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Project title: **“Pilot Action for Securing a Sustainable European Next Generation of Efficient RE-free magnets”**
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PASSENGER
FOR SUSTAINABLE AND EFFICIENT RE-FREE MAGNETS

Deliverable No. 1.4

Fabrication of high-coercive ferrite loose powder (jHc > 5.5 kOe) accompanied by a moderate decrease (<8%) in remanence for improved performed when decreasing operating temperature

Due date of deliverable	31/10/2023
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Author name(s)	Enrico Forlin, Sergio Salviati, Paolo Bernardi, Rodolfo Miranda
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This project has received funding from the European Union`s Horizon 2020 Research and innovation programme under grant agreement no. 101003914

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LIST OF ACRONYMS

Abbreviation	Unit	Meaning
Br	G and emu/g	Remanence
jHc	Oe	Coercivity
Sr	-	Strontium
XRD	-	X-Ray Diffraction
SEM	-	Scanning Electron Microscopy
VSM	-	Vibrational Sample Magnetometer
TEM	-	Transmission Electron Microscopy

SUMMARY

Deliverable goal is to create improved Strontium Ferrite Powder based on an increase in coercivity and limited remanence loss with no use of critical raw materials (e.g. lanthanum and cobalt, as it is the case for high-quality commercial Sr-ferrite). Two strategies were therefore taken into consideration: ferrite powder for compounding/injection molding and for sintering, similarly to previous work reported in D1.3. The production process involves mixing of raw materials (iron oxide, strontium carbonate and additives) with water to form a slurry, which is then pumped in the high temperature ovens. The powder is then cooled, transported to the milling equipment and delivered to other PASSENGER partners for further characterisations and testing. Table 1 summarises the deliverable goals and the achieved values for the two applications.

Table 1. Achieved magnetic values compared to deliverable target

		Target values	Ferrite for sintering	Ferrite for compounding
Remanence [kG]	Br	>1.84	2.7	2.6
Coercivity [kOe]	jHc	>5.5	5.7	5.4

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1. INTRODUCTION

The aim of the deliverable is the production of ferrite loose powder with coercivity jHc > 5.5 kOe and moderate decrease of remanence, <8%, in comparison with former target of 2.0 kG, i.e. Br > 1.84 kG. Regarding practical applications, one of ferrite's key limitations is the tendency for coercivity to decline with operating temperature (e.g., motor performance in cold weather), which may lead to a serious demagnetization risk of the magnets during operation. The main objective is therefore to create a ferrite powder with improved magnetic characteristics while maintaining an increased coercivity to compensate for temperature-related losses. ILPEA's approach to achieve the deliverable is to modify both powders formulations and process parameters in order to improve magnetic performance in close collaboration with IMDEA, concerning the application of the self-developed flash-milling method and use of specific additives, and also with MBN, concerning the scale-up of laboratory processing of powder to industrial scale. JSI and TUDA supported such activities, providing advanced characterization techniques and assessment of magnetic properties with temperature respectively. Given that the production parameters (in particular the raw materials cost and temperatures) do not surpass those commonly employed in the routine manufacture of Sr-ferrite, ILPEA's techniques are also feasible from an economic perspective at industrial scale.

IMDEA, JSI, TUDA and MBN have collaborated with ILPEA for the achievement of the deliverable.

2. METHODOLOGY

2.1. FLASH-MILLING METHOD

IMDEA has applied its self-developed flash-milling method and a novel approach consisting on the pre-addition of Fe_2O_3 powder to the Sr ferrite produced by ILPEA for successful development of coercivity, while maintaining a well-balanced value of magnetization based on the chosen milling time [1, 2]. Both approaches have gone through an optimization process of the heat treatment (include both temperature and duration).

The resulting samples have been characterized at IMDEA by XRD, SEM and VSM (the latter for magnetic characterization at room temperature). TEM characterization has been carried out by JSI. TUDA has been responsible of making the magnetic characterization at different temperatures.

MBN has applied its proprietary technology based on High Energy Ball Milling (HEBM) to replicate at larger scale the effect of flash milling on loose powder properties, providing representative samples to IMDEA for magnetic characterizations.

Based on former developments and achievements, a group of selected materials has been identified by ILPEA and IMDEA, to match application targets, detailed in the following. The different names refer to different aim (final magnet fabrication) and to different commercial grades (all of them with nominal composition $SrFe_{12}O_{19}$, and with different grades).

- *P07-PSG* *[Aim: sintered magnets]*
- *P07 PSG 2M* *[Aim: sintered magnets]*

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- *P21B* *[Aim: injected magnets]*
- *P41B* *[Aim: injected magnets]*

2.2. Sr-FERRITE FABRICATION

The industrial production process flow followed by ILPEA can be outlined as follows, similarly to previously reported flow (D1.3):

Mixing of raw materials (Iron Oxide, Strontium Carbonate and additives) with water to form a slurry (Fig 1).

Figure 1. Mixers used for raw materials dispersion in water at ILPEA

Treatment of the slurry in a cylindrical oven (Fig. 2), where water evaporates (colder zone) and subsequent sintering occurs (hotter zone).

Figure 2 High temperature cylindrical oven at ILPEA

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Once cooled, powder is transported in the milling systems (Fig. 3) using a selection grid to sort out the particles size. Milling process is carried out in a closed environment to avoid dispersion of finer particles.

Figure 3. Milling systems at ILPEA fabrication of high-coercive ferrite loose powder results

PASSENGER has focused on the development two different materials, based on two major ferrite applications: injection molding/compounding and sintering.

2.3. ILPEA'S FERRITE FOR SINTERING IN PASSENGER

ILPEA has successfully managed the achievement of the target values (remanence and coercivity) based on former achievements previously reported.

P07 PSG material family has been developed at ILPEA using IMDEA concept, i.e. milling until a very low particle size, adding additives and heating the blend at 1000ºC. Loose powder milled has a particle size in the range 0.3-0.5µm and properties measured by VSM satisfied the project targets of D1.3. Further improvements based on the same material led to the development of "P07 PSG 2M". The sintered powder was characterized by ILPEA with hysteresis-graph measurement (Fig. 4), finding values of Br = 2.795 kG and jHc = 5.699 kOe that meet the deliverable targets. Compared to former powder variant, the Br decrease was limited to -4.5%, while jHc exceeds the 5.5 kOe target value. This powder has been developed for sintering applications.

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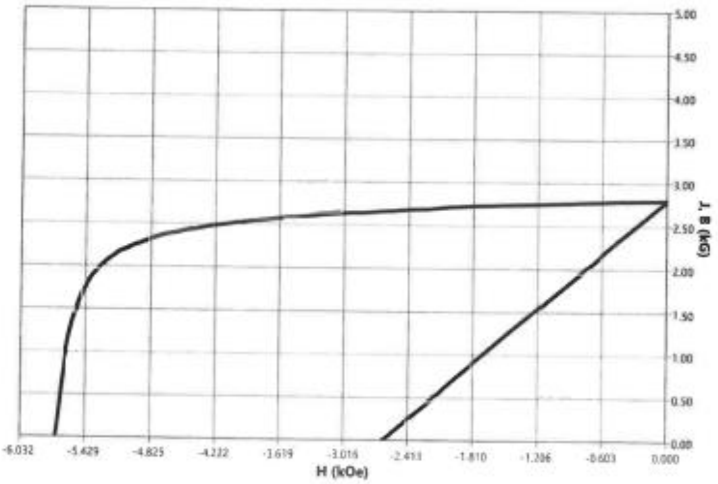

Temperature	20.0 °C
Br	(2.795 ± 0.028) kG
HcB	(2.658 ± 0.040) kOe
HcJ	(5.699 ± 0.085) kOe
BHmax	(1.881 ± 0.038) MG·Oe
BA	(1.391 ± 0.014) kG
HA	(1.352 ± 0.020) kOe
Hknee	(4.036 ± 0.061) kOe

Figure 4. Hysteresis graph curve and values for P07PSG2M

P07PSG2M was additionally characterized by IMDEA using VSM (Fig. 5). The analysis with a different technique confirmed the aforementioned properties.

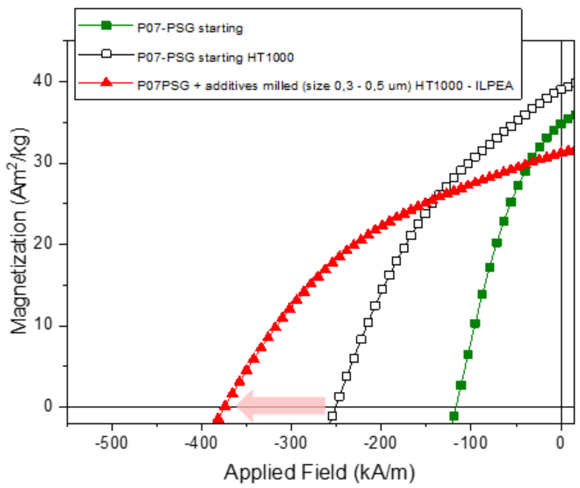

Figure 5. VSM curve for P07PSG2M

2.4. ILPEA'S FERRITE FOR INJECTION MOLDING/COMPOUNDING IN PASSENGER

Considering the need to integrate final magnets in PASSENGER's products, ILPEA improved strontium ferrite loose powder for compounding, in order to advance even further towards an improvement of the quality: P41B is the result of such activities, starting from P02Sr followed by a next generation named P21B. Figure 6 shows images of the ferrites produced at industrial scale by ILPEA in PASSENGER.

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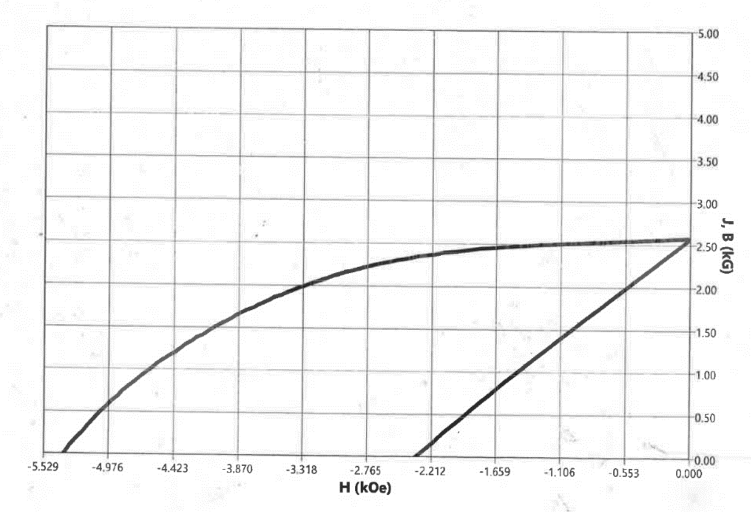
Figure 6. Improved Sr-ferrite material P41B produced by ILPEA in the framework of PASSENGER, achieving a production of 4 tones already at this stage.

The powder was enclosed in a polymeric matrix and calendered in order to study its properties for compounding application. It was characterized with hysteresis-graph measurement by ILPEA (Fig. 7), finding values of Br = 2.576 kG and jHc = 5.360 kOe that approach the deliverable targets. Compared to former P21B powder variant showing 2310 x 2641 jHc x Br (as calendered magnetic compound), coercivity was increased with a reduction of Br by just -2.4%.



Temperature	20.0 °C
Br	(2.576 ± 0.026) kG
HcB	(2.346 ± 0.035) kOe
HcJ	(5.360 ± 0.080) kOe
BHmax	(1.562 ± 0.031) MG·Oe
BA	(1.289 ± 0.013) kG
HA	(1.212 ± 0.018) kOe
Hknee	(2.441 ± 0.037) kOe

Figure 7. Hysteresis-graph curve and values for P41B

2.5. TEMPERATURE DEPENDENCE OF THE MAGNETIC PROPERTIES

In order to demonstrate the functionality of this improved ferrite at high and low temperatures, TUDA did measurements on the samples produced by IMDEA, as previously

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reported in D1.3. Figure 8 shows the results. Based on these results, we can confirm that the objective of developing a Sr-ferrite permanent magnet material with excellent magnetic properties at high and low temperatures (the latter being critical for hard ferrites) has been successfully managed. This has been possible by application of IMDEA's flash-milling and further improved, through the use of additives and with no use of any critical raw materials (e.G. La and Co as typically used in commercial ferrites to achieve this objective). Results are obvious when comparing coercivity evolution for the commercial starting ferrite powder (ILPEA's P02Sr) and the ferrite powders processed by IMDEA.

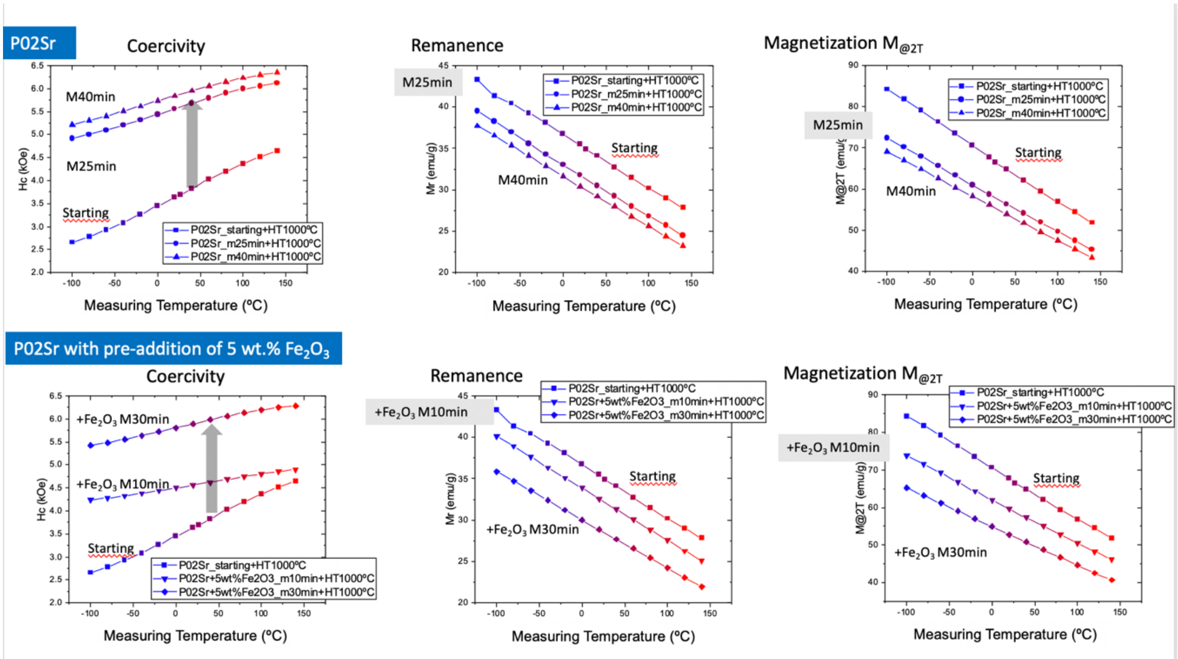

Figure 8. Evolution of coercivity, remanence and maximum magnetization as a function of the measurement temperature for the precursor (starting) and same material + Fe_2O_3 pre-addition

1. CONCLUSIONS

The target values specified for the D1.4 are:

- Remanence Br>1.84 kG
- Coercivity jHc>5.5 kOe

After improving both ferrite formulations and production process parameters based on the collaborative work carried out with IMDEA, JSI, MBN and TUDA in the framework of the PASSENGER project, ILPEA has been able to produce loose powders with the following properties:

- P07PSG2M for sintering applications with Remanence Br =2.795 kG and Coercivity jHc=5.699 kOe
- P41B for compounding/injection molding with Remanence Br =2.576 kG and Coercivity jHc=5.360 kOe

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The former material fulfils all the objectives considered in Deliverable 1.4.

The latter material fulfils the remanence target, while approaching the coercivity target, for compounding applications and subsequent integration in PASSENGER products, in collaboration with BARLOG and KOLECTOR for the preparation of ferrite/polymer compounds, in WP2 and WP3 activities.

Remarkably, a production scale at ILPEA of 4 tons of the improved ferrite material -to share among the partners to continue working in WP2 and WP3- illustrates a successful up-scaling in PASSENGER.

2. REFERENCES

[1] A. Bollero *et al.*, ACS Sustainable Chem. Eng. 5, 3243 (2017).

[2] A. Bollero *et al.*, PCT/EP2018/063222: “Ferrite type materials and process for the production thereof”.

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