

Article

Challenges in Automotive Fuel Cells Recycling

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Abstract: Fuel cell driven cars belong to the ‘zero emission’ vehicles and should contribute to lower CO₂ emissions. However, they contain platinum, which is known as a critical material in the European Union. This study investigated the potential contribution of recycling fuel cell vehicles (FCV) to satisfy the platinum demand arising from a widespread deployment of fuel cell vehicles in Europe. Based on a qualitative examination of the four consecutive steps in the recycling chain (collection, dismantling, disassembly and pre-processing, material recovery) of fuel cell vehicles, two recycling scenarios were developed. Using dynamic material flow analysis, these two recycling scenarios were applied to two scenarios for the market penetration of fuel cell vehicles in nine European lead markets to deliver both the associated impact on platinum demand and the contribution of recycling for meeting this demand. The diffusion of FCV in Europe will not cause a depletion of platinum resources in the short term, as the calculated 537.06 t and 459.24 t in cumulative platinum requirements are far below the currently estimated global reserves. However, concerns regarding the future development of platinum supply and demand remain.

Keywords: fuel cell; end-of-life vehicle; electric vehicle; recycling; material flow assessment; STAN

1. Introduction

In 2011, the European Council reconfirmed its determination to meet the 2 °C-target for global warming through European Union(EU)-wide reductions of greenhouse gas (GHG) emissions of 80% to 95% (compared to 1990 levels) by 2050 [1]. As road transport alone makes up one-fifth of the European Union’s total emissions of carbon dioxide (CO₂) [2] in order to meet the overall reduction targets by 2050, transport-related GHG emissions must be cut by up to 67% [1]. Despite ongoing improvements to the emission standards of internal combustion engine vehicles, under the current European vehicle portfolio, 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 [3]. 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. Fuel cell vehicles, on the other hand, combine the emission benefits of battery electric vehicles with almost the same ranges and refuelling times 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 [4], a market segment which accounts for around half of all passenger cars in the EU and is responsible for 75% of transport-related CO₂ emissions [5].

However, potential trade-offs and unintended rebound effects resulting from the endorsement of novel powertrain technologies must be considered carefully. As highlighted by a number of

publications, electromobility not only entails environmental benefits, but also has substantial impacts on the demand for non-renewable resources, in particular for so-called special or technology metals [6–12].

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 CO₂ equivalents in the range of 13,000 tons per ton of platinum group metals [13], excessive water and energy consumption [14], as well as habitat destruction, air and water pollution and generation of dust, particulate matter and solid waste [15]. 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 [16]. 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 [17].

While various publications discuss the diffusion of fuel cell vehicles and the resulting platinum demand [6,7,9,12,17–20] 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. While industrially used catalysts are recycled with rates close to 90%, passenger cars represent one application with relatively low platinum recycling rates, as the recycling quotas for exhaust gas catalysts reach a mere 50% to 60% [21]. This already makes the automotive industry the largest net consumer of platinum today, even when growth of vehicle sales is ignored [17]. With fuel cell vehicles containing more platinum than combustion engine vehicles by a factor of >10, a widespread deployment could severely aggravate the temporal and structural scarcity already perceptible in the global platinum market. It is therefore in the interests of both fuel cell and car manufacturers to close the existing knowledge gaps and focus on the contribution of recycling for meeting their future platinum demand.

Hence, this study aims to investigate to what extent the recycling of platinum from fuel cell vehicles may contribute to satisfying this industry's future platinum demand. In order to provide an enhanced understanding of the current proton exchange membrane fuel cell technology and its material compositions, the paper sets out with a review of the state-of-the-art. The subsequent part then 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. At the core of this study is a dynamic material flow analysis, 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.

2. Theoretical Context

2.1. Proton Exchange Membrane Fuel Cells

From a structural perspective, fuel cell vehicles can be considered a type of hybrid vehicle, in which the fuel cell replaces the internal combustion engine (Figure 1) [22]. Using atmospheric oxygen and compressed gaseous hydrogen supplied from the onboard tank, the fuel cell generates electricity, which powers the vehicle's electric motor. Due to their favourable attributes, such as low operating temperature, fast start-up and fast response to varying loads, fuel cells installed in current automotive applications are of the Polymer Electrolyte Membrane fuel cell (PEMFC) type.

PEMFC consist of a membrane electrode assembly (MEA) framed by bipolar flow field plates (Figure 2). The MEA comprises a compound structure of a polymer electrolyte membrane, a gas diffusion layer (GDL) and a catalyst layer. Serving as the fuel cell's electrolyte, the gas-proof but proton-conductive membrane consists of a 50 to 150 µm thick foil, which is known mainly by its

registered trade name Nafion[®]. The membrane separates the cell's electrodes, which are formed of a gas diffusion layer and a catalyst layer that is usually composed of carbon-supported platinum or platinum-alloys [23].

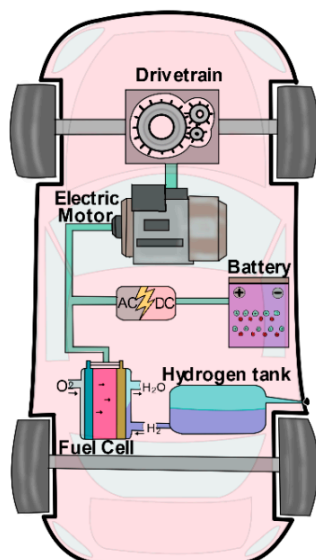


Figure 1. Simplified schematic view of a fuel cell driven vehicle (own figure).

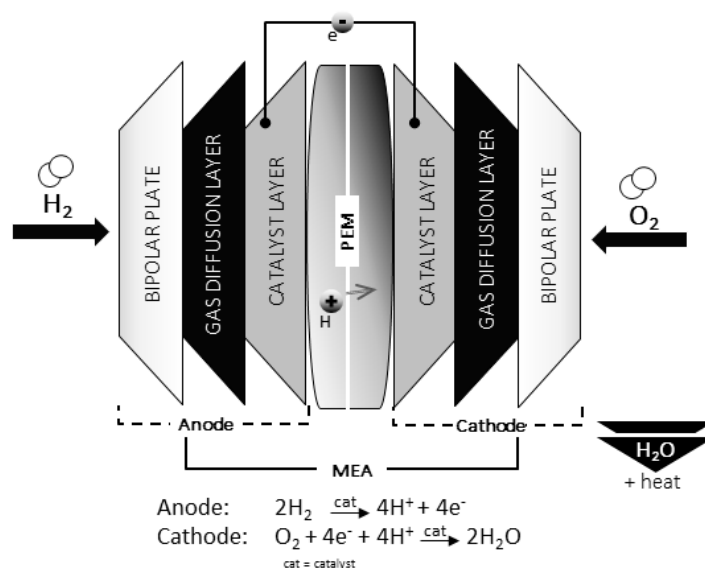


Figure 2. Simplified schematic view of Polymer Electrolyte Membrane fuel cell (PEMFC) (own figure).

At present, the most widely used membrane material continues to be Nafion[®], which is made of a hydrophobic polytetrafluoroethylene (PTFE, also known as Teflon[®]) backbone with hydrophilic side groups containing ether units and sulphonic acid end groups. Besides these fluorocarbon-containing membranes, intense research is also done on polybenzimidazole (PBI) membranes which are soaked with phosphorous acid. Such membranes do not suffer as much from swelling and water loss at temperatures above 100 °C, and thus allow operation temperatures of up to 200 °C. At temperatures above about 130 °C, the adsorption of carbon monoxide (CO) on the platinum surface decreases, and thus the catalyst poisoning effect by CO is strongly reduced. CO is an unwanted by-component in hydrogen fuel, if H₂ is obtained from methane conversion. It was reported for PBI/phosphorous acid fuel cells that they can tolerate 3 vol % CO in H₂ at 200 °C while at 125 °C only 0.1 vol % are acceptable.

for reaching the same current density [24]. However, this type of high-temperature fuel cell is still in the research phase and not available on the commercial market.

In PEMFC, the GDL commonly consists of carbon cloth or carbon paper, hydrophobically treated with PTFE in order to prevent water from accumulating in the pores. An additional micro-porous layer consisting of carbon or graphite particles mixed with PTFE is typically adhered to the GDL to improve both electrical contact and water transport [25].

A platinum-based catalyst is generally used for both the oxidation reaction taking place at the anode and the reduction reaction occurring at the cathode. Particles of the catalyst (10 to 100 nm in size) are finely dispersed on a porous substrate, which usually consists of high surface area carbon powders, e.g., carbon black. Commercial production today typically employs procedures similar to those of the printing industry, in which the supported or unsupported catalyst material is mixed with solvents, binder (perfluorosulfonic acid or other ionomers in protonated form) and other additives to form a 'catalyst ink' and then applied in wet form. The resulting catalyst layer is between 10 and 30 μm thick [25–27].

2.2. Development of the Platinum Content

Platinum offers superior catalytic properties with regard to activation of the desired reactions, stability in the fuel cell's harsh chemical environments and long-term durability. However, its high costs in particular, have sparked research aimed at reducing or replacing platinum loadings while maintaining or even improving PEMFC performance. Dedicated research programs have resulted in significant technology advances, allowing a considerable reduction of PEMFC platinum loadings without compromising performance in recent years [28]. While research aimed at eliminating the use of platinum group metals as catalyst materials altogether and replacing it with alternatives is ongoing, it is unlikely to produce any viable options for the coming decade at least [17,29]. Hence, a further reduction of platinum load remains a vital prerequisite for commercialisation of FCV.

Table 1 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 [30]. Note that the data presented in Table 1 refers to so-called nanostructured thin-film catalysts, which are based on platinum alloyed with, for example, nickel, cobalt, manganese, iron or titanium and which currently remain at a laboratory stage.

Table 1. Technical targets for electrocatalysts in 80 kW automotive fuel cell systems (adapted from [31] (p. 8)).

Characteristics	Unit	Status 2013	Target 2020
Total platinum content	$\text{g}/\text{kW}_{\text{rated}}$	0.14	0.125
Total platinum loading	mg/cm^2 electrode area	0.15	0.125

Assuming a fuel cell stack of 80 kW, the platinum loading reported for 2013 in Table 1 translates into a total platinum requirement of around 11.2 g per FCV. However, as these are fuel cell stacks produced and tested under laboratory conditions only, the platinum load of fuel cell stacks installed in fuel cell vehicles currently 'on the road' is likely to be considerably higher. Estimates given by expert assessments range from 30 to 42 g of platinum per fuel cell vehicle (depending on the assumed stack power), though no official figures have been published by vehicle manufacturers [17,32,33].

3. State-Of-The-Art in the Recycling Chain of Fuel Cell Vehicles

With up to 42 g of platinum per vehicle, fuel cell vehicles could hence constitute a significant secondary resource in the case of further market penetration. The concurrent implementation of an effective recycling concept would not only serve to counteract the aggravation of scarcities, but also secure resource access and reduce raw material costs for the automotive industry. The co-development

of efficient recycling systems is hence considered a prerequisite for successful market integration of fuel cell vehicles.

In order to determine the state-of-the-art in the recycling of fuel cell vehicles as well as anticipating challenges that could lead to suboptimal recycling rates, we performed a systematic literature search of scholarly databases, patent databases as well as online repositories of the US Department of Energy. To supplement scientific literature with information on practice-related projects, a further exploration using a combination of relevant search terms was performed using an online search engine. As the focus of our research lies on the impact of fuel cell vehicle diffusion on the European market, all legal and regulatory aspects refer to conditions within the European Union. The presentation of the literature search results follows a common partitioning of the recycling chain into consecutive steps.

3.1. Collection

In the European Union, end-of-life vehicle recycling is governed primarily by Directive 2000/53/EC, which will presumably also apply to the various powertrain technologies classed as electric vehicles. Under the Directive, automobile producers are liable for take-back, treatment and recycling of end-of-life vehicles (ELV) and incur the costs involved with these requirements; the end-of-life treatment is thus free of charge to consumers [33]. 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, all states certify 'Authorised Treatment Facilities' that issue a 'Certificate of Destruction' to consumers when their deregistered vehicle enters the ELV process [34,35]. 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 [34]. While the issue of ELV's unknown whereabouts is discussed on a European level, it is not the focus of this paper and will only be considered in so far as implications for the future fate of FCV can be deduced.

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 [36]. Only a small number of vehicles are exported directly to non-EU countries; the majority of these vehicles, however, end up in countries lacking the infrastructure required for environmentally sound recycling. Issues such as illegal exports of ELV falsely classified as used vehicles, lack of legal rigorousness, unlicensed scrap operators, abandoned and garaged ELV represent further significant barriers to maximising collection rates [35].

While collection deficits represent the single largest cause for platinum-group metal losses in the recycling chain of automotive catalysts [36] and the issues 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. Not only is the efficiency of ELV collection schemes likely to improve in future years as a result of corrective actions initiated 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 [20].

Nonetheless, the need for highly efficient logistics chains and economic incentives is postulated throughout the literature. Concepts discussed in current research projects include producer-led schemes, such as a reclaimable deposit or lease program for fuel cells, a platinum lease program, in which ownership of the contained platinum remains with the (possibly governmental) lessor

throughout the fuel cell vehicle's life cycle, and vertical integration of collection, dismantling and recovery businesses, which would lead to an increase in recycling efficiency by reducing the number of actors involved [37–41].

3.2. Dismantling

As is the case with battery electric vehicles, fuel cell vehicles will presumably follow the same end-of-life processing routes as conventional ELV, with the provision that the fuel cell system must be removed prior to shredding [42]. Instructions for the salvaging and dismantling of FCV have been devised in the United States, 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 [43]. These components generally necessitate manual removal, discharge or other treatment before the vehicle can be further dismantled, which is likely to add significant costs.

The extent to which incorrect removal of fuel cell stacks may lead to platinum group metal (PGM) losses in future recycling activities 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, as fuel cells are generally concentrated in a relatively compact stack (cf. Figure 1) 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.

As mentioned above, failure to remove the fuel cell stack and other components manually prior to further dismantling involves serious safety hazards. 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 disassembly of exhaust gas catalysts, of which 4% fail to be removed prior to shredding [37]. Since the stack can be removed and transported intact, dust losses as described, for example, for exhaust gas catalysts are also considered insignificant [41]. It is, however, uncertain in how far fuel cell stacks of future FCV will be accessible and removable by independent workshops and ELV dismantlers. It may be assumed that for some time following the wider market penetration of FCV, only few, branded workshops will have the specialist knowledge, personnel and equipment required for removal of the fuel cell stacks, as is the case with battery electric vehicles [42].

3.3. 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 [25]. 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. 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. However, during operation of the fuel cell, small platinum particles may diffuse into the membrane. Excluding the membrane and other parts of the fuel cell from the recycling process may thus limit recycling efficiency in addition to making the recycling process more complicated.

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 end-of-life fuel cells and have hence not been developed to date [44].

Schiemann et al [44] provide exemplary information on the manual disassembly procedure of a PEM fuel cell stack into its respective fractions, an overview of which is given in Table 2. 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.

Table 2. Disassembly process for PEM fuel cells and fractions produced (adapted from [44] (pp. 28–31)).

Disassembly Step	Action Taken	Fraction Produced
Step 1	Removal of tie rods and casing	Connecting elements: high-grade steel
Step 2	Removal of end plates	Connecting elements: high-grade steel;
		Insulation: plastics; End plates: high-grade steel or aluminium alloy
Step 3	Withdrawal of the stack's individual layers, each consisting of bipolar plates, seal, gas diffusion layer and membrane electrode assembly	Seal: plastics
		Bipolar plates: graphite, polymeric binders, additives, separating agents
		Gas Diffusion Layer (GDL): non-woven fabric, carbon paper Membrane Electrode Assembly (MEA): polymeric membrane coated with graphite and platinum

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 [44]. 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, making separation of the GDL and catalyst coated membrane difficult. Attempting to strip the GDL off such 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 [45].

In addition, the fuel cell stacks of some manufacturers are moulded rather than screwed shut and therefore cannot be disassembled by simply removing the casing and individual layers [46]. 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. It is conceivable that minimal dust losses occur when the stack is forcefully broken into individual units. When applied to high volumes of end-of-life fuel cells, the platinum losses resulting from such processes could be significant.

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. Of the nine platinum recovery processes analysed in the following chapter, 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.

Shore [45] gives a more detailed description of the applied procedures and also mentions potential drawbacks of the employed technologies, such as matting together of MEA fragments and subsequent obstruction of milling equipment, overheating of equipment and failure to successfully comminute MEAs due to the elastic properties of the membrane. In order to counteract these problems, Shore reports two possible approaches: cryogenic milling and granulation at room temperature followed by milling [45]. 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.

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 [41,44,47].

In a detailed analysis of the different types and brands of fuel cells available, Schiemann et al stress that it will likely be tremendously difficult for disposal companies to recognise the exact type of fuel cell system they are dealing with [44]. 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.

3.4. Material Recovery

As the presence of fluorine compounds in all three layers is customary according to the state-of-the-art, it is especially the MEA materials that present established recycling practices with considerable challenges. Given higher volumes, the feed of end-of-life PEMFC to established pyrometallurgical recycling processes for the recovery of platinum is not feasible, although lower volumes may be tolerated. Incineration of the fluoropolymers contained in Nafion[®], the hydrophobically treated gas diffusion layer and the catalyst layer's binding agent leads to the formation of hydrogen fluoride (HF), a toxic compound that is both harmful to health and highly corrosive [46,48]. The application of established pyrometallurgical procedures would thus require the costly and time-consuming 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 [49]. Despite such investments, any fluorine constituents remaining in the PGM-containing slag could hamper the later separation [50]. In addition, through incineration the valuable membrane material Nafion[®] is essentially destroyed in the process and cannot be recycled [44]. From a recycling viewpoint, the absence of fluorine in the membranes would thus be a major advantage of alternative fuel cell systems, such as the PBI/phosphorous acid fuel cells.

Jha et al. [51] give an extensive overview on the hydrometallurgy recovery and recycling of platinum by leaching of spent catalysts. Despite the standard route of using aqua regia as a leachant, they are focusing on platinum leaching using acidic and alkaline solutions in the presence of oxidizing agents such as nitric acid and hydrogen peroxide, sodium cyanide and iodide solutions. In addition, they also highlight the environmental concerns. Since most publications deal with platinum recovery in general and do not address the PEMFC specifications, few papers are dealing directly with the membrane characteristics in fuel cells [52–55]. Most researchers already identify the potential of not only recovering the platinum from fuel cells; they focus on recovering the whole membrane for reusing it in new applications. However, this is still on laboratory scale and not available on the market today. From the environmental point of view, it would be the best way.

The intention of this paper is not to add another literature review to the community. Instead we performed a patent search in order to determine to what extent the aforementioned issues with regards to the formation of HF have been acknowledged by relevant actors and resulted in the development of alternative procedures. This search included patents registered under the European Patent Office, the US Patent & Trademark Office and the World Intellectual Property Organization (WIPO) in English language since the year 2000, which focus on the recovery of platinum group metals from PEMFC while avoiding HF emissions. Note that with a view to the research focus of this paper, 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, were neglected. The results of the patent search including a short description of the respective procedures are summarised in Table 3.

Table 3. List of patents for recovery of Platin Group Metals (PGM) from PEMFC avoiding hydrogen fluoride (HF) emissions.

Patent Number	Inventor/s	Proprietor	Materials Recovered (and Rate in %)	Procedure	Source and Technology
WO/2004/102711	Hagelüken, Kayser, Romero-Odeja, Kleinwächter	Umicore AG & Co. KG	PGM (95%)	Binding of HF through inorganic additive	[56], thermal pretreatment followed by hydro-metallurgy
WO/2006/024507	Koehler, Zuber, Binder, Baenisch, Lopez	Umicore AG & Co. KG	PGM and/or fluorine-containing constituents (no rate provided)	Separation of membrane and catalyst layer by treatment with medium in supercritical state	[50], thermal pretreatment (supercritical status) followed by hydro-metallurgy
WO/2006/115684	Shore, Robertson, Shulman, Fall, Matlin, Heinz	BASF Catalysts LLC	PGM and/or ionomer (up to 99%)	Leaching using aqua regia	[57–61], hydrometallurgy
WO/2007/149904					
WO/2009/029463					
WO/2009/149241					
WO/2010/132156					
EP2700726A1	Romero, Meyer, Voss	Heraeus Precious Metals GmbH	PGM (no rate provided)	HF-resistant furnace	[62], thermal treatment only
WO/2015/010793	Paepke	Daimler AG	PGM and/or ionomer (>98%)	Separation of membrane and catalyst layer through ultrasound and solvent addition	[63], pretreatment by ultrasonic bath followed by pyrometallurgy

As shown in Table 3, a number of recovery procedures are available that allow the recovery of platinum from end-of-life MEA incurring only minimal losses. However, while avoiding HF emissions, all procedures described above continue to entail certain drawbacks.

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 [62]. Using water in its supercritical state, the process proposed by [50] relies on a high-pressure operating environment, thus posing concerns for safety as well as untenable equipment costs. Similar reservations may be voiced for the HF-proof furnace and scrubbing process invented by [62], as its cost advantage compared to refurbishment of existing precious metal refining facilities is questionable. Finally, while the procedure proposed by [45] 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.

Insufficient yields of platinum may be problematic in those cases, where removal of the GDL is required in the pre-processing stage 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. The platinum nanoparticles have to be recovered by dissolution in acidic solutions forming Pt^{2+} ions. In a subsequent step the platinum ions are electrochemically reduced [57]. 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 [45]. 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 [45] describes, the conventional combustion of MEA produces a platinum-rich ash, which must be further refined to remove impurities.

Finally, it should be noted that as of 2016, all processes have been implemented and tested at a laboratory level only. Although [45] 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 end-of-life fuel cells arise.

4. Material Flow Analysis for Fuel Cell Vehicles

Building on the insights gained in the qualitative investigation of the recycling chain of fuel cell vehicles, we use dynamic material flow analysis (MFA) to quantitatively assess the impact of a widespread diffusion of fuel cell vehicles in Europe on the global demand for platinum as well as the potential of recycling for meeting this demand.

4.1. Research Design and Calculation Approach

The dynamic MFA for this study is performed using the software tool STAN (short for SubSTance flow ANalysis), a freeware available from the Technical University Vienna, Austria (<http://www.stan2web.net/>). The calculations performed as a preliminary basis to create the model of platinum flows arising from the market penetration of fuel cell vehicles are derived from a top-down approach as summarised in Figure 3. An explanation of the parameters selected that govern both flow values and transfer coefficients within the MFA diagram is given below.

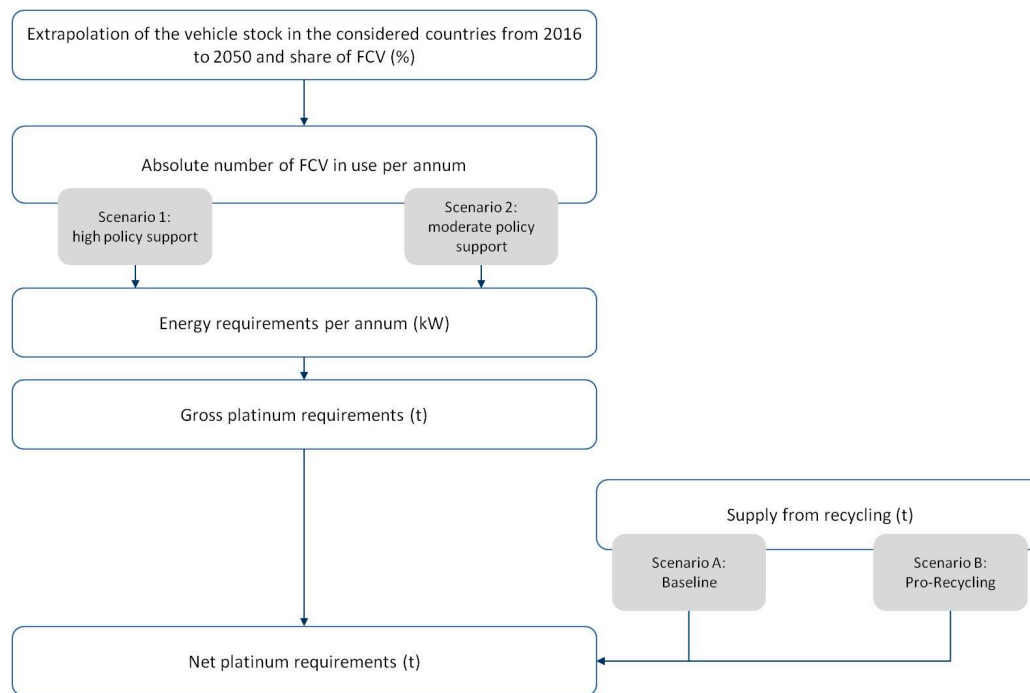


Figure 3. Calculation approach in preparation for the Material Flow Assessment.

4.2. System Boundaries and Assumptions

In order to narrow down the scope of the MFA, a number of system boundaries and parameters were set.

Several studies are available projecting possible scenarios of FCV market diffusion, some of which also include predictions of associated platinum requirements [6,7,9–11,17–20]. Aiming to evaluate possible recycling pathways for FCV for a closed European system, 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.

From the remaining publications, the HyWays study of the European Commission [64] 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. Based on 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 four on FCV. In doing so, the study focuses on the following ten European countries, which hence also form the geographical focus of the MFA presented in this paper: Finland, France, Germany, Greece, Italy, the Netherlands, Norway, Poland, Spain and the United Kingdom. Of these, Greece was later excluded in this study due to a lack of data on vehicle ownership.

In view of technological developments since the publication of the study, two scenarios assuming moderate technological progress were selected, which in turn differentiate between high policy support (Scenario 1) and moderate policy support (Scenario 2). Scenario 1 is hence based on the assumption that market barriers, such as high investment costs, can be counteracted due to a political endorsement of particularly FCV thus pushing the market penetration of FCV. In scenario 2, on the other hand, political endorsement applies to sustainable mobility in general rather than to a specific powertrain technology, i.e., it puts no focus on FCV.

Applying the market penetration ratios given in [64] to an extrapolation of the vehicle stock in the nine lead markets delivers a projected development of the total vehicle stock as well as the FCV market shares as illustrated in Figure 4. The four FCV market penetration scenarios provided by [64] 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 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”. [64] (p. 16)

Given the current state of the FCV market, the assumptions underlying the two more conservative scenarios appear closer to reality. Hence, only the two conservative scenarios, will be considered for this MFA.

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 of 0.3% will be used. Based on the vehicle stock reported by Eurostat (2014) and the above mentioned average annual growth rate, the vehicle fleet in the nine analysed EU member states will develop as illustrated in Figure 4. 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.

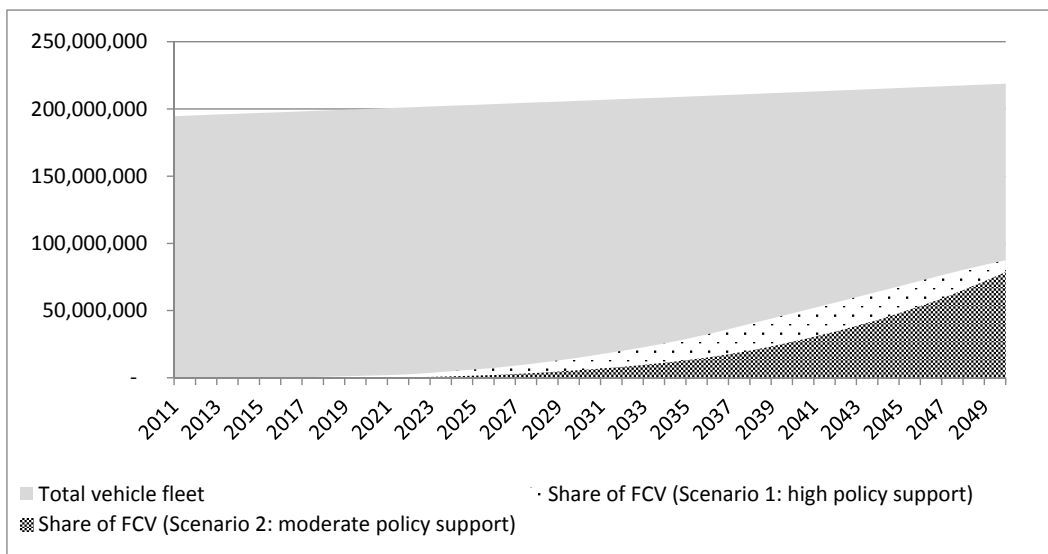


Figure 4. Development of the vehicle stock from 2011 to 2050 and share of fuel cell vehicles.

In addition, various other parameters were gained from a review of the relevant literature as well as expert interviews and are summarised in Table 4.

Table 4. Parameters and assumptions for material flow analysis.

Parameter	System Boundaries/Assumptions	Sources
Geographic boundary	Finland, France, Germany, Italy, the Netherlands, Norway, Poland, Spain, the United Kingdom	[64]
Time scale	2016 to 2050	Authors' choice based on data availability
Market penetration scenarios	High policy support, modest learning rate (Scenario 1); modest policy support, modest learning rate (Scenario 2)	[64]
Average annual growth rate of vehicle fleet	0.03%	Mean value of [5,65–67]
Recycling scenarios	Baseline (Scenario A), Pro-Recycling (Scenario B)	Authors' choice
Type of vehicle	Light-duty passenger vehicle	Authors' choice based on vehicle segmentation and GHG emissions
Average vehicle lifetime	10 years	[25]
Average vehicle power	80 kW	[30]
Average vehicle weight	1447 kg	[25]
Average weight of fuel cell stack	40.8 kg	[25]
Platinum load per kW	Continuously declining from 2020: 0.125 g, 2030: 0.07 g, 2050: 0.07 g	[31,68]
Loss rate of catalyst ink in manufacturing	Continuously declining from 2020: 15%; 2030: 5.2%; 2050: 1.1%	Authors' estimation based on [69–74] and average annual improvement rate of ~0.8%
Use phase loss	0.67%	[41,75]

As evident from the preceding chapter, exact figures 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 developments, two recycling scenarios with diverging assumptions, as listed in Table 5, were developed using information gained during the preceding qualitative analysis of the recycling chain. These are to be interpreted as a “best case” and “worst case” scenario, with the latter based on recycling rates of platinum-rich exhaust gas catalysts, as reported by [37], but supplemented with the limited factual information on the recycling of fuel cells that is available. The Baseline-Scenario (A) therefore represents the actual recycling possibilities, including the whole recycling chain, as collection, processing, etc. The Pro-Recycling Scenario (B), on the other hand, applies the most optimistic estimates for each parameter as derived from the preceding literature review and includes very little losses and very few so-called “unknown whereabouts” in car recycling. The efficiency of the entire recycling chain depends on the efficiency of each step. For example, in the Baseline Scenario (A) total recycling chain efficiency can be calculated as follows: $33\% \times 96\% \times 96\% \times 95\% = 28.9\%$. Despite efficiencies of above 95% for all remaining steps, the low rate of ELV collection thus delivers a low overall recycling efficiency, stressing the fact that careful management of each step in the recycling chain is necessary. In reality, a realistic number might lie between these two scenarios.

Table 5. Parameters of the two recycling scenarios (based on expert knowledge and own assumptions).

Parameter	Baseline Scenario (A)	Pro-Recycling Scenario (B)
Share of End-of-Life Vehicles collected by recycling facilities	33%	85%
Share of fuel cell stacks 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 recycling chain efficiency	28.9%	80.8%

All parameters as well as results of the preliminary calculations were entered into the STAN software to deliver annual and cumulative gross platinum requirements from 2016 to 2050, as well as the share of demand met by recycling of fuel cell vehicles. The material flow diagram can be viewed in the Supplementary Materials section.

5. Results and Discussion

Based on the parameters described above, the material flow analysis delivers the following results with regards to the development of platinum demand and recycling potential arising from a deployment of fuel cell vehicles in Europe.

The results of Table 6 indicate that the diffusion of FCV in Europe will not cause a depletion of platinum resources in the short term, as the calculated 537.06 t and 459.24 t in cumulative platinum requirements are far below the currently estimated global reserves of 66,000 t of platinum group metals [76], even though any contribution from the recycling of FCV is not yet included in this extrapolation.

In contrast, a comparison of the maximum annual gross demand established by the two scenarios, as illustrated in Figures 5 and 6, 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, although US and Asian vehicle markets are not even considered in this extrapolation. With 23.42 t and 39.66 t, respectively, by 2050 the European FCV industry alone would require a maximum

of 14.5% and 24.5% of the 161.74 t produced in 2014 [77]. 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.

Table 6. Cumulative platinum demand and recycling potentials.

Task	Scenario 1 (High Policy Support)	Scenario 2 (Moderate Policy Support)
Cumulative platinum demand for Fuel Cell Vehicle (FCV) production	537.06 t	459.24 t
Cumulative platinum content of end-of-life FCV	293.51 t	155.23 t
Cumulative amount of platinum recovered (Scenario A: Baseline)	84.8 t (208.71 t lost)	44.85 t (110.38 t lost)
Cumulative amount of platinum recovered (Scenario B: Pro-Recycling)	230.02 t (63.49 t lost)	119.17 t (36.06 t lost)

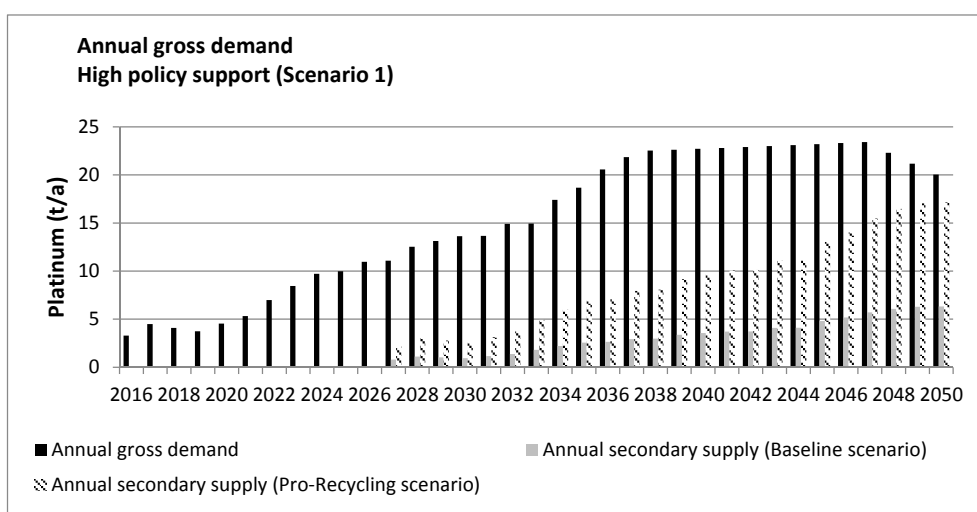


Figure 5. Scenario 1—Annual gross demand for platinum and contribution of secondary supply.

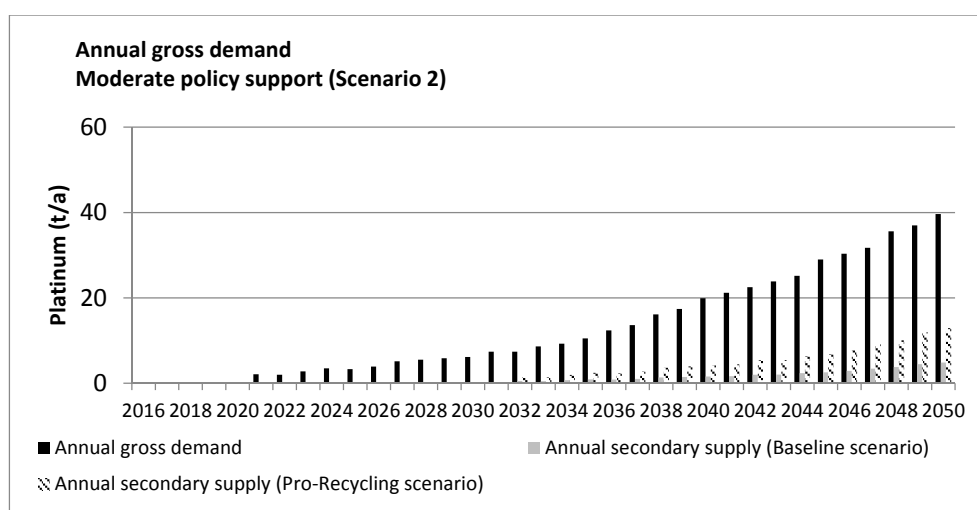


Figure 6. Scenario 2—Annual gross demand for platinum and contribution of secondary supply.

With regards to the role of recycling in meeting the platinum requirements of a growing FCV fleet, Figures 7 and 8 show a significant recycling potential, the exploitation of which could greatly

reduce this industry’s dependence on the volatile platinum market. Up to 85.6% (17.14 t) of the platinum required for European FCV production could be obtained from recycling of end-of-life FCV by 2050 (Scenario 1; Pro-Recycling), while the other three scenario combinations deliver much more conservative results of 31.5% (6.32 t) (Scenario 1; Baseline) (cf. Figure 7), 32.6% (26.74 t) (Scenario 2; Pro-Recycling) and 12.3% (4.86 t) (Scenario 2; Baseline) (cf. Figure 8). Although the latter three estimates appear less promising in terms of meeting the industry’s platinum requirements, even the lowest of these values still represents 4.86 t of platinum (worth £179,820,000 at today’s market price in 2015 [78]) that would otherwise have to be produced from primary sources.

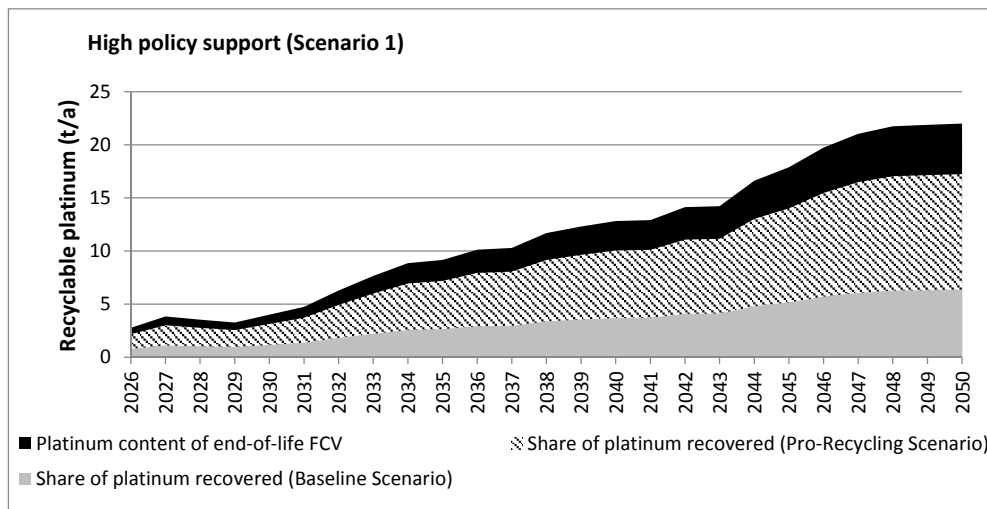


Figure 7. Scenario 1—Recycling potential and share of platinum recovered.

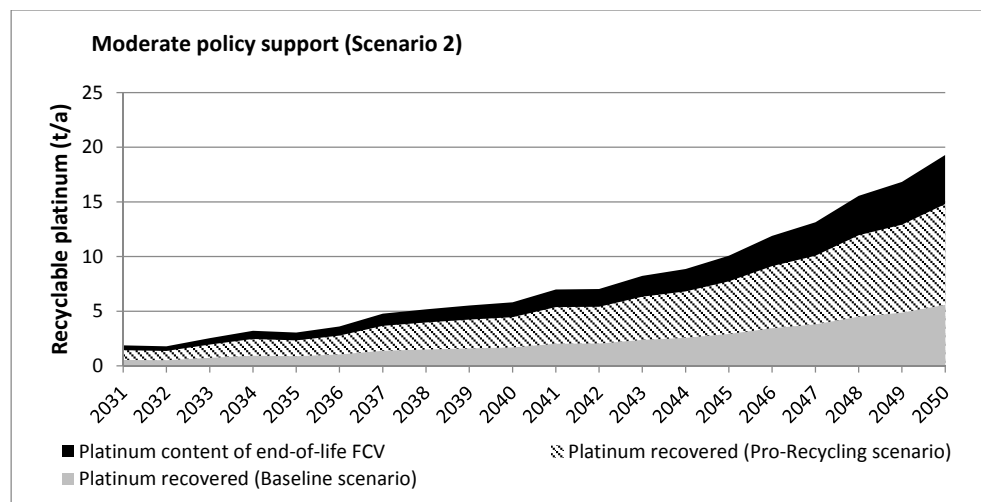


Figure 8. Scenario 2—Recycling potential and share of platinum recovered.

Nonetheless, in both Scenario (1) and (2), the discrepancy between the two recycling scenarios becomes apparent. This is underlined in Figures 7 and 8, 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.

Considering the entire time span from 2016 to 2050, Table 6 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 and thus cast doubts on the endorsement of fuel cell vehicles for sustainable development.

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 and quantitative assessment of this study with previous publications, one may conclude that several studies tend to overestimate the platinum supply achievable from FCV recycling. For example, Råde, Tiax, Saurat and Bringezu [6,19,20] assume recycling rates of 90%, 95% and 75%, respectively. This paper, on the other hand, upon examining each step in the recycling chain, proposes much more conservative recycling rates between 28.9% and 80.8%.

As analysed in Chapter 3, highly efficient recycling chains as the ones cited by Råde, Tiax, Saurat and Bringezu [6,19,20] 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 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% [19]. Without major changes to the current collection and reporting system, such rates appear unattainable.

As shown in Chapter 3, 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. 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.

6. Conclusions and Implications for Further Research

This study investigated the potential contribution of recycling to satisfy the platinum demand arising from a widespread deployment of fuel cell vehicles in Europe. Based on a qualitative examination of the four consecutive steps in the recycling chain (collection, dismantling, disassembly and pre-processing, material recovery) of fuel cell vehicles, two recycling scenarios with “best case” and “worst case” future recycling rates were developed. Using dynamic material flow analysis, these two recycling scenarios were applied to two scenarios for the market penetration of fuel cell vehicles in nine European lead markets to deliver both the associated impact on platinum demand and the contribution of recycling for meeting this demand. As demonstrated by the highly diverging results, 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 study’s MFA imply that well-devised recycling systems could contribute quantities of secondary platinum in an order of magnitude that could mitigate existing raw material dependencies and possible future supply shortages considerably.

It appears as though stakeholders of the precious metal industry that are already involved in the recycling of platinum-containing applications have acknowledged both the opportunities and challenges arising from a further diffusion of fuel cells and fuel cell vehicles in particular. A number of procedures for recovering platinum from end-of-life fuel cells with high efficiencies and under the premise of avoiding HF emissions exist, yet it must be stressed that none have been implemented at an industrial scale. Future research should thus not only investigate the large-scale applicability of such procedures, but also environmental impacts and profitability in comparison to established pyrometallurgical recovery processes. Hydrometallurgical recovery processes seems to show promising results, especially relating to the recovery rate of more than 90% but further research

is needed to achieve the best environmental performance. A big advantage in some hydrometallurgical processes is the fact that the membrane could be recovered as a whole membrane and might be available for reuse purposes, whereas the pyrometallurgical process recovers only materials and not components. From a recycling perspective, further research on PBI-based fuel cell systems that lower the presence of fluorine in the membrane also appears to be promising, although their efficiency and stability remain unsatisfying at the current development status. In the gas diffusion layer (GDL), some polytetrafluoroethylene (PTFE) will always be necessary to ensure the necessary hydrophobicity. However, compared to Nafion[®] membranes, the total fluorine content will be reduced by about 80%.

Even more problematic is the fact that less is certain about the preceding stages in the recycling chain, which appear to have received little attention by producers, recycling businesses, researchers and other stakeholders. This is especially concerning when considering that deficits in the initial stages of the recycling chain (above all collection) account for a major share of material loss in the recycling chains of other platinum-rich consumer goods and could potentially do so for fuel cell vehicles as well. 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.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2313-4321/1/3/343/s1>, Figure S1: STAN material flow diagram including flow codes, Figure S2: STAN use phase sub-system of the material flow diagram including flow codes.

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Abbreviations

The following abbreviations are used in this manuscript:

CO	Carbon Monoxide
CO ₂	Carbon Dioxide
ELV	End-of-Life Vehicle
EU	European Union
FCV	Fuel Cell Vehicle
GDL	Gas Diffusion Layer
GHG	Greenhouse Gas
H	Hydrogen
HF	Hydrogen Fluoride
MEA	Membrane Electrode Assembly
MFA	Material Flow Analysis
PBI	Polybenzimidazole
PEM	Polymer Electrolyte Membrane
PEMFC	Polymer Electrolyte Membrane Fuel Cell
PGM	Platinum Group Metals
PTFE	Polytetrafluoroethylene
US	United States of America
WIPO	World Intellectual Property Organization

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