Recovery of platinum group metal value via potassium iodide leaching

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Platinum recovery from secondary sources such as end-of-life polymer electrolyte fuel cells (PEMFCS) via electro-winning and chemical dissolution in deoxygenated 4 M potassium iodide with varying added iodine content was investigated. Previous research in this field has shown that complete platinum recovery from model systems is possible, but further study was necessary to determine the process viability with Pt containing devices. The work presented here investigated the leach rate of platinum black deposited on an electrochemical quartz crystal microbalance (EQCM) as well as the effective recovery of Pt from untested and end-of-life polymer electrolyte membrane fuel cells. Platinum black dissolution rates were found to be dependent on added iodine content, with higher concentrations accelerating the reaction. Platinum recovery from leached materials, as determined by aqua regia digestion, was found to be 98.7% and 96.7% for untested and end-of-life PEMFCs, respectively. Results indicate that higher iodine concentrations continuously improved recovery efficiency, but increasing iodine concentration above 5 mM resulted in comparatively minor improvements.

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1. Introduction

Fuel cells are electrochemical devices which generate current capable of performing useful work in an external electrical circuit by separating two half cell reactions with an electrolyte. Proton exchange membrane fuel cells (PEMFCS), which oxidize hydrogen to water, are of particular interest due to their characteristically high current density and consequent suitability for application in automotive power. Currently the majority of these devices use platinum nanoparticles as the principal catalyst, and with a typical PEMFC automotive stack consisting of greater than a hundred individual cell estimates which state that 34% of the total stack cost in 2010 was directly due to platinum content (Papageorgopoulos, 2011). Despite efforts to find means of either reducing platinum content or eliminating it entirely using alternative catalysts, platinum content and cost are likely to remain significant issues for the fuel cell industry for the foreseeable future (Proietti et al., 2011; Serov et al., 2012; Wee et al., 2007; Wilson and Gottesfeld, 1992). Global demand for platinum group metals (PGMs) is anticipated to continue to increase whilst supply from refineries is likely to remain stagnant putting additional pressure to effectively use the metal as well as incentivising Pt metal values recovery from end-of-life devices. Within the European Union (EU) this is acutely recognised and platinum groups metals are part of the 14 strategic materials which are considered vital for continued economic development and as such receive special attention through EU grants and schemes. Recycled platinum currently represents approximately 25% of the global platinum supply stream, and total output has increased annually since 2004 (Matthey, 2014). Existing recycling methods are well established and have been shown to be capable of achieving total metal value recovery, but economic and environmental considerations continue to be problematic.

Recovery processes can typically be classified in three different ways: selective chlorination or gas phase volatilization, pyrometallurgical, and hydrometallurgical (Jha et al., 2013). Selective chlorination processes make use of differing vapour pressures of metal chlorides in order to selectively remove target metals via gas phase adsorption on a sacrificial matrix. Kim et al. demonstrated that it was possible to recover over 90% of PGMs with a chlorination process from spent automotive catalytic converters (Kim et al., 2000). Whilst these processes are highly tuneable and are capable of producing high purity metals, gas phase volatilization does require the use CO and Cl₂, both hazardous gases (Kim et al., 2000). The hazards associated with using these gases for any process of appreciable capacity lead to some significant health and environmental risks. In the UK the storage and use of chlorine on site requires special registration and working procedures on the UK Control of Major Accident Hazards (COMAH) legislation which adds significant costs to a project (HS&E, 2015).

Pyrometallurgical processes operate at temperatures between 1500 and 1700 °C and are used to separate PGM from other metals; however these methods are ill-suited for devices containing significant fluorine content, such as PEMFCs, due to the possible formation of hydrofluoric acid, carbonyl fluoride, and other COF compounds under oxidative conditions (Ario and Soda, 1977; Benson et al., 2000; Conesa and Font, 2001; Samms et al., 1996). Finally hydrometallurgical processes are
the most diverse, but can be generally characterised by leaching Pt in oxidising solutions. Whilst capable of attaining yields in excess of 95% these processes can be improved upon significantly (Angelidis, 2001; Angelidis and Skourakis, 1996; Bautista et al., 1988; Bonucci and Parker, 1984; Kim et al., 1994; Tyson and Bautista, 1987; Woo et al., 2000).

Most hydrometallurgical methods reported require extremely aggressive conditions immersing the spent material in highly acidic and oxidising chloride containing media such as aqua regia (Baghalha et al., 2009; Barakat and Mahmoud, 2004; Jimenez de Aberasturi et al., 2011; Schreier and Edtmaier, 2003). Barakat and Mahmoud investigated the use of aqua regia to recover PGMs from spent reforming catalyst, and obtained a 97.5% yield of the metals in high purity forms (Barakat and Mahmoud, 2004). Jafarifar et al. and Niemelä et al. investigated PGM dissolution in aqua regia with the addition of a microwave digester. Both groups were able to achieve comparable yields and purities to Barakat and Mahmoud but at significantly reduced time scales and leachate volumes (Jafarifar et al., 2005; Niemelä et al., 2012). Finally Baghalha et al. examined the kinetics of Pt dissolution in aqua regia and determined that the rate is limited by the ratio of aqua regia volume to solid content whilst the process itself is solely a surface chemical reaction (Baghalha et al., 2009). Despite being a highly efficient means of recovering metal value, aqua regia is an extremely corrosive solution with well documented biological and environmental effects making it unattractive for large scale recycling processes. Other groups have investigated means of replacing aqua regia with cyanide extraction (Bruckard et al., 1992; Chen and Huang, 2006; Han and Meng, 1994, 1996). These groups have shown that it is possible to recover Pt, Pd, and Rh via cyanide extraction, however the proposed system operated at temperatures and pressures exceeding 200 °C and 10 bar, making it highly energy intensive (Chen and Huang, 2006; Han and Meng, 1996).

A less studied, but highly promising alternative to both aqua regia and cyanide extraction is the use of iodine to complex PGMs. Zanjani and Baghalha investigated platinum extraction in acidified 0.12–0.48 M iodine–iodide solutions at temperatures between 25 and 95 °C (Zanjani and Baghalha, 2009). Their work showed that Pt could be leached from spent materials with sufficient time and iodine oxidant content; however Pt recovery reached a maximum of only 80%, possibly due to limited iodine concentration. Furthermore Zanjani and Baghalha acidified the iodine–iodide solution with HCl, thus reducing the benefit of using a less corrosive solvent. Conversely Dawson and Kellsall investigated the benefits of near neutral iodine–iodide based PGM dissolution, and they were able to show a clear trend stating that higher triiodide concentrations increased Pd and Pt dissolution rates (Dawson and Kellsall, 2007, 2013). By immersing a Pt black flag electrode in neutral 4 M KI solutions with varying triiodide content, they were able to determine the effective dissolution rate of Pt by measuring the dissolved metal content through UV–vis Spectroscopy (Dawson and Kellsall, 2013). Of the methods previously discussed, this process is viewed as having the greatest potential because of its mild operating conditions, reduced hazardous chemicals usage, and significantly lower economic investment in the required process plant. Furthermore the platinum iodo complexes generated by this process are reported to be less sensitising than their chloro counterparts reducing the associated risk of processing PGM salts. Despite the numerous benefits there is very little published work in the use of concentrated iodide solutions for Pt recovery, and further investigation was required to validate this process for recovery from commercial devices. The work presented here is comprised of two separate but related studies. The first used electrochemical quartz crystal microbalance (EQCM) in order to monitor in situ mass changes of Pt black electrodes. These represent a model platinum based catalyst used in electrochemical devices such as PEMFCs. The second investigated the potential to leach platinum nanoparticles from untested and end-of-life (EOL) PEMFCs. The results published here will be used in the development of a more economical, safe and environmentally benign PGM recovery process.

### 2. Material and methods

#### 2.1. Electrode preparation

Platinum black electrodes were produced using 1-inch platinum quartz monitor crystals (Inticon 149240-1) as the substrate using a method adapted from literature (Feltham and Spiro, 1971, 1972; Ilic et al., 2000). The crystals used in this study were manufactured for use at 90 °C, and have a resonant frequency of 5 MHz. The crystals were placed in an EQCM holder (Inticon CHT-100), and functioned as the working electrode in a 3-electrode system. Prior to an experiment the crystals were exposed to air and the electrolyte solution heated to 90 °C each for 30 min, to monitor nascent frequency and resistance fluctuations. This was done as a calibration process in order to ensure accurate and reliable measurements could be obtained. The platinum black working electrode and platinum wire counter electrode were placed in a 0.072 M Chloroplatinic acid solution and a saturated calomel reference electrode (SCE) was placed in 1 M H2SO4 solution with an agar salt bridge connecting both electrolytes. The working electrode was held at 0 V vs standard hydrogen electrode (SHE) for 50 s resulting in a visually even Pt black deposit, and the mass of the electrodes was between 100 and 275 μg. Fig. 1 shows a representative SEM micrograph of the electrode surface.

Hydrogen adsorption was used to determine the active surface of the platinum black electrode, and the method described here is adapted from the referred text (Kinoshita et al., 1973). The working electrode was placed in deoxygenated 0.5 M H2SO4 solution and 5 cyclic voltammetric sweeps between 0 and 1.5 V (SHE) at 0.1 V s−1 were performed. The charge passed between 0 and 0.4 V (SHE) was calculated and divided by 210μC, the charge necessary to form a 1 cm2 monolayer of hydrogen, in order to determine the surface area (Conway et al., 1973). The surface areas of the Pt black electrodes were found to be between 44.5 and 77 cm2.

#### 2.2. Platinum black dissolution

Potassium iodide (ReagentPlus®) was supplied by Sigma-Aldrich, and was used to produce 0.15 dm3 of 4 M KI solutions in 18.2 MΩ DI water. For some experiments iodine was added to the reaction vessel solution by using a 1 N iodine solution (Sigma-Aldrich 319007—100 mL) to produce 5, 10, and 15 mM solutions. Experiments conducted without additional iodine will be labelled as 0 mM throughout this work however it should be noted adventitious iodine was present under these conditions due to the oxidation of iodide by atmospheric oxygen dissolving in the solutions during preparation.

![Fig. 1. SEM micrograph of Pt black electrode (50,000 times magnification, 15 kV, JEOL 5600 SEM). Image courtesy of Dr. Nigel Fullwood (n.fullwood@lancaster.ac.uk).](image-url)
The 4 M KI solution was heated to 90 °C in a flat-bottomed reactor vessel and sparged with nitrogen for 30 min in order to remove dissolved oxygen content. The reactor was only open to atmosphere during experiments via a water cooled condenser attached to the vessel. This minimized evaporative losses whilst elevated temperatures were maintained without the complexity and safety concerns of pressurisation. The EQCM was then placed in the reactor vessel where mass changes were monitored for 20 min. Additionally the open circuit potentials of these electrodes were monitored constantly during the dissolution process in the varying iodine concentrations.

To understand further the kinetics of the Pt dissolution at applied potentials the dissolution rate of Pt black at varying potentials in 4 M KI with 0 mM iodine was also investigated. The platinum black electrode and platinum wire counter electrodes were placed in the reaction vessel, and the reference electrode was connected via an agar salt bridge whilst placed in a separate, ambient temperature 4 M KI solution. Using a computer controlled potentiostat (Compactstat, Ivium Technologies, Netherlands) cyclic voltammetry was performed between 0 and 1.23 V (SHE) at 20 mV s⁻¹ for a total of 5 sweeps.

2.3. PEMFC leach

PEMFCs were provided by a fuel cell manufacturer as both untested and EOL samples. These materials were cut into strips measuring approximately 0.5 cm by 2 cm prior to leaching in order to allow them to fit in the reaction vessels used but would also be representative of possible comminution processes which could be applied in practice. The untested materials consisted of a catalyst coated membrane (CCM) whilst the EOL material was comprised of the CCM compressed to approximately 0.5 cm by 2 cm prior to leaching in order to allow them to be cleanly delaminated. Due to the difference in materials 0.1 g of untested material was used per experiment whilst 0.3 g of EOL material was leached per experiment. Iodine was also added to the reactor volume to produce 5, 10, 15, and 20 mM concentrations. The solution was heated prior to leaching. Iodine was also added to the reactor volume to produce 0.080 dm³ of acid. The acid was placed in a at-bottomed reactor vessel in a 3:1 ratio to produce 4 M KI solution. 0.2 dm³ of 4 M KI solution was prepared per experiment whilst 0.3 g of EOL material was leached per experiment. 0.2 dm³ of 4 M KI solution was prepared per experiment using 18 M quality DI water and sparged with N₂ for 30 min prior to leaching. Iodine was added to the reactor volume to produce 5, 10, 15, and 20 mM concentrations. The solution was heated to 90 °C and continuously stirred during the leach process. The reactor was closed and a water circulating condenser was attached to the vessel in order to minimize evaporative losses. The experiment ran for 120 min, with 1 cm² samples of the solution being taken after predetermined intervals. These samples were then treated with ascorbic acid to reduce excess iodine before being analysed using a UV–Visible spectrometer (Evolution 220, Thermo Scientific, UK) to determine dissolved Pt content (Balcerzak and Kaczmarczyk, 2001; Balcerzak and Pergol, 2003). The leached materials were filtered from the solution, then washed and dried before being analysed with X-ray photoelectron spectroscopy (XPS) and digested in aqua regia.

2.3.1. Aqua regia digestion

A final digestion phase in extremely aggressive conditions was used to determine any residual Pt left on the iodide leached materials. As stated previously aqua regia digestion is a well-documented and established method for PGM recovery, and was viewed as the most reliable method for accurately determining residual platinum content in the PEMFC materials. Aqua regia was prepared prior to each digestion by combining 37 wt.% HCl and 65 wt.% HNO₃ (obtained from Sigma-Aldrich) in a 3:1 ratio to produce 0.080 dm³ of acid. The acid was placed in a flat-bottom reactor vessel and heated to 105 ± 2 °C whilst stirred continuously. PEMFC material was added to the reactor and allowed to digest for 2 h after which the acid was decanted and stored. A sample was then diluted with 18 M quality DI water to approximately 2.2 wt.% aqua regia and analysed for Pt content via ICP-OES (Thermo Fisher Scientific ICAP Duo 6300). Dilution was required to be compatible for use with the instrument set up.

2.3.2. XPS analysis

XPS measurements were conducted at the NEXUS facility located at Newcastle University, and were performed using a Thermo Scientific K-Alpha XPS. The instrument produced X-rays with an energy of 1486.6 eV with an equipped microfocused monochromatic Al-Kα source. The membrane layer for each sample was analysed in three distinct spots, measuring 400 μm by 800 μm. Survey scans were taken in order to determine elemental composition and were carried out with pass energies of 200 eV and dwell times of 10 ms. Charge neutralisation was used throughout the analysis; however spectra were calibrated using the binding energy of graphitic carbon, 284.45 eV, in order to negate the possibility of charge build-up (Smith and Black, 1984; Wertheim et al., 1980).

3. Results and discussion

3.1. Platinum black dissolution

Fig. 2 shows the E-pH diagram for the platinum-iodine system, and shows the available window for platinum dissolution and triiodide formation (Bard et al., 1985). From previous work it has been established that two possible complexes can be formed through the following oxidation reactions:

\[
\text{PtI}_4^- + 2e^- \rightarrow \text{Pt} + 4\text{I}^- \quad E = 0.400 \text{ V(SHE)} \quad (1)
\]

\[
\text{PtI}_6^{2-} + 2e^- \rightarrow \text{PtI}_4^- + 2\text{I}^- \quad E = 0.329 \text{ V(SHE)}. \quad (2)
\]

The complete oxidation of Pt metal to Pt⁴⁺ is the most likely in the conditions used for this study, and can be visibly observed by the distinctive purple of the PtI₂⁻ (aq) complex formed during the experiments. High iodide concentrations are used to suppress the dissolution potential and to help prevent the formation of any products with limited solubility such as Pt₄ (Dawson and Kelsall, 2013).

The cyclic voltamogram and associated mass losses for a platinum black electrode in 4 M KI and adventitious iodine heated to 90 °C are shown in Fig. 3. Fig. 3A shows the electrode gained mass initially during the anodic sweep before reaching 0.4 V (SHE) at which time the deposit began to steadily lose mass. The initial increase in mass is assumed to be due to iodine adsorption as has been established in literature (Schartd et al., 1989; Shu and Bruckenstein, 1991). The mass loss observed continues during the cathodic sweep before reaching 0.4 V (SHE), at which point the electrode mass remains constant. This pattern is repeated during the remainder of the experiment but with each subsequent scan removing less material; the reduced dissolution rate is most likely to be due the diminished surface area of the platinum black electrode.

![Fig. 2. E-pH diagram for Pt-iodo complexes which shows the potential and pH window of PtI₂⁻ and PtI₄⁻ formation (Bard et al., 1985). Complete oxidation of platinum to Pt⁴⁺ was the desired product for the dissolution process, however iodine concentration in solution must be sufficient to inhibit the formation Pt₄, an insoluble precipitate.](image-url)
The voltammogram in Fig. 3B shows a clear oxidation process occurring from 0.4 to 1.23 V (SHE), achieving a maximum of 64.75 mA, and is a combination of the dominant iodide oxidation reaction and platinum dissolution. From Fig. 3A it can be assumed that the dissolution reaction achieves a near steady-state rate of mass loss at potentials above 0.5 V (SHE), indicating that Faraday’s Law of Electrolysis can be used to determine the current the dissolution process would produce. Knowing the sweep rate was 20 mV s\(^{-1}\) and assuming the dissolution reaction is the oxidation of Pt to Pt\(^{4+}\), approximately 0.342 mA of the observed current can be attributed to the mass loss during the first cycle. This steady state mass loss with increasing potential suggests that the dissolution rate limit is highly dependent on a diffusion limited process such as film formation or surface oxidation.

As stated previously the effect of iodine concentration on Pt dissolution was also investigated. The initial surface areas of the platinum black electrodes as well as the total mass removed during each experiment is shown in Table 1. Additionally Fig. 4a shows the recorded mass losses during the course of the experiment; however the data shown has been truncated to 2000 s for clarity as some of the conditions required significantly longer time frames to reach completion. The figure shows a trend indicating increasing iodine concentration accelerates the dissolution process, but the level of the effect is less apparent due to surface area inconsistencies between electrodes. This complication is most obvious when comparing the results between 5 and 10 mM which show approximately the same dissolution rate (−110 μg/2000 s) but with significantly different surface areas. In addition, though the 0 mM experiment had the slowest rate, losing 70.39 μg over the selected time frame, the electrode had the highest surface area (77 cm\(^2\)) which may have increased the apparent dissolution rate. Furthermore during these experiments the open circuit potential was monitored and was found to sit between 0.40 and 0.42 V (SHE) for all conditions. This result is in good agreement with those obtained from cyclic voltammetry and with previously established literature values, and supports the conclusion that oxidant concentration has limited impact on the dissolution rate.

In order to separate the relative effects of both electrode surface area and iodine concentration it was necessary to calculate a molar flux, \(\nu_\theta\), and thus provide a more rigorous conclusion regarding Pt black dissolution. The electrodes were then approximated as a collection of identical spheres shrinking at the rate prescribed by \(\nu_\theta\). To find the molar flux second-order polynomials were fit to the 4 curves to generate relationships correlating recorded mass loss with time. These equations were then differentiated and solved at \(t = 0\), and the resulting value divided by the initial surface area and molar mass of platinum to give a unique molar flux coefficient. To determine the number and radius of the spheres necessary of the model, Eqs. (3) and (4), equations for the surface area and mass of identical spheres, were solved simultaneously for \(r\), radius, and \(n\), number of spheres.

\[
A = 4\pi r^2 n
\]  
\[
M = \frac{4}{3}\pi r^2 np
\]

Using these values it is possible to calculate the dissolution of Pt from the electrode over time, the results of which can be seen in Fig. 4b. Due to the simplicity of the approximation of the model it should be noted that it has been assumed that the reaction rate remains constant and that there are no switches between kinetic and transport control regimes during the dissolution process. Comparing the predicted results to those obtained experimentally shows good agreement, however the approximation typically underestimates the actual dissolution rate. Additionally, though the method used predicts Pt to exist as nanoparticles, Fig. 1 clearly shows the surface is composed of aggregates on the sub-micrometre scale. In practice some pore diffusion limitation might be expected but the closeness of the fit from the simple model suggests that increasing complexity by accounting for this is not necessary to adequately describe the dissolution rates.

Finally, it remains that despite accounting for surface area, the tabulated molar flux grows drastically from 10 mM to 15 mM and there is little explanation for why this discontinuity, if real, exists. It is possible that the dissolution rate observed is being affected by a switch between a kinetic and transport control process further compounded by the difference in the initial surface area of the electrode but this seems unlikely given the cyclic voltammetry results. Further investigation is required before a more definitive conclusion can be made.
3.2. PEMFC dissolution

Fig. 5 plots the dissolved platinum content as measured by UV–vis spectrometer against time for both untested and cycled PEMFC materials. Fluctuations in the results are likely due to minor experimental error in this study, rather than changes in Pt concentration. For both sets of materials a clear trend between iodide concentration and dissolution rate is apparent with 0 mM samples containing significantly lower Pt content than all other conditions. Furthermore experiments with additional iodine content all showed very similar behaviour with increasing concentrations leading to more rapid dissolution; however increasing iodine concentration above 5 mM only led to incremental changes. This is consistent with the EQCM results shown in Fig. 3A. For the untested materials the leaching process was able to achieve near total dissolution after approximately 20 min. Additionally the final dissolved Pt concentration was similar for each experiment with added iodine, indicating that the dissolution process for 0 mM may have been oxidant limited. Furthermore because the materials were leached without being subjected to destructive testing protocol it is probable that the platinum was present in an idealized form, sub-5 nm nanoparticles decorating the electrode surface, which would contribute favourably to the observed dissolution rates.

Whilst the cycled materials exhibit the same trends as the untested samples, dissolved Pt content rises at a much slower rate for the cycled samples, achieving near complete dissolution after 60 min. This delay is most likely due to physical changes caused by the testing protocol used, and suggests that future investigation should be targeted toward improving leach rate rather than yield. The exact source of the difference is unclear though nanoparticle sintering, oxidation of the carbon support leading to restricted pore diameters, and subsurface particles are believed to be the most likely cause. In addition whilst increasing reactor iodine concentration led to more effective Pt dissolution, the cycled material leached with 15 mM iodine shows markedly inferior results to the 10 and 20 mM experiments, most likely caused by localized variations in the PEMFC tested.

Table 2
Platinum concentration in solution as determined by ICP.

| Iodine concentration (mM) | Mass (g) | Reactor concentration (PPM) | PPM (platinum)/g(material) | Residual |
|---------------------------|---------|----------------------------|-----------------------------|----------|
| Standard                  | 0.106   | 150.2                      | 1417.0                      | -        |
| 0                         | 0.101   | 82                         | 811.9                       | 57.3%    |
| 5                         | 0.091   | 8.6                        | 945                         | 6.7%     |
| 10                        | 0.078   | 5                          | 641                         | 4.5%     |
| 15                        | 0.083   | 6.4                        | 77.7                        | 5.4%     |
| 20                        | 0.079   | 1.4                        | 17.7                        | 1.3%     |
| Standard                  | 0.096   | 32.8                       | 341.7                       | -        |
| 0                         | 0.419   | 47                         | 112.2                       | 32.8%    |
| 5                         | 0.372   | 10.4                       | 280                         | 8.2%     |
| 10                        | 0.362   | 10                         | 27.6                        | 8.1%     |
| 15                        | 0.344   | 20.8                       | 60.5                        | 17.7%    |
| 20                        | 0.372   | 4.2                        | 11.3                        | 3.3%     |
3.2.1. ICP results

Table 2 lists the residual platinum content from the leached materials as determined by aqua regia digestion. The residual content is calculated by dividing measured platinum concentration in solution by mass of metal digested. By comparing calculated values of the leached materials to the control samples, it can be shown that the leaching process removed 42.7–98.7% of Pt from the untested samples and 67.2–96.7% of Pt from the cycled samples during the 120 min leaching process. These results are in good agreement with those obtained from UV–vis spectrophotometry, and indicate that increasing iodine concentration in solution improves Pt leaching in 4 M KI; however, the data suggests that a threshold iodine concentration is required to achieve near total recovery and that more concentrated solutions only provide incremental benefit. For the samples used in this work a concentration of 5 mM iodine was sufficient to remove approximately 93% of the platinum present in the untested samples, but that value could differ for other materials.

3.2.2. XPS results

XPS analysis of the samples shows that the materials go through significant chemical changes during both cycling and leaching protocols. Survey scans for the control samples can be seen in Fig. 6, and the atomic weight percentages of 4 elements, carbon, oxygen, fluorine, and platinum, are listed for each sample in Table 3. Comparing the two control samples it is clear that the cycling protocol used to test the cells has caused a significant reduction in the fluorine content, suggesting breakdown of the ion-exchange membrane, as well as removing platinum from the surface. These two changes are believed to be the principal causes for the reduced Pt leach kinetics. Additionally because both sets of experiments achieved comparable dissolved platinum concentrations it has been assumed that the total platinum of experiments achieved comparable dissolved platinum concentration and dissolution rate was observed. This can be accounted for by transport limitations in the cases of very low oxidant concentration and limitation due to film build up at higher concentrations. Additionally the dissolution process was shown to occur spontaneously with the rate being determined by the electrode’s chemically accessible surface area. Through cyclic voltammetric scans and application of Faraday’s law of electrolysis it was shown that the principal reaction which would occur during electrowinning procedures is the formation of iodine/triiodide, not platinum dissolution, making direct electrochemical methods ineffective. A practical application of the process through recovery of platinum from PEMFCs showed excellent potential; for both untested and EOL materials more than 95% of the platinum content was removed after 120 min. As with the platinum black electrodes, increasing iodine concentration improved recovery rates and yields; however, the necessary iodine concentration is likely sample dependent with concentrations above this value providing only minor improvements to dissolution kinetics.

Data access management

Due to confidentiality agreements with research collaborators, supporting data can only be made available to bona fide researchers subject to a non-disclosure agreement. Details of the data and how to request access are available at Lancaster University research portal: http://dx.doi.org/10.17635/lancaster/researchdata/19.

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