Rare earth-based permanent magnets: A proposed way to the circular economy

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ABSTRACT

Critical raw materials, especially the rare earth metals like Dy, Nd, Sm, and recently also the transition metal Co are becoming more and more important to Europe's future energy independence, and offer the ability to be competitive in smart mobility and renewable energy innovation. The primary goal of the efforts from the Department for nanostructured materials from Jožef Stefan is to implement the state-of-the-art laboratory-developed & economically efficient technologies for the recycling and reprocessing of critical metals from end-oflife products. The aim is to integrate them into industrially relevant processes in order to reduce Slovenia and Europe's dependence on economically and strategically sensitive supplies and to increase their competitiveness on international markets.

This article depicts a strategic issue of the European Union in the field of technology transfer, which should benefit the research community and the economy. However, this issue is not being addressed at the proper level: the scientists and industry are working to solve the technical problems, but are not supported sufficiently on the political level.

Keywords

Critical raw materials, rare earth elements, permanent magnets, Nd-Fe-B, Sm-Co

POVZETEK

Kritične surovine, zlasti redke zemeljske kovine, kot so Dy Nd, Sm in v zadnjem času tudi prehodna kovina Co, postajajo vse pomembnejše za prihodnjo evropsko energetsko neodvisnost in ponujajo sposobnost konkurenčnosti na področju pametne mobilnosti in inovacij iz obnovljivih virov energije.

Primarni cilj prizadevanj Oddelka za nanostrukturne materiale Jožefa Stefana je uvajanje najsodobnejših laboratorijsko razvitih in ekonomsko učinkovitih tehnologij za recikliranje in predelavo kritičnih kovin iz izrabljenih izdelkov. Cilj je vključiti jih v industrijsko pomembne procese, da bi zmanjšali odvisnost Slovenije in Evrope od ekonomsko in strateško občutljivih zalog ter povečali njihovo konkurenčnost na mednarodnih trgih.

Članek prikazuje strateško vprašanje Evropske unije na področju prenosa tehnologije, ki bi moralo biti v korist raziskovalni skupnosti in gospodarstvu. Vendar se to vprašanje ne obravnava na ustrezni ravni: znanstveniki si skupaj z industrijo prizadevajo rešiti tehnične težave, vendar na politični ravni še niso dovolj podprti.

Ključne besede

Kritične surovine, redkozemeljski elementi, trajni magneti, Nd-Fe-B, Sm-Co

1. INTRODUCTION

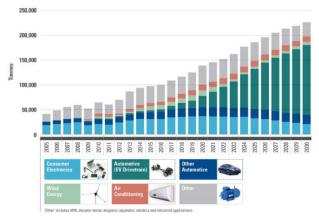
EU plans for the transition to a low-carbon society and energy efficiency by 2050 (the so-called European Green Deal) [1] will require radical solutions, especially with the aim of reducing greenhouse gas emissions, which are projected to reduce by as much as 80%. The segments that will contribute the most are the development of green energy and electric mobility. The latter will require highly efficient electric motors to achieve this goal. The efficiencies of electric motors (mass versus efficiencies) based on permanent metal magnets of rare earth elements (such as Nd₂Fe₁₄B and SmCo₅, Sm₂Co₁₇) both sintered and bonded are known to be higher than induction motors, which contributes mainly in terms of miniaturization of devices with preserved or even improved efficiencies. From this point of view, permanent magnets are a hot subject to further research with the aim of improving their state-of-the-art properties. However, rare earth metals based on rare earth metals are on the list of the most Critical Raw Materials (CRM) important for the EU, which will require their comprehensive treatment in the form of their complete use without and waste

and their efficient recycling of both systems using novel recycling processes that are being developed on the department.

2. CRITICAL RAW MATERIALS

2.1 EUs dependency on critical raw materials and their applications in permanent magnets

One of the major problems EU has been facing since 2011 is ensuring a sustainable access to Critical Raw Materials [2], in particular elements of the lanthanide group, i.e. rare earths. The group understands 15 + 2 elements, the most characteristic and useful of which are permanent magnets: Neodymium, Samarium, Dysprosium and Terbium with lately also Co, that is a transition metal. A key factor influencing that is their natural abundance and related production in only a few countries, such as China, Brazil, Russia, Australia and the Democratic Republic of Congo. Limited access and the political manipulations concerning the CRM issues are attributed to the way some of these countries use trade and tax policies to reserve their natural resources exclusively for their own use. China for the moment controls as much as 84% of the world's rare earth mineral production. Although the trade restrictions that have peaked in 2011 have declined at the moment, fear of a new material crisis still persists.



A Figure 1: Current consumptions of Nd-Fe-B PMs by application and future predictions [3]

A key component of the Europe Green Deal is to accelerate the "transition to sustainable and smart mobility", as transport accounts for a quarter of CO2 emissions. That is why the electrification of the transport system is receiving large investments and research at the global level. Company Tesla, as the first mass producer of electric vehicles alone, is increasing production to 500,000 vehicles by the end of 2020 and with the expansion of its production plant in Shanghai and the opening of a new one in Berlin in the coming years reached as many as one million new e-vehicles on the market. Also, other major car manufacturers such as Toyota, Honda, Kia, Renault e.g. invest significantly in development and e-production. Volkswagen alone is expected to produce as many as 1.5 million e-vehicles by 2025. In 2011, the EU gave priority to rare earths as the most critical CRMs, but in the years since, it has focused mainly on permanent magnets made out of them based on two alloy systems, namely neodymium-iron-boron (Nd-Fe-B) and samarium-cobalt (Sm-Co). The latter systems are given the highest priority, as they are crucial in e-vehicles in their drive motors, servo controllers, starting motors and regenerative brake generators. The projected consumption and use of permanent magnets based on Nd-Fe-B and Sm-Co is shown in Figure 1. Today, the industry consumes 50,000 tons of these

magnets for powertrains in e-vehicles, and consumption is expected to grow to 150,000 tons in the next 10 years. Here, the EU is in a difficult position, as it has no active rare earth mines, so it has to import up to 90% of rare earth-based permanent magnets, while European producers of permanent magnets can be counted on the fingers of one hand. Here, Slovenia is strongly represented by two manufacturers of permanent magnets, namely Magneti Ljubljana d.d. and Kolektor Group d.d., which have managed to maintain a competitive advantage to this day, that gives Slovenia and enormous potential and advantage.

2.2 Novel solutions in Rare earths-based permanent magnets circular economy

2.2.1 The state of the art of the technology

From SICIRS it is evident that, diverse methodologies for recycling Nd-Fe-B magnets have been summarized in detail by many authors [4,5,6]. The recycling approaches can be broadly classified into physical/mechanical processing, pyrometallurgical and hydrometallurgical separation & recovery. Physical/mechanical processing, including resintering [7,8], hydrogenation disproportionation desorption and recombination [9-11], of sintered Nd-Fe-B magnet scrap will typically have a smaller environmental footprint compared to recycling routes, which rely on stripping of the REEs. The pyrometallurgical routes can be used to remelt the REE alloys and extract the different REE in the form of oxide, halide, fluoride or other metallic compound which can then be reduced to metallic form [12-17]. However, these pyrometallurgical processes operate at a temperature of around 750-950 °C and are thus energy intensive. Hydrometallurgical recycling processes designed for Nd-Fe-B magnets are promising due to the mild operating temperature, relatively simple equipment and the continuous separation ability [18, 19]. In hydrometallurgical processes, however, Nd-Fe-B magnets are completely dissolved with an acid. The roasting pretreatment at 900 °C is generally required. Iron, which is the major component of Nd-Fe-B magnets (60-70%) consumes large amount of acid, alkali and other precipitation agents that cannot be recycled in the whole process [5, 19]. REEs are concentrated by solvent extraction and then are precipitated with either oxalic or carbonic acid. The precipitate is further calcined at 950 °C to form REOs, which can then be returned to the initial manufacturing process for Nd-Fe-B magnets [20]. We also reviewed the patent documents using the queries below, from Patbase document system. Results were the following: The most populated field is the one including ((Nd₂Fe₁₄B or NdFeB or Nd-Fe-B) as earth particulate material)) in the title or abstract AND FT=(grain boundary*) anywhere in the text. This yielded 97 patent families. On the other hand, using earth particulate material)). Yielded some less, 74 families. On the other hand the search showed that ((Nd2Fe14B or NdFeB or Nd-Fe-B) and single crystal anodic etching is a rather unpopulated field with 0 patent families present at the moment and that our technology is not only operational, but worth exploring in the sense of novelty.

In the proposed method the Nd₂Fe₁₄B grains are recovered by electrochemical etching of the bulk sintered Nd-Fe-B magnets or magnet scraps using an anodic oxidation process presented in Fig. 2 [21,22]. In this process the metallic Nd-rich phase in the grain boundaries is oxidized to Nd³⁺ as ions on the anode. The liquid electrolyte used in this process is formed of a non-aqueous solvent in order to prevent the Nd₂Fe₁₄B grains from oxidation. This allows direct reuse of the collected Nd₂Fe₁₄B grains for new magnet making.

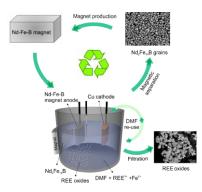


Figure 2: Selective electrochemical etching for recycling of Nd-Fe-B permanent magnets

2.2.2 The economics of the recycling

One of the purposes of the paper is show that although the magnets are needed in Europe, the fact that the rare-earth elements come mostly come from outside Europe presents an intriguing moment in the development of the technology transfer processes in Europe, in line with the recycling recommendations.

The economics of the process of recycling in the field of the rare earth magnet shows that investing into some local technology that would enable extraction from recycled components would benefit the environment and the countries of the EU that do not poses rare earth material sources. However, also such advanced recycling would still carry costs that result in 'virgin materials' being cheaper. Thus, considering the economics of the processes and the recommendations of the EU, we must conclude that the changes into a sustainable economy will remain impossible without legislative changes within the EU that are crucially needed to encourage this activity and contribute to the circular economy.

Thus, as a result of our research, we would like to propose some concrete measures to improve the position of the recycling processes of the rare earth metal components in the EU.

A novel recycling route for end of life (EoL) Nd-Fe-B magnets is thus proposed based on the electrochemical etching. Electrolyte can be recovered by distillation and re-used in a closed-loop thus minimizing safety risks and environmental impacts. Upon that the overall REEs mass balance from the initial magnet is 100% preserved that forms a circular economy. The total energy consumption of the magnet-manufacturing process using the proposed electrochemical recycling route is estimated to be ~2.99 kWh kg-1, which is much lower than hydrometalurgy (30.0-33.4 kWh/kg) and directly comparable to direct reuse (3.0 kWh/kg) [8], if we consider the conventional additive of the Nd-Pr hydride (4 wt.%), inclining to as feasible possible production, albeit very green and sustainable. We have shown that recycling process costs are actually a barrier in enlarging the usage of such processes industrially in the EU. Thus we propose a more targeted intervention that would tip the balance towards the recycling processes not only in regard to the rare earth materials, but all that are not applicable in significant enough amounts to be economically viable. The situation could greatly be improved if the EU could import the relevant waste from other regions of the world, which would enable cost reductions of the processes, based on the quantity. On the other hand, the EU could even - maybe - become selfsufficient in the supply of the rare earth materials. This is also a policy that would provide a significant and a wide-ranging impact in other European recycling technologies, dealing with

economically negligible amounts of waste that do not prove economically sustainable for recycling.

2.2.3 *The solutions to be used for sustainability*

Persistent measures to achieve greater sustainability and independence from external suppliers, thus include, among other things, the recycling of industrial wastes and end-of-life products. Permanent magnets based on Nd-Fe-B and Sm-Co systems (as Co, as it is similar to rare earths subjected to major political and economic manipulations) due to the high content of these metals represent the most valuable secondary source of these raw materials. Currently, less than 1% of all rare earths used are recycled, mainly because they are dispersed in many applications, and are difficult to extract. A lot of labor force is therefore needed and the economic calculation does simply not add up.

Currently, the only way to recycle rare earth-based permanent magnets from waste streams of electrical and electronic equipment is by crushing and recycling using physical, chemical or pyrometallurgical pathways, which are costly, energy consuming and environmentally unfriendly. Upon that the developed novel feasible and green solutions for recycling REEs-based permanent magnets are of tremendous impact. The proposed technology for selective PMs leaching [22] and a related technology (EP 019 197 716.4) for complete electrochemical PMs leaching and REE recovery are in the patent procedure at EU Patent office. Efforts are also being made towards permanent magnets circular economy also on the national (ARRS L2-9213, L2-1829) with Magneti Ljubljana Ltd and Kolektor Group Ltd and international level via several European projects that encompasses the mentioned industrial partners in Slovenia and all over EU (ETN-DEMETER, H2020 SUSMAGPRO, ERA MIN II MAXYCLE, EIT RAW MATERIALS INSPIRES). Within H2020 proposal SUSMAGPRO TRLs of 7-9 are aimed via three pilot plants for recycling of EoL permanent magnets that are planned in Europe. Recently we got awarded the EIT RAW MATERIALS proposal on recycling REEs-based permanent magnets from white goods, where we collaborate also and also with Slovenian companies Domel Ltd, Gorenje Ltd, Surovina Ltd and Zeos Ltd.

3. CONCLUSIONS

Despite success stories, the challenge still persists when transferring the technologies form lab scale to functioning production lines, as the requests from the industry are strictly connected with the economic feasibility.

However, the proof of concept of the novel technology is shown on the lab scale reaching TRLs 3-4 and represents only an initiation that a technology could be feasible. Upon that much more investments would have to be made for "technology transfer" projects, to bridge the exact TRL gap between 5-7 like SUSAGPRO. In order for EU to become CO2 zero efficient in to compete with the far East when it comes to be CRMs independent, the investments in the whole value chain on recycling of PMs have to be made. Slovenia for example has an extreme potential to act as a role model or as a feasible permanent magnet circular economy closed loop example, as it holds a geographical, professional and economical potential as to serve as a central location for the collection of waste magnets and their remanufacturing based on rare earths from the central and eastern parts of the European Union. The later has been recently successfully recognized by the EIT RAW materials scheme via funded INSPIRES project. The use of local

suppliers would significantly reduce carbon emissions and it is expected that in a few years Slovenia could produce 10 to 40 tons of Nd-Fe-B alloy magnets per year on the basis of recycling within the European SUSMAGPRO project. And the successful model could be later applied in different EU countries, using the recourse from EoL wind mills for example (like Scandinavian countries). However, this is not going to be possible without legislative changes within the EU that are crucially needed to encourage this activity and contribute to the circular economy, not to forget the most important thing the stimulations from the local governments and European Investment Bank.

This would strongly encourage local productions of rare earth secondary minerals and permanent magnets. Otherwise, the European rare earth industries i.e. permanent magnets will remain exposed to fluctuations in open market prices, making them very vulnerable and consequently uncompetitive.

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