




# Investigating the properties of recycled NdFeB magnets

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**Abstract** We report investigations on the processing by Spark Plasma Sintering (SPS) of RE<sub>2</sub>Fe<sub>14</sub>B (RE = Nd, Pr. . .) powders obtained by hydrogen decrepitation of decommissioned magnets and the magnetic properties of the consolidated magnets. First experiments have been carried out with a commercial powder to make clear the mechanisms occurring during the powder densification. The magnetic properties of the sintered bulks were measured to determine the SPS cycle resulting in the best saturation and remanent magnetization ( $M_s$  and  $M_r$ , respectively), coercive field ( $H_c$ ) and stored magnetic energy (BH<sub>max</sub> product). The optimized cycle was subsequently applied to a recycled powder. The properties of the magnets obtained after processing the recycled powder are compared to those made with the commercial one.

## 1 Introduction

There is a growing demand in the world for RE<sub>2</sub>Fe<sub>14</sub>B (RE = Nd, Pr. . .) magnets. They are used to build the rotors of modern electric engines and for many other applications. However, the production is unevenly distributed in the world since more than 90% of the Rare-Earth material required for the manufacturing of magnets is produced in China [1–3]. In addition, the extraction of Rare-Earth ores and their purification is very polluting processes [4]. A consequence of their growing use is that the quantity of decommissioned RE<sub>2</sub>Fe<sub>14</sub>B magnets is also growing quickly, to the point that they are referred to as urban mines. The objective of the work reported here was to investigate the possibility to consolidate new magnets by Spark Plasma Sintering using as a precursor the powder obtained by hydrogen decrepitation from decommissioned magnets. Hydrogen decrepitation consists of heating in a hydrogen atmosphere the decommissioned magnets. Hydrogen enters the material and reduces it to a powder [5–7]. This part of the work was carried out at Institut Néel in Grenoble (France) using decommissioned magnets provided by Valeo Powertrain Systems. Spark Plasma Sintering is a rapid consolidation method that results in a good understanding and control of the sintering kinetics. The heat source is not external but is an electric current (AC, DC or pulsed) that flows across the die containing the powder to the sinter. Simultaneously, an uni-axial pressure is applied. The main difference

between SPS and conventional or hot-pressing methods is that SPS allows to prepare highly dense samples with grain growth control and to save processing time [8, 9]. Previous works have shown that SPS is well-suited to the consolidation of NdFeB magnets [10, 11]. SPS sintering was carried out at CRISMAT in Caen (France). The process was optimized by sintering a commercial powder provided by the AICHI company (*Aichi Europe GmbH Immermannstr.65b 40210 Dueseldorf, Germany*). According to the information provided by the supplier, the good properties of the powder result from hydrogenation-dehydrogenation steps included in its fabrication route [12]. No other information was provided. We have selected the process resulting in the best saturation and remanent magnetization ( $M_s$  and  $M_r$ , respectively), coercive field ( $H_c$ ) and stored magnetic energy (BH<sub>max</sub> product) for the bulks consolidated with the commercial powder. Then, we used this process to sinter bulks made with recycled powder.

We firstly report on the precursor's characteristics and the selected sintering process, then we present the structural, microstructural and finally magnetic properties of the consolidated magnets.

## 2 Precursors and consolidating process

The composition of the commercial powder as provided by the supplier is given in Table 1.

Several batches of recycled powder were produced from decommissioned magnets provided as scrap material to Institut Néel. As a result, the performances of the

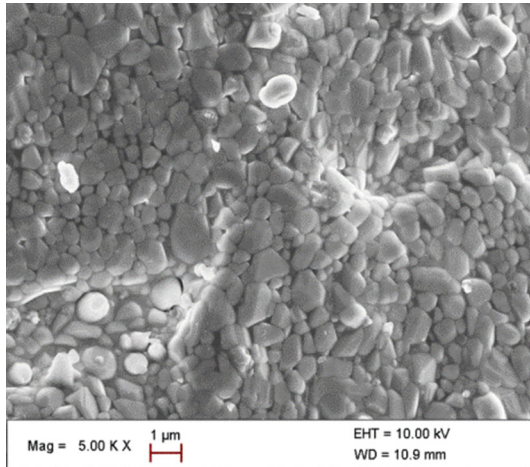
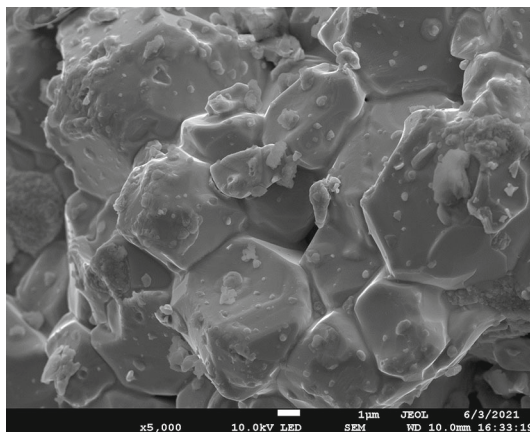
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**Table 1** Chemical composition of the supplied AICHI powder

Element	B	Cu	Al	Pr	Nd	Fe
Mass content (%)	1.0–1.2	0.3–0.6	0.3–0.7	0–9	20–31	65–75

**Table 2** Mass content in Nd, Pr and Fe of one of the batches of recycled powder

Element	Pr	Nd	Fe
Mass content (%)	6.5	17	76.5

**Fig. 1** Micrograph of the commercial powder**Fig. 2** Micrograph of the recycled powder

initial magnets were not known. The powder was dehydrogenated after decrepitation. The chemical composition of the recycled powder could slightly change from one batch to another one but, was near stoichiometry with some additives such as Al. As an example, Table 2 shows the mass content in Nd, Pr and Fe of one of the batches.

Figure 1 shows a micrograph of the commercial powder and Fig. 2, that of a recycled one. The commercial powder consists of large clusters including grains the

size of which is around 1  $\mu\text{m}$ , while the clusters in the recycled powders include 4–6  $\mu\text{m}$  grains.

The powders have been consolidated by Spark Plasma Sintering (SPS). For this purpose, they have been inserted in a graphite mold and sintered in an HP D25 machine provided by *FCT System GmbH Rauenstein, Germany* without performing any magnetic alignment of the powder. Figure 3 shows the thermal cycle applied to the samples. As compared to the other sintering techniques, the temperature is lower and the processing time is very short [8, 9]. In addition, no subsequent annealing or hydrogen desorption step of the obtained samples was necessary to obtain good magnetic properties (see below). These observations suggest that consolidating NdFeB powders by SPS is a less expensive process than the conventional techniques.

### 3 Structural and micro-structural properties of the consolidated bulks

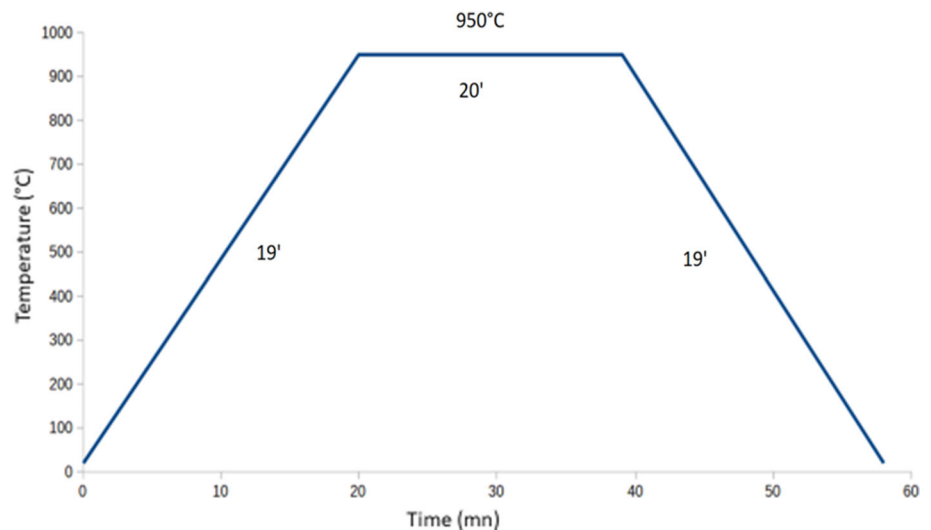
Figure 4 shows the X-rays diagrams of consolidated bulks obtained with the commercial powder (in blue) and the recycled powder (in orange). Most peaks are common to both types of samples and correspond to the  $(\text{Nd, Pr})_2\text{Fe}_{14}\text{B}$  phase. An Nd-rich phase different from  $\text{Nd}_2\text{Fe}_{14}\text{B}$ , possibly  $\text{Nd}_2\text{Fe}_{17}$  [13] was also identified in both samples.

Micrographs of the bulks (1) after cleaving the bulks and (2) after surface polishing are displayed in Fig. 5. The micrographs after cleaving show that the grains in the bulk fabricated with the recycled powder are much larger than those in the bulk made with the commercial powder. After polishing the surfaces, the presence of a bright material along the grains boundaries of the bulk made with the commercial powder (shown as a red line) is visible. This material was identified as the Nd-rich phase detected in the X-rays diagrams. It includes Neodymium and Oxygen but, as mentioned above, the presence of  $\text{Nd}_2\text{Fe}_{17}$  cannot be excluded [13]. It is not visible in the bulk fabricated with the recycled powder, the bright phases of which were not identified. They could be intermetallic phases, but not crystallized rare-earth oxides because this would have deteriorated strongly the magnetic properties of the sample.

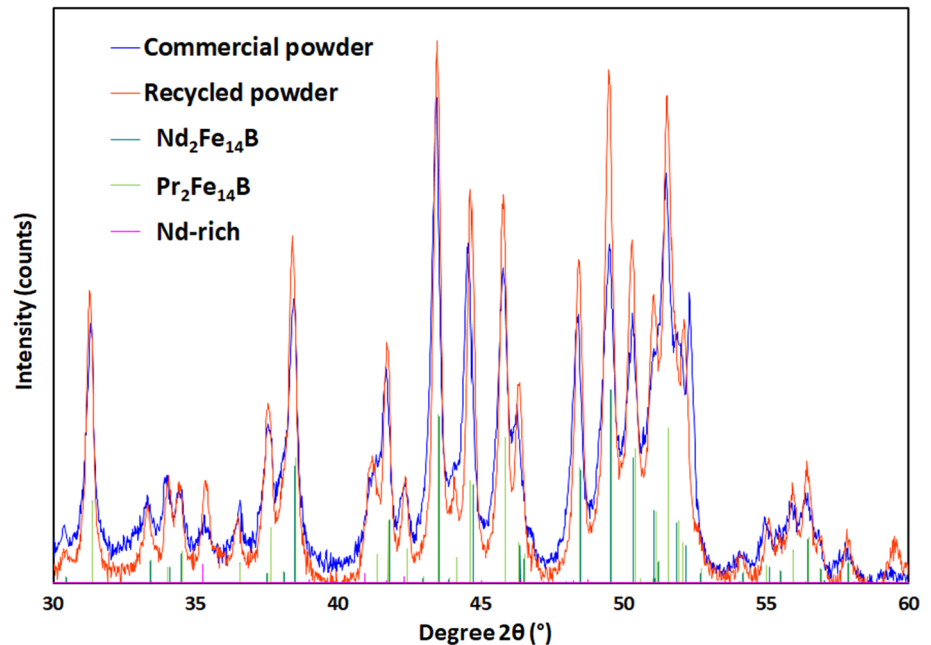
### 4 Magnetic properties

Hysteresis cycles were measured with a SQUID magnetometer (*Quantum Design, model MPMS, San Diego, USA*) on parallelepipeds with dimensions in the 1 mm

**Fig. 3** SPS thermal cycle applied to consolidate the bulks fabricated with both the commercial and the recycled powders



**Fig. 4** X-rays diagrams of the consolidated bulks obtained from (i) the commercial powder (in blue) and (ii) the recycled powder (in red)

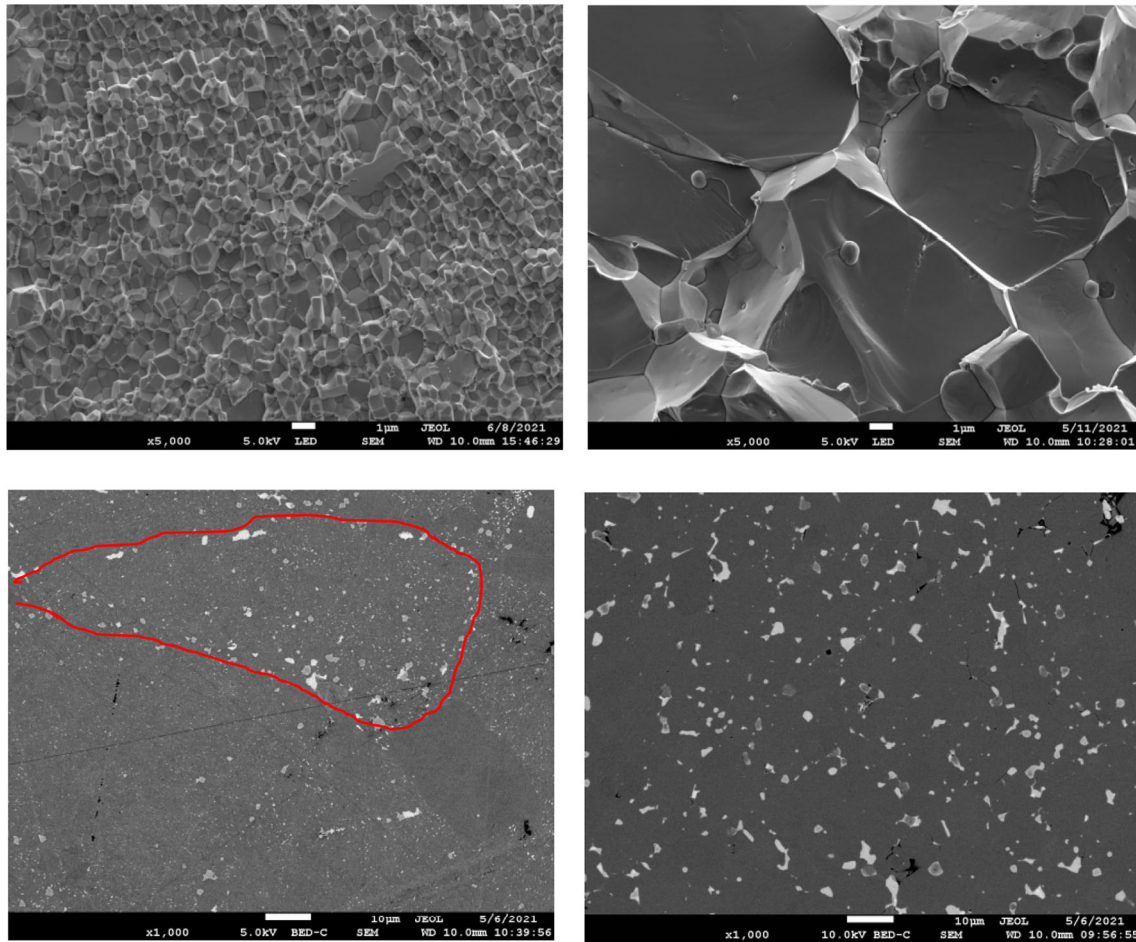


range cut from bulks fabricated at different pressures in the SPS apparatus. Cycles corrected for the demagnetizing field according to the expressions proposed in [14] are visible in Fig. 6 for bulks fabricated with (1) the commercial and (2) the recycled powder. Pressures above 100 MPa are detrimental to the magnetic properties of the samples made with the commercial powder, while pressure has little effect on those of the bulks made with the recycled one. At 65 MPa and 100 MPa, the bulks made with the commercial and the recycled powder show similar remanent magnetizations ( $\mu_0 M_r \sim 0.7$  T), coercive fields ( $\mu_0 H_c \sim 1$  T) and stored energy ( $BH_{\max} \sim 20$  MGOe). Figure 7 compares mappings of the vertical component of the magnetic field of magnets consolidated with the commercial and the recycled powders applying a 100 MPa pressure on the one hand to that of an off-the-shelf NdFeB magnet (N45) with

the same dimensions on the other hand. The mappings were obtained by scanning the area located 1 mm above the samples surface with a Hall probe after magnetizing them with a 5 T field. The field above the magnet sintered with the commercial powder is more homogeneous than that above the recycled magnet and is 15% larger than this last one. However, it is 40% lower than that of the off-the-shelf NdFeB magnet.

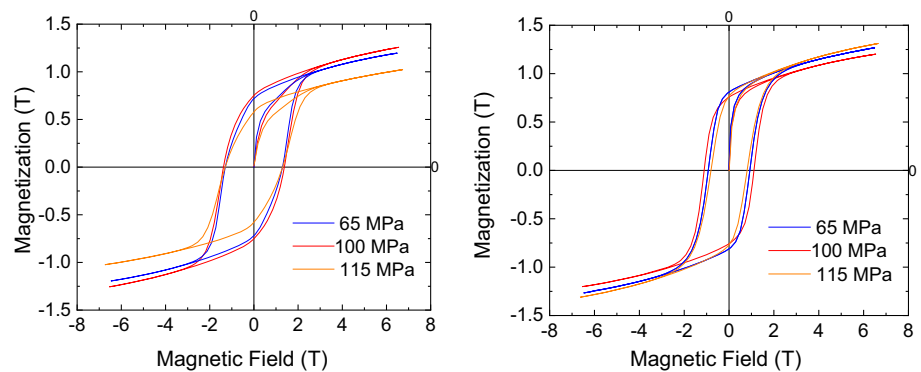
## 5 Conclusions and perspectives

In a large range of pressures, the magnetic properties of the samples sintered with the recycled powder are very similar to those consolidated with the commercial powder, although the microstructures of both the precursors and the bulks are different. In addition, SPS



**Fig. 5** Left (right) column: micrographs of a bulk obtained from the commercial (recycled) powder; upper line: after cleavage of the bulks; lower line: after polishing the surfaces

**Fig. 6** Hysteresis cycles of bulk magnets fabricated at different pressure in the SPS apparatus with the commercial powder (on the left) and the recycled powder (on the right)

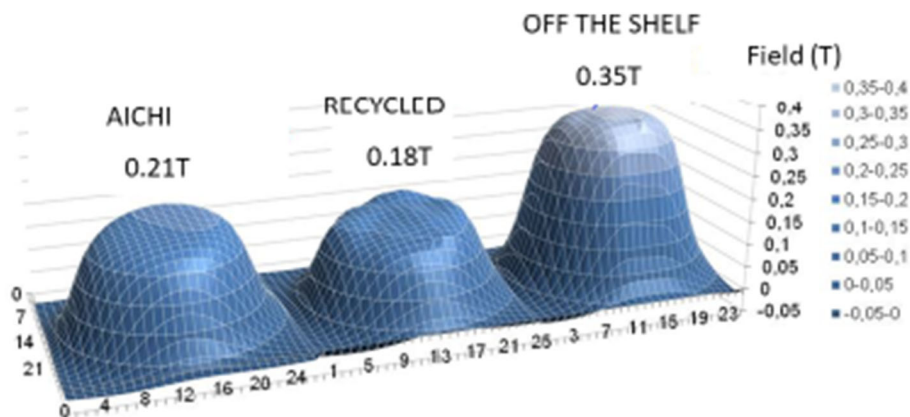


sintering results in a coercive field and a remanent magnetization, which are near or in the range of the performances expected of NdFeB magnets. However, the field generated by the magnets made by SPS is lower than that of a typical off-the-shelf one with the same dimen-

sions. The performances of the SPS magnets could be enhanced by orientating the grains of the precursors before or during the sintering process in an easy magnetization direction. This will be the focus of future studies.



**Fig. 7** Mappings of the magnetic field above the AICHI (left) and the recycled (middle) SPS samples compared to that above an off-the-shelf N45 NdFeB magnet (right). The maximum field above the samples is indicated for each magnet



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