with a stirrer together with 1500 ml of deionised water (i.e. a 100- to 1000-fold amount based on the mass of the fluorine-containing compounds).

This mixture is heated for seven hours, reaching a maximum temperature of 384°C and a maximum pressure of 240 bar. The solution obtained from this treatment is filtered using commercial laboratory filters, which produces a filter cake that resembles carbon black as well as a transparent, greenish filtrate. Analysis of the filter cake shows that it consists of platinum and carbon-containing residues only, while fluorine-containing compounds are not present. From this filter cake, platinum can be recovered using conventional hydrometallurgical processes. In addition, it is possible to concentrate the filtrate and recover the ionomer from the concentrate using a recasting process.

Apart from deionised water, the following supercritical media listed in Table 7 may be employed as solvents; however, this requires further research on appropriate process parameters.

Table 7: Supercritical media for the extraction of fluorine-containing compounds (Koehler et al. 2006)

Medium	Temperature (°K)	Pressure (10 <sup>5</sup> Pa)
Nitrogen (N <sub>2</sub> )	> 126	> 33.9
Carbon Dioxide (CO <sub>2</sub> )	> 304.2	> 73.8
Water (H <sub>2</sub> O)	> 647.2	> 220.6
Ammonia (NH <sub>3</sub> )	> 405.6	> 113.7

While the inventors state that platinum may be “obtained in very good yield” (Koehler et al. 2006), data on the efficiency of the recovery process is not provided.

### 5.3.2.2 Procedures developed by BASF Catalysts LLC

Since the several patents applied for by Shore et al. (cf. Table 6) were all developed as part of one perennial research project and in some cases represent enhancements of the initial process, a summary of the invention as provided in the project’s final report will be given only. Details for the respective procedures can be obtained from the patents listed in Table 6.

Adapted to problems encountered in earlier versions of the recovery method, the process published in the project’s final report separates precious metals from fluorine-containing compounds by leaching, as illustrated in Figure 21.

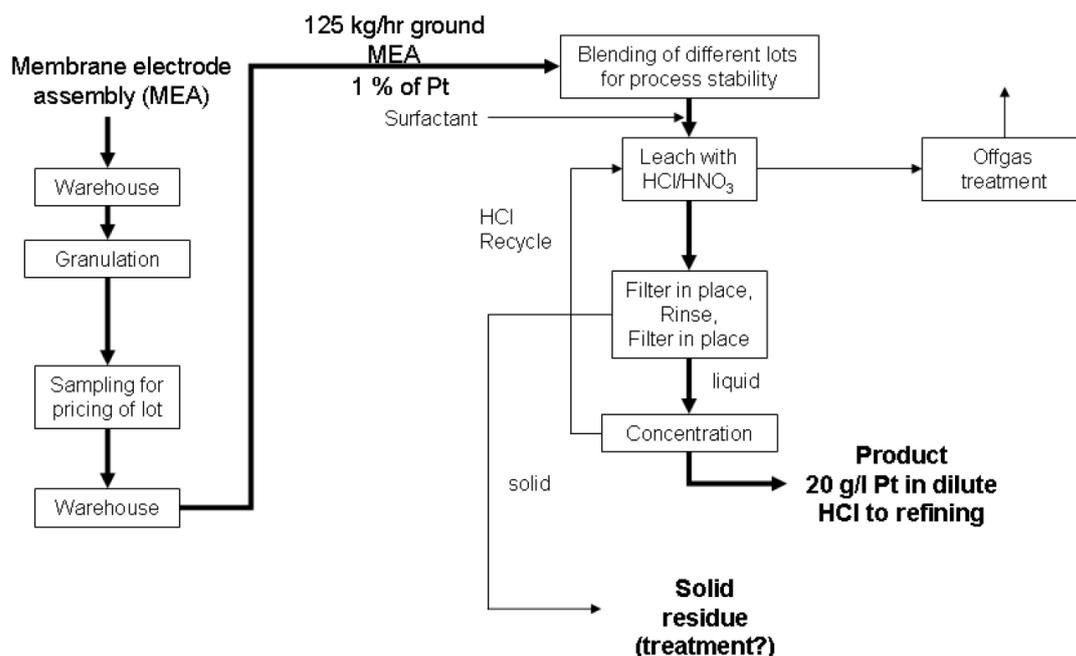


Figure 21: Process diagram for the recovery of platinum from MEA (Shore 2009, p. 16)

Since leaching is only viable if enough surface area is available for the acid to contact the precious metal sites, first of all 5-layer MEA with GDL of either carbon cloth or carbon paper are granulated at room temperature using a Press Side Granulator (Shore et al. 2009a, p. 1101). Due to the hydrophobic nature of the resulting MEA powder, an aqueous surfactant<sup>19</sup> must be added to stop the powder from floating on the acid's surface (Shore 2009, p. 14-17). Noting that the surfactant acted as an agglomeration agent, an alternative approach was also tested by the inventors, which involves exposing the MEA powder to 10-fold reduced amounts of surfactant and facilitating material dispersion by sonication. Adding only insignificant costs but substantially improving platinum recovery in the subsequent leach, the latter procedure constitutes the preferred option (Shore et al. 2009a, p. 1101).

The leach is then performed by adding aqua regia, a mixture of concentrated hydrochloric acid (HCl) and concentrated nitric acid (HNO<sub>3</sub>) in a 3:1 ratio (by volume) and heating the mixture. The resulting reaction produces water, nitrogen oxides (NO<sub>x</sub>), chlorine and aqueous hexachloroplatinic acid (H<sub>2</sub>PtCl<sub>6</sub>) and can be described by the following equations (Shore et al. 2009b):

- 1)  $\text{HNO}_3 + 3\text{HCl} \rightarrow \text{NOCl} + 2\text{Cl}_2 + 2\text{H}_2\text{O}$
- 2)  $\text{Pt} + 2\text{HNO}_3 + 4\text{HCl} \rightarrow (\text{NO})_2\text{PtCl}_4 + 3\text{H}_2\text{O} + \frac{1}{2}\text{O}_2$
- 3)  $(\text{NO})_2\text{PtCl}_4 + 2\text{HCl} \rightarrow \text{H}_2\text{PtCl}_4 + 2\text{NOCl}$
- 4)  $\text{H}_2\text{PtCl}_4 + \text{Cl}_2 \rightarrow \text{H}_2\text{PtCl}_6$

However, it was noted that adding HNO<sub>3</sub> in the ratio described above produces excessive amounts of NO<sub>x</sub> as well as chlorine oxidants, which increases vessel pressure, necessitates extensive off-gas cleaning requirements, limits the possibility to recycle HCl and thus adds costs. Alternative approaches were therefore tested, which showed that decreasing the amount of HNO<sub>3</sub> to approximately 15% of the aqua regia stoichiometry, as well as adding concentrated HCl before adding HNO<sub>3</sub> improved the efficiency of platinum recovery. In addition, other oxidants, such as hydrogen peroxide, chlorine gas and sodium chlorate were trialled, but preference was given to using aqua regia. When adding HCl first, platinum yields of 99% were consistently achieved with one leach step (Shore 2009, p. 18).

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<sup>19</sup> in this case, Plurafac LF 120 by BASF Corporation and Strodex PK-90 by Ashland Chemicals were used

In order to achieve the desired platinum concentration of 20 g per litre (cf. Figure 21), the leachate needs to be distilled (Shore 2009, p. 28). Following the distillation, above 80% of the HCl added in the leach step can be recycled from the residues; however, since the leaching process requires concentrated rather than azeotropic HCl, a process must be chosen to yield concentrated HCl (ibid., p. 29). Other waste streams include NO<sub>x</sub> emissions and solid MEA residues, which must be treated according to environmental regulations (ibid., p. 38). However, since employing a conventional combustion method for platinum recovery would lead to the liberation of 400 g hydrogen fluoride and 2 kg CO<sub>2</sub> per kg of Nafion®-based MEA, the inventors stress the procedure's environmental benefits (ibid., p. 38).

Although the process described above has so far only been tested under laboratory conditions, its potential and economic viability for large-scale utilisation at a plant treating MEA from around 500,000 FCV per year was confirmed as part of the research project. Based on the standard platinum load per MEA and platinum price at the time of publication, the variable costs of this process amount to only a few percent of the platinum value (ibid., p. 26; p. 38).

#### 5.3.2.3 Procedure developed by Heraeus Precious Metals GmbH

- a) Device and method for the thermal treatment of products containing fluorine and precious metals (Romero et al. 2014)

Referring to the process described by Hagelüken et al. (2005b), the inventors claim that this procedure is unsuitable for materials which release hydrogen fluoride at low temperatures, as the inorganic additive's absorption capability has proven insufficient or not fast enough and hydrogen fluoride gas is thus still released. Rather than providing a novel recovery process, their invention is therefore aimed at providing a plant that is able to handle thermal treatment of fluorine-containing components without any damage to the equipment or release of harmful substances. The plant consists of an ashing furnace and a post-combustion chamber made of at least 85 wt. % aluminium oxide, making it resistant to hydrogen fluoride, as well as several exhaust gas treatment steps in order to prevent harmful emissions.

Since no information can be obtained on potential cost advantages of this approach compared to the refurbishment of existing recycling plants with protective linings and other specialised equipment, the details of this invention will not be analysed further. If such cost advantages exist, however, the construction of separate furnaces at existing precious metal refiners could be a more viable approach than implementation of the laborious, multi-step procedures described above.

#### 5.3.2.4 Procedure developed by Daimler AG

##### a) Method for recycling membrane/electrode units of a fuel cell (Paepke 2015)

Claiming that the process invented by Hagelüken et al. (2005b) is unsuitable for materials which release hydrogen fluoride at low temperatures and the procedure by Koehler et al. (2006) involves considerable safety requirements due to the high-pressure operating conditions, this invention proposes an alternative process for separating fluorine- and precious metal-containing compounds using ultrasound and a solvent.

First of all, the CCM or 5-layer MEA are shredded to fragments measuring 0.1 mm<sup>2</sup> to 9 cm<sup>2</sup>. Care must be taken to ensure that the comminution does not lead to the formation of smaller particles, as this would result in the creation of an undesirable suspension or colloidal solution when mixed with the solvents. The shredded MEA are then placed into an ultrasonic bath filled with ethanol and water at a ratio of 4 to 1. During the ultrasonic treatment at a frequency of 20 to 110 kHz and a temperature of 5 to 40°C, the mixture is continuously circulated by stirring or oscillating of the container.

After around three minutes, the catalyst layer made of carbon black and precious metals is solved almost entirely from the membrane. As the precious metal-loaded substrate particles transition into the solvent, the precious metal-loaded and fluorine-containing fractions can be separated by a two-step filtration process. The PGM-loaded filters can then enter a conventional pyrometallurgical recovery process, while the remaining membrane material may be supplied to a subsequent material or energy recovery process and the solvent is reused. Overall, PGM recovery rates of 98% and above can be achieved.

### 5.3.3 Potential deficits in material recovery

Judging from the number of recovery procedures available that achieve yields of 95% and above it appears as though platinum recovery from EoL MEA is possible incurring only minimal losses. However, upon closer investigation it becomes clear that while avoiding HF emissions, all procedures described above continue to entail certain drawbacks.

As mentioned previously, mixing the MEA with an inorganic additive, such as calcium carbonate, has shown to be insufficient in binding HF, especially for materials which release hydrogen fluoride at low temperatures (Romero et al. 2014). Using water in its supercritical state, the process proposed by Koehler et al. (2006) relies on a high-pressure operating environment, thus posing concerns for health and safety as well as untenable equipment costs.

Similar reservations may be voiced for the HF-proof furnace and scrubbing process invented by Romero et al. (2014), as its cost advantage compared to refurbishment of existing precious metal refining facilities is questionable. Finally, while the procedure proposed by Shore (2009) avoids HF emissions, it uses hydrochloric acid and nitric acid, both of which are harmful if skin contact occurs, and generates nitric oxides as well as solid MEA residues, which then require further treatment to avoid environmental impacts.

Furthermore insufficient yields of platinum may be problematic in those cases, where removal of the GDL is required in the pre-processing stage (cf. Chapter 5.2) or the recovery process is aimed at separating membrane and catalyst layer. This is due to the fact that over a fuel cell's lifetime, platinum particles tend to migrate from the catalyst layer into the membrane and / or GDL, leading to the formation of nanocrystallites. Since these are less than 200 nm in size, the platinum-loaded nanocrystallites cannot be recovered by filtration and are hence lost in the solvent (Shore & Matlin 2009). In addition, some loss of theoretically recoverable platinum in waste streams, such as the process water and solid waste from leaching, appears to be unavoidable (Shore 2009, p. 39).

Several of the above mentioned patents also fail to elaborate on any further refining needs in order to obtain platinum of a purity level that is suitable for commercial use. As Shore (2009, p. 37) describes, the conventional combustion of MEA produces a platinum-rich ash, which must be further refined to remove impurities.

Although except for Shore et al., all of the above-mentioned patents assume a further downstream treatment in the form of hydrometallurgical and / or pyrometallurgical recovery and refining processes, the potential for and magnitude of platinum losses is not mentioned. In contrast, the leaching process proposed by Shore (2009) produces a dilute platinum solution, which is concentrated by distillation and can then be refined directly, thus already integrating a major part of the standard refining process (*ibid.*, p. 37).

Finally, it must be noted that to date, all processes have been implemented and tested at a laboratory level only. Although Shore (2009, p. 38) provides a cost analysis and comes to the conclusion that large-scale operations based on the invented process as described above would be feasible from a financial perspective, no such information is available for any of the other recovery methods. It is therefore not possible to determine whether industrial-scale implementation is possible at all, nor how any of the procedures mentioned above compare to a HF-proof refurbishment of existing precious metal recovery facilities with special protective linings for the furnaces and off-gas treatment in terms of costs. Further research on recovery methods focussing on process efficiency and costs is likely to only take place once sufficient volumes of EoL fuel cells arise.

To conclude, this chapter has shown that the recycling of platinum from fuel cell vehicles is still in its very early stages. While methods for recovering platinum at a high efficiency are available at least at a laboratory level, it appears as though the preceding stages in the recycling chain have received little attention by the research community and other stakeholders so far. However, as demonstrated in this chapter, deficits in the stages of disassembly, dismantling and above all collection are often responsible for major losses and valuable materials becoming unavailable for recycling. Since the efficiency of the entire recycling chain is determined by its weakest link, it is therefore vital to research and devise efficient strategies for collection, dismantling, pre-processing and material recovery before high volumes of end-of-life fuel cell vehicles arise.

## 6 Platinum flows from the diffusion and recycling of fuel cell vehicles

By evaluating the progression of platinum demand from the fuel cell vehicle industry, to what degree this demand can be satisfied by primary production and what challenges may be present in a future recycling chain, the preceding chapters form the qualitative basis for the research question of this thesis. In order to assess the potential contribution of recycling to meeting the future platinum demand of the fuel cell vehicle industry in quantitative terms, this chapter will build on the insights gained and develop a dynamic material flow analysis.

### 6.1 Methodology

#### 6.1.1 Material flow analysis

*“Increased consumption has led to an accumulation of significant stocks of metals in the anthroposphere, and the collection and recycling of metals from these secondary resources has become more and more important. These activities rely on knowledge of anthropogenic material cycles regarding quantities, qualities and locations of metal-containing goods that have accumulated”* (Müller et al. 2014, p. 2102).

Providing this required knowledge of material cycles in the anthroposphere, material flow analysis (MFA) constitutes a useful tool for analysis, planning and forecasting of resource consumption. It helps to identify the depletion and accumulation of resource stocks in both natural and anthropogenic environments, models the shift of materials from natural reserves to anthropogenic stocks and thus aids in forecasting the scarcity of resources (Brunner & Rechberger 2004, p. 16).

MFA is based on the principle of conservation of matter or mass balancing (i.e. input equals output) and is closely linked to the concept ‘metabolism of the anthroposphere’, which can be understood as an analogy to the human or natural metabolism. While a more detailed review of this idea is beyond the scope of this thesis, suffice it to note that the concept imitates the continuous cycling of material and energy present in biological processes and has been used to analyse and describe urban and regional material balances, especially for the purpose of environmental protection and waste management. The term was coined by Baccini and Brunner (1991), although similar ideas have

emerged in various disciplines throughout the 20<sup>th</sup> century (cf. Baccini & Brunner 1991; Ayres & Ayres 2002; Brunner & Rechberger 2004).

Depicting and quantifying the material stocks and flows of this anthropogenic metabolism, MFA includes “*the analysis of the throughput of process chains comprising extraction or harvest, chemical transformation, manufacturing, consumption, recycling and disposal of materials [...] based on accounts in physical units*” (Bringezu & Moriguchi 2002, p. 79). This way, MFA delivers a systematic assessment of the sources, pathways, intermediate and final sinks of a material within a system defined in space and time. By balancing inputs and outputs, flows of waste and environmental burdens as well as depletion and accumulation of material stocks can be identified (Brunner & Rechberger 2004, p. 3).

Most MFAs on metal cycles in the anthroposphere use static models, thus providing only a snapshot in time but failing to deliver information on the dynamics of resource consumption and its impacts on stocks and flows (Müller et al. 2014, p. 2103). Since future changes in platinum stocks and flows are of particular interest for the purpose of this thesis, this chapter will instead focus on a *dynamic* MFA to model future platinum flows and thus provide information about the behaviour of this system over a time interval. Various methods and principles for preparing dynamic MFAs have been developed in recent years (cf. Ayres & Ayres 2002; Müller et al. 2014), but most are based on the generic system depicted in Figure 22.

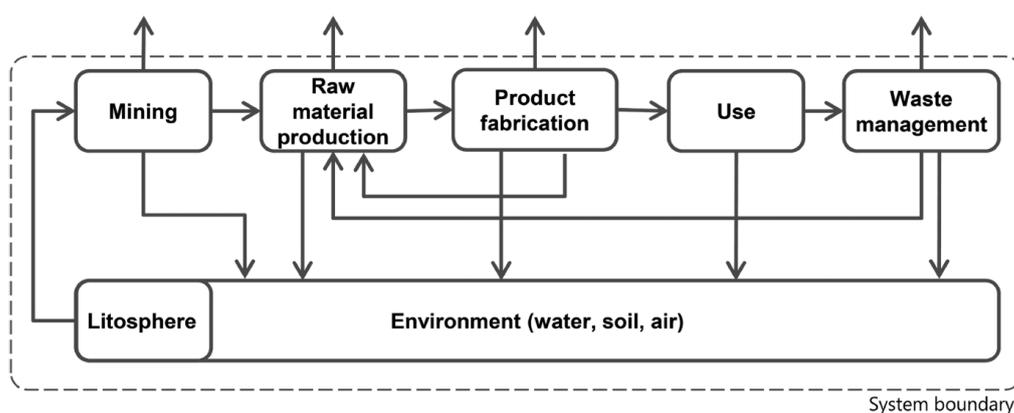


Figure 22: Generic dynamic material flow model of metals (Müller et al. 2014, p. 2105)

In terms of terminology, this thesis will follow the definitions established by Baccini and Brunner (1991) and developed further by Brunner and Rechberger (2004), which are summarised in the following.

First of all, it is necessary to distinguish between substances, materials and goods. For the purpose of MFA, ‘substance’ refers to “*any (chemical) element or compound composed of uniform units. All substances are characterized by a unique and identical constitution and are thus homogenous*” (Sax & Lewis 1987 in Brunner & Rechberger 2004, p. 36). In contrast ‘goods’ are defined as “*economic entities of matter with a positive or negative economic value [...] made up of one or several substances*” (ibid.). Finally, ‘materials’ is used as an umbrella term for both substances and goods (ibid., p. 37).

Modelling the movement of materials through the system, ‘processes’ are defined as “*transformation, transport or storage of materials*” (Brunner & Rechberger 2004, p. 37) and can describe both natural and man-made processes. Encompassing the material that is stored within a process, ‘stocks’ form part of this process and are defined as “*material reservoirs (mass) within the analyzed system*” (ibid., p. 4). Stocks are essentially dynamic and can be accumulated or depleted, but also remain constant over time. With regards to processes, MFA assumes a ‘black box’ approach, meaning that sub-processes within a process are neglected<sup>20</sup>, as only inputs and outputs are of interest. Disclosing the quantity of materials stored within a process, stocks form the only exception to this approach. If the residence time of a stock exceeds 1,000 years, it is generally considered a ‘final sink’ (ibid., p. 38).

The processes within the MFA are connected by ‘flows’ and ‘fluxes’ of materials. These terms are often used interchangeably although their actual denotation differs. While a flow describes the mass flow rate, i.e. the mass of a specific material that flows per time, in physical units (e.g. tons per annum) a flux is used to describe the mass flow rate per ‘cross section’. This can be, for example, a studied region, city or inhabitant. A flux can thus be considered a specific flow; integration of all fluxes for the cross section then delivers the total flow. All flows/fluxes require both origin and destination, which is represented by an import and export process that crosses the system boundaries (ibid., pp. 39-40).

Finally, a ‘system’ consists of “*a set of material flows, stocks and processes within a defined boundary*” (Brunner & Rechberger 2004, p. 4). This boundary is defined in space and time and must be chosen carefully considering data availability, objectives, applied balancing period and residence time of materials.

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<sup>20</sup> Otherwise the process must be divided into several sub-processes.

### 6.1.2 Software tool

The dynamic MFA for this thesis is performed using the software tool STAN (short for SubSTance flow ANalysis), which is a freeware developed by the Institute for Water Quality, Resources and Waste Management at the Vienna University of Technology in cooperation with INKA software. The software is available in its third version (STAN 2.5), which was published in 2012, from the TU Vienna's dedicated website.<sup>21</sup> STAN not only remediates some of the issues of earlier MFA instruments, such as the need for laborious and error-prone use of several software products, but also addresses the issue of data uncertainty and inconsistency. The software performs data reconciliation by automatically using redundant information to support uncertain data across time periods and data layers. In addition, statistical tests are used to detect gross errors, while corresponding data uncertainties are calculated by error propagation. With its 'period feature', STAN is particularly useful for performing dynamic MFA covering a perennial time span (cf. Cencic & Rechberger 2008).

### 6.1.3 Calculation approach

The calculations performed as a preliminary basis to create the model of platinum flows arising from market penetration of fuel cell vehicles are derived from a top-down approach as summarised in Figure 23. While this chapter provides a general overview of the employed calculation approach, the specific calculations can be reproduced by conferring to Appendix 3. Furthermore, the various data sources for the specific assumptions underlying these calculations will be explained further in the subsequent chapter 6.2.

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<sup>21</sup> <http://www.stan2web.net/>

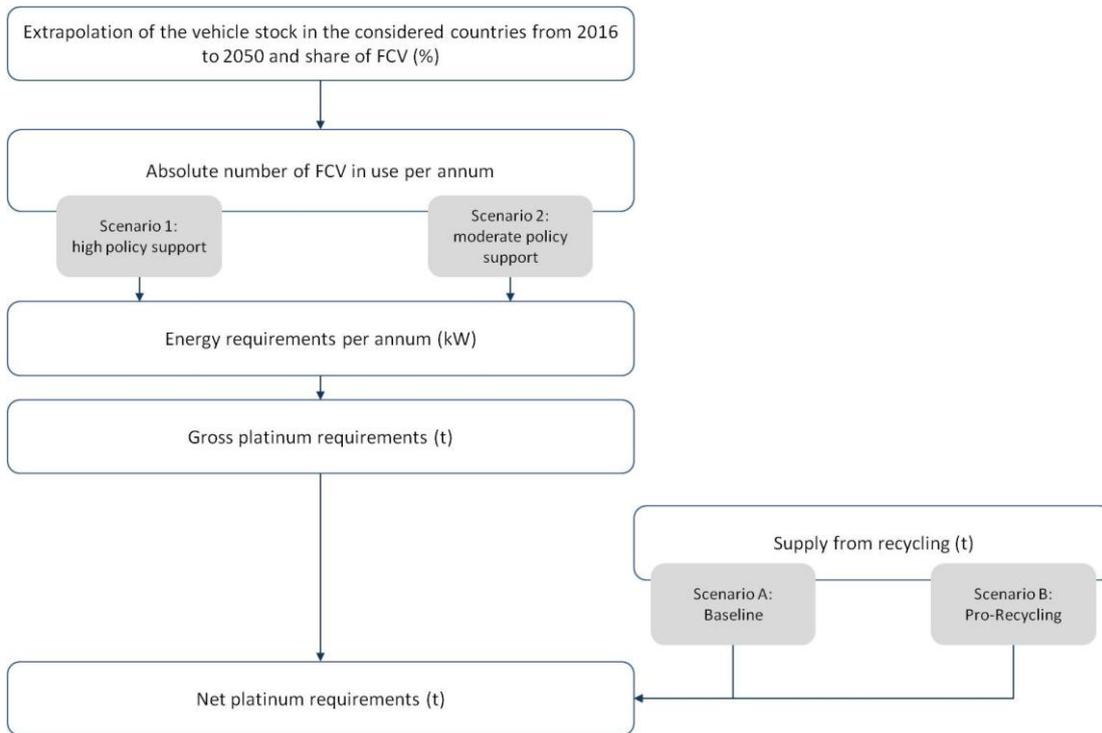


Figure 23: Calculation approach in preparation for the MFA (own source)

The calculation originates from the FCV market penetration ratios given by the two scenarios in EC (2008, p. 17), which are applied to the analysed EU member states' extrapolated vehicle stock to derive the absolute number of FCV in use per annum over the considered time scale of 35 years (cf. Chapter 6.2.1). In the next step, the annual energy requirements (kW/a) are calculated from the number of newly registered FCV per annum. Applying to this the platinum load per kW of the respective year as well as any losses occurring in the production stage, one can then determine the gross platinum requirements per annum. In order to determine the net platinum requirements, the potential supply from recycling is then calculated. Based on the two recycling scenarios given in Chapter 6.2.3, any platinum losses throughout the use phase and within the recycling chain are subtracted from the potentially available platinum content of end-of-life FCV assuming a vehicle lifetime of 10 years. The various material efficiency ratios of production, use phase and recycling are used as transfer coefficients and transferred into the MFA software, together with the values obtained from the aforementioned calculations.

## 6.2 System boundaries and assumptions

A number of assumptions and system boundaries have to be set in order to narrow down the scope of the MFA. While the complete list of parameters is given in Appendix 4, some of the assumptions made require a further examination and are thus explained in more detail in the following sub-chapters.

### 6.2.1 Market penetration scenarios

Since the projected level of market penetration provides the first step in the calculation and thus ultimately determines the mass and volume of platinum flows, a careful choice of scenario is critical for the validity of the later analysis. In this regard it must be stressed once more that projections, by definition, only forecast *possible* developments and must not be misunderstood as precise predictions of the future. Based on a set of specific assumptions, different scenarios may provide a fan of projections that, hopefully, capture the actual development if any set of assumptions comes true. Despite these limitations, projections provide a valuable tool by indicating the drivers and influences of future developments (cf. Angerer et al. 2009, p. 7).

A number of studies are available projecting possible scenarios of FCV market diffusion, some of which also include predictions of associated platinum requirements. As summarised in Appendix 1, these have come to highly diverging conclusions, depending on the time horizons, geographical locations and technologies considered, as well as on the applied assumptions concerning supporting aspects, such as infrastructure, political measures and technological developments. Aiming to evaluate possible recycling pathways for FCV for a closed European system, for the purpose of this thesis it was especially important to select a study focusing on Europe specifically. This requirement already precluded a number of publications, as the majority take on a global perspective, with some also considering specific lead markets such as the US, Canada and Japan.

From the remaining publications, the “HyWays” study, published by the European Commission in 2008, was chosen because it covers the most far-reaching time horizon (2010 to 2050) and for the equally important reason that the contained data is easily accessible. While this study provides four scenarios outlining possible rates of market penetration, depending on the level of policy support for hydrogen technologies as well

as the rate of technological progress, the overall study may be considered overly optimistic with regards to FCV market penetration when compared to, for example, Råde (2001) or Carlson et al. (2003). It must therefore be noted that selecting this study as this thesis' MFA basis involves the risk of adopting false assumptions on market diffusion of FCV. In principle, however, this drawback applies to any study and cannot be eliminated.

The HyWays report was published as part of an integrated project co-funded by research institutes, industry and the European Commission, which ran from 2004 to 2007 with the objective of developing a validated roadmap for the introduction of hydrogen technologies in Europe. Based on technology databases, sociological, technological and economic analyses as well as extensive interaction between science and industry experts from a range of member states, the study produced market penetration scenarios for various hydrogen-based technologies, including FCV. In doing so, the study focuses on the following ten European countries, which will also form the geographical focus of this thesis' MFA: Finland, France, Germany, Greece, Italy, the Netherlands, Norway, Poland, Spain and the United Kingdom.

The four FCV market penetration scenarios provided by EC (2008, p. 17) include two rather optimistic scenarios that are characterised by high to very high policy support and fast technological progress and assume that mass production of FCV will begin by 2013. In contrast, the two more conservative scenarios based on modest rates of technical learning delay the earliest mass-market rollout to 2016 and are characterised as follows:

*“the hypothetical start of mass production has been shifted to 2016 and the number of first movers<sup>22</sup> reduced to 4 which will ramp-up their plant utilization rate from 5% to 90% within a five year time frame (maximum production capacity of each of the four plants 100,000 units per year). After reaching full utilization of the production capacities of the first movers [...] after 5 years [...], it was assumed that followers are entering the market in a similar way and the first movers are doubling their production capacity” (European Commission 2008, p. 16).*

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<sup>22</sup> i.e. the first companies to enter a specific market or industry (author's note)

Given the current state of the FCV market and the recent launch of the Toyota Mirai (cf. Chapter 3), the assumptions underlying the two more conservative scenarios appear closer to reality. Hence, only the two conservative scenarios, which are pictured in Figure 24, will be considered for this thesis' MFA.

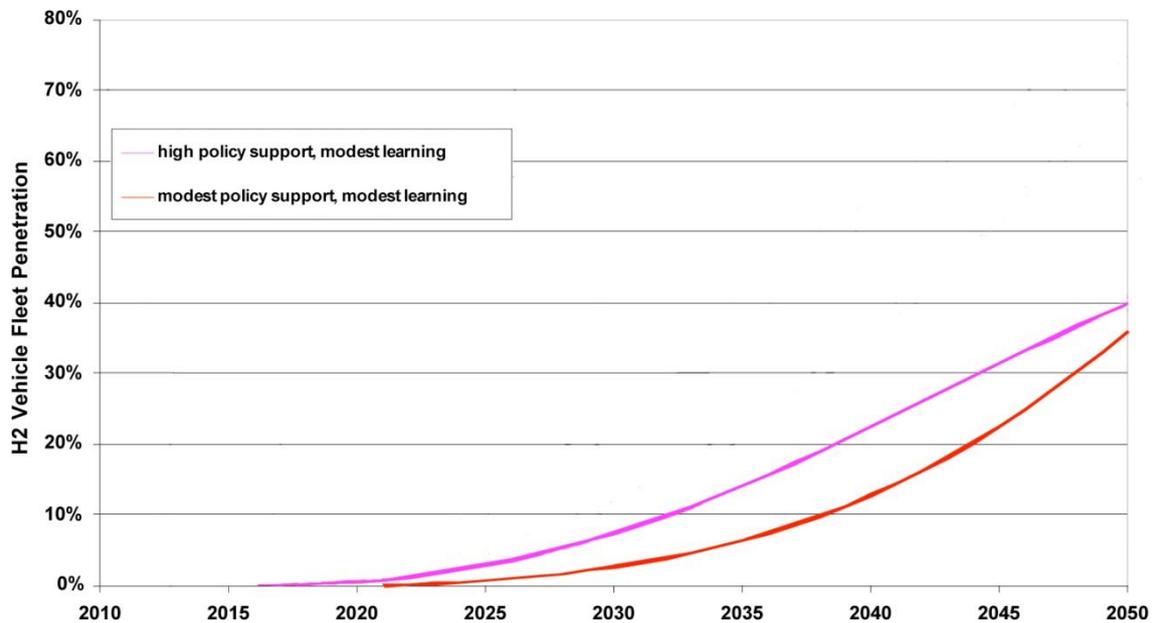


Figure 24: Market penetration rate of hydrogen vehicles for passenger transport (adapted from EC 2008, p. 17)

It must be noted that the study does not differentiate between fuel cell vehicles and other hydrogen-based power-train technologies. However, as examined in Chapter 3, hydrogen-based combustion engine technologies and other options have not been pursued by vehicle manufacturers in recent years. A market introduction in the near future thus appears highly unlikely and hence, technologies other than FCV as described in Chapter 3.2 will be neglected in this thesis.

In addition, the study not only includes light-duty passenger vehicles, but also public transport buses in its analysis. Public transport is likely to represent an important market for fuel cell vehicles, thus failing to differentiate the penetration rates given in Figure 24 by type of vehicle bears the potential of significantly overestimating the market potential of light-duty fuel cell vehicles. However, since this information is not disclosed in the report and the level of market penetration is given in ratios rather than absolute numbers, applying these ratios to the projected future numbers of light-duty passenger vehicles in the EU in order to deduce the number of FCV is considered a viable approach.

Historical and current data on car ownership in the EU can be obtained from the ACEA<sup>23</sup> website; however, limited information is available on expected vehicle fleet growth rates. The car ownership growth rate of 2.18% supplied by the World Business Council on Sustainable Development appears to be a commonly used value (cf. Fulton & Eads 2004, Annex A 6; Albach 2014, p. 78). While this value may be true for the world as a whole, given the declining demand for passenger transport documented by the European Environmental Agency (EEA 2014) in recent years, such assumptions appear somewhat unrealistic when focusing on the EU specifically. Other publications come to much more conservative estimates, as summarised in Table 8.

With a view to the geographical focus of this thesis, the mean value of the growth rates suggested below will be applied, with the latest publication on passenger cars in use in the EU (Eurostat 2014) used as the baseline amount. It must be noted that several studies forecast the level of vehicle ownership to peak at some point in the future and then decline. While the MFA would undoubtedly benefit from an accurate calculation of the vehicle fleet's growth per year, for simplicity reasons only an average annual growth rate will be used.

Table 8: Estimates for growth rate of vehicle fleet in literature (own source)

Source	Geographic focus	Time Horizon	Average annual growth rate of vehicle fleet <sup>24</sup>
BMVBS (2006)	Germany	2002 – 2050	-0.06%; -0.02%; 0.24%
McKinsey & Company (2010)	EU 27, Norway & Switzerland	2010 – 2050	0.32%
Shell Germany (2014)	Germany	2014 – 2040	-0.11%
World Energy Council (2011)	OECD countries	2010 – 2050	0.81% to 0.91%
Mean value			0.30%

The average annual growth rate is then applied to the number of vehicles in use reported by the ten member states in 2011, which represents the latest data available via Eurostat (2014). However, no vehicle stock figures have been reported by Greece in the past few years, hence this country will be excluded from further analysis due to a lack of data. In addition, no data is available on the vehicle stock of France prior to 2012; hence this figure has been used to calculate vehicles in use in 2011 instead of actual 2011 data.

<sup>23</sup> European Automobile Manufacturers Association

<sup>24</sup> if not stated explicitly, this was calculated based on latest data on vehicle ownership given by Eurostat (2014)

Based on the vehicle stock reported by Eurostat (2014) and the above mentioned average annual growth rate, the vehicle fleet in the 9 analysed EU member states will develop as illustrated in Figure 25. In 2050, the market penetration rates given by the two scenarios are equivalent to 87,521,198 fuel cell vehicles (=7,001,695,862 kW) in Scenario 1 and 78,769,078 fuel cell vehicles (=6,301,526,276 kW) in Scenario 2, respectively.

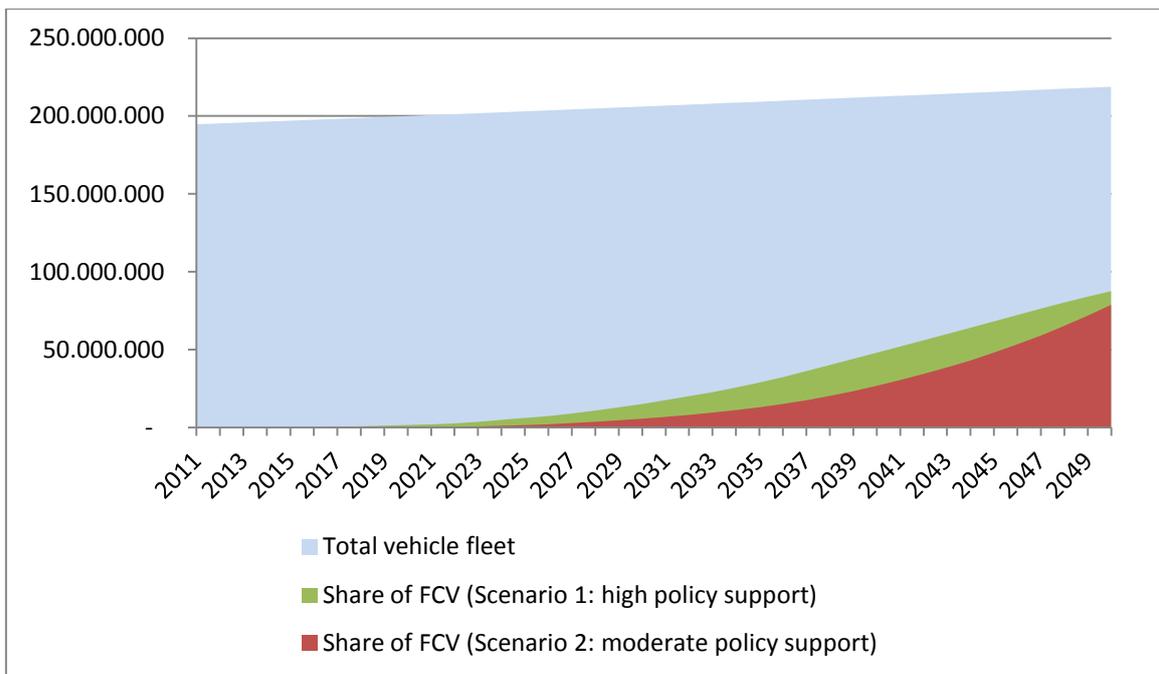


Figure 25: Development of the vehicle stock from 2011 to 2050 and share of fuel cell vehicles (own source based on EC (2008); Eurostat (2014))

### 6.2.2 Technical assumptions

In addition to the market penetration scenarios described above, certain assumptions have to be made concerning technical aspects. These include the current and future platinum load per kW installed power, material leakage during the FCV use phase, as well as production and assembly of fuel cells and FCV and the respective efficiencies of these processes.

As mentioned in Chapter 2.3, the required platinum load per kW has been reduced continuously. Future research efforts are likely to result in further reductions of the catalyst quantities required and possibly even in ultimately replacing platinum by a cheaper material altogether. Nonetheless, for the purpose of this MFA it is assumed that platinum will continue to constitute the main catalyst material until 2050.

As mentioned previously, little is known about the actual platinum content of current FCV rather than the platinum load documented in laboratory experiments. However, Simons and Bauer (2015, p. 3) assume a platinum content of 0.25 g per kW in 2012, a value which will be also be used in this thesis's MFA, although the authors base their assumption on a much lower vehicle power. In light of the significant technological improvements in the past decade, the already achieved platinum reductions, and further progress towards ultralow-platinum catalysts since publication of the target values cited in Chapter 2.3, this thesis will also conform to researchers of the US Department of Energy and assume a platinum loading of 0.125 g/kW and 0.125 mg/cm<sup>2</sup>, respectively, for the year 2020 (cf. US Drive FCTT 2013, p. 8).

From 2020 onwards, the required platinum load is likely to be reduced further and reach a level of approximately 0.07 g/kW by 2030, as forecasted by Adamson (2015, p. 37). This platinum load is roughly equivalent to the loading required in diesel exhaust gas catalysts in 2015. While further reductions of platinum requirements from 2030 onwards are conceivable, any technology development forecasts reaching even further into the future may be considered idle speculation.

This thesis thus complies with the hypothesis of Gerboni et al. (2008, p. 44), who assume that the rate of technological progress eventually levels out as the technology becomes more established. This idea is supported, for example, by Saurat and Bringezu (2009, p. 409), who also assume a decreasing rate of material efficiency gains. For the purpose of the MFA it is therefore assumed that from 2030 onwards, a platinum load of 0.07 g per kW will continue to be applied (cf. Figure 26).

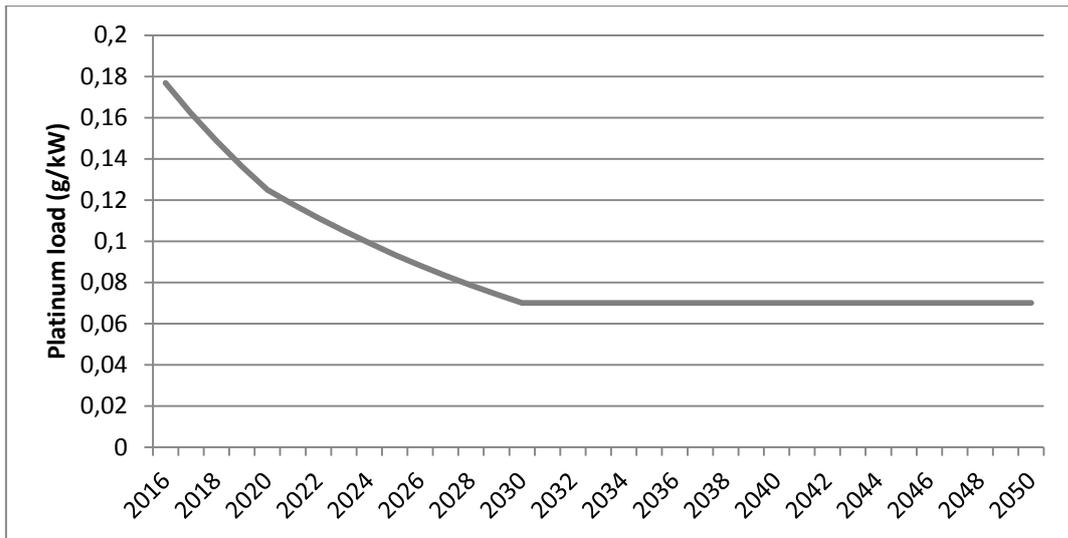


Figure 26: Development of platinum load per kW (own source based on US Drive FCTT (2013, p. 8); Adamson (2015, p. 37))

With regards to the manufacturing process of fuel cells and FCV, the MFA describes the production of a CCM-style MEA using a spray-coating method. Detailed information concerning the efficiency of this process and possible material losses cannot be obtained, which is likely due to the fact that the research focus currently lies on improving performance parameters rather than production efficiency. Since a large proportion of the fuel cells in use today is produced manually in small-scale laboratories, options for automated assembly and the associated process efficiencies have likely not been investigated yet. In addition, such information may also be considered highly relevant for competitors and therefore closely guarded by fuel cell manufacturers.

Manufacturers of coating machinery praise their equipment for ‘minimising wasteful overspray<sup>25</sup>’ and ‘maximising coating precision’ (cf. Ultrasonic Systems Inc. 2014; Sonner Ultrasonics 2015; Sonotek 2015). However, the efficiency of any application process depends on a number of factors, including geometry of the workpiece, viscosity, composition and particle size of the applied material, handling of application equipment and other process parameters. Experience with coating methods for other applications has shown that material loss caused by overspray cannot be avoided entirely, especially if using manual coating procedures.

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<sup>25</sup> that part of the applied material that does not deposit on the workpiece, caused by spraying past the workpiece as well as by lateral flows of finely dispersed material droplets (cf. Betrieblicher Umweltschutz Baden-Württemberg 2015)

According to Wagner (P. Wagner, Fuel Cell Division Next Energy, pers. comm. 22 July 2015), process losses in the region of 5% to 20% can be expected for automated, industrial-scale coating of membrane rolls, while Bernhart et al. (2014, p. 13) propose a catalyst ink scrap rate of 10%. Ehrenberger (S. Ehrenberger, Deutsches Zentrum für Luft- und Raumfahrt, pers. comm. 01 October 2015), assuming future NSTF catalysts are applied using a magnetron sputter system, deems catalyst material losses so low they can be neglected altogether. Fraunhofer IPA (as quoted by Betrieblicher Umweltschutz Baden-Württemberg 2015), on the other hand, suggests the typical overspray losses for different coating methods given in Figure 27, depending on the specific method used as well as the workpiece structure.

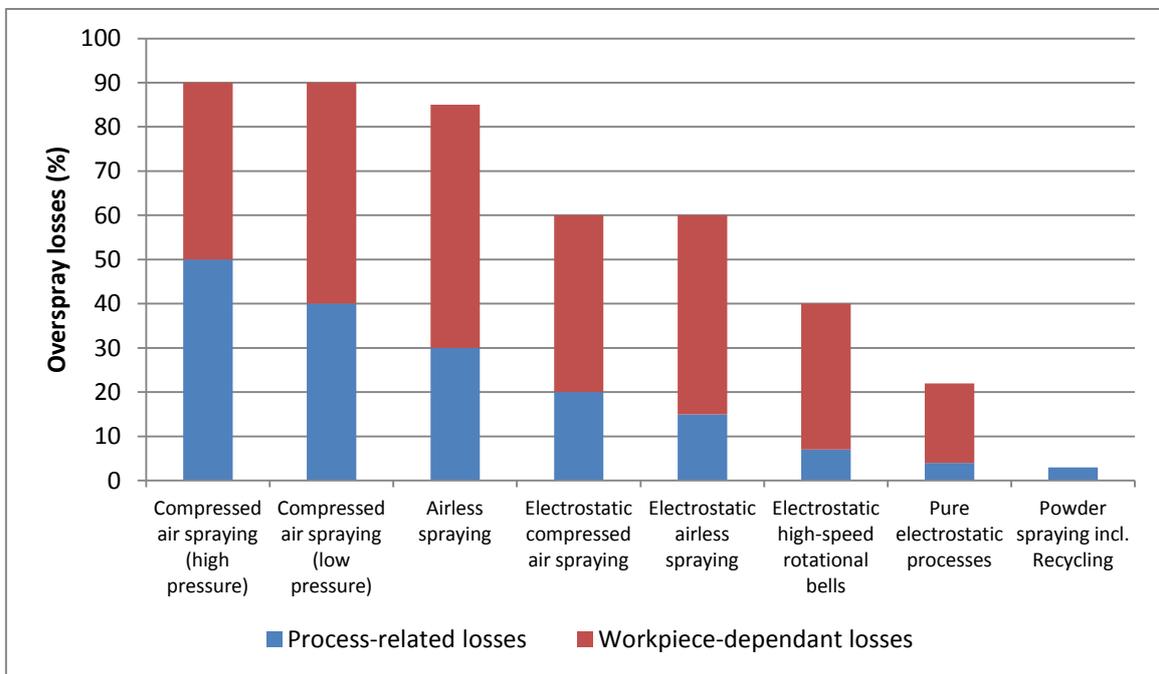


Figure 27: Typical overspray losses of various coating methods (adapted from Fraunhofer IPA, in Betrieblicher Umweltschutz Baden-Württemberg 2015)

It must also be noted that overspray occurring in electrostatic coating methods, which generally use a dry powder instead of an ink or emulsion, can be recycled immediately if collection mechanisms are in place, as the powder does not change its characteristics during the coating process (Kaz 2008, p. 30).

Based on the estimates given above and an application of the concept of ‘learning curves’ (cf. Yelle 1979; Stump 1987), which suggests that production processes become more efficient as experience with a process increases, the MFA assumes that losses of the catalyst ink’s solid content in the coating process will decrease over time, as illustrated in Figure 28. Note that this figure may appear rather optimistic when considering the estimates given in Figure 27, however, it only considers material that is no longer available for use, while any amounts that can presumably be collected and recycled internally have been incorporated.

In addition, it is assumed that the solid content is made up of 28 wt.% Nafion (dry) and 72 wt.% Pt/C (Simons & Bauer 2015, p. 5). These substances, in turn, are assumed to consist of 57.4 wt.% tetrafluorethylene (TFE) and 42.6 wt.% perfluoroalkyl sulfonyl fluoride (PSF) (ibid.) and the Pt/C of 19.8 wt.% platinum and 80.2 wt.% carbon, as given by Wilson and Gottesfeld (1992, p. 3). Regrettably, a more recent publication citing the latter details could not be obtained. In addition, solvents, water, additives and any other possible constituents have been neglected. However, since overspray losses usually refer to the loss of solid contents, this should not affect the conclusions drawn.

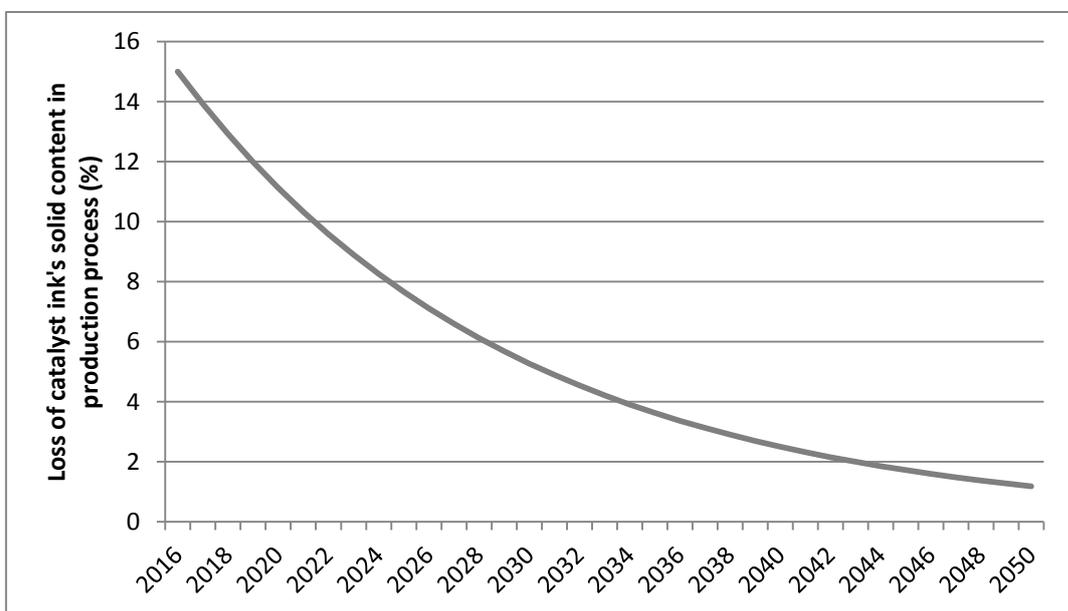


Figure 28: Development of catalyst ink loss in coating process (own source)

Since alternative application methods as well as the production of gas diffusion electrode-type MEA<sup>26</sup> are likely to involve different levels of material loss, these will be neglected for the purpose of the MFA. In addition, it is assumed that any material losses or defects arising during the assembly of the fuel cell stack, manufacturing of FCV and associated transport processes will be recycled internally. While this bears the potential for significantly underestimating both primary platinum requirements and the contribution of fuel cell recycling to meeting the industry's platinum needs, due to a lack of available data these additional processes cannot be incorporated into the MFA.

Finally, with regards to the FCV use phase, according to Kromer et al. (2009, p. 8283) and Dyck (A. Dyck, Fuel Cell Division Next Energy, pers. comm. 22 April 2015) material leakage in the form of platinum leaving the FCV (as is the case with exhaust gas catalysts, for example) is below 1%. For the purpose of the MFA, use phase losses of 0.68% of the contained platinum are thus assumed, which represents the (rounded) mean value between the 0.35% estimated by Kromer et al. and the 1% estimated by Dyck.

### 6.2.3 Recycling scenarios

As evident from Chapter 5, exact numbers for platinum losses arising from deficits in the respective steps of the recycling chain cannot be given before real-world experience with a recycling system for fuel cells is gained. In order to capture a range of possible recycling chain developments, the MFA will thus model two recycling scenarios with diverging assumptions, as listed in Table 9.

The first of these scenarios will be a baseline scenario providing a snapshot of the current state of platinum recovery from exhaust gas catalysts. This scenario is based on Hagelüken et al. (2005a) but adapted to account for factual information concerning fuel cell recycling efficiency that is already available.

The second scenario, the pro-recycling scenario, can be considered a more optimistic case based on the idea that due to both political measures and industry support effective policies are in place that ensure a high collection rate as well as limited platinum losses during the dismantling, pre-processing and material recovery stages. Thus, for each step

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<sup>26</sup> in which catalyst particles are applied to the gas diffusion layer rather than the membrane, cf. Chapter 2.2

of the recycling chain, this scenario assumes the most efficient values of the ranges portrayed in Chapter 5.

Table 9: Parameters of the two recycling scenarios (own source)

Parameter	Baseline Scenario	Recycling Scenario
Share of EoL vehicles collected by recycling facilities	33%	85%
Share of fuel cell stack recovered in dismantling step	96%	99%
Share of platinum-containing components recovered in pre-processing step	96%	98%
Share of platinum recovered in material recovery step	95%	98%
Total efficiency	28.9%	80.8%

As evident from Table 9, the improvements within the recycling chain assumed in the pro-recycling scenario apply mostly to the efficiency of collection. As elaborated in Chapter 5.1, it is rather likely that not only will ELV collection systems improve considerably, but more importantly, FCV will be of use only in those countries, where a suitable hydrogen infrastructure is in place, making the issue of illegal ELV exports largely irrelevant. The quantities of platinum gathered through recycling are assumed to be available for FCV production in the subsequent year.

### 6.3 Limitations

It must be emphasized that despite all due care being exercised in preparing the material flow analysis and the corresponding calculations, certain limitations persist that may question the results of this thesis.

First of all, the analysis rests on the market penetration ratios suggested by EC (2008, p. 17). As mentioned previously, such projections must not be misinterpreted as precise predictions of future developments. Rather, the diffusion of fuel cell vehicles will be governed by a range of influencing factors that cannot be foreseen at this point in time, such as political support, consumer wants, manufacturer choices, private investment, market trends and technological progress. It is therefore possible that the diffusion of FCV in Europe will follow paths entirely different to those assumed in this thesis.

Similar objections apply to the two recycling scenarios. As explained in Chapter 5, material losses arising from deficits in the respective steps of the recycling chain are likely to be considerably lower with regards to fuel cells from FCV than is the case for exhaust gas catalysts from ICE vehicles. The baseline scenario is therefore likely to significantly overestimate platinum losses in the recycling chain. At the same time, the

recycling scenario may be considered overly optimistic with regards to the efficiency of collection, dismantling, pre-processing and material recovery and is thus likely to underestimate platinum losses in the recycling chain. As mentioned previously, projections must be understood as providing a range of possible outcomes; the actual outcome of a real-world recycling chain for fuel cells is thus likely to lie somewhere in between the two scenarios outlined as part of this thesis. In addition, the efficiencies of material recovery adopted as part of the MFA, have only been reported in literature but not been re-confirmed in practice.

As stated previously, the platinum loading per kW achieved at a laboratory stage represents the best-available technology, but must not be confused with the technology available on the road. It is hence possible that the platinum load targets set by the US DoE are under real-world conditions not achieved on time, if ever. Furthermore, it is assumed that certain processes, such as the application of the catalyst layer but also the required platinum content, are going to improve continuously. However, this does not generally apply to the manufacturing of vehicles, which are usually produced in series, meaning that improvement will rather take place in steps.

Also, even if the assumed rate of improvement is accurate, the assumed initial values for process losses may significantly under- or overestimate the actual losses. At the same time, the catalyst ink composition used to calculate production losses stems from 1992, and may thus well be outdated, although this does not affect the resulting platinum demand. Additionally, the MFA incorporates efficiency gains for the production processes, but not for the recovery process.

Finally, the MFA only considers platinum used as the catalyst material of PEMFC in fuel cell drivetrains, while any PGM alloys (e.g. ruthenium) within the fuel cell as well as other components of the fuel cell vehicle that may contain additional quantities of platinum and other PGM have been neglected.

While the results' applicability would obviously benefit from addressing the limitations listed above, due to the given time and space constraints as well as issues concerning data availability this cannot be accomplished as part of this thesis.

## 6.4 Results

The Material Flow Analysis prepared in STAN yields the diagram given in Appendix 5 and four separate data sets for the four market penetration/recycling scenario combinations, each modelling the flows of goods and of the substance platinum separately. Note that in Appendix 5, flows containing platinum are depicted in red. The use phase's sub-system, pictured in blue, is shown in Appendix 6.

Based on the assumptions described in Chapter 6.2, the material flow analysis delivers the following results with regards to the development of platinum demand arising from a market diffusion of fuel cell vehicles:

As shown in Figure 29 and Figure 30, annual platinum requirements for the production of FCV alone could rise to a maximum of 23.42 t in 2047 in the high policy support scenario (1) or 39.66 t in 2050 in the moderate policy support scenario (2).

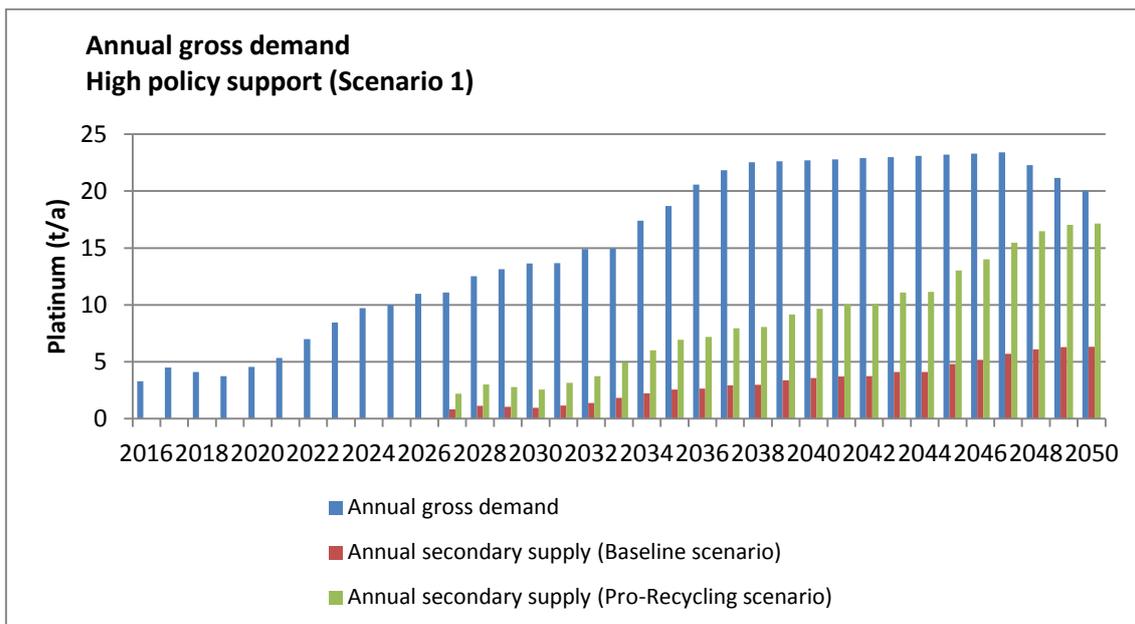


Figure 29: Scenario 1 - Annual gross demand for platinum and contribution of secondary supply (own source)

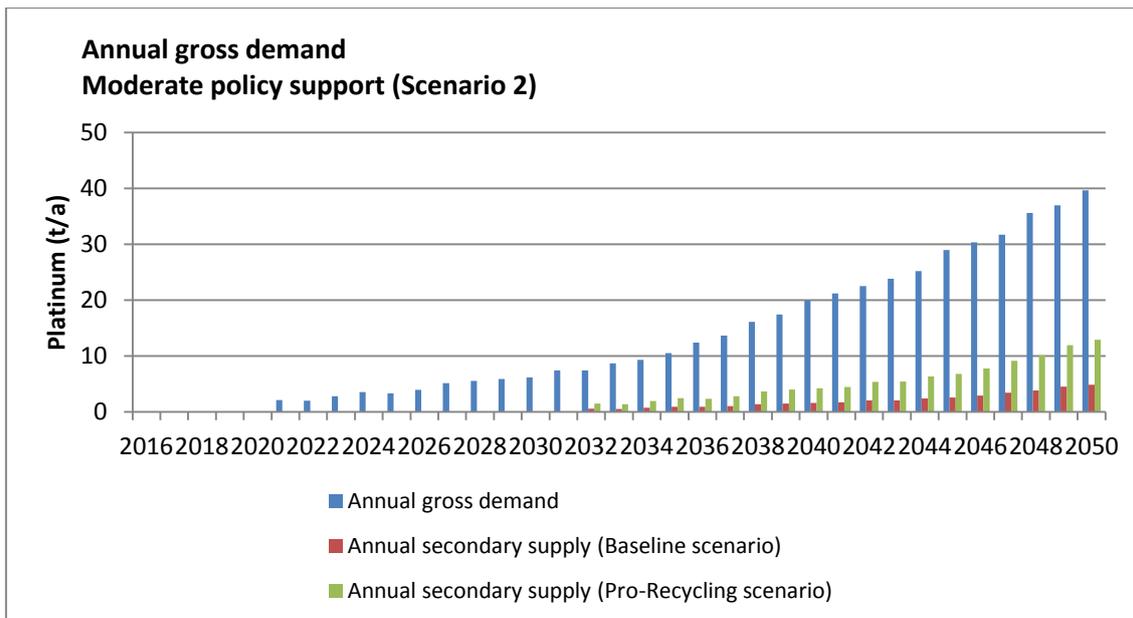


Figure 30: Scenario 2 - Annual gross demand for platinum and contribution of secondary supply (own source)

These results may appear somewhat astonishing at first, as in either scenario the platinum demand does not increase continuously, but rather rises in leaps. However, this observation can be explained by the continuously decreasing platinum load and production losses assumed as part of the MFA. Thus, while the rising numbers of FCV produced drive platinum requirements up, these factors have an opposite effect and therefore cause the platinum demand to actually fall in years with only small increases in the number of FCV. Note that Scenario (1) also assumes a state of market saturation and thus declining numbers of newly produced FCV from 2047 onwards.

Furthermore, it is conspicuous that the maximum gross platinum requirements documented in Scenario (2) exceed those of Scenario (1). This is due to the fact that the final market penetration rates in 2050 do not deviate too strongly between the two scenarios (40% in Scenario 1 and 36% in Scenario 2, respectively), but in Scenario (2), this high market penetration rate is achieved in a shorter time scale (30 years rather than 35 years). Thus the numbers of newly produced FCV increase faster, leading to a steep increase in platinum demand. Considering the cumulative platinum demand illustrated in Figure 31, it becomes apparent that the overall demand arising from Scenario (1) certainly exceeds that of Scenario (2) (537.06 t in Scenario (1) and 459.24 t in Scenario (2), respectively).

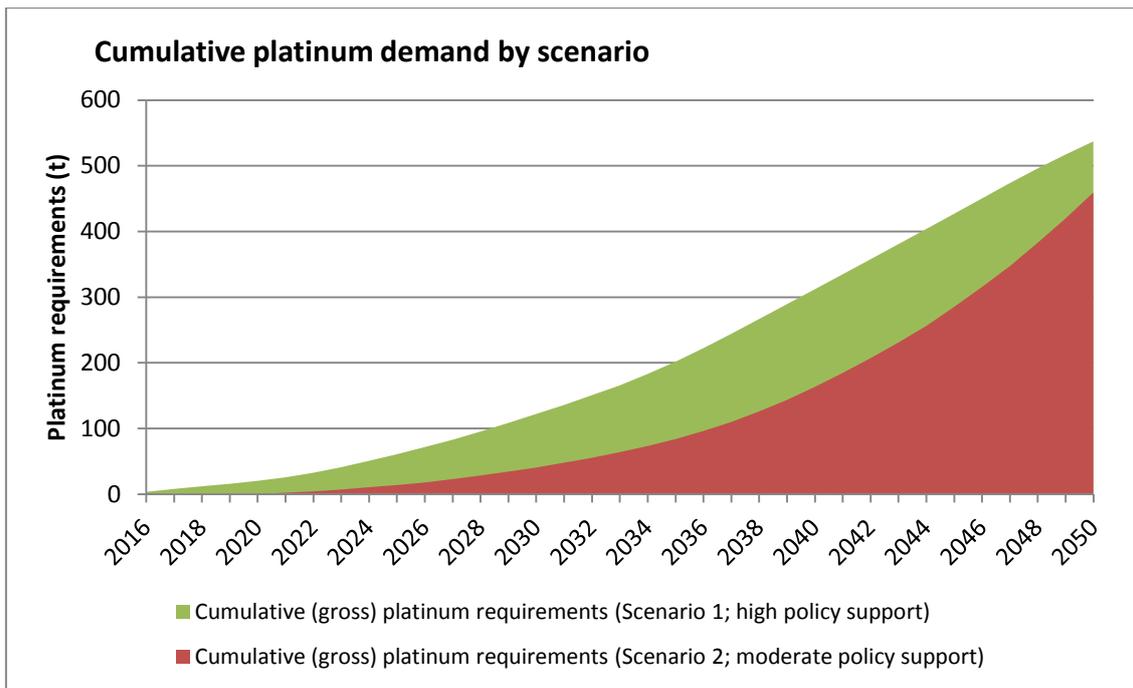


Figure 31: Cumulative platinum demand, not including contribution of secondary supply (own source)

Both the annual platinum requirements and the cumulative platinum requirements shown above indicate that the diffusion of FCV according to either scenario will have a significant impact on global platinum demand. As mentioned in Chapter 4.3, it is impossible to establish at this point in time to what extent any platinum demand from the fuel cell industry will be additional to or replace the demand from other areas of application, such as that of automotive exhaust gas catalysts. The results of Figure 31 also demonstrate that the diffusion of FCV will not cause the depletion of platinum resources, as the calculated 537.06 t and 459.24 t cumulative platinum requirements are far below the currently estimated reserves of 66,000 t of platinum group metals, even though any contribution from the recycling of FCV is not yet included in this extrapolation. At the same time, it must be emphasised that these figures consider only nine European lead markets. A similar level of market penetration in other parts of the world could therefore increase the required quantities of platinum dramatically.

In contrast, a comparison of the maximum annual gross demand established by the two scenarios with the current global supply shows that the future platinum requirements for European FCV production only could place a significant strain on the global platinum market. With 23.42 t and 39.66 t, respectively, the European FCV industry alone would require a maximum of 14.5% and 24.5% of the 161.74 t produced in 2014 (cf. Chapter 4.2). This could not only lead to significant price increases, but also raises the

possibility of supply shortages as well as the dependency of a number of industries on a critical resource and emphasises the importance of recycling.

With regards to the role of recycling in meeting the platinum requirements of a growing FCV fleet, Figure 29 and Figure 30 show a significant recycling potential, the exploitation of which could greatly reduce this industry's dependence on the volatile platinum market. As demonstrated in Figure 29, the secondary supply of platinum could eventually cover up to 85.6% (17.14 t) of the platinum requirements in 2050 assuming the optimistic Pro-Recycling scenario's efficiencies. While the Baseline scenario falls short of such values, even under these rather conservative assumptions, recycling could contribute up to 31.5% (6.32 t) of annual gross demand in 2050, thus underlining the need for research and investment in fuel cell recycling.

In Figure 30, however, the benefits of recycling are much less obvious, as secondary supply contributes a maximum of 32.6 % (26.74 t) in the Pro-Recycling scenario in 2050 and 12.3% (4.86 t) in the Baseline scenario in 2050, respectively. This is due to the fact that as mentioned previously, the FCV fleet is built up much faster in Scenario (2) than in Scenario (1), which already assumes a stage of market saturation by 2050. With an assumed lifetime of 10 years, the first (relatively few) end-of-life FCV will only be recycled in 2031 and thus demand continues to outpace secondary supply. The true benefits of the high recycling efficiencies assumed in the Pro-Recycling case may only become apparent in years later than 2050, when higher volumes of end-of-life FCV arise and the rise in production output may decline.

Nonetheless, in both Scenario (1) and (2) the discrepancy between the two recycling scenarios becomes apparent. This is underlined in Figure 32 and Figure 33, which plot the recycling potential (in terms of platinum content available in end-of-life vehicles) of Scenario (1) and (2) against the realised quantities of secondary supply under the assumptions of the Baseline and Pro-Recycling cases. As the results show, with a maximum of 4.76 t and 4.48 t, respectively, significant quantities of platinum are lost every year even in the Pro-Recycling scenario. These amounts are, however, far below the maximum annual losses of 15.64 t (Scenario 1) and 13.72 t (Scenario 2), respectively, that fail to be recovered from end-of-life FCV under the Baseline scenario.

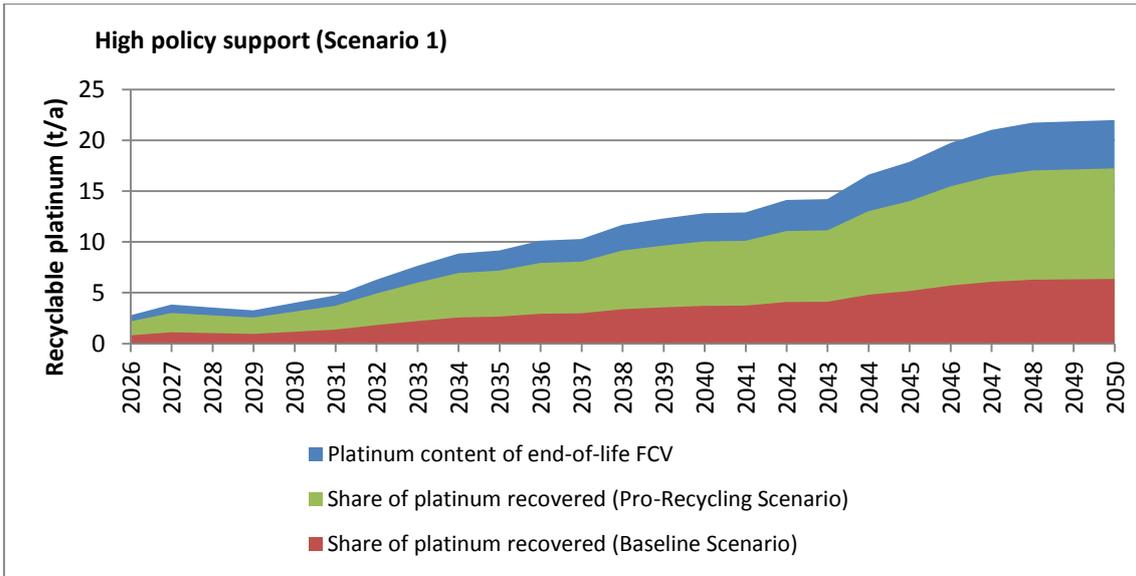


Figure 32: Scenario 1 - Recycling potential and share of Pt recovered (own source)

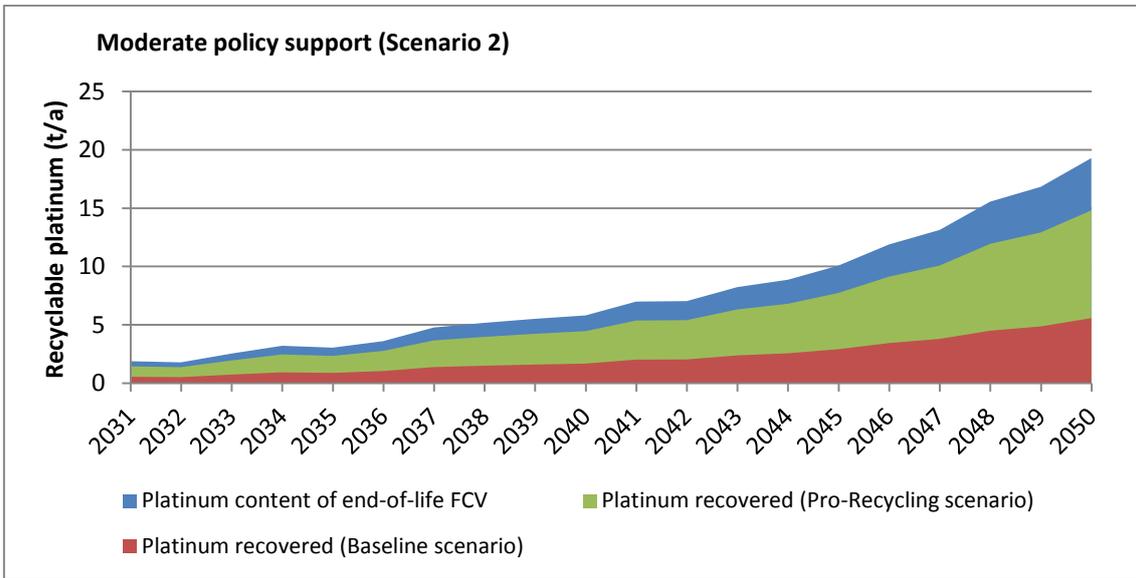


Figure 33: Scenario 2 - Recycling potential and share of Pt recovered (own source)

Considering the entire time span from 2016 to 2050, Table 10 lists the cumulative platinum contents of end-of-life FCV as well as the shares recovered under the four scenario combinations. While the platinum losses are substantial even in the Pro-Recycling scenarios, the losses documented as part of the Baseline scenario are excessively high.

Table 10: Recycling potential from end-of-life FCV and amount of Pt recovered per scenario (own source)

	Scenario 1 (high policy support)	Scenario 2 (moderate policy support)
Cumulative platinum content of end-of-life FCV	293.51 t	155.23 t
Cumulative amount of platinum recovered (Baseline scenario)	84.8 t (208.71 t lost)	44.85 t (110.38 t lost)
Cumulative amount of platinum recovered (Pro-Recycling scenario)	230.02 t (63.49 t lost)	119.17 t (36.06 t lost)

In general, the results of the MFA point out the high uncertainty involved in projecting metal demand and flows arising from any application in the future. It is neither possible to give precise predictions of the future platinum requirements arising from the diffusion of FCV in Europe, nor of the quantities of secondary platinum supply becoming available through recycling, which is demonstrated by the wide range of estimates given in the figures above.

In addition, it is difficult to compare the results of this MFA to other estimates and extrapolations of platinum demand resulting from the diffusion of FCV, as all are based on diverging assumptions, time scales and geographical locations (cf. Appendix 1). Nonetheless, when comparing the results of this MFA to the estimates given by Bernhart et al. (2014), which can be viewed as applying the most similar assumptions, it becomes apparent that the results do not differ drastically. Bernhart et al. (2014) assume a global FCV stock of 300,000 to 5 million vehicles in 2020 and a platinum load of 10 to 16 g per vehicle, which results in a cumulative platinum demand of 4.8 t to 50 t. This MFA, on the other hand, is based on a European FCV stock of 0 to 1.5 million vehicles and a platinum load of 10 to 14.1 g per vehicle, which results in platinum requirements of 0 to 20.14 t. Other publications, however, arrive at much higher estimations of the platinum demand resulting from a diffusion of fuel cell vehicles (cf. Appendix 1).

## 7 Discussion

In spite of the significant differences between the four market penetration/recycling scenario combinations, the results supplied in the previous chapter indicate that while exact quantities are difficult to predict, the adoption of fuel cell vehicles in Europe will have a significant impact on global platinum demand.

It is common for resource availability to be assessed simply in terms of matching geological deposits against the required quantities of the material in question. As concluded in Chapter 6.4, the increased demand for platinum projected by this thesis can likely be met by a raise in primary production capacity from a geological perspective, since the known PGM reserves and resources are large enough to cover demand for the foreseeable future. However, while platinum depletion may not be an issue in the short to medium term, with a view to the Brundtland Commission's<sup>27</sup> definition of sustainable development that grants future generations equal rights to resource access, this non-renewable resource base must not be considered inventory for the current generation to consume.

Even if the currently known reserves indicate that sufficient quantities of platinum are available (for a certain scenario), problematic levels of resource depletion and scarcity cannot be ruled out in the long term. In addition, as stressed previously, not only does this thesis restrict itself to the future platinum demand resulting from a single application in a single world region and within a well-defined time span, it is further not known to what extent this demand will be additional to or replace demand from other areas of application. Any advice on the long-term availability of sufficient quantities of platinum must therefore be considered premature.

At the same time, not only is the projection of future resource demand afflicted with uncertainties, but future production capacities are similarly difficult to predict. While according to USGS (2011, p. 121) those PGM reserves whose extraction is economical at current prices and technology levels amount to at least 66,000 t, it is unclear which of the cited deposits are actually in production at the time of writing. Due to the long lead times required by mining businesses to pursue exploration activities, go through the

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<sup>27</sup> „Sustainable development is development that meets the needs of the present without compromising the ability of future generations to meet their own needs.” (WCED 1987, p. 41)

respective legal and political procedures and set up the required infrastructure and equipment, responses by the mining industry to increases in platinum demand have been sluggish in the past.

As mentioned previously, this could lead to supply shortages, price surges and inventory build-up. Adding to this the highly concentrated supply base in countries with a relatively poor political stability and governance, as well as the labour issues and infrastructure insecurities experienced in the past, an increase in the vulnerability of platinum supply is inevitable.

On a similar note, the primary production of platinum involves not only excessive carbon emissions, but also other environmental burdens, such as excessive energy consumption or air and water pollution (cf. Chapter 4.5). This could impact on the diffusion of FCV in several ways.

First of all, it remains uncertain whether fuel cell vehicles may actually be regarded a 'greener' option, let alone a 'zero-emission vehicle', when the environmental aspects of both raw material production and hydrogen production are factored in (cf. Chapter 3). Although a number of life cycle assessments (LCA) have been produced in recent years (cf. Pehnt 2000; Pehnt 2003; Gerboni et al. 2008; Simons & Bauer 2015), these have come to ambiguous conclusions and again rely on a number of assumptions that are difficult to compare. However, conclusions drawn from such publications may not only impact on public perception, but also on political support for FCV and hydrogen infrastructure and can thus have significant impacts on the market potential of FCV.

In addition, possible stricter environmental regulations in the prime supplier countries of platinum could have serious impacts on resource availability in the future. Imposition of stricter regulations could both make mining more expensive, rendering some deposits uneconomical, and prohibit the exploitation of others entirely. Such developments could, in turn, restrict mining businesses' ability to supply the demanded quantities of platinum and thus increase the vulnerability of the platinum market once more.

The diffusion of fuel cell vehicles in Europe may thus significantly raise dependence on a critical resource that is not available for production in the EU itself – not only for fuel cell and car manufacturing, but also for the various other platinum-dependant industries. Given that these include vital applications such as chemical and petrochemical industries as well as pharmaceuticals, an increase in the criticality of platinum cannot be desirable from a European perspective.

In order to ease such dependencies, stabilise the platinum supply base and minimise risks, costs and environmental impacts, the co-development of recycling procedures for recovering the platinum from end-of-life FCV may hence be considered a prerequisite.

As the results of Chapter 6.4 have demonstrated, the quantitative assessment of future platinum flows from FCV recycling is afflicted with uncertainties and may be considered highly speculative. Nonetheless, the results of this thesis' MFA imply that well-devised recycling systems could contribute quantities of secondary platinum in an order of magnitude that could ease the dependencies and possible supply shortages described above considerably. As analysed in Chapter 6.4, up to 85.6% of the platinum required for FCV production could be obtained from recycling of end-of-life FCV in 2050 (Scenario 1; Pro-Recycling), while the other three scenario combinations deliver much more conservative results of 31.5% (Scenario 1; Baseline), 32.6% (Scenario 2; Pro-Recycling) and 12.3% (Scenario 2; Baseline), respectively. Although the latter three of these estimates appear much less promising, even the lowest of these values still represents 4.86 t of platinum (worth € 179,820,000 at today's market price<sup>28</sup>) that would otherwise have to be produced from primary sources.

Their presumed high recycling potential is one of the reasons that a number of studies portray FCV and the impacts of their market diffusion on platinum demand in a rather positive way. However, when comparing the results of both the qualitative (cf. Chapter 5) and quantitative assessment (cf. Chapter 6.4) of this thesis with previous publications, one may conclude that several studies tend to overestimate the platinum supply achievable from FCV recycling. For example, Råde (2001), Tiax LLC (2003) and Saurat & Bringezu (2009) assume recycling rates of 90%, 95% and 75%, respectively. This thesis, on the other hand, proposes much more conservative recycling rates of 28.9% and 80.8%, respectively.

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<sup>28</sup> approx. € 37 per gram; as quoted by Johnson Matthey (2015a) on May 11, 2015.

As analysed in Chapter 5, highly efficient recycling chains as the ones cited above may be conceivable in theory in the future, but are incommensurable to the quotas realised for vehicles today and exceed those achieved in the majority of platinum applications by far. Such assumptions, however, also impact on the platinum demand arising from a diffusion of FCV as suggested in these studies, and hence involve the potential for significantly underestimating the net platinum requirements. Considering the fact that 98% is the highest recovery rate reported by any of the patents analysed in Chapter 5.3 for the final step of material recovery, all the preceding stages in the recycling chain would need to collect the respective material handled with 99% efficiency in order to achieve an overall recycling rate of 95% (as suggested by Tiax LLC 2003). As shown in Chapter 5, the recycling of platinum from fuel cell vehicles is still in its very early stages, largely due to the fact that significant volumes of end-of-life fuel cells do not exist yet. Any values stated for possible recycling chain efficiencies, fractions recovered and amounts lost must therefore be understood as an indication only and the underlying assumptions considered with due circumspection.

It appears as though stakeholders of the precious metal industry that are already involved in the recycling of platinum-containing applications, including amongst others Umicore & Co KG and Heraeus Precious Metals GmbH, have acknowledged both the opportunities and challenges arising from a further diffusion of fuel cells and FCV in particular. A number of procedures for recovering platinum from end-of-life fuel cells with high efficiencies and under the premise of avoiding problematic HF emissions exist, yet it must be stressed that none of these have been implemented at an industrial scale. This fact raises a number of questions, concerning not only the commercial applicability of such procedures, but also with regards to environmental impacts and profitability when compared to established pyrometallurgical recovery processes.

Nonetheless, Umicore, for example, claim that their recycling plant in Hoboken, Belgium, is already set up to handle the volumes of end-of-life fuel cells likely to occur in the medium term, stating that “... [the recovery] *process will be upscaled into a production plant when volumes from the upcoming mass application of fuel cells especially from the automotive sector are expected*” (Zuber et al. 2004a, p. 223). While it is hardly possible to verify such statements, it is likely that recycling businesses in general will hesitate to invest in dedicated plants and technologies as long as it remains uncertain which type of powertrain technology will prevail. The great uncertainty that persists not

only regarding the political endorsement of specific powertrain technologies, but also concerning the future direction of technological advances in the FCV industry as well as of the current fuel cells' material compositions pose further barriers for investment by recycling businesses.

However, what appears even more problematic is the fact that even less is certain about the preceding stages in the recycling chain, which seem to have received much less attention than the material recovery stage by producers, recycling businesses, researchers and other stakeholders. This is especially concerning when considering, as shown in Chapter 5, that deficits in the preceding steps (i.e. collection, dismantling and pre-processing) account for a major share of PGM losses in the recycling chain of other platinum-rich consumer goods and could potentially do so for FCV as well.

Deficits in the collection system for end-of-life conventional vehicles in the EU have been made public and counteracting strategies discussed in the research community for a number of years, yet collection rates have not improved notably (cf. Hagelüken 2005a; Fergusson 2007; Lucas et al. 2010; Wilts et al. 2011; Wäger et al. 2012; Wilts & Bleischwitz 2012; Heiskanen et al. 2013). This emphasises not only the complexity of this issue, but also demonstrates that the effects of any measures taken will likely not become visible for a number of years.

Recycling businesses, such as Axion Recycling and Umicore, have acknowledged a need for action and are researching options for closing the loop in fuel cell recycling by means of, for example, a producer-led take-back or lease system (cf. Chapter 5.1). As mentioned previously, the high number of actors involved in the life cycle of consumer goods represents one of the major reasons for inefficient interfaces within the recycling chain, thus the aforementioned strategies do well in trying to address this issue.

However, Wilts et al. (2011) state “*As there is and will be no single person or institution who owns processes and produces throughout the whole cycle of extraction, production, consumption, recycling, and disposal, the question is how responsibility [...] can be attributed to the actors along the chain in a way that favours sustainable management of the substances involved*” (p. 905).

This indicates what the authors title a ‘responsibility gap’, which means that even concepts such as Extended Producer Responsibility<sup>29</sup> (EPR) for vehicles, already implemented within the EU and one of the cornerstones of the European waste legislation, fail to prevent open systems and a loss of secondary resources (ibid.).

Several aspects that contribute to this issue can be identified. First of all, the high mass-based recycling targets prescribed within the EU (cf. Chapter 5.1) as well as the associated high environmental compliance and labour costs have created incentives not only for legal used vehicle exports, but also for illegal shipments of waste end-of-life vehicles. Naturally, producers will base their choice of disposal option on economic criteria. Recycling targets that refer to the total weight of the vehicle thus give no incentive for the recycling of platinum (or other critical metals), which makes up only a small percentage of the overall weight in both conventional ICE vehicles (here present mainly in the catalytic converter) and fuel cell vehicles.

Additionally, secondary raw material markets have failed because “*an asymmetrical distribution of information between recyclers and industry purchasing secondary raw materials has been impairing efficient agreements*” (Wilts & Bleischwitz 2012, p. 2). Even though in the case of platinum, recycling should be economical in principle, transaction costs<sup>30</sup> caused by the spatial and temporal distribution of secondary sources, the unpredictability of volumes and a knowledge gap concerning the contained materials, may lead to a preference for primary platinum (Wilts et al. 2010, pp. 32-33).

In addition, both material and waste streams increasingly take place on a global scale, rather than being restricted to specific countries or world regions. Environmental regulations at a national or EU level are therefore relatively limited in their effectiveness, or as Wilts et al. (2011) put it: “*In order to actually set effective incentives for resource conservation and recycling of raw materials by EPR, the manufacturers’ responsibility cannot be allowed to end at the border*” (p. 906). Practice-oriented environmental regulations will therefore require new forms of governance that also include stakeholders in the countries of raw material extraction and in the destination countries of used vehicle

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<sup>29</sup> „An environmental policy approach in which a producer’s responsibility for a product is extended to the post-consumer stage of a product’s life cycle” (OECD 2015)

<sup>30</sup> A concept of economic theory; i.e. costs associated with the exchange of goods and services incurred by market participants for overcoming market imperfections, including, for example, costs for information, communication, legal fees etc. (Johnson 2005).

exports (*ibid.*). Furthermore, governments lack the knowledge on material flows required to effectively correct and regulate the consistently failing markets (Wilts & Bleischwitz 2012, p. 2).

In order to counteract the abovementioned limitations of the existing ELV regulations, an international covenant of the stakeholders involved within a vehicle's value chain has been proposed. This voluntary but binding contract is to be based on set recycling targets for specific materials, recycling facilities according to European environmental standards in the destination countries, sanctions and enhanced monitoring and reporting. Despite the benefits of both raw material access and partnership deriving from such a contract, the substantial costs of both negotiating and maintaining this kind of voluntary agreement must not be underestimated (*cf.* Wilts et al. 2010; Wilts et al. 2011; Wäger et al. 2012; Wilts & Bleischwitz 2012). In addition, it must be noted that similar voluntary environmental self-regulations have failed in the past, which signifies the need for political endorsement of and participation in this matter. While the European Commission (2014, p. 10) already acknowledges the need for international cooperation to secure access to critical raw materials, this target should hence be extended to secondary resources and actively pursued.

While this could lead to a significantly improved collection quota of end-of-life vehicles of any powertrain technology, much less advice appears to be available on remedying deficits in the dismantling and pre-processing stages of the recycling chain. Integrated research efforts that include all stages and actors of a potential fuel cell recycling chain and are aimed at devising practice-oriented strategies would therefore be highly beneficial in order to achieve a closed European system and minimise losses of a critical resource. While the first projects addressing such aspects have been announced recently (*cf.* European Commission 2015a), many open questions remain to date.

As mentioned in Chapter 5, similar issues persist with regards to the end-of-life treatment of lithium-ion batteries from BEV. However, although by far not all questions have been resolved satisfactorily, at least research projects have recently begun looking at aspects such as design of collection systems, cost-benefit analyses of manual and automated dismantling procedures, health and safety aspects, infrastructure and equipment requirements, investment needs and legal classification of components, all of which will prove crucial knowledge once significant volumes of end-of-life lithium-ion batteries arise (*cf.* Kwade & Bärwaldt 2012). Although these developments have only

taken place on a laboratory scale so far (if at all), several universities and research institutions are also engaging in setting up actual automated disassembly lines, which will likely prove necessary for handling the larger volume of lithium-based traction batteries that will presumably arise in the coming decades (cf. Weyrich & Natkunarajah 2013; Markowski et al. 2014; TU Braunschweig 2015).

Regrettably, such comprehensive analyses and, above all, practice-oriented evaluations are not (yet) available for FCV. Instead, most publications dealing with this topic have only focused on isolated aspects of fuel cell recycling from a theoretical perspective, with the majority pursuing the final stage of material recovery but neglecting the preceding stages in the recycling chain (cf. Chapter 5).

Another aspect that could not be addressed sufficiently in this thesis due to a lack of data is the potential for internal recycling of production scrap. The MFA conducted as part of this thesis assumes that a maximum of 15% of the catalyst material is lost in the coating process in 2016, with the ratio of process losses improving continuously. This is likely to severely underestimate the level of platinum losses experienced under current production methods, as currently these tend to employ a catalyst *ink* rather than dry powder and thus require entirely different coating methods. This uncertainty is also demonstrated by the range of estimates given in Chapter 6.2.2. In how far such process losses are already being recycled today is beyond the knowledge of this author. However, given that Zuber et al. (2004b, p.17), for example, specifically mention the opportunity for recycling production scrap, while other sources imply material scrap rates of up to 90% (cf. Chapter 6.2.2), this could be an even more important point for intervention in order to prevent platinum losses.

Should the diffusion of FCV actually proceed as projected in either of the two scenarios used in this thesis, the first significant volumes of end-of-life FCV will arise in around ten years time. At the current rate of scientific and technological progress, however, it is likely that the build-up of recycling capacities will lag behind substantially.

## 8 Conclusion and Outlook

As established in this thesis, the platinum requirements arising from a diffusion of fuel cell vehicles in Europe may have significant impacts on the global platinum market. While the exact quantities demanded by this application in the future are difficult to predict, it is unlikely that they will lead to a depletion of primary platinum deposits in the short term. However, at levels of up to a quarter of the global platinum demand in 2014, an aggravation of the already persisting structural and temporal scarcity of platinum supply appears inevitable.

This demonstrates that an unequivocal endorsement of fuel cell vehicles with the objective of reducing GHG emissions from the European transport sector may be uncalled for, as it causes concerns with regards to other aspects of sustainable development. In the long term, this applies above all to problematic levels of resource depletion, but also to other aspects of primary platinum production, such as environmental impacts of mining activities. In the short term, however, increased structural and temporal scarcity of platinum supply could not only raise the EU's dependence on this critical resource and hinder the diffusion of fuel cell vehicles, but also impede the development of other platinum-dependant future-oriented technologies. This must be considered antithetical to the EU's paradigm of sustainable development.

Such concerns could be eased considerably with the co-development and implementation of effective recycling strategies. Ideally, once FCV are well-established, recycling of platinum from end-of-life FCV should cover the basic demand of this application, while primary production would only need to replace inevitable losses as well as cover growth in this sector. However, as shown in this thesis, the recycling of FCV is still in its initial stages of development and many questions remain unsolved. In particular, this applies to the recycling logistics accompanying material recovery, i.e. collection, dismantling and pre-processing of the fuel cell stacks. An international metal covenant, as well as producer-led schemes, have been proposed to tackle deficits in collection, in addition to further practice-oriented research on the entire recycling chain.

While the step of platinum recovery appears to be the most thoroughly analysed, uncertainties remain here as well. Not only is it debatable to what degree the procedures outlined as part of this thesis are suitable for large-scale application, it is further not known whether recycling of platinum will have monetary benefits under the given circum-

stances. Without economic incentives, however, actual future recycling rates could be much lower than proposed in this thesis. A timely specification and analysis of costs would thus be highly useful, especially if such a report were to come to the conclusion that governmental incentive structures are necessary.

In addition, not only is the environmental advantage of fuel cell vehicles debatable when aspects such as resource extraction and fuel production are factored in, the environmental advantage of the recycling technologies presented as part of this thesis is equally uncertain. Further holistic life cycle assessments that cover a fuel cell vehicle's entire life cycle from 'well to wheels', respectively compare the various recycling procedures available with each other and with primary production, are thus recommendable.

As mentioned previously, the potential for internal production scrap recycling was largely ignored in this thesis due to a lack of data. This could represent an important point for promoting resource efficiency, which should be investigated more thoroughly. Furthermore, the achievable recycling rate will depend to a large degree on whether recyclability is considered in the design of fuel cell vehicles. While this aspect was alluded to only briefly in this thesis, design for dismantling or design for recycling, respectively, should be considered in more detail.

In general, platinum may be considered one of the most thoroughly researched elements with regards to material flows in the anthroposphere (cf. Hagelüken et al. 2005a; UNEP 2012). Nonetheless, as this thesis has demonstrated, the spatial and temporal distribution of the platinum contained in consumer goods and in particular, the lack of knowledge thereof, poses a challenge for effective exploitation of these secondary resources. Here, comprehensive material flow analysis could provide an important tool to gather information on timing, location and expected quantities of end-of-life vehicles, in order to lower the transaction costs involved in their recycling.

Finally, it must be emphasised that even under the most ideal circumstances, some dissipative losses of platinum are inevitable, for example in the use phase, but also through human errors and technical limitations in recycling. Neglecting any growth of platinum-dependant applications, simply maintaining the status quo will therefore continue to require additional primary resource input to replace these losses. While recycling thus

cannot resolve the issue of resource depletion entirely, it can contribute to prolonging resource availability considerably, as demonstrated in this thesis.

However, the time span by which the depletion of primary platinum deposits may be postponed depends also on the strategies devised and implemented today, before significant volumes of end-of-life vehicles arise. Uncertainty concerning the market adoption and technological development of fuel cells prevails and prevents recycling businesses from exploring and investing in recycling options. Further research should hence be promoted to resolve the issues listed above and initiate expedient recycling strategies.

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## Appendix 1: Projections of fuel cell vehicle diffusion and resulting platinum demand in literature

Source	Assumptions	Year of projection	Projected platinum demand from FCV
<b>Angerer et al. (2009)</b>	- platinum load of 0.8g/kW - total installed capacity 430 GW	2030	Cumulative demand: 344 metric tons
<b>Arthur D. Little (2001), as cited in Yang (2009)</b>	- platinum load 200g/vehicle - 500,000 FCV at 50kW ( $\approx$ 25 GW installed capacity)	n/a	Cumulative demand: 100 metric tons
<b>Bernhart et al. (2014)</b>	- conservative scenario: platinum load of 16g/ vehicle; 300,000 FCV - optimistic scenario: platinum load of 10g/vehicle; 5 million FCV	2020	Cumulative demand - conservative scenario: 4.8 metric tons - optimistic scenario: 50 metric tons
<b>Buchert et al. (2011)</b>	- platinum load of 10g/vehicle - 4.2 million new FCV registrations	2030	Annual gross demand: 42 metric tons
<b>Carlson et al. (2003)</b>	- platinum load of 30g/ vehicle - 5 million FCV in US, Japan, Western Europe, China, India	2030	Cumulative demand: $\approx$ 150 metric tons
<b>Gordon et al. (2006)</b>	- platinum load of 0.4mg/cm <sup>2</sup> - 500 million FCV at 75kW average power ( $\approx$ 37,500 GW installed capacity) - recycling rate of 45%	n/a	- Cumulative: 15,000 metric tons - Annual net demand: 1,000 metric tons
<b>Rade (2001)</b>	- platinum load of 19g/vehicle - 5,328 million FCV at 50kW ( $\approx$ 266,400 GW installed capacity) - recycling rate of 90%	2100	- Cumulative demand: 101,000 metric tons - Annual net demand: 2,700 metric tons
<b>Saurat &amp; Bringezu (2009)</b>	- platinum load of 16g/vehicle - no. of FCV not disclosed but market penetration ratio represented graphically - recycling rate of 75%	2030	Annual net demand: 84 metric tons
<b>Tiax LLC (2003)</b>	- platinum load of 15g/vehicle - 35 million FCV at 75kW ( $\approx$ 2625 GW installed capacity) - recycling rate of 95%	2050	- Annual net demand: 250 metric tons

Note that all units were converted to metric tons in order to improve comparability. Cumulative demand = total platinum content of FCV fleet in the projected year; annual gross demand = annual platinum demand from new FCV registrations in the projected year; annual net demand = annual platinum demand from new FCV registrations minus supply from FCV recycling in the projected year.

## Appendix 2: Cost estimates of various fuel cell components in literature

Source	Cost Estimates		Assumptions		Technology Level of Year	Cost share
	Component	Cost	Power Output	Production volume		
McKinsey & Company (2010)			85 kW	20.000 units	2015	
	MEA (incl. Catalyst)	€ 6.164				33%
	Catalyst	€ 3.194				17%
	Structure and other components	€ 3.212				17%
	Balance of Plant	€ 9.516				50%
	Total Fuel Cell System	€ 18.892				
McKinsey & Company (2010) (as summarized by Greene & Duleep 2013, p. 14)			85 kW	200.000 units	2020	
		€ 2.465				33%
	Catalyst	€ 1.420				19%
	Structure and other components	€ 1.050				14%
	Balance of Plant	€ 3.960				53%
	Total Fuel Cell System	€ 7.475				
James et al. (2013)			80 kW	1.000 units	2013	
	MEA (incl. Catalyst)	€ 10.592				51%
	Catalyst	€ 2.063				10%
	Structure and other components	€ 2.676				13%
	Balance of Plant	€ 7.276				35%
	Total Fuel Cell System	€ 20.632				
James et al. (2013)			80 kW	10.000 units	2013	
	MEA (incl. Catalyst)	€ 3.224				42%
	Catalyst	€ 1.075				14%
	Structure and other components	€ 761				10%
	Balance of Plant	€ 3.562				46%
	Total Fuel Cell System	€ 7.669				
James et al. (2013)			80 kW	100.000 units	2013	
	MEA (incl. Catalyst)	€ 1.774				36%
	Catalyst	€ 995				20%
	Structure and other components	€ 613				12%
	Balance of Plant	€ 2.429				49%
	Total Fuel Cell System	€ 4.908				

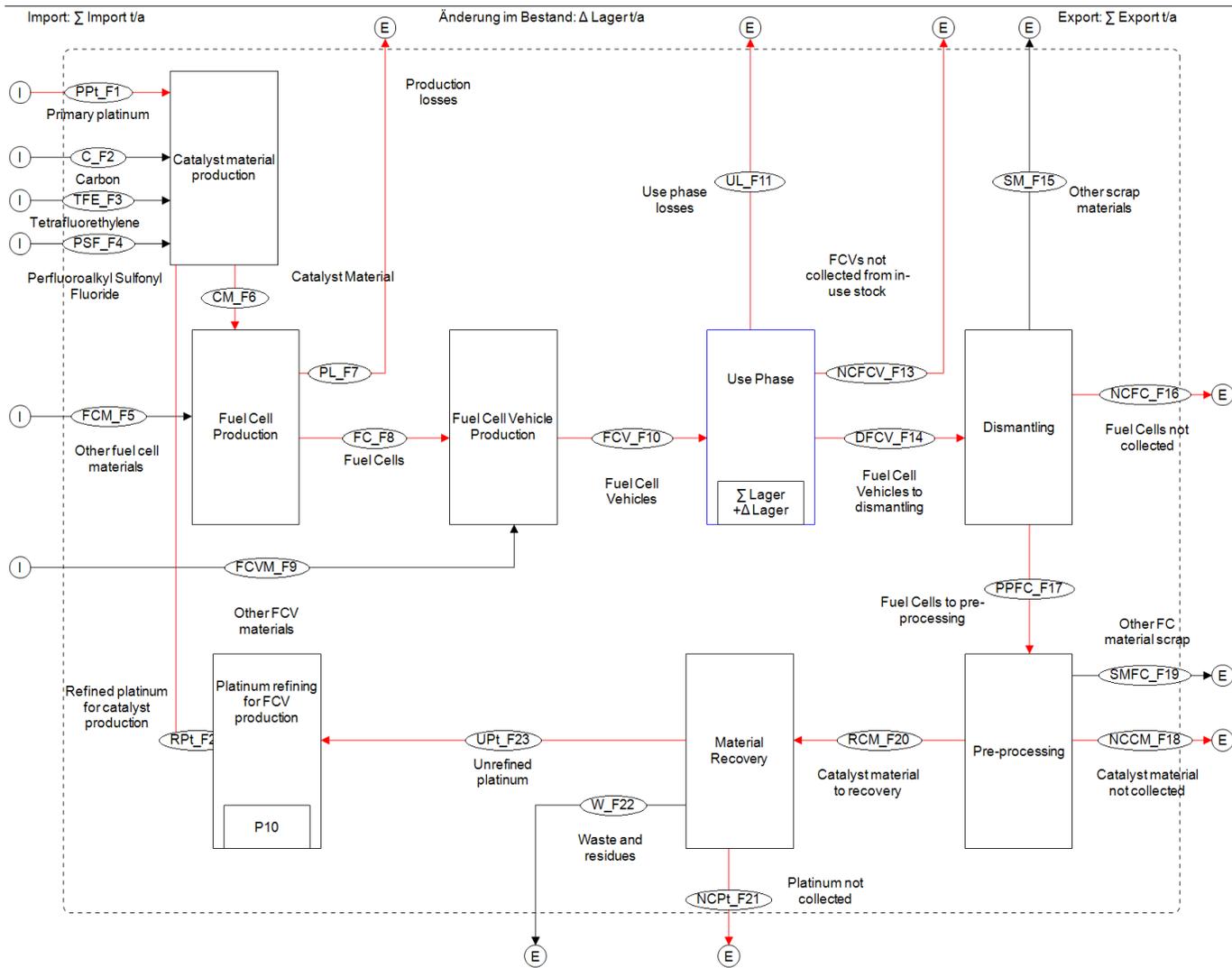
### Appendix 3: Detailed calculation approach

Step	Explanation	Calculation
1	Extrapolation of vehicle fleet in the countries examined	vehicle fleet 2011 * average annual growth rate
2	Annual market share of FCV in absolute numbers	vehicle fleet of year X * market share of year X according to scenario (1) and (2)
3	Number of new FCV per year	number of FCV in year X - number of FCV in year X-1
4	Energy requirements per year (kW)	number of new FCV per year * 80 kW
5	Platinum requirements per year	energy requirements of year X * platinum load of year X
6	Mass concentration of Pt per FCV	platinum requirement in year X / (weight of FCV * number of new FCV in year X)
7	Mass concentration of Pt per fuel cell stack	platinum requirement in year X / (weight of fuel cell stack * number of new FCV in year X)
8	Catalyst material requirements per year	platinum requirement in year X / Platinum content of catalyst ink
9	Catalyst material loss per year	catalyst material requirements of year X * production loss
10	Number of end-of-life FCV	end-of-life FCV of year X = FCV fleet of year X-10
11	Platinum content of end-of-life FCV	end-of-life FCV of year X = platinum requirements of Year X-10 * (1- use phase loss)
12	End-of-life FCV collected per year	end-of-life FCV of year X * collection rate according to scenario (A) and (B)
13	Fuel cell stacks dismantled per year	end-of-life FCV collected in year X * dismantling rate according to scenario (A) and (B)
14	Catalyst material processed per year	fuel cell stacks dismantled in year X * pre-processing rate according to scenario (A) and (B)
15	Platinum recovered per year	catalyst material processed in year X * recovery rate according to scenario (A) and (B)
16	Secondary platinum available for catalyst ink preparation	secondary platinum available for production in year X = platinum recovered in year X-1

## Appendix 4: Assumptions and parameters of Material Flow Analysis

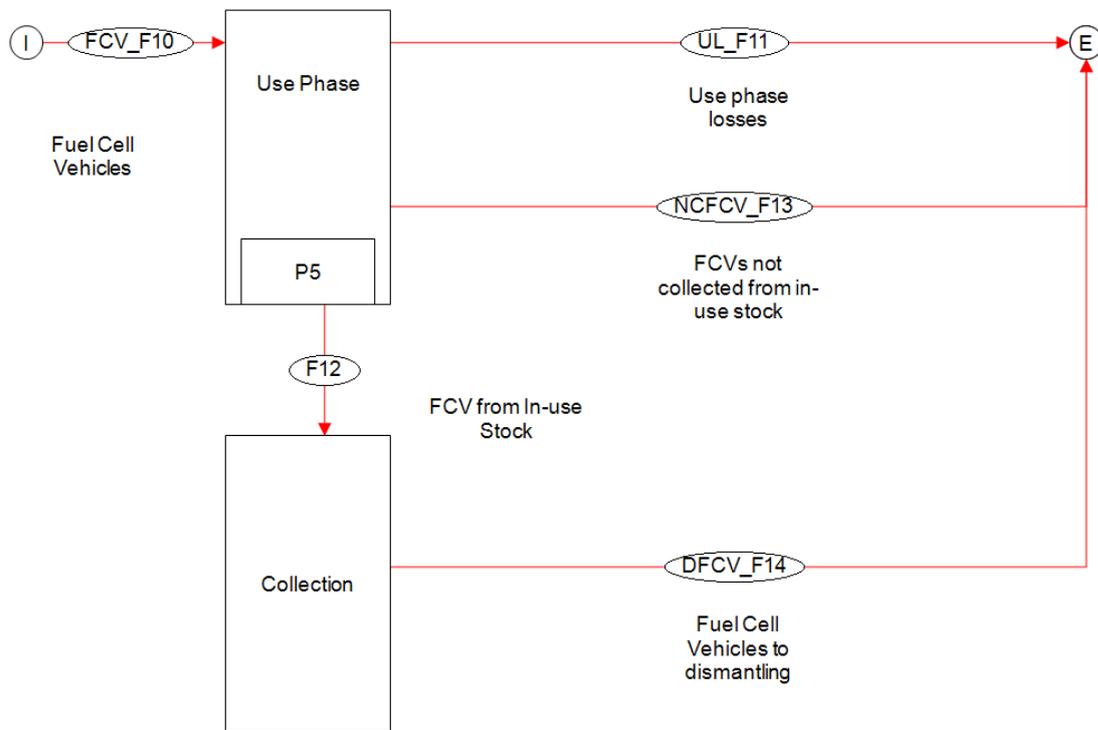
Parameter	System Boundary / Assumption	Source/s
<b>Geographic boundary</b>	Finland, France, Germany, Italy, the Netherlands, Norway, Poland, Spain, the United Kingdom.	European Commission (2008); Greece excepted due to lack of data
<b>Time Scale</b>	2016 to 2050	Own choice based on data availability
<b>Market penetration scenarios</b>	High policy support, modest learning rate; modest policy support, modest learning rate	European Commission (2008)
<b>Type of vehicle</b>	Light-duty passenger vehicle	Own choice
<b>Average annual growth rate of vehicle fleet</b>	0.3%	Mean value of estimates in literature
<b>Vehicle lifetime</b>	10 years	Simons & Bauer (2015)
<b>Average vehicle power</b>	80 kW	US DOE
<b>Average vehicle weight</b>	1447 kg	Simons & Bauer (2015)
<b>Average fuel cell stack weight</b>	40.8 kg	Simons & Bauer (2015)
<b>Platinum load per kW</b>	2020: 0.125 g, 2030: 0.07 g, 2050: 0.07 g	US Drive FCTT (2013), Adamson (2015)
<b>Catalyst ink loss in manufacturing</b>	2020: 15%; 2030: 5.2%; 2050: 1.1%	Own calculation based on literature estimates and average annual improvement rate of ~ 0.8%
<b>Use phase material loss</b>	0.67%	Pers. Comm. (Dyck 2015); Kromer et al. (2009)
<b>Recycling scenarios</b>	Baseline, Pro-Recycling	Own preparation, in parts based on Hagelüken et al. 2005a

## Appendix 5: STAN material flow diagram including flow codes (own source)



Platinum-containing flows depicted in red, use phase subsystem depicted in blue

**Appendix 6: STAN use phase sub-system of the material flow diagram including flow codes (own source)**



Platinum-containing flows depicted in red

### **Selbstständigkeitserklärung**

Hiermit versichere ich, dass ich diese Arbeit selbstständig verfasst und keine anderen als die angegebenen Quellen und Hilfsmittel benutzt habe. Außerdem versichere ich, dass ich die allgemeinen Prinzipien wissenschaftlicher Arbeit und Veröffentlichung, wie sie in den Leitlinien guter wissenschaftlicher Praxis der Carl von Ossietzky Universität Oldenburg festgelegt sind, befolgt habe.

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