Characterization of Bulk Rare-Earth Permanent Magnets with Complex Geometries

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Abstract

Rare-Earth (RE) permanent magnets are ubiquitous across a wide range of market sectors, from energy to information technology. The security of supply for the raw materials used in the production of these permanent magnets is of critical importance to the low carbon future envisioned by the European Union. The Horizon 2020 project Resource Efficient Production of Magnets (REProMag http://www.repromag-project.eu/), looks to address the problem of the sustainability of RE permanent magnets by developing an innovative automated manufacturing route. The Shaping, Debinding and Sintering (SDS) process being developed will allow economically efficient production of net shape magnetic parts with complex structures and geometries, whilst being 100% waste free through the use of fully recycled raw material. As part of this project, a series of demonstrator magnets will be produced, each with a unique complex geometry, making traditional material characterization impracticable.

Standard electromagnet methods suffer from limitations on specimen shape and size and flux leakage from induced air gaps. The National Physical Laboratory (NPL) is working towards a new measurement standard to overcome these limitations and allow material characterization throughout the production process of the complex components. Pulsed Field Magnetometers (PFMs) discharge a current from a capacitor bank at different frequencies to generate large magnetic fields inside a solenoid, with pick-up coils for the measurement of magnetic field strength and polarization. Their large sample space allows for the non-destructive characterization of complex, real-world components with a maximum lateral dimension of 50 mm at operational temperatures up to 200 °C. In comparisons against an electromagnet, PFMs have shown good agreement for the measurement of remanence, but discrepancies with other important parameters such as coercivity and energy product. This paper will discuss NPL's investigation of the physical effects, such as self-demagnetization and magnetic viscosity, behind these discrepancies.

Keywords

Permanent Magnet, Rare-Earth, Characterization, Pulsed Field Magnetometry, Additive Manufacturing

1. Introduction

1.1 REProMag

As the preferences of consumers shift towards hi-tech, miniaturised and green products, the material security regarding the supply of rare-earth elements becomes more of a concern. The price spike of these rare elements in 2011 highlighted the precarious position Europe held. To reduce Europe's dependence on foreign supply, technologies must be developed not only to use the resource more efficiently through optimization of design, but also to have zero wastage throughout production.

REProMag is looking to develop a novel processing route that allows the production of net shape permanent magnets with 0% wastage. These hard magnetic parts will be produced with specific applications in mind, such as motors, actuators and fixation in the fields of energy, medical and aerospace. The Shaping, Debinding and Sintering (SDS) process will be implemented with additive manufacturing techniques to produce complex components surpassing the performance of isotropic resin bonded magnets currently on the market. Energy savings of up to 30% compared to current sintering production and the ability to add geometric features (such as cooling channels) or structural optimizations (such as fins and fixtures) without additional material wastage will improve the competitiveness of the European magnet production industry.

1.2 Material Characterization

Once the SDS process and additive manufacturing are optimised to deliver anisotropic, high energy product samples, a series of demonstrators will be produced spanning a wide range of permanent magnet applications. The characterization of these components for material properties and alignment is the final stage of REProMag and, much like the other parts of the project, requires innovation. Standard techniques for the characterization of permanent magnets are covered by IEC 60404 part 5 [1], which instructs the determination of the key material parameters such as intrinsic coercivity (H_{cJ}), coercivity

(H_{cB}), magnetic remanence (B_r) and maximum energy product (BH_{max}) with the use of an electromagnet. The electromagnet technique is a closed-circuit measurement, meaning the sample is enclosed by a high permeability yoke that carries the flux through an infinite path. This reduces the complexity of the analysis with corrections limited to the thermal and electrical drift of the instrumentation. There are, however, limitations to this method due to sample geometry, the generated magnetic field strength and operational conditions. The optimal geometries for characterization in an electromagnet are 10 mm diameter by 10 mm length cylinders or 10 x 10 x 10 mm cubes. These geometries are appropriate for grade classification used in the purchase and sale of material, but are not representative of actual end use geometries and may not reflect the magnetic performance of actual components.

The tolerances on the length of the samples are also very strict as the faces must be parallel, otherwise an air-gap is induced, which negates the use of the yoke and allows for flux leakage out of the magnetic circuit. The saturation of the pole faces is another source of flux leakage. At low supplied current levels, the current to magnetic field relationship of the electromagnet is linear, but becomes non-linear at higher current levels due to the reduced permeability of the pole faces, eventually resulting in saturation. This negates the effectiveness of the yoke and dramatically affects the measurement of high coercivity samples. Measurements at operational conditions, such as elevated temperatures are also impractical using the electromagnet method due to significant uncertainty arising from temperature gradients at the pole face-sample interface and throughout the sample itself. As the goal of REProMag is to produce components with complex geometries and structural optimization, it is apparent that an alternative technique is required for the material characterization.

Considering the limitations of the electromagnet technique it was concluded that a closed-circuit technique would not be appropriate for this project, as the yoke is the source of the discussed limitations. Open-circuit techniques include vibrating sample magnetometers, vibrating coil magnetometers and superconducting quantum interference devices [2-4], but these techniques are not suitable for the direct characterization of material parameters for large bulk samples. Another, relatively new, open-circuit technique is pulsed field magnetometry [5-7], which utilises a capacitor bank to discharge a large damped pulse through a magnetizing solenoid. The applied magnetic field strength, H, and the magnetic polarization, J, are detected by secondary pick-up coils. The coil used to detect magnetic polarization is wound with auxiliary windings to compensate for air flux, as the cross section of the pick-up coil is larger than that of the sample, and so encloses a significant air flux contribution. Figure 1 gives a schematic of the two possible configurations of different PFM's.

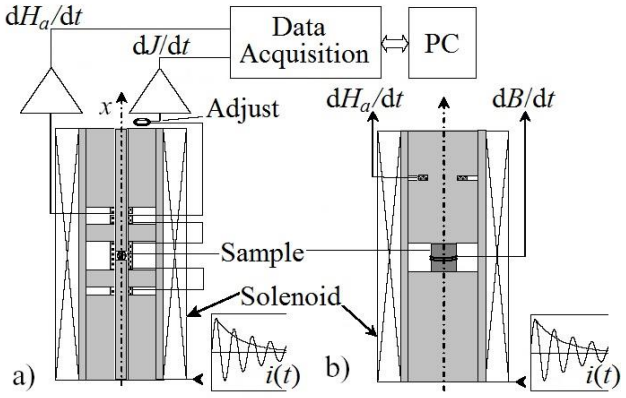

Fig. 1. Schematic of a PFM [5] where in a) the pick-up coils couple to the flux at the ends of the sample and in b) the pick-up coils couple to the flux at the mid-plane of the sample.

A significant advantage a PFM has over the traditional electromagnet techniques for permanent magnet characterization is the increased sample volume. The PFM at the NPL has a maximum bore size of 50 mm, suitable for the measurement of components with real geometries, instead of standard material samples. Another advantage is the maximum field achievable, ±6.5 T, which allows simultaneous magnetization and characterization. This removes the need to pulse the sample prior to the measurement. Taking away the yoke also allows for measurement at operational conditions with a temperature capability covering the range from 20 up to 200 °C.

1.3 PFM corrections

The disadvantages of the technique are related to the physical nature of the materials being characterized. Aside from the practical corrections required for the instrumentation, there are two dynamic and one magnetostatic corrections. Magnetic viscosity and eddy current corrections must be applied as the PFM is a dynamic (AC) measurement. The changing magnetic field will induce eddy currents in a material, which generate a field that opposes the applied magnetic field. To obtain the intrinsic DC properties, an f/2f [8] technique is implemented to correct for the eddy-currents. Two measurements are performed using pulses of different frequencies and then used to extrapolate towards the true material properties.

Magnetic viscosity is also caused by the transient field and is due to the thermal activation of magnetization reversals over energy barriers.[9] There is a "lag" between the changing applied magnetic field strength and the resulting change in magnetic induction. This

manifests as a measured value of H_{cJ} greater than the true value, giving the appearance of a magnetically harder material.

The magnetostatic correction, which is related to the geometry of the sample, is arguably the most significant correction related to the application of the PFM for the samples of interest in this project. Self-demagnetization is not a recent phenomenon and is well documented in texts dating back to Maxwell. A magnetized sample that has a boundary, without an infinite flux path, will have an internal field in opposition to the magnetization. This manifests as a shallow curved knee of the demagnetization curve which will impact the value reported for $BH_{max.}$ The demagnetizing field, H_d is defined as:

$$H_d = -NM \quad (1)$$

where M is the magnetization ($M = \mu_0 J$, where μ_0 is the permeability of free space) and N is the demagnetization factor with a value of $0 < N < 1$.

The demagnetization factor has two forms, the magnetometric and the fluxmetric demagnetization factors, dependent on the measurement method used. The fluxmetric demagnetization factor, N_f, is the ratio of the average demagnetizing field to average magnetization at the mid-plane of the specimen and the magnetometric demagnetization factor, N_m, is the ratio of the average demagnetizing field to average magnetization over the volume of the specimen. These definitions are important for the different configurations of PFM discussed previously. Configuration a) in Figure 1 uses the magnetometric demagnetization factor and b) uses the fluxmetric demagnetization factor.

The PFM at NPL has the configuration shown in a) and therefore requires the investigation of the magnetometric demagnetization factor. The value of N is dependent on the geometry of the sample and the susceptibility of the material. Existing determinations of N have commonly involved finite element modelling or complex numerical models. This is due to a non-homogenous internal field for geometries other than ellipsoids. The most widely used determination of the demagnetization correction in industry is that of Chen *et al* [10], which gives the theory behind the calculations and the historic development of the models. Tables for both magnetometric and fluxmetric demagnetization factors, for cylinders covering a wide range of aspect ratios[1] and susceptibilities, χ, are quoted. Though widely accepted, the numerical model suffers from necessary simplifications such as a constant susceptibility, with each value of N calculated at a different susceptibility. This simplification is erroneous when one takes into account the non-linear nature of the susceptibility of ferromagnetic materials, where χ ranges from 0 to infinity through the hysteresis curve.

This paper introduces the work being performed at NPL to investigate the magnetic viscosity and the magnetometric demagnetization factor. Once these phenomena are understood they can be properly corrected for to give true intrinsic properties of the demonstrator magnets produced as part of the REProMag project.

2. Experimental

2.1 Magnetic Viscosity

Magnetic viscosity was investigated by performing a series of measurements on an NdFeB magnet with an H_{cJ} value of approximately 1330 kA/m. For each measurement the polarization reversal time (PRT) was changed to investigate the effect of different applied magnetic field strength sweep rates (dH/dt) on the measured coercivity. The PRT is defined as the time taken for the polarization to reduce from 90% of B_r to zero. The longest PRT was > 665 s equating to a dH/dt of 0.002 kA/m/s and the shortest was < 5 s, equating to a dH/dt of 1.4 kA/m/s. The measurements were performed on a metrological grade electromagnet in accordance with IEC 60404 – 5, set up as shown in Figure 2.

The applied magnetic field strength was measured using a transverse Hall probe connected to a Bell 9640 gaussmeter. The magnetic polarization was measured using a two hundred turn Magnet Physik J coil attached to a Magnetech MIF-1 integrating fluxmeter. The outputs of both the gaussmeter and fluxmeter were connected to digital multimeters and were logged by a validated Labview data acquisition programme. The Hall probe and gaussmeter were calibrated against a nuclear magnetic resonance device, which gains traceability via frequency. The integrator was calibrated by passing a known current through the primary winding of a calibrated mutual inductor and then breaking the current to generate a known flux across the secondary. During the measurement of the sample a load of 7.8 Ω was used over the integrating fluxmeter to match the loading of the mutual inductor used in the calibration.

[1] The aspect ratio is the ratio of diameter to the length of the cylinder. A 10 x 10 mm cylinder has an aspect ratio of 1 whereas a cylinder of 5 mm diameter and 10 mm length would have an aspect ratio of 2.

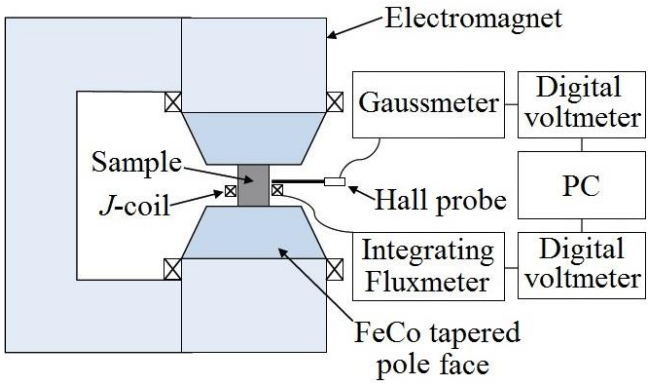

Fig. 2. A schematic of the electromagnet set-up taken from [11].

2.2 Demagnetization Factor

The determination and investigation of N_m was undertaken using a relatively simple method developed at NPL, when compared to the complex modelling. A complete discussion of the method can be found in [12], so only a brief summary will be presented here. An identical set up to the viscosity measurement was used for the measurement of the second quadrant demagnetization curve. Unlike the viscosity measurements however, the PRT was kept high (> 100 s) in order to approach quasi-static conditions, as is best practice. The Kennelly dipole moment, m_k, of the sample was then measured using a Helmholtz coil [13] and the working point polarization, J_m, of the sample calculated from m_k and the volume, V.

$$J_m = {}^{m_k}/_{V} \tag{2}$$

By locating the calculated working point polarization on the demagnetization curve, a gradient line can be drawn from this point to the origin and the magnetometric demagnetization factor can be determined. This method is limited to only the magnetometric demagnetization factor as the moment measurement takes the volume average of the sample. The experimental set-up for the moment measurement can be seen in Figure 3, with the induced e.m.f across the coils being measured as a volt-second product by an integrating fluxmeter. The Helmholtz coil used during the measurement has an internal radius of 130 mm, with a coil constant determined by electron-spin resonance.

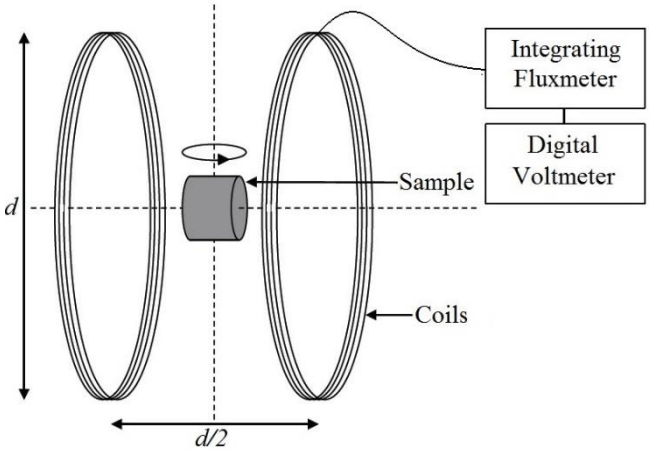

Fig. 3. Schematic of the Helmholtz coil used for the determination of m_k. d/2 is equal to 130 mm for this set up.

The samples chosen were a series of NdFeB cylinders with aspect ratio ranging from 1 up to 2.5. These aspect ratios cover a significantly smaller range than the modelled values but are optimised for the experimental measurement set-up. The shortest length of the magnets was 10 mm with a maximum length of 18.4 mm. The dimensions of the sample were selected to keep the volume of each sample the same. This choice was deliberate to limit the number of calibrations required of the integrator and also to show that the changes in J_m were a consequence of the geometry and not due to the overall volume.

3. Results and discussion

3.1. Magnetic Viscosity Results

The results from the magnetic viscosity measurement can be seen in Figure 4. The graph plots the measured coercivity against the natural logarithm of the sweep rate of the applied magnetic field. The H_{cJ} vs Ln(dH/dt) relationship does not immediately seem relevant, however if one takes viscosity effects to be from thermally activated processes then the magnetization must relax according to:

$$M(t) = A + Bexp(-t/\tau) \tag{3}$$

where A and B are constants and τ is the relaxation time, following the Arrhenius law. If one also assumes that there is no distribution of relaxation times the dependence of the coercivity on sweep rate can be deduced [9]:

$$H_c(\dot{H}) = H_f \ln(\dot{H}) + constant \tag{4}$$

where $\dot{H}$ is the time derivative of the field and H_f is a fictitious field, labelled as the thermal fluctuation field dependent on the Boltzman constant, k_B, temperature,

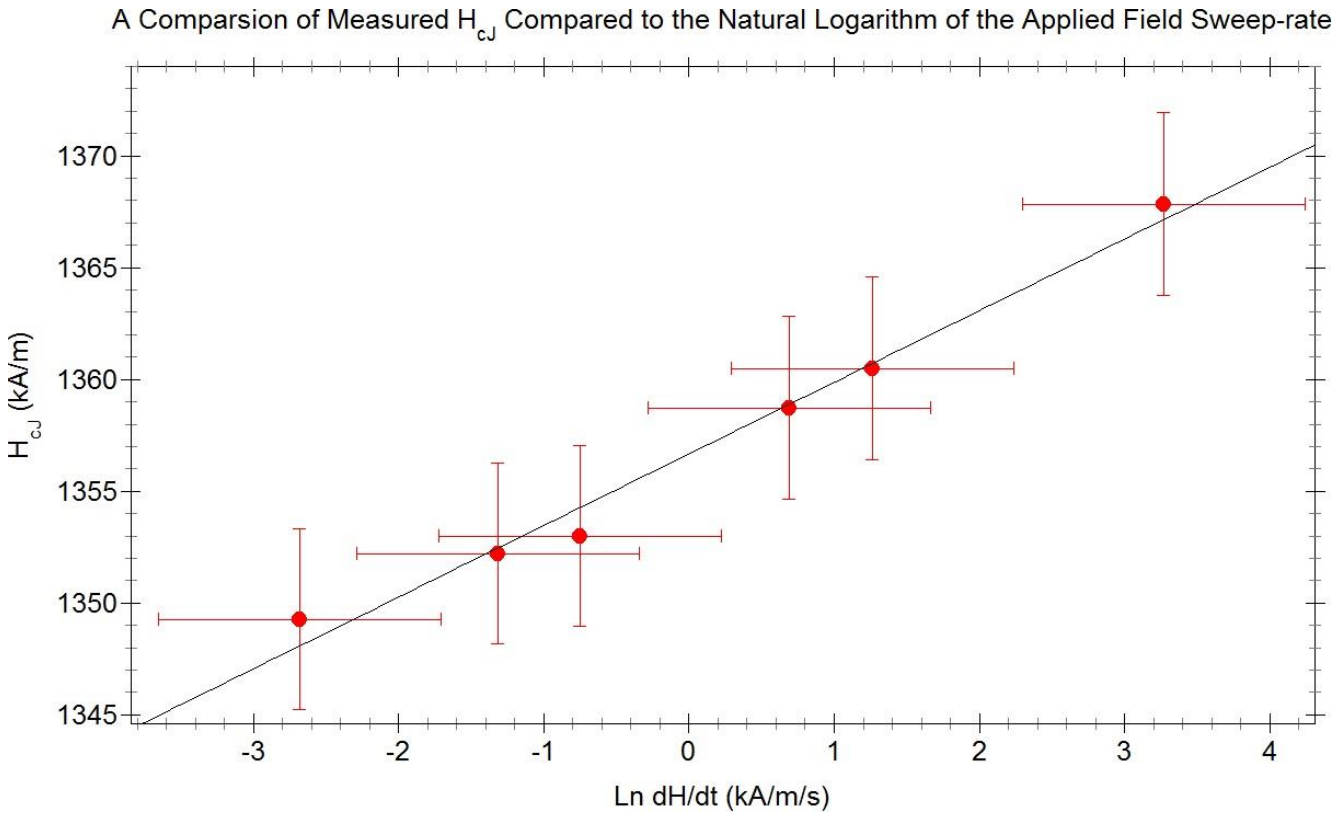

Fig.4. The plot of Ln(d*H*/d*t*) against measured coercivity displaying the expected linear behaviour

T, magnetization saturation, M_s and activation volume, *V**.

$$H_f = {}^{k_B T}/_{V^* M_s} \quad (5)$$

Figure 4 displays the expected linear behaviour between sweep rate and coercivity. This linear relationship allows for the direct determination of the thermal fluctuation field. Further work extending the range of sweep rate is required to truly explore the relationship, and to confirm linearity as the sweep rate approaches a quasi-static state.

3.2. Magnetometric Demagnetization Factor Results

The results of the study of N_m are given alongside the theoretical results from Chen *et al* in Table 1. The experimental data for aspect ratios of > 1.5 all show results lower than the modelled results, whereas results for aspect ratios of 1 and 1.5 are higher. Further tests of samples with aspect ratios between 1.5 and 2 will allow investigation of this crossing of the two data sets and whether there is a physical significance of the point where the two sets are equal. The differences seen between the data sets is hypothesised to be related to the material assumptions, such as isotropic and homogenous behaviour of the material and a constant susceptibility, used by the model.

The benefit of the experimental technique is that true material data is used, which takes into account the non-linear hysteretic behaviour of ferromagnetic materials. As the cylinder geometry tends towards a rod it is expected that the demagnetization factor should reduce, which is seen in both data sets, but far more quickly in the experimental data. The method of calculating the distribution of magnetic poles in the numerical method may have caused an overestimate at the boundaries, leading to the large discrepancies observed in longer samples. Further studies of different aspect ratios and different materials are required to formally conclude this topic. The results from this study give confidence that this method can be extended to more complex shapes (provided the surfaces are parallel). This will allow investigation of real-world shapes such as hollow tubes, and will directly feed into the REProMag project.

Tab.1. The experimental results compared to the widely accepted numerically modelled results for the magnetometric demagnetization factor.

Aspect Ratio	Theoretical N_m	Measured N_m	Estimated Uncertainty ±%	Difference w.r.t theoretical values %
1	0.312	0.332	4.8	6.41
1.5	0.230	0.235	7.1	2.17
2	0.182	0.164	9.3	-9.89
2.5	0.150	0.093	8.1	-38.00

4. Conclusion

This paper has reviewed pulsed field magnetometry, outlining the advantages and disadvantages. The two most significant correction factors for the measurement of permanent magnets with complex geometries have been discussed and the work to evaluate them presented. The large discrepancy seen between the experimentally determined magnetometric demagnetization factor and the theoretically derived value illustrates the need for further investigation of these phenomena. Currently the PFM at NPL is capable of performing measurements on complex geometry components as part of a batch variation test. This capability allows rapid evaluation of new material innovations and developments. This gives immediate feedback to the materials engineers developing the material synthesis and binding solutions. With continued work, this technique will be able to provide metrological standard characterization of the demonstrator magnets, which is currently impractical with any other magnetic measurement technique.

Acknowledgement

The authors would like to thank the National Measurement System of the Department for Business, Innovation and Skills that provides funding to develop and maintain the measurement systems at NPL. Research has been done as part of the project “Resource Efficient Production for Magnets – REProMag” financed by the European Commission under the Horizon 2020 FoF-Framework under grant agreement 636881.

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