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Elements of circular economy in the manufacture of NdFeB permanent magnets IN ICPE-CA

GEORGETA ALECU*, WILHELM KAPPEL

*National Institute of Research and Development for Electrical Engineering ICPE-CA,
313 Splaiul Unirii, 3, Bucharest, Romania*

Abstract. In the life cycle of the product, the link between the initial phase and the final stage in its realization is the recycling through the rehabilitation of materials in the production process. This gives circularity to the economy, thus minimizing consumption of natural resources, optimizing manufacturing costs, creating new jobs, developing business. The paper describes in a conclusive example the defining elements of the circular economy. We present this example for the simplicity of our demonstration. This mode allows a direct and edifying treatment of the defining elements of the circular economy. We have chosen as a product the permanent magnets based on rare earths of the NdFeB type. The choice is targeted because NdFeB permanent magnets are expensive products, so the benefits being obvious. We demonstrate the manufacturing of NdFeB permanent magnets with defined magnetic properties from raw materials, their use in products (for ex. in synchronous electric motors) and at the end of the life cycle of motors, the recovery of permanent magnets with the possibility of obtaining the powders used initially in the technological process to manufacture new NdFeB permanent magnets with similar magnetic properties and the possibility of their use in electrotechnical products. Even if, following a piece to piece sorting of the permanent magnets after magnetic parameters of the resulting products would remain scrap, they could be recycled in the same way so that the amount of waste would ultimately be minimal. Finally, we present a recycling technology of NdFeB permanent magnets based on one of our patents filed in 1998.

Key words: circular economy, recycling, waste, permanent magnets NdFeB

1. Introduction

At European and global level, the concept of circular economy has a good visibility, [1]. The focus is on the recycling of waste and products at the end of their life (EOL) cycle but also on the way the products are made, so that the resulting waste is in the

*Correspondence address: georgeta.alecu@icpe-ca.ro

smallest quantity, to waste as little as possible and to reuse as much as possible. Basically, the circular economy implies the efficiency of the resources in order to produce more economic value with the same or less resources. In opposition to the basis of the consumer society, the circular economy aims to redefine the economic growth and gradually, partially or totally, is given up on the consumption of resources: urban mining, [2], aims to recover the resources from waste and from the products reached at the end of their cycle. life, then reusing or recycling them. Thus, the proportion of the raw materials from recycling in the total of the raw materials used is increased, as well as the degree of recyclability of the products at the EOL cycle. A complete life cycle should improve the use of secondary materials and create the appropriate economic incentives to avoid creating waste and reusing it. It is realized the so-called "industrial symbiosis" that can use the waste from one production process as a raw material for another.

For the EU raw materials are essential, but in the world their availability become more and more difficult. After the EU Raw Materials Initiative, [3], it was decided to identify a list of critical raw materials at EU level. Also we underline that the production of many materials is concentrated in a small number of countries: more than 90% of rare earths (RE) and antimony (Sb), and more than 75% of Ge and W from the world productions take place in China, 90% of Nb in Brazil and about 77% of Pt in South-Africa. Many times, metallic elements which has an important role in modern technologies are by-products of the mining and production process of other important metals used in the industry: for example, Zn, Al and Cu can be here mentioned. For such a mining of important technological metallic elements, it must be taken in the account, that the opening of an Al, Cu or Zn mine is a long time process (up to 20 years could be the necessary time period), so that the availability of rare metals, which can be called as "critical", is many times maladaptive to quick changes. In order to avoid such challenges, the European Commission has launched an integrated strategy in November 2008: the EU Raw Materials Initiative, which address measures in three areas: to secure sustainable access from outside Europe, improving framework conditions for extracting minerals within Europe and promoting the recycling and resource efficiency of such materials, [4]. Another situation is the deposits in which metals may occur as "coupled elements" without a real carrier metal: as an example is the group of rare earth elements (REE). With regards to geological availability, the Group of the EC observes that, as geological scarcity is not considered as an issue for determining criticality of raw materials, global reserve figures are not reliable indicators of long term availability, [4].

Of greater relevance regarding the attribute to be critical of a raw material are changes in the geopolitical-economic situation. that can have an impact on the supply and demand of raw materials. A raw material is labelled "critical" when the risks of supply shortage and their impacts on the economy are higher compared with most of the other raw materials.

The mentioned study considers that the REE are critical and that means that the products using such raw materials will be manufactured with increasing difficulties, [5]. In this category are included permanent magnets (PM) which contain such

elements; today the most powerful PM has in their composition REE. Nowadays the most used PM are the performant PM based on NdFeB, [6]. The high demand increases the expected price. The new prices might be a starting point to build up recycling systems for rare earth compounds, [7]. Only a few industrial recycling activities are currently implemented for rare earths. Until now, there has been no large scale recycling of rare earths from PM. Principally, the recycling processes for the rare earths are quite complex and expensive if re-use is not possible and a physical and chemical treatment is necessary. Most of the recycling procedures are energy-intensive processes. The main post-consumer activities – the recycling of rare earths from electric motors and hard disks and other electronic components – will require intensive dismantling. Several constraints for a wider recycling of rare earths were identified: the need for an efficient collection system, the need for sufficiently high prices for primary and secondary rare earths compounds, losses of post-consumer goods by exports in developing countries and the long lifetime of products such as electric motors in vehicles and wind turbines of 10 - 20 years before they could enter the recycling economy, [7]. The potential of the EU to recycle some very important REE for PM, is shown in Fig.1a for Nd and in Fig. 1b for Dy, [8], both REE has an around 30% weight content in PM based on the system NdFeB.

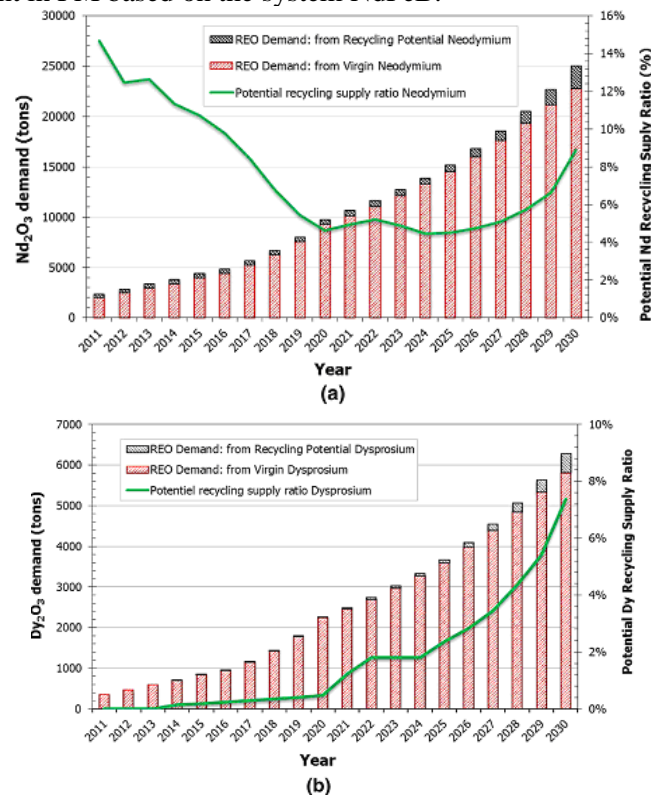


Fig. 1. Potential of recycling in the EU of important REE in the composition of NdFeB PM, Nd and Dy in 1b (after [8])

Considering the quantities of REE which are included in the up today produced PM of the type NdFeB, approximately 65 000 t of Nd and 20 000 t of Dy, results from the Fig.1 that recycling of Nd and Dy could be an important source of REE in the NdFeB based manufacturing of PM, [9]. The use as raw materials of the recycled ones are also beneficial in terms of CO₂ emission, as can be seen from Fig. 2, [9]. The use of recycled raw materials is also convenient for the reduction of the CO₂ concentration in the Earth atmosphere, Fig.2, [9] (the mining and the separation processes in the RE industry is responsible for around 90% of the whole energy consumption in the NdFeB PM manufacturing process).

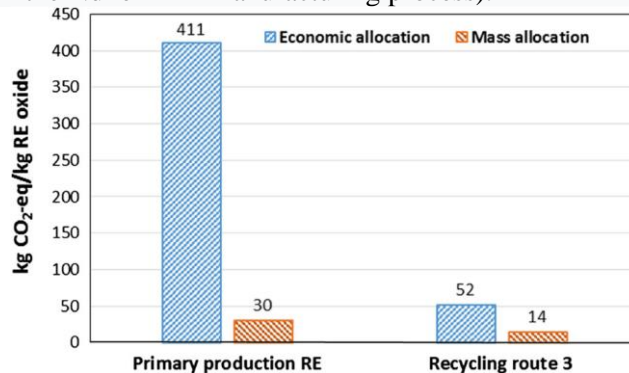


Fig. 2. Comparison between the CO₂ emissions of the extraction of Nd and Dy from ore and the emissions caused by using recycled Nd and Dy, [9]

2. Elements of the manufacturing technology of NdFeB PM

A basic technology scheme for NdFeB PM is represented in Fig. 3, [6]. There are many variants of technological processes against that basic shown in Fig.3 [10,11]. Some from these processes are based on the so-called hydrogen decrepitation (HD) and on hydrogen decrepitation followed by a decomposition and of a recombination of some phases (HDDR) in order to form magnetic anisotropic phases, used in bonded anisotropic PM manufacturing, [12, 13, 14, 15].

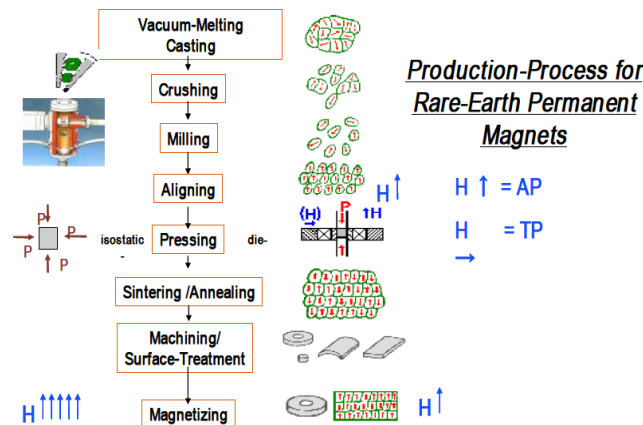


Fig. 3. Technology process of the NdFeB PM, [6]

The place of NdFeB PM in the space of the magnetic materials is shown in Fig.4, [6]. Here we represented in different colours the main used technological processes to manufacture PM.

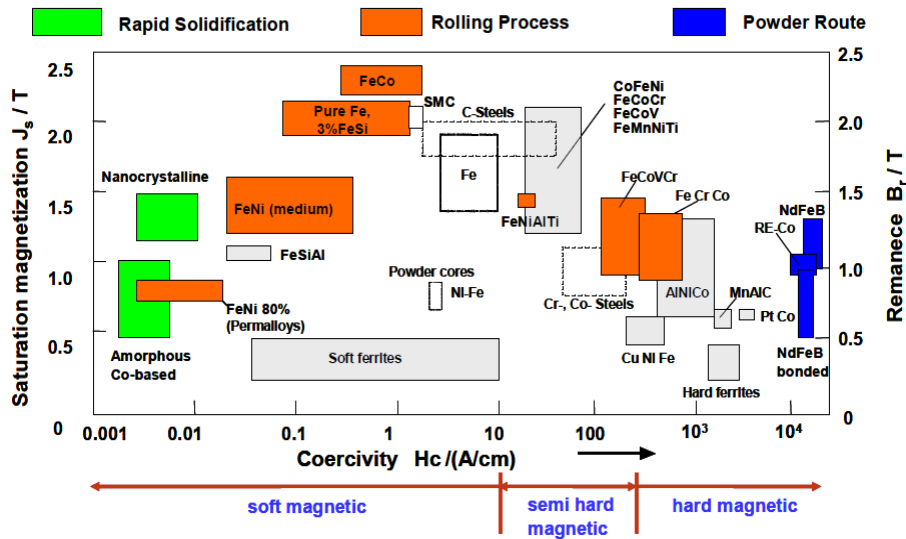


Fig. 4. The place of NdFeB PM in the family of magnetic materials, [6]

As it can be seen from Fig. 4, today there are NdFeB PM with extremely high coercive field, PM that can work at temperatures above 200°C without suffering significant irreversible losses, [11]. PMs have also been developed that can withstand extreme corrosive conditions, as is the case of offshore wind generators. Such an example of a magnet is shown in Fig. 5: the wet heat test at 2.6 bar pressure - the so-called HAST (Highly Accelerated Stress Test) test - at 130°C, 100% RH for 14 days shows smaller losses of 1 mg/cm², [6].

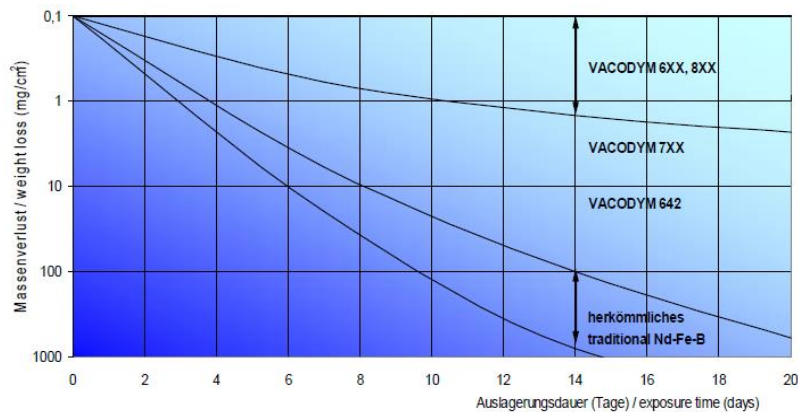


Fig. 5. HAST test for some NdFeB PM, [6]

Regarding the destination of NdFeB type PM production, we can see in Fig. 6 the distribution of this production for different economic fields, [16].

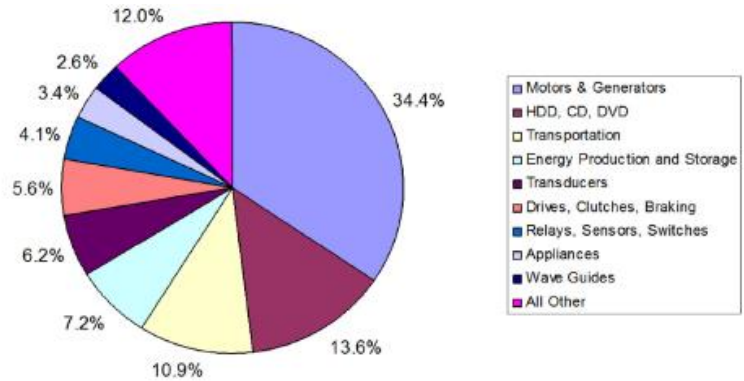


Fig. 6. Distribution of NdFeB PM after application type, [16]

From Fig. 6 it is observed that the majority of produced PM are intended for electric cars. Some applications consume from 10-20 g for small engines, about 1 kg of PM for engines used in electric vehicles and up to about 250 Kg/MW for wind turbines, [17]. This use advertises high operating temperatures, often above 200°C. But the high temperatures of use impose expensive PM (see Fig.7, [17]) due to the elements Dy and Tb, which partially replace Nd in NdFeB PM and thus increase the intrinsic magnetic coercive field. A limited possibility to increase this field is also the use of ferromagnetic phase grains with magnetic monodomain dimensions, also from the control of the free surface of these grains, so that in them no domains with reversed magnetization are formed, [18]. The maximum theoretical value of the specific energy for the NdFeB type PM with $\mu_0 M_0 = 1.61$ T the saturation magnetization of the Nd₂Fe₁₄B ferromagnetic phase is about 516 kJ/m³, but for some structures of these PM with exchange interaction, [19], 1000 kJ/m³ were predicted, [20].

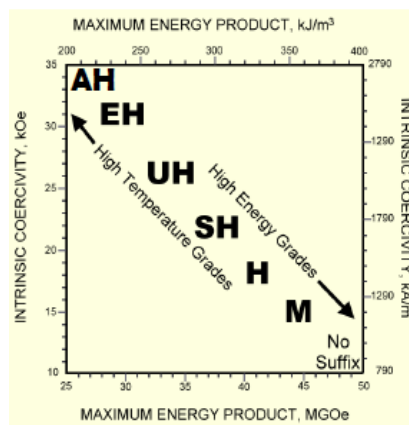


Fig. 7. The effect of working temperature increase of the NdFeB PM on their magnetic characteristic, [17]

The current world consumption of rare earths expressed in rare earth oxides (REO) is about 200 000 t/year, as can be seen from Fig. 8, [17].

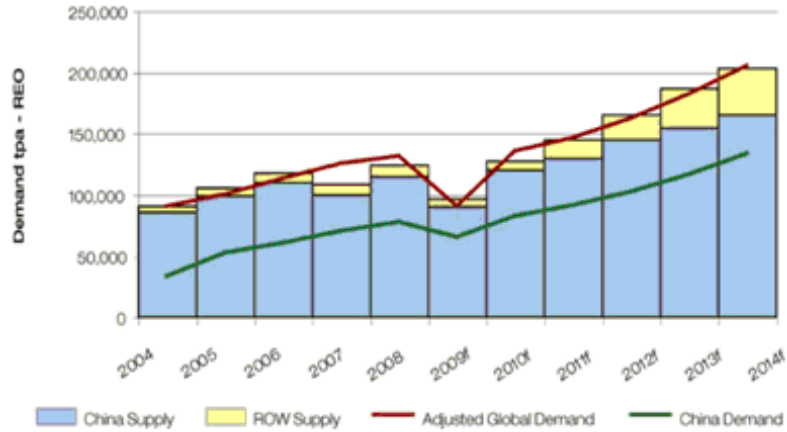


Fig. 8. Demand and production of REE measured in REO, [17]

From Fig. 8 it can be seen that the biggest supply and the biggest demand of REE is coming from China.

The distribution by different categories of users of these REO is shown in Fig. 9, [16]. Here the highest weight is owned by the NdFeB PM. Also the highest value.

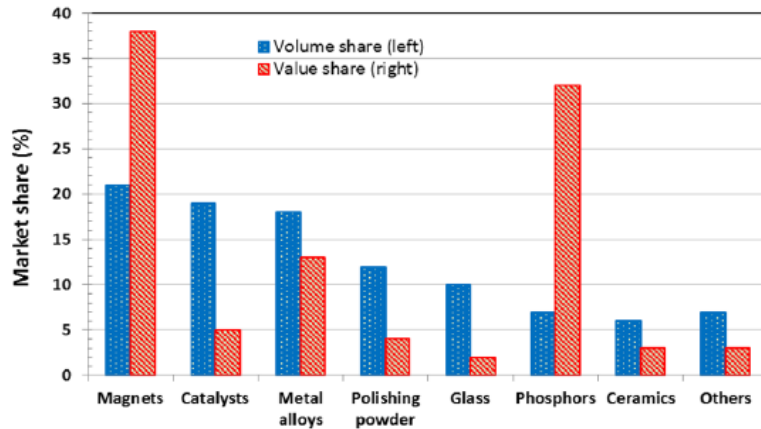


Fig. 9. Distribution of REO, as volume and value, over different domains

3. Circular economy and NdFeB PM

The rational use of natural resources was one of the first environmental concerns underlying the first European treaties.

Since 2015 there is an action plan adopted by the European Union with a set of multisectorial measures related to waste, waste from electrical and electronic

equipment, waste water and other measures that should lead EU Member States to some well-established targets.

The concept of circularity is closely related to the efficiency of the use of natural resources at the system level, respectively throughout the whole life cycle of the products, as well as the transformation of waste into new resources for other industries.

The Framework Directive 2008/98/EC on waste establishes, inter alia, the so-called "waste hierarchy", as well as the criteria for defining by-products, important aspects in promoting the circularity and revaluation in the internal market of new products resulting from the processing of waste.

In this context, the European Commission has established the "2017 list of EU-critical raw materials", a list that is subject to periodic review, at least every three years, to take account of market developments and production technologies. The number of raw materials evaluated increased with each update. Economic importance and supply risk remain the two most important parameters used to determine the critical character of a raw material.

In these studies, and legal provisions, the "rare earths" comprise a group of 17 considered critical elements composed of the 14 elements 4f, to which are added Y, La and Sc, La, Ce, Pr, Nd, Sm, Eu, Sc are "light" rare earths, the others are called "heavy".

China holds the supremacy by supplying 97% of the REE needed worldwide. Other countries that can profitably exploit RE are the U.S., with 13% of the world's total reserves, Russia, Australia, Canada, Greenland and South Africa. Difficulties in sourcing raw materials have a negative impact on industrial and economic performance.

These supply difficulties are one of the component criteria for appreciation of critical raw materials, raw materials of which are rare earths that are present in a proportion of around 30 % (mass) in the composition of PM of NdFeB type. These elements are Nd, Dy, Tb, Pr, of which Nd is the most used.

As shown above, until now they have been used in the PM of NdFeB type approx. 65 kt Nd and 20 kt Dy, so that a recycling of these rare earths, even with a low recycling potential, [21, 22], will have an important contribution in supplying the PM industry with valuable raw materials. Moreover, since the consumption of critical elements has the highest weight for PM: in 2008, we have for PM 76% of total consumption of Nd₂O₃ and even 100% for Dy₂O₃, [22].

Recycling of NdFeB PM can take place from the production scrap or from waste after an expensive and, in many situations, a complicated dismantling process. As scrap it is supposed that 20-30% from the produced PM are scrapped, [7]! In Europe, between the first patent files, which relate to the recycling of NdFeB PM, is the Romanian Patent 115996, [23], from 1998. The process allows a complete recovery of the NdFeB magnetic alloy with additions of Dy, Al, V by its use in the manufacture of other PMs. From the technical point of view, there are many references in the specialized literature that deal with the recycling of NdFeB family waste, emphasizing either the technological aspects or the specializations on certain

products. From the first category we can cite the work, [9], which, based on its recycling on HD, similar to the patent, [23], reaches the conclusion “The magnetic properties of recycled magnets made of regained scrap magnet powders come close to those made of primary materials. In the cases of the sintered magnets and hot-deformed magnets that contain various amounts of scrap powders, remanence and coercivity systematically score slightly lower than those of magnets made from virgin materials”.

After [23] it can be recycled also Nd-Fe-B permanent magnets with high coercivity, Fig. 10, used mainly in electrical machines.

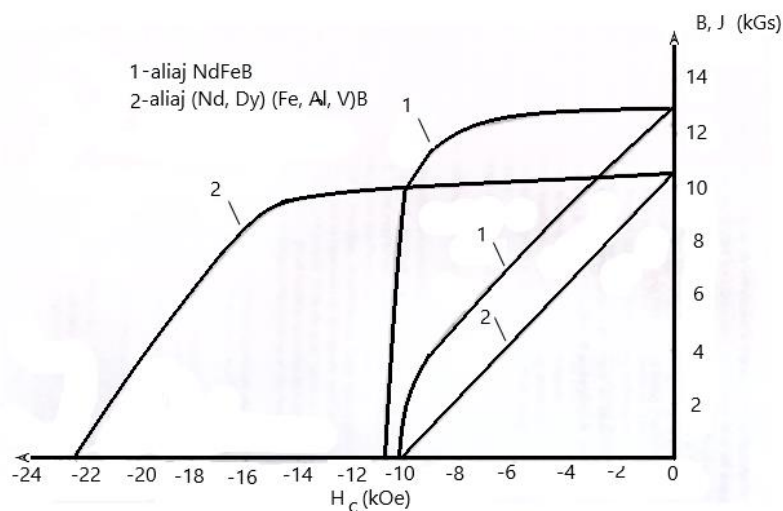


Fig. 10. Demagnetisation curves of NdFeB permanent magnets which can be recycled after [23]:
1 = normal PM, 2 = high coercivity PM

The flow chart in Fig. 11 of the established recycling technology is similar to that in the above Fig. 3: we suppose that the PM scrap is collected and divided in different grades, the scrap in each grade is supposed to have the same chemical composition, then the first two steps are replaced by step 1 in the Fig. 11 - surface cleaning of the magnet scrap, chemically or mechanically if the magnet surface is not free from some epoxy resins used to fix the PM and 2=hydrogen decrepitation (HD) in order to transform the scraps in the well-known onion like structure. HD take place with hydrogen pressures up to 5 bar with the temperature of the scraps of 100-150°C. The temperature or the hydrogen pressure could be a little higher for high coercivity grades (having Dy, Co and other alloying elements in the composition). This second step could be followed by a very easy crushing process. Many times the HD treatment is followed in the same step, at temperatures around 600°C by a dehydrogenation of the main magnetic phase in order to restore the initial magneto-crystalline anisotropy of the $\text{Nd}_2\text{Fe}_{14}\text{B}$ phase, which play an important role in the aligning process before and during field pressing.

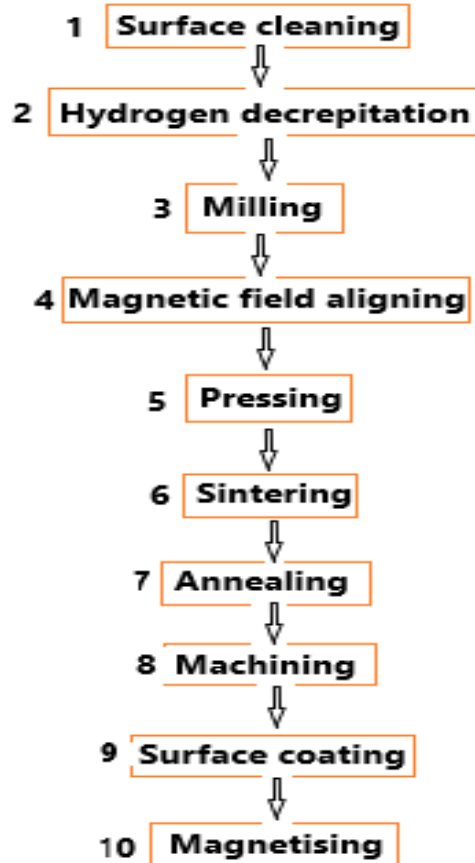


Fig. 11. Flow chart of the PM recycling technology.

1=Surface cleaning; 2= Hydrogen decrepitation (including a light crushing of the treated alloy and possible dehydrogenation at around 600°C); 3= Milling (jet mill, planetary mill) up to 2-8 micrometers ; 4= magnetic field aligning; 5= pressing (isostatic, or - in a die perpendicular to the magnetic field direction); 6= Sintering in vacuum or Ar at typically 1080°C; 7= Annealing (vacuum or Ar atmosphere at around 650°C); 8= machining to desired dimensions; 9= surface coating in order to protect the PM against chemical agents; 10= magnetising in order to be delivered.

Mainly the finished PM, made from recycled alloys, could have a little lower magnetic properties, often done by the increase of the oxygen content during the recycling. This can be seen in the Fig. 12, in which it is shown the intrinsic demagnetising curve of a recycled PM starting from scrap having very high intrinsic coercivity (number 2 in Fig. 10).

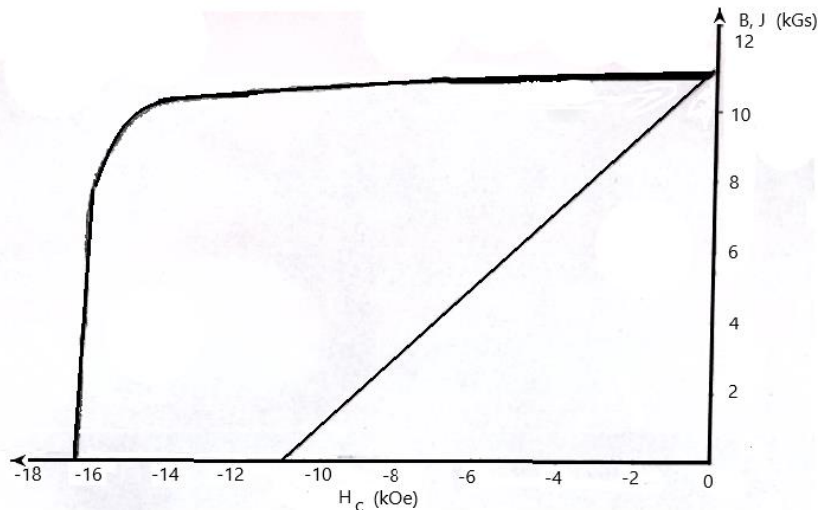


Fig. 12. Demagnetising curves of recycled PM scrap from the type 2 (in the notation after Fig. 10)

The reduction of the intrinsic coercivity observed in Fig. 12 reduces also the highest working temperature of the PM: such permanent magnets cannot work without irreversible losses at the same temperature as the PM from type 2 in Fig. 10.

Similar to [23], [16] presents physical recycling methods, including HD and separation from shredded products containing PM of the NdFeB type, hydro- and pyrometallurgical methods, also in extension methods based on chemical processes, Moore et. al., [24], propose to recycle NdFeB scrap using copper melts. In the paper [25] it is analysed the barriers of effective recycling of REE from NdFeB PM: current commercial recycling technologies cannot recover the small amounts of the metals in question, that are present in modern products, end-products to be recycled contain a complex mixture of metals that change over time as a result of technological advances, prohibitive cost of the collection and end-products to recycling facilities, the recycling process is not part of a collection chain. Product-oriented recycling was discussed in [7], where it is shown that Hitachi developed a machine to dismantle PM of the type NdFeB from hard disk and compressors with the capacity of 100 PM/h, also Habib et al., [22], shows an approximation of REE recovery rates for NdFeB PM used in wind turbines, Peeters, [26], describes a recycle method for PM used in HDD, Lixandru et. al., [27], from general electric equipment and Goonan, [21], from PM and other products containing REE. But in all situations, we have only a very low recycling proportion of REE. A real increase of the recycling coefficient for REE from PM is, after [7], possible only to build up an European recycling plan (Fig. 13).

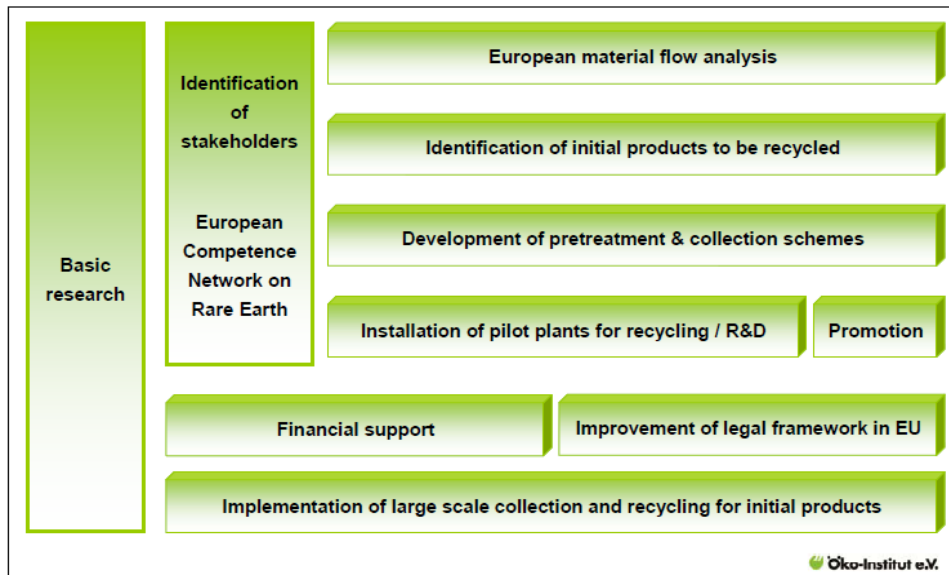


Fig. 13. Proposed recycling plan of NdFeB PM, [7]

4. Conclusions

In a circular economy, the value of the products and materials incorporated therein is maintained as much as possible. In other words, we can specify that the circular economy is a resource efficient economy, finally reaching a circular economy of regeneration. Waste and resource utilization are minimized as much as possible, and the resources reached at the end of their life do not leave the economic flow, but are reused further developing revenues to meet national / international requirements for certain metals.

There were presented aspects regarding the role and the global use of some important rare earth elements from the constitution of permanent magnets in the economic development of the society. The recycling of REE from PM is up today only possible in a small extent, although today there are technologies that allow the recycling of these rare earths. It is necessary to build up a real recycling scheme of REE from PM. The magnetic properties of the magnets obtained from recycled magnetic powders are close to those made from primary materials. In the case of sintered magnets and hot deformed magnets, which contain different amounts of residue powders, the remanence and coercivity are systematically slightly lower than those of magnets obtained from virgin materials.

In terms of ecological footprint, all recycled PM are superior to that obtained from primary raw materials.

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