

Impact of the European Union vehicle waste directive on end-of-life options for polymer electrolyte fuel cells

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Abstract

Polymer electrolyte membrane fuel cells (PEMFCs) may well be powering millions of cars by 2020. At its end-of-life, each car will have a redundant PEMFC stack. The EU vehicle waste directive sets tough recycling and re-use requirements for the cars of the future. The criteria for assessing the end-of-life options are based on technical, economic and environmental feasibility. The optimum strategy will require stack dismantling and separation of the major components. Steel and aluminium parts can enter the general recycling stream, but the membrane electrode assembly and bipolar plates will require a specialised recycling process. One option is to shred the MEA, dissolve and recover the membrane, burn off the carbon, and recycle the platinum and ruthenium catalysts using solvent extraction. The heaviest part of the PEMFC stack is the bipolar plates. If carbon fibre based, the bipolar plates could enter a fluidised bed recovery process where the constituent materials are recovered for re-use. The EU vehicle waste directive sets high recycling targets based on weight, and thus it is strongly advisable for the relatively heavy bipolar plates to be recycled, even though energy recovery by incineration may be a cheaper and possible more environmentally benign option. The EU vehicle directive will put pressure on the end-of-life options for the PEMFC stack to be weighted towards recycling and re-use; it will have a significant impact on the design and end-of-life options for the PEMFC. The overall effect of this pressure on the end-of-life treatment of the PEMFC and the consequential contribution to environmental life cycle impacts is discussed. It is concluded that a range of external pressures influence the selection of a suitable end-of-life management strategy, and while opportunities for re-use of components are limited, all components of the PEMFC stack could in principle be recycled. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Polymer electrolyte fuel cells; End-of-life; Life cycle analysis; Recycling; EU vehicle waste directive

1. Introduction

Polymer electrolyte membrane fuel cells (PEMFCs) are an emerging technology, which offer many advantages over conventional methods of electricity generation. They are under development for both transportation and stationary power applications. Research efforts are presently focused on issues such as stack performance, durability and cost. Information on the present status of PEMFC development can be found in e.g. [1,2].

The increasing emphasis on fuel cells as a candidate power generation system of the future [3] means that there is a growing need to look at the environmental impact of the whole life cycle of the system. This includes the manufacturing, in-use, and end-of-life stages. Such an approach is

termed a life cycle assessment (LCA). LCA is an analytical environmental management tool (see for example [4]) used to inform decision making within environmental product, process and systems design, as well as a recommended step in the implementation of environmental management systems [5]. It is used to assess the environmental burden of a product, process or activity over its entire life cycle starting with raw materials extraction and ending with the final waste disposal. The full process of making this assessment involves: a goal definition and scoping phase, an inventory analysis, an impact assessment and finally an improvement assessment [4].

While the potential environmental impacts of PEMFCs in-use are well documented [6] there is a great deal of uncertainty concerning the environmental impact of the manufacturing and end-of-life stages. A first step towards exploring the potential environmental impacts of the manufacturing stage has been reported by Karakoussis et al. [7] who analysed the materials and energy flows associated with the manufacture of a PEMFC system using an LCA approach. A similar approach has been reported for assessing

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Table 1
Materials inventory of a representative 70 kW PEMFC stack [7]

Component	Material	Weight (kg)	Wt. %
Electrode	Platinum	0.06	0.1
	Ruthenium	0.01	0.0
	Carbon paper	4.37	6.3
Membrane	Nafion membrane	5.64	8.1
Bipolar plate	Polypropylene	16.14	23.1
	Carbon fibres	16.14	23.1
	Carbon powder	21.52	30.8
End-plate	Aluminium alloy	2.80	4.0
Current collectors	Aluminium alloy	1.14	1.6
Tie-rod	Steel	2.05	2.9
Total		69.87	100.0

the environmental impact of manufacturing a solid oxide fuel cell system [8–10]. The study reported in this paper extends this earlier work, and then seeks to set it in the context of the likely impact of the European Union vehicle waste directive.

The PEMFC considered was one suitable for use in light duty vehicles, with a power output of 70 kW. The stack construction was based on that detailed in [11]. Table 1 summarises the materials inventory used for this study. End-of-life management options were considered for the individual components of the stack; the Nafion-type membrane, the platinised electrodes, the bipolar plates and the ancillary components (tie-rods, end plates and casing). For each component a range of options was examined, following the waste hierarchy, which provides a framework for the discussion of end-of-life options and their environmental impact. Using the waste hierarchy, which suggests that re-use is (normally) environmentally better than recycling, recycling better than incineration with energy recovery, and incineration better than disposal, an assessment of the options identified was made and an end-of-life strategy suggested. Data for the study was gathered via a series of interviews with key players in the field, together with a review of relevant literature.

2. Results and discussion

2.1. Electrolyte

The fluorinated Nafion-type membrane is the most frequently used membrane in the PEMFC. This type of membrane has a high production cost, with projected Nafion costs of US\$ 50 m⁻² [12]. Hence, both economic and environmental factors drive membrane re-use and recycling.

Re-use of the membrane would be preferred, both in environmental and economic terms. However, there are many barriers to re-use. In particular, membrane dehydration and pin-holing [13] are a common cause of failure.

Dehydrated membranes are unlikely to be re-usable as they are weak, and as they would probably be damaged when the membrane electrode assembly was removed from the stack. Furthermore, contaminants will accumulate on the membrane during use and the structure will be degraded, thus reducing the efficiency of the membrane. A process to mend pin-holes in Nafion exists [14], but could be difficult to apply to a membrane electrode assembly as the resin injected could well damage the gas diffusion layer.

Recycling the membrane is therefore likely to be more feasible than re-using it. To recycle, the membrane must first be removed from the MEA. The membrane is sandwiched between two electrodes so mechanical separation from the carbon structure would be difficult. Chemical extraction of the membrane for recycling is a more viable option. To remove the membrane, it must be dissolved and then returned to its ionomer form.

Dissolution of the membrane and re-casting as a polymer film [15] is one possible method of membrane recycling. The MEA would need to be shredded, the ionic groups on the membrane converted to the Li⁺ salt, and the membrane dissolved in a water:ethanol mixture at 250 °C under pressure. The soluble membrane could then be separated from the other MEA components, and the solvent evaporated to produce powder. This could then be converted to the Nafion ionomer powder using nitric acid. To re-form a Nafion membrane the Nafion powder would be dissolved in ethanol and dried at ambient temperature. While the resultant film has been described as brittle [15], this is nonetheless a potential method of reproducing the Nafion membrane from an MEA at its end-of-life. The recycled Nafion powder could be re-entered into the Nafion membrane production process.

Major uncertainties in analysing the recycling of Nafion lie in the amount of energy consumed, the cost of the process, and the purity of the recycled Nafion. However, it has been shown that the energy requirements for recycling should be below the 14 kJ kg⁻¹ used in the production of Nafion powder from its raw materials [7].

A possible problem when trying to recycle the membrane is that the degraded parts of the polymer may contaminate the recyclate. It may then be difficult to recover the monomer after polymer degradation. Hence, the recyclate may need to be purified during the process, or the membrane boiled in nitric acid prior to recycling to remove the degraded areas.

If the above method can efficiently and cost effectively recycle the membrane, then this appears to be an attractive end-of-life management route. In comparison, incineration is not a favourable option, as highly toxic hydrogen fluoride would be emitted, and a costly HF recovery plant would need to be built alongside the incinerator. Dissolving the membrane out of the MEA is therefore the alternative preferred to incineration or re-use.

Other options to perfluorinated membranes are under development, and are briefly considered here from an end-of-life perspective. Hydrocarbon based membranes offer

lower cost than the fluorinated forms, but are less resistant to oxidation. However, absence of fluorine in the membrane means that they can be readily incinerated without hydrogen fluoride emission. Recycling however is likely to be more difficult, due to high levels of chemical degradation within the membrane likely at its end-of-life. One developer has reported a membrane that is both non-fluorinated and offers high performance [26]. In this case recycling may be possible. However, few details of the membrane are available, precluding discussion of end-of-life options. Fig. 1 summarises the end-of-life options for all three membrane types.

2.2. Platinum/ruthenium electrocatalysts

There are fundamental economic and environmental reasons for recycling the platinum and, if present, ruthenium, in fuel cells. The value of platinum and ruthenium in a 70 kW stack is around US\$ 1000 at precious metal prices \approx US\$ 16 g⁻¹.

The Department of Energy and the USGS minerals information team in USA agree that it will be essential to recycle the platinum in the fuel cell for the product to be sustainable in the long term [16]. The environmental argument for recycling platinum is strong. Emissions of sulphur dioxide (SO₂) are decreased by a factor of 100, and the primary energy demand is reduced by a factor of 20, when the platinum is recycled in comparison to its production from primary sources [17].

Both platinum and ruthenium can be recovered with high yield using a chemical recovery process. The use of solvent extraction to recycle both platinum and ruthenium is well established [18]. The electrolyte membrane must be removed before solvent extraction, as every catalyst particle is in contact with the membrane in the MEA. After electrolyte removal, residual organics must be removed, which would otherwise cause problems in the extraction process, such as crud formation and poor phase separation. Given that the fluorinated polymer has already been removed, it would then be possible to burn-off residual organic material to leave an incinerator ash with high platinum and ruthenium content suitable to enter the extraction process.

A detailed review of the solvent extraction process for the recovery of platinum and ruthenium has been given by Barnes and Edwards [18]. Ruthenium would be extracted before platinum by distillation of the tetraoxide. Platinum is extracted into tri-*n*-butyl phosphate in 5 M HCl, using a counter-current process of extraction, scrubbing, and stripping of platinum. Residual scrub liquor is recycled back to the extraction stage, or piped away for waste liquor treatment depending on its composition, thus reducing waste emissions. The final stage uses water to strip the platinum from the organic phase and the resultant strip liquor contains pure H₂PtCl₆. Ammonium chloride is used to precipitate out the platinum as 99.95% pure (NH₄)₂PtCl₆. Emissions to air from the process include ammonia, chlorine, nitrogen dioxide and hydrogen chloride. Any base metals released during

platinum recycling are precipitated out using lime and then landfilled.

Platinum recycling is crucial to the sustainable future of PEMFCs due to limited platinum reserves, coupled with the saving in energy by a factor of 20 in comparison to extraction from the ore. The platinum industry will see a large increase in recycling if the prediction that the world fuel cell market will exceed 2.1 million fuel cell passenger cars by 2010 is realised. At the current level of platinum loading of 60 g per stack [7], this would result in 126 tonne of platinum becoming available for recycling by 2020 (assuming a 10 year car/fuel cell life). At US\$ 16 g⁻¹ this would be a US\$ 2 × 10⁹ market for platinum and ruthenium recycling.

2.3. Bipolar plates

There are several different materials under development for the fuel cell stack's bipolar plate, including stainless steel, graphitic carbon, and carbon composites.

Graphite-based bipolar plates are used by a number of manufacturers. To reduce cost, volume and weight, resin impregnated graphite plates are also being developed [19]. Graphite is resistant to corrosion, so the bipolar plate should have a lifetime well beyond that of other components. However, even if the bipolar plate was undamaged after use, improvement in fuel cell design will mean that the plate design will be obsolete, such that re-use would not be possible. The only practical end-of-life option for a graphite plate would therefore be to burn it for energy recovery.

Other fuel cell developers are pursuing the use of steel bipolar plates. This is a fairly low cost option. The drawbacks are the potential for corrosion and subsequent damage to the cell. Makkus et al. [20] has suggested that steel bipolar plates are re-usable once cleaned and the outer oxide layer removed. This work also confirms that the steel could be recycled. The recycling option is the most likely, as it is probable that the design of the bipolar plate will be obsolete at cell's end-of-life, such that re-use would not be possible.

Given that the end-of-life options for these two materials appear straightforward, this study focussed on carbon composite bipolar plates. It has been suggested that these have a potential for low cost, large scale manufacture [11]. However, an end-of-life management route is not straightforward. Recycling, and incineration to generate energy, are again the likely two options for the end-of-life management of the carbon composite bipolar plates.

Given that the bipolar plates comprise 70–80 wt.% of the stack, the European Union vehicle waste directive [21] will push the manufacturer to recycle rather than incinerate. This is discussed further in Section 4. Table 1 shows that the carbon composite material comprises carbon fibres, carbon powder and polypropylene polymer. Carbon fibres are the most costly material in the plates, and the carbon powder contributes the largest mass to the plates. Three processes have been identified which could be used for recycling:

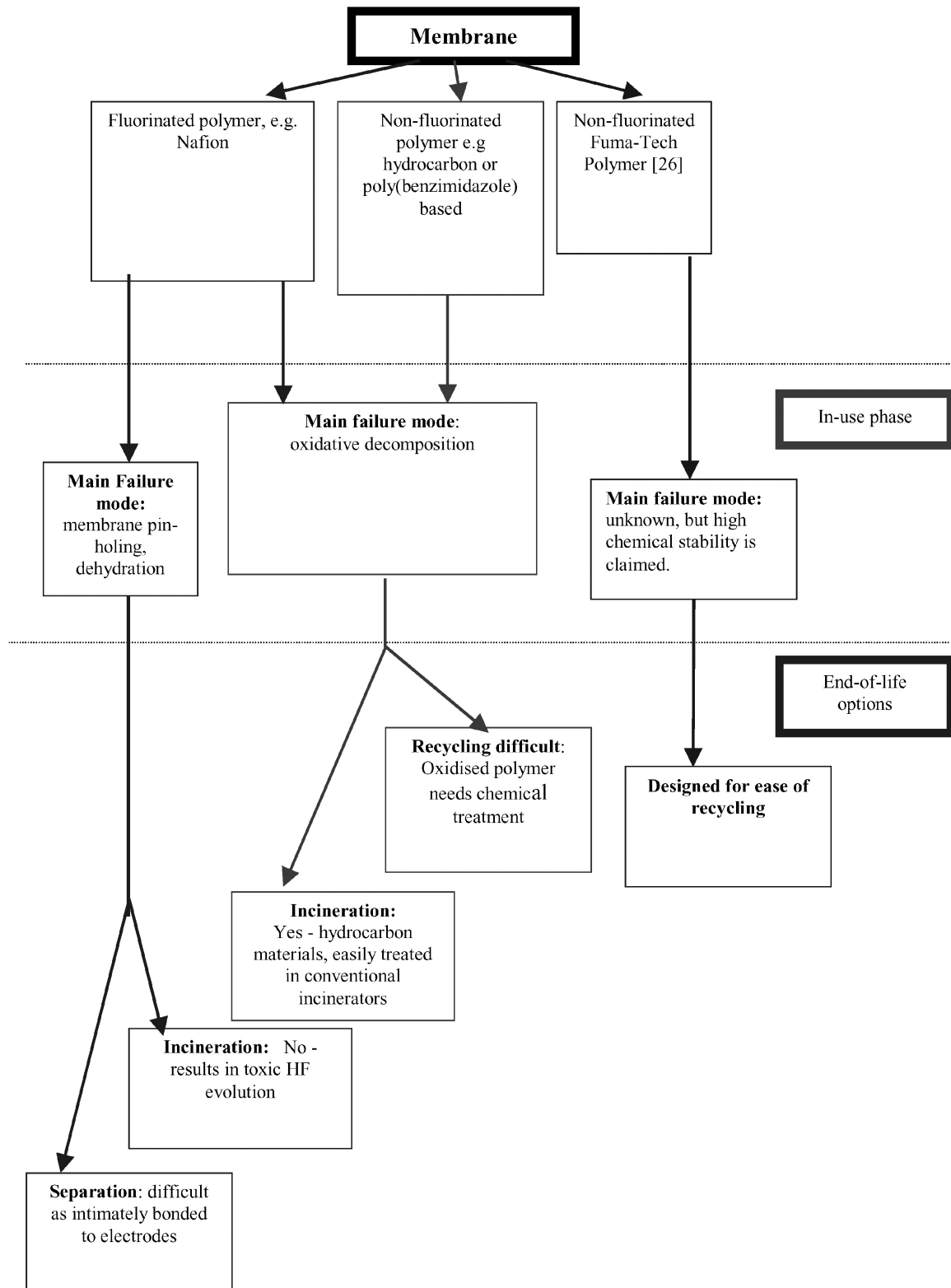


Fig. 1. Summary of end-of-life options for the electrolyte membrane.

catalysed low temperature pyrolysis [22], reverse gasification [22] and fluidised bed fibre recovery [23].

Comparison of these three methods suggests that the fibre quality from the reverse gasification and catalysed low

temperature pyrolysis routes may be impaired, which could effect their potential for re-use. Reverse gasification produces high carbon monoxide levels (a major process by-product), which will need to be treated before release. Low

temperature pyrolysis operates in a closed system, which minimises environmental emissions, and generates sufficient heat to drive the process without the need for additional fuel.

Fluidised bed fibre recovery appears to be an attractive option. The process is the only one of the three to recover both the carbon fibres and the carbon filler. It has been reported that the quality of the recovered fibres is high [23], which could enable re-use. Avoiding combustion of the carbon filler also reduces carbon dioxide emissions from the process. Given these potential advantages, the use of fluidised bed fibre recovery to recycle carbon composite bipolar plates is considered in further detail in the following paragraph.

The carbon composite bipolar plates are first broken down using a hammer mill. After sizing, the material is fed into a fluidised bed at 450 °C, which breaks down the composite. The filler and the fibres are recovered and the exhaust gases are burnt (with heat recovery). Acid gas scrubbing is used to remove any acid or halogen in the off-gas. It has been estimated that such a processing plant would need to take in around 9000 tonne of composite per year from scrap collected within an 80 km radius [23]. This high tonnage would initially cause a problem if the plant were used solely for bipolar plates—to break even it would require over 160,000 end-of-life stacks per year. Inputting a mixture of carbon composites is an option, although this would mean the carbon fibres could not be recycled directly back into bipolar plates. However, with increasing pressures on car companies to recycle a large proportion of their car, and incorporate recyclates into new cars, there is a potential growth market for recovered carbon fibres.

As stated previously, all three components in the carbon composite bipolar plates are combustible, and thus it would also be possible to burn the plates for energy recovery. The energy value for the composite is 35.6 MJ kg⁻¹, as shown in Table 2, higher than that of coal (26–30 MJ kg⁻¹ [22]). As the plates are made of such a high-energy material, the value of energy from waste must not be ruled out.

Certainly, the option of recovering energy from the composite waste would be a straightforward, low cost, flexible option with minimal incinerator ash generated. However, the drawbacks are many, from public opinion to environmental impact. Another problem with energy recovery is the residues of metal and, more importantly, Nafion that may be left

on the plates. Emissions of HF in incinerator plumes would be serious and may well stop the plates from being incinerated, as costs of installing a HF recovery plant may be prohibitive.

In the short term, when only small numbers of fuel cell vehicles are reaching their end-of-life every year, it is likely that carbon composite bipolar plates will be incinerated as this capacity already exists. They could also enter into an existing general carbon fibre recovery process, although the grade of fibre produced by a wide variety of scrap would make the re-sale value low and the fibres could not be used in new bipolar plates. The option of transporting low value bipolar plates across the country to a single fibre recovery plant could possibly produce more CO₂ than burning the plate locally in a combined heat and power plant.

Fig. 2 presents a summary of the end-of-life options for the three types of bipolar plate materials discussed in this section.

2.4. Ancillary components

The ancillary components of the fuel cell stack are the non-repeat items. They are significant when considering recycling as they contribute to nearly 16% of the weight of the stack. They consist of the polypropylene housing, the PTFE insulators, the steel tie-rods, the aluminum alloy end-plates and the soft aluminum alloy current collectors.

The optimal end-of-life management option for both the steel and aluminium parts of the fuel cell stack is recycling. There are large energy savings if the metals are recycled in comparison to extraction. The polypropylene housing could also potentially be recycled, the energy saving would need to be investigated to assess the feasibility and effectiveness.

The first stage in recovering the steel tie-rods involves removing them from the stack. The rods would be unbolted and, based on present designs, the rods, MEA and bipolar plates should be relatively straightforward to separate. The rods would then be transported to a recycling plant—it is unlikely they could be re-used as after 10 years the dimensions of the stacks will change.

The Boustead Model [24] gives an energy value of 22.4 MJ kg⁻¹ to produce steel from iron ore. The International Council on Metals and the Environment, states that recycling steel requires 30–35% of the energy required to manufacture steel from the raw materials [25]. Taking the

Table 2
Energy content of a typical carbon composite bipolar plate^a

Composition	30 (wt.%) Carbon fibres	40 (wt.%) Carbon powder	30 (wt.%) Polypropylene polymer	Total
Mass per 70 kW stack (kg)	16.14	21.52	16.14	53.8
Mass per kW (kg of fuel)	0.23	0.31	0.23	0.77
Energy recoverable by combustion (MJ kg ⁻¹)	32 (31)	32 (31)	44 (32)	35.6
Energy recoverable per FU (MJ per FU)	7.36	9.92	10.12	8.19

^a FU: functional unit (material required to manufacture each kW capacity of PEMFC stack).

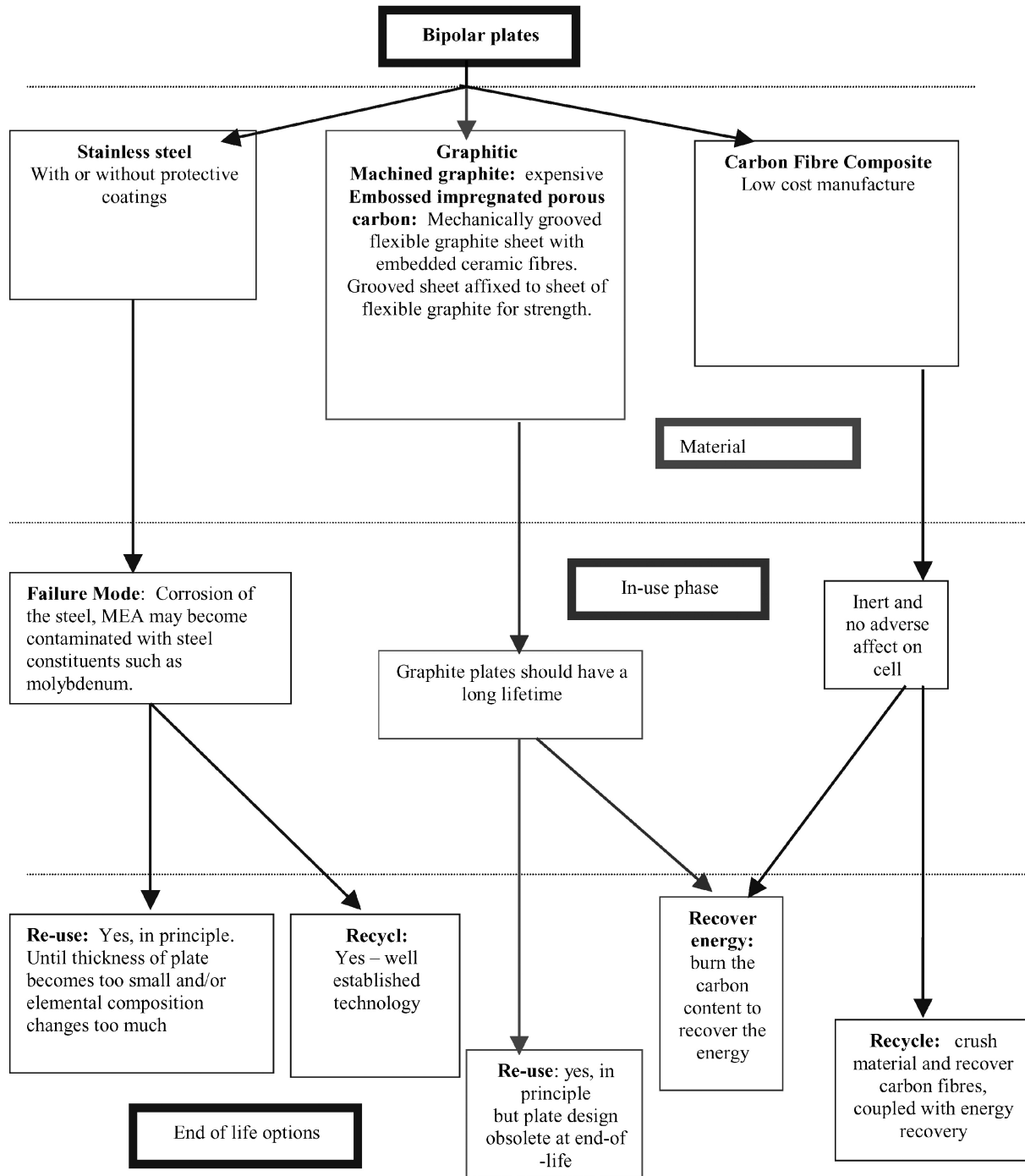


Fig. 2. Summary of end-of-life options for bipolar plates.

mid-value, this gives an energy requirement of 7.3 MJ kg^{-1} for recycling the steel rods. The additional energy to manufacture the steel is 0.03 MJ kg^{-1} , so the entire scrap to new steel rod in the stack process should take around 7.33 MJ kg^{-1} . The infrastructure for recycling scrap car steel already exists along with an excellent market for the recycled product, so these factors will facilitate steel rod recycling. The energy saved also offers a clear advantage from an economic and environmental resource perspective.

The steel rods should not be contaminated with the fluorinated polymer, minimising problems during smelting.

The aluminium end-plates are well positioned for fast removal; they need unscrewing from the stack before the other components can be removed. It has been estimated that recycling requires 5% of the energy required to produce primary aluminium from the ore [17]. This results in the end-plate alloy requiring 28.2 MJ kg^{-1} for recycling and the soft alloy for the current collectors 14.1 MJ kg^{-1} .

Polypropylene is commonly used for the stack casing. This material is already recycled within the automotive industry. This system of cascade recycling, as compared to closed loop recycling—where the recycle will have the same quality, function and thus value as the primary material—could be used for the fuel cell polypropylene casing, and would allow the integration of recycle into a new vehicle. The casing would be removed, granulated, compressed and then used as splash shields, wheel guards etc. in new vehicles. To facilitate removal of the casing, design for easy removal should be considered.

3. An end-of-life management route for the PEMFC stack

A proposed end-of-life management strategy for the PEMFC stack is shown in Fig. 3. This assumes a Nafion-type electrolyte and composite carbon fibre bipolar plates. Bipolar plates, platinum electrocatalysts, membrane and

ancillary components could all be recycled. The flow chart details the order and the links in the chain for the PEMFC stack at end-of-life, from dismantling to recycling the platinum.

The first step in the dismantling procedure is to remove the fuel cell stack. The next step is to dismantle the stack by removing the casing and unscrewing the steel tie-rods. The rods would go to a steel recycling plant with other steel car scrap, and an equivalent route would be followed for the aluminium end plates. The bipolar plates would enter a fluidised bed treatment process to extract the carbon filler and carbon fibres, potentially for re-use in new bipolar plates.

The MEA would enter a recycling stream different to that of the other components as it contains the fluorine in the polymer, which would emit toxic fumes if heated. It would be advisable to have a specialised treatment plant for the comparatively light but very valuable MEA. Firstly, the MEA needs shredding into small pieces to aid its dissolution. Dissolving the membrane would require ethanol at high

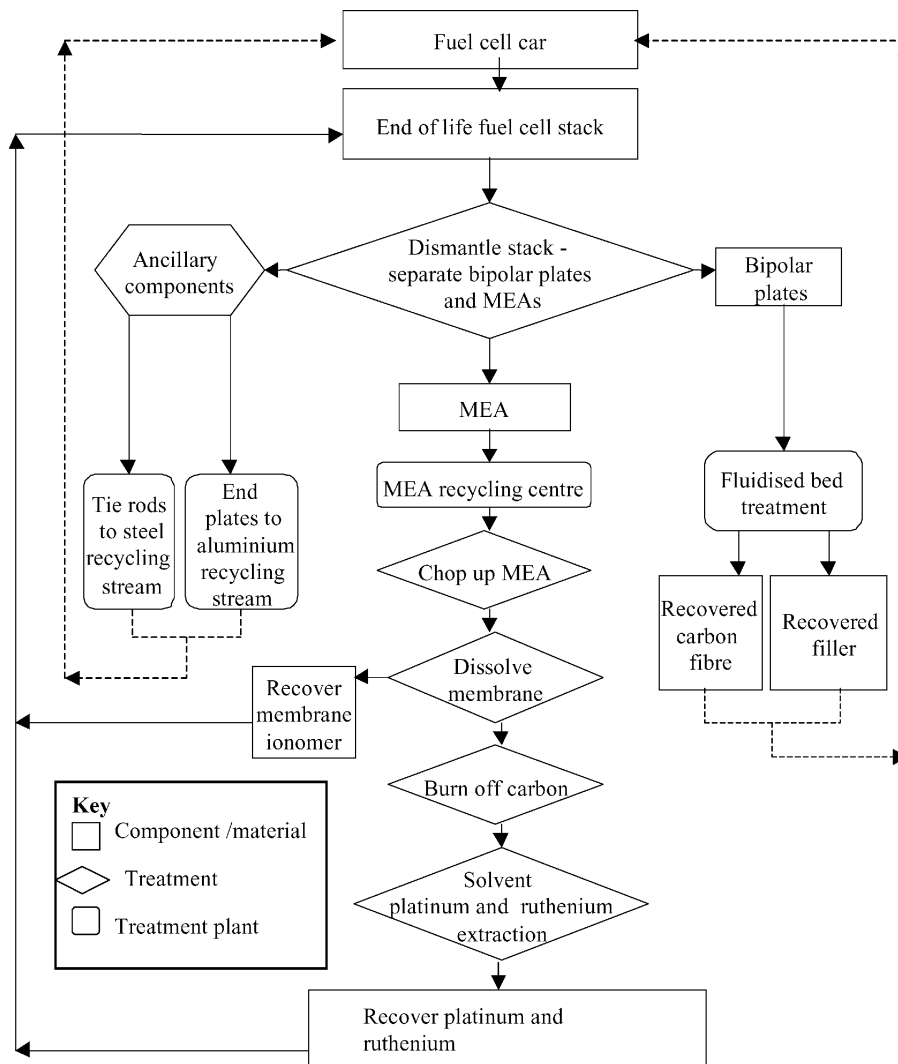


Fig. 3. Proposed flowsheet for the end-of-life treatment of PEMFCs.

temperature and pressure, so it would have to be carefully controlled and is likely to be an energy intensive process.

Once the membrane is removed, residual carbon would be burnt off to produce a precious metal bearing ash. This would then enter the solvent extraction process, which in principal is capable of recovering more than 99% of the platinum and ruthenium as a high purity product. This high recovery is essential due to the high demand that fuel cells will put on both platinum and ruthenium resources. It is likely that all of the recovered metal will be required for the manufacture of new fuel cell MEAs.

4. Impact of European Union vehicle waste directive

The end-of-life vehicles directive imposes recycling and re-use regulations on vehicles that will be sold in the future, and also on the vehicle at its end-of-life. The aim of the directive is to harmonise measures concerning end-of-life vehicles, in order to minimise impact on the environment, and to avoid any impact on competition between member states [21]. The directive sets out strict targets, such that vehicles type-approved and put on the market after 1 January 2005 must be re-usable and/or recyclable to a minimum of 85 wt.% per vehicle.

The responsibility for ensuring vehicle take-back and covering the costs of end-of-life process lies with the vehicle manufacturer (OEM). The Society of Motor Manufacturers and Traders estimate the total liability from the take-back scheme at a cost of UK£ 4 billion for the UK Motor industry and UK£ 30 billion for Europe [27]. These high take back costs are largely because the scrap value of recycled IC engine cars is only ~UK£ 20 and the cost of returning the cars, dismantling them and recycling them will be higher. The value of the fuel cell car may well be increased because of the valuable elements, such as platinum, in the fuel cell. The authors suggest that a future scenario is that a specialist tier 1 or 2 company will be contracted by the OEM to recycle the fuel cell stack.

In conclusion, the directive re-enforces the need to recycle and re-use components of the fuel cell stack. With such high recycle and re-use targets, it will put pressure on the car manufacturer to ensure the smaller components, such as the end plates, are recycled even though they make a small contribution to the total mass. The recyclability of every part of the car must be considered in order to reach the target. Issues such as design for recyclability and component labelling will also be important to minimise cost.

5. Conclusions

End-of-life options for the electrolyte, electrocatalysts, bipolar plates and ancillary components of a polymer electrolyte membrane fuel cell (PEMFC) have been considered, and an end-of-life management strategy proposed, in the

context of the European Union vehicle waste directive. The optimum strategy will require stack dismantling and separation of the major components. Steel and aluminium parts can enter the general recycling stream, but the membrane electrode assembly and bipolar plates will require a specialised recycling process. One option is to shred the MEA, dissolve and recover the membrane, burn off the carbon, and recycle the platinum and ruthenium catalysts using solvent extraction. The heaviest part of the PEMFC stack is the bipolar plates. If carbon fibre based, the bipolar plates could enter a fluidised bed recovery process where the constituent materials are recovered for re-use. The EU vehicle waste directive sets high recycling targets based on weight, and thus it is strongly advisable for the relatively heavy bipolar plates to be recycled, even though energy recovery by incineration may be a cheaper and possible more environmentally benign option.

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