Electrochemical routes for environmentally friendly recycling of rare-earth-based (Sm–Co) permanent magnets

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Abstract

The consumption of critical raw materials, especially those in permanent magnets of Nd–Fe–B and Sm–Co-type, has significantly grown in the past decade. With predictions on further electrification growing exponentially the demand for these materials will even increase. This implies that efforts in assuring sustainability must involve recycling from secondary resources. In recent years the electrochemical approaches in recycling have been extensively investigated and applied owing to their advantages of high efficiency and selectivity, easy operation, low energy consumption, and environmental friendliness. In this paper, we investigate the Sm2(Co,Fe,Cu,Zr)17 permanent magnet leaching process using the anodic oxidation to be paired with the metal deposition on the cathode. Linear sweep voltammetry was performed from −0.15 to 1 V versus Pt quasi reference electrode that indicated current peaks that would correspond to some preferential leaching of the crystal phases contained in the magnet. The latter was confirmed using the SEM/EDXS analysis. The continuous leaching of the Sm2(Co,Fe,Cu,Zr)17 magnet was performed at a direct current density of 2, 4 and 8 mA cm−2 at the time period of 0–240, 240–480 and 480–720 min, respectively. The ICP-MS results confirmed the leaching of all the metals from the original Sm2(Co,Fe,Cu,Zr)17 permanent magnet. The concentration of Sm3+, Cu2+, Fe2+ and Zr2+ increases linearly along with the leaching time. Reversely the concentration of Co2+ decreases linearly due to its consumption by electrodeposition of Co, Fe and Cu on the cathode. The presented paired electrochemical process could serve as a starting point for the recycling and recovery of critical raw materials without any acid usage and waste generation.

Graphical abstract

Extended author information available on the last page of the article
1 Introduction

Today’s unsecure market of raw materials i.e., in particular the rare earths (REs) that are used for processing of the RE-transition metal permanent magnets (PMs) (like Nd–Fe–B and/or Sm–Co); pose a recycling challenge, that strives beyond the technical aspects, as it is more on the geopolitical and economic nature. Current applied recycling solutions mainly depend on the technology’s development that may or may not be acceptable in certain countries due to national and/or local regulations. The challenge of recycling the REs persists, as it is mainly connected to extensive energy usage, the consumption of large amounts of chemicals, the production of large amount waste waters that need to be discharged and a lot of disposal waste [1]. However, to achieve the raw materials circular economy and sustainability REs recycling is a necessity with the REs-based PMs being an extremely valuable REs source. Many studies have focused on trying to solve the Nd–Fe–B PMs circular economy, mainly because of their outstanding performance and also due to very high demand for critical applications in electric vehicles, traction and energy conversion motors, and wind turbines [2]. But Sm–Co magnets (SmCo5, Sm2Co17) have not been investigated much so far mainly because of their low market share. However, there are certain technological advantages of Sm–Co over Nd–Fe–B PMs. Sm–Co magnets have an exceptionally high magnetocrystalline anisotropy (Hₐ), which means they are very difficult to demagnetize. Sm–Co magnets are also more resistant to corrosion and can operate at higher temperatures than Nd–Fe–B magnets because of their high Curie temperature (T_C(Sm₂Co₁₇) > 747 °C), which is much higher than that of Nd–Fe–B (312 °C) [3, 4]. This makes Sm–Co magnets the first choice for vehicle-drivertrain applications because they are the most resistant to thermal demagnetization. They are also indispensable for specific application such as traveling wave tubes (TWTs), accelerometers, aeronautical gyroscopes and aircraft-engine micro-motors and micro-generators [5, 6]. Even though the market share of the Sm–Co PMs is more than ten times smaller in comparison to Nd–Fe–B PMs, the current driving force for recycling of Sm–Co is mainly driven by the price volatility of the Co [7] and not so much of the Sm that is one of the RE element. For recycling Sm–Co magnets, widely used methods include direct re-use methods that enables regenerating the recycled magnets utilizing hydrogen decrepitation process to break away the bulky magnet into fine powders for further processing. This process is simple and easy for up-scale without consumption of other chemical reagents. However, the regenerated magnet’s remanence and maximum energy product are generally lower than the original magnet before recycling [8]. Pyrometallurgical routes, such as gas-phase extraction methods have been developed as an alternative for the hydrometallurgical routes. Gas-phase extraction methods transform rare-earth and transition metal from Sm–Co magnets into volatile chloride by chlorination and separate them based on differences in volatility at high temperatures. The anhydrous rare-earth chlorides are concentrated in the deposits in the higher temperature zone (800–900 °C), while cobalt and nickel chlorides are in the lower temperature zone (500–700 °C). The purity of each recovered chloride can be higher than 99%. The chlorides of other metals, such as iron, copper, zirconium, and aluminum, are condensed at the outlet of the reactor (below 350 °C) [9]. Extraction of the rare earths by chlorination requires relatively high temperatures and long reaction times. Additionally, a major problem associated with this method is the high corrosivity of metal chloride with large amount consumption of chlorine gas which is toxic.

Hydrometallurgy by acid leaching process has also been widely used for rare-earth permanent magnet recycling [10]. The conventional way includes the dissolution of the Sm–Co in particular Sm₂(Co,Fe,Cu,Zr)₁₇-based magnets in commercial mineral acids such as nitric, sulfuric, hydrochloric or hydro perchloric. All the contained metal values dissolve. Fe is later precipitated out as Fe-hydroxide, jarosite or goethite via the pH control however, none of the precipitates has a particular value and reduce the final rare-earth yield. So, however, widely applicable, the hydrometallurgical process comes with some drawback, especially in the case of acid usage, wastewater generation, and process waste. There are suggestions on wastewater reduction by saponification [11], but still, large amounts of energy are required during the acid regeneration process after stripping.

The whole hydrometallurgical recovery route thus consumes large amounts of mineral acids and energy and has severe environmental impacts with regards to chemical usage and waste water discharge. Theoretically, the transfer of e⁻ from RE-transition metal-based PMs during leaching can be utilized for metal deposition electrochemically. Consequently, large amounts of mineral acid can be avoided as was already shown for the case of Nd–Fe–B by our group. Our previous studies have shown that via fine tuning of the electrochemical conditions and baths, we can steer the electrochemical leaching from selective to complete leaching of the permanent magnets in question [12, 13]. Herein, the Sm₂(Co,Fe,Cu,Zr)₁₇ magnet scrap (from magnet producing company Magneti Ljubljana Ltd) was used as an example...
to show the feasibility of the electrochemical leaching and deposition to contribute to full recycling route for this magnet type that was performed in a closed loop.

2 Experimental

Dimethylformamide (DMF, > 99%, Sigma-Aldrich, Germany) was purchased from Sigma-Aldrich, Germany. To remove the water from the DMF, molecular sieve (4A, Sigma-Aldrich, China) was added into DMF after being dried under vacuum at 160 °C for more than 24 h, prior to use. CoCl₂ (> 99%, Alfa Aesar) was used as received without further purification.

All dried chemicals were stored inside a closed bottle in a glove box operating under Argon (Ar) gas, with water and oxygen content below 1 ppm. The water concentration determined by Karl Fischer titration (C20S, Mettler-Toledo, Switzerland) in the electrolyte was less than 50 ppm. The Sm₂Co₁₇ (Sm₂(Co,Fe,Cu, Zr)₁₇ magnet scrap used in this study was supplied by Magneti Ljubljana, (Slovenia). After coating removal, the bulk magnets were demagnetized prior to any electrochemical treatment. A three-electrode cell (Fig. 1) was used for all the electrochemical measurements with a Potentiostat (Gamry, Reference 600, USA) device. The electrochemical behavior of Sm₂Co₁₇ magnet in 15 mL DMF containing 0.25 M CoCl₂ at room temperature was evaluated by linear sweep voltammetry (LSV).

A polished Sm₂Co₁₇ magnet flake (2 × 15 × 30 mm³) or a Pt wire (ϕ = 0.5 mm) was used as a working electrode, respectively. A Pt wire (ϕ = 0.5 mm) was used as the quasi reference electrode (QRE), because the potential obtained using a Pt QRE is stable and reproducible in DMF at mid-range temperature. All the potentials reported herein were referred to Pt QRE. A Pt plate (30 × 10 mm²) was selected as a counter electrode. The electrode area of the working electrode and counter electrode immersed in the electrolyte was 0.05 mm² and 80 mm², respectively. For the electrochemical leaching experiments, a Cu foil substrate (40 × 20 mm²) was employed as the cathode and a polished Sm₂Co₁₇ magnet flake (2 × 15 × 30 mm³) was used as the anode (the physical area of the Sm₂Co₁₇ magnet was 2.5 cm²). A Pt wires (ϕ = 0.5 mm) was used the quasi reference electrode. All experimental procedures were carried out in an Ar-filled atmosphere at room temperature. The current of 5, 10 and 20 mA was applied at the time period of 0–240, 240–480 and 480–720 min, respectively. The concentrations of Sm³⁺, Cu²⁺, Co²⁺, Fe²⁺ and, Zr²⁺ in the solution were measured by mass spectrometry with inductively coupled plasma (ICP-MS, Agilent 7700, Agilent Technologies, Tokyo, Japan) operating at 20 kV accelerating voltage, 0.25 nA beam current and at working distance of 15 mm. The crystal structure of the leached magnet was determined by X-ray diffractometer (XRD, PANalytical, Netherlands).

3 Results

3.1 Microstructural investigation of Sm₂Co₁₇ magnet scrap

Figure 2a shows the back-scattered electron (BSE)-SEM image of the initial polished Sm₂Co₁₇ magnet scrap. At low magnetizations three phases with some pores are observed on the polished surface in the inset of Fig. 1a which is very typical as reported elsewhere [5, 14]. The dark grey phase is the “matrix phase” (labeled as “1”) which contains 11.1 at. % Sm, 58.5 at. % Co, 22.4 at. % Fe, 5.8 at.% Cu and 2.2 at.% Zr confirmed by EDXS. The white phase (labeled as “2”) consists of 65.1 at.% O and 34.9 at.% Sm which is supposed to be Sm₂O₃ phase. The void-like phase (labeled as “3”) only contains Zr which is metallic Zr. The grain boundaries are not distinguishable in this case similar to reported literature [14]. There were indications that the phase is hexagonal with Th₂Ni₁₇ structure but it was later reported to be hexagonal TbCu₇ phase, implying
that the material consists of supersaturated TbCu₇ phase with partial Tb₂Ni₁₇ order [16]. They anticipated that 2:17 cells nucleate and grow out of 1:7 phase that induces an increase in Cu concentration. It was stated that the solubility of Cu 2:17 at 850 °C is ~4 at.% [17], and Sm content in the surrounding 1:7 matrix, assumes 1:5 stoichiometry and forms the cell boundaries [14]. Sm₂Fe₈.5Co₈.5 (2:17) phase which has the highest peak intensity is the main component of the magnet. Combining the XRD result with the microstructure of the magnet, the grey phase (1) is a mixture of 2:17 phase/1:7 phase and 1:5 phase which agrees with the literature [18–20]. The peaks for Sm₂O₃ phase and metallic Zr phase are not labeled because they are overlapping by the peaks of the matrix phase in the XRD pattern due to their low content in the original magnet.

3.2 Electrochemical behavior of Sm₂Co₁₇ magnet scrap

Figure 3a shows the linear sweep voltammetry (LSV) of a Pt electrode (black curve) and the initial Sm₂Co₁₇ magnet scrap (red curve) in DMF containing 0.25 mol L⁻¹ CoCl₂ with the scan rate of 20 mV s⁻¹. In the black curve where Pt was used as the working electrode, current density is stable at around 0 mA cm⁻² from −0.15 to 1 V indicating no reduction or oxidation reaction. When Sm₂Co₁₇ magnet was used as the working electrode, the current density starts to increase at the potential of −0.1 V along AB portion shown in the red curve in Fig. 3a which is caused by the oxidation reactions on the anode. From the point B on, the current density increases regularly along BC.
For all the electrolysis experiments, the following oxidation reactions of the 1:5, 1:7 and 2:17 phases are possible at the anode:

\[
\begin{align*}
\text{Sm} - 3e^- & \rightarrow \text{Sm}^{3+} \quad (1) \\
\text{Sm(Cu, Co)}_5 & - 13e^- \rightarrow \text{Sm}^{3+} + 5(\text{Cu}^{2+}, \text{Co}^{2+}) \quad (2) \\
\text{Sm}_{0.8}\text{Zr}_{0.2}\text{Co}_{5.36} & - 13.6e^- \rightarrow 0.8\text{Sm}^{3+} + 0.24\text{Zr}^{2+} + 5.36\text{Co}^{2+} \quad (3) \\
\text{Sm}_2\text{Fe}_{8.5}\text{Co}_{8.5} & - 40e^- \rightarrow 2\text{Sm}^{3+} + 8.5\text{Fe}^{2+} + 8.5\text{Co}^{2+} \quad (4)
\end{align*}
\]

The most possible reduction reactions according to element’s standard reduction potentials on the cathode are as following:

\[
\begin{align*}
\text{Co}^{2+} - 2e^- & \rightarrow \text{Co} \quad (5) \\
\text{Fe}^{2+} - 2e^- & \rightarrow \text{Fe} \quad (6) \\
\text{Cu}^{2+} - 2e^- & \rightarrow \text{Cu} \quad (7)
\end{align*}
\]

The peak (P1) of 5 mA cm\(^{-2}\) recorded at 0.17 V is speculated for the oxidation of Sm-based phases, most probably Sm(Cu,Co)\(_5\) phase (1:5, reaction 2) and Sm\(_{0.8}\)Zr\(_{0.2}\)Co\(_{5.36}\) phase (1:7 (2:17), reaction 3). Because Sm is preferentially prone to be oxidized due to its highly negative electrode potential (reaction 1). Therefore, higher Sm content (with the standard reduction potential \(E^0 = −2.68\) V vs SHE) in the crystal phase possibly prioritize the leaching / oxidation. The continuous increase of current density in BC portion is probably caused by the oxidation (reaction 4) of the Sm\(_2\)Fe\(_{8.5}\)Co\(_{8.5}\) phase in the magnet.

Figure 3b–d show a representative microstructure of the Sm\(_2\)Co\(_{17}\) magnet surface with different magnifications after electrochemical leaching by LSV at room temperature. It can be observed that the grain boundary phase Sm(Cu,Co)\(_5\) located between the matrix grains was leached away, leaving behind the Sm\(_2\)O\(_3\) phase (white phase in Fig. 3b) which is not prone to be electrochemically oxidized i.e., leached. The dark grey crystals shown in Fig. 3c are metallic Zr which is less prone to be leached (\(E^0 = −1.45\) V vs SHE), compared to the 1:5 phase, that contains the highest amount of Sm, among all the Sm containing phases in the magnet. It can be observed that leaching took place leaving behind hollow spaces and exposing a 3D network of an unleached structure which is shown in Fig. 3d. The diameter of the hollow spaces in the network structure varies from 50 to 200 nm. Thus, the network structure shown in Fig. 3d is formed by preferentially leaching away the Sm(Cu,Co)\(_5\) grain boundary between the micron sized grains and leaching away the Sm–Co 2:17 phase in the nanocells, leaving behind the nanoscale cell boundaries in stoichiometry 1:5 i.e., Sm(Cu,Co)\(_5\)* (dotted here with *, to distinguish it from the micro Sm(Cu,Co)\(_5\) grain boundary).

### 3.3 Anodic leaching of Sm\(_2\)Co\(_{17}\) magnet scrap by direct current

The leaching of Sm\(_2\)Co\(_{17}\) magnet scrap was performed using direct current of 2, 4 and 8 mA/cm\(^2\) at the time period of 0–240, 240–480 and 480–720 min, respectively. The leached metallic phases dissolve in the electrolyte in the ionic form. Figure 4 shows the concentrations (measured using ICP-MS) of Co\(^{2+}\) (Fig. 4a), Cu\(^{2+}\) (Fig. 4b), Fe\(^{2+}\) (Fig. 4c), Sm\(^{3+}\) (Fig. 4d) and Zr\(^{2+}\) (Fig. 4e) that are monitored in the electrolyte solution under different leaching current along with the leaching time.

The concentration of Sm\(^{3+}\), Cu\(^{2+}\), Fe\(^{2+}\) and Zr\(^{2+}\) increases linearly along with the leaching time under the current of 5, 10 and 20 mA, respectively. Reversely the concentration of Co\(^{2+}\) decreases linearly due to its consumption on the cathode by electrodeposition. The leaching rate of Sm\(^{3+}\) increases linearly from 0.0117 to 0.0797 mM L\(^{-1}\) min\(^{-1}\) with an increasing current from 5 to 20 mA. The concentration increases of Fe\(^{2+}\) and Zr\(^{2+}\) become faster with 10–20 mA than 5–10 mA, possibly ascribed by its consumption of cathodic deposition for the Fe case. The concentration of Cu\(^{2+}\) doubles with increasing current from 5 to 10 mA, however, it goes back to the same rate with a further increase of current to 20 mA. This can possibly be attributed to the selective leaching of Cu from the 1:5 and 2:17 phases and then from the 1:5* phase, accompanied with and increased deposition of Cu at the cathode with time which consequently reduces the amount of the ionic Cu\(^{2+}\) in the solution.

The consumption of Co\(^{2+}\) by electrodeposition on the cathode is faster at the beginning of leaching due to its initial high concentration. The consumption rate of Co fluctuates from 5 to 20 mA which might be caused by the composition change of the electrolyte along with time where the deposition of Fe and Cu happened instead of the pure Co deposition.

### 3.4 Metal deposition on the cathode

During anodic leaching, metal deposition occurs on the cathode in parallel in order to keep the charge balance. Figure 5 shows the elementary composition of the deposit on the cathode at the current of 5, 10 and 20 mA for the first 60 min. As can be seen, the deposits contain Co, Fe and Cu. Sm and Zr were not co-deposited which may be because of the much higher concentration of transition metal ions (TM, Co\(^{2+}\) and Fe\(^{2+}\)) which could not trigger the co-deposition and negative standard electrode potentials of the metals in question. Figure 5 shows that Co is the main content of the
deposit (85–93 at.%). The Fe content varies from 5 to 11 at.%). The fluctuation of Co and Fe in content is caused by the concentration changes of Co²⁺, Fe²⁺ and Cu²⁺ in the electrolyte. The content of Cu in the deposit increases from 0 to 4 at.% with the increasing current. Iron concentration in the electrolyte directly depends on the anodization rate of Sm–Co, i.e., the applied current. While the iron concentration in the electrolyte influences the iron content in the deposited alloys. As the major ion species of the ions in the electrolyte is Co²⁺, therefore, the influence of the iron concentration on the iron content in the deposit is limited, which is in a good agreement with the literature [21]. Because Cu³⁺ is the most prone ion to be deposited in the electrolyte, in terms of its electrode potential.

### 3.5 Morphology and crystal phases of the leached magnet

Figure 6a–c shows the microstructure of the magnet after leaching for 720 min. It can be seen from Fig. 6a that the cell boundaries were partially leached which results in the porous structure. This indicates that there is some preferential leaching of the phases in the Sm₂Co₁₇ magnet obeying the leaching priority order Sm(Cu,Co)₅ > Sm₂Co₁₇ > Sm(Cu,Co)₅* that is an interplay between the Sm content that is the least noble metal and Cu which is the most noble metal in the investigated magnet. Figure 6b shows the front surface morphology of the leached magnet after leaching for 720 min. Some particles containing Sm, Cu, Zr and O with the size
from 100 nm to 3 μm were found. Going deeper into the magnet, we can observe a network structure that consists of the 1:5*-type cell boundaries in Fig. 6c which is similar as shown in Fig. 3d.

Figure 6d shows the XRD pattern of the leached magnet after 720, from which the Sm2O3 (PDF# 01-080-7746), CuO (PDF# 01-078-0428) and Zr (PDF# 04-006-2825) phases are observed. It could be understood that Sm2O3 and CuO remain unleached since both of them are electrochemically inert. Sm2O3 and metallic Zr may come from the original magnet shown in the inset of Fig. 2a. The appearance of metallic Zr in the leached magnet might because of its incomplete leaching since the Zr2+ ions were detected in the electrolyte by ICP-MS. CuO is possibly a component of the cell boundaries, according to the reported literature [20, 22, 23], but peaks overlap with Zr, so it is hard to claim that.

4 Discussion

The anodic dissolution i.e., the leaching of the Sm2Co17 scrap magnet, has its route in the complex Sm2Co17 magnet microstructure, the chemical, and the electrochemical nature of the phases. Sm2Co17-type magnets have peculiar microstructure, both on micro and nano levels. According to the literature [24], the Sm–Co 1:5 phase generally precipitates in the micro-grain boundaries. Therefore, it could be speculated that Sm(Cu,Co)5 phase sitting in the grain boundaries was leached away preferentially as seen from Fig. 3. Further it is known that the cellular microstructure of the Sm2Co17 magnet is the key feature for the high coercivity in these kinds of magnets [18, 20, 22], with the average cell size of the 2:17 phase is around 100 nm and the average cell boundary width is around 10 nm. Upon that it could thus preliminary concluded that the 1:5 microscale cell boundary and the 2:17 cells are preferentially leached, compared to the nanoscale cell 1:5* boundaries. The first assumption was that the amount of Sm, which is the least noble metal in the system, dictates the leaching regime, with leaching starting at the magnet surface attaching the Sm-rich phases.
However, the leaching sequence of the phases observed is not following the Sm content in the phases. Thus, the explanation most probably lies in the phase’s nominal compositions. As stated in [25] these phases vary in the Cu content with an average Cu content reported to be 5.4 wt% in microscale 1:5 grain boundary, 18.4 wt% in the nanoscale grain 1:5* boundary in-between the 2:17 grains and 5.3 wt% in the interior of the 2:17 nanograins. As, it is known that alloying of the elements with more noble metals increases their stripping potential [26], the most reasonable explanation would be that the phases that contain less Cu preferentially strip in contrast to the phases that contain more Cu (that is the nanoscale cell 1:5* boundary in our case). To better understand the leaching process in three-dimensional space, a mechanism for the electrochemical leaching of the sintered Sm₂(Fe,Co,Cu,Zr)₁₇ magnet with 1:5 micro-grain boundary, 1:5* nanograin boundary, and 2:17 cell interiors is the following. At the very beginning, the anodization begins with the leaching of the 1:5 micro-grain boundary phase, as the electrochemical potential is always higher at the electrode edges [6]. Leaching of the magnet proceeds until a certain time, when a certain thickness of 1:5 micro-grain boundary phase has been leached away, now exposing the noncellular grains of 2:17 phase to be leached—the leaching depth d for the increased time of leaching increases.

Consequently, the exposed area of the anode/magnet becomes longer times, which decreases the current density and the over-potential for leaching. As a result, the leaching rate 1:5 micro-grain boundary reduces with leaching time. When a constant current is applied and to keep the charge-balanced, the leaching of the 2:17 grains needs to occur at the leached thickness at prolonged times, leaving the 18.4 wt% Cu containing 1:5* boundary phase to be leached last. Upon that, we can conclude that the electrochemical leaching of the Sm₂Co₁₇ magnet can convert all the metallic phases into an ionic form that has potential for further REEs separation and extraction. The deposition of the transition metals like Co, Fe, and Cu is simultaneously done on the cathode. As the electrochemical potentials of Co²⁺/Co (− 0.22 V vs. HER) and Fe³⁺/Fe (− 0.447 vs. HER) are close to each other, it is hard to deposit single metal elements individually. Selectively extracting the metal elements could be realized by ionic liquid-based selective precipitation process [27] or solvent extraction process [28].

In summary, the current state-of-the-art routes for the recycling of the end-of-life (EOL) Sm–Co magnets or Sm–Co swarf use dilute mineral acids (H₂SO₄, HCl, HNO₃) [1] to convert the metallic phases into ionic form. These chemical processes convert all the metallic elements from scrap Sm–Co magnets to their corresponding ionic species by electron (e⁻) transfer. However, after the separation, reducing these ions requires additional e⁻ sources, resulting in additional energy usage and wastewater generation.

This work introduced an electrochemical recycling route for Sm–Co magnet that avoids using any mineral acid and produces transition metal alloys (on the cathode) as high-value products under room temperatures. We shorten the magnet dissolution and acid regeneration cycle and completely reduce the amount of wastewater and Fe-based waste. Compared to current hydrometallurgical and pyrometallurgical routes, this work developed an environmental-friendlier route that could be potentially applied in rare-earth industry. Some issues would have to be resolved before the process gets developed further towards commercialization, with investigations focusing on (1) volatility of the electrolyte. DMF used in this work is volatile, because of which, the electrochemical cell has to be completely sealed in order to keep the volume of the electrolyte constant. Only batch processing could be applied in a sealed system for practical purposes. An organic electrolyte with a high boiling point needs to be developed. (2) The process so far is developed only for bulk Sm–Co magnets to be used as the anode. However, for recycling the Sm–Co magnet swarf, for example, generated from the polishing and cutting process cannot be directly used as the anode. The surface of this sludge is already oxidized to form a non-conductive insulation layer. The current oxidation rate of the Sm–Co magnet can further be accelerated via using different oxidative chemicals. However, using an oxidant possibly introduces other elements that may negatively influence the deposition process on the cathode. More importantly, one more step might be needed to remove the added oxidant, leading to a longer recycling route.

5 Conclusion

Sm₂Co₁₇ magnet scrap was leached by anodic oxidation. LSV study combined with SEM/EDXS nad XRD study revealed that there is some preferential leaching of the phases in the Sm₂Co₁₇ magnet obeying the leaching priority order Sm(Cu,Co)₁₅ − microintergranular phase, Sm₂Co₁₇ nanocells > Sm₂Cu₁₅* − nanogranular phase, that is an interplay between the Sm content that is the least noble metal and Cu content in these phases which is the most noble metal in the investigated magnet system. On the cathode part, metallic Co–Fe–Cu alloy was deposited in parallel among which Co content was more than 85 at.%. The current study shows novel aspects in recycling of rare-earth-based permanent magnets (Sm₂Co(Fe,Cu,Zr)₁₇-type in our case) using electrochemical anodization. This process is used to overcome the issues of the current hydrometallurgical processing, where significant amounts of acids are primary used to leach the magnets.
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Author contributions All authors contributed to the study conception and design. Material supply and preparation was performed by [MK, JB, XX], the data collection and analysis was performed by [XX, SK, ZS, KM, JS, SS, KZR]. The first draft of the manuscript was written by [XX and KZR] and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript. Conceptualization: [XX, KZR]; Methodology: [XX, MK, JB]; Formal analysis and investigation: [XX, SK, ZS, KM, JS]; Writing—original draft preparation: [XX, KZR]; Writing—review and editing: [KZR]; Funding acquisition: [KZR, SS, MK, JB]; Resources: [-]; Supervision: [KZR].

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