HYDROCYCLONE SEPARATION OF HYDROGEN DECREPITATED NdFeB

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Abstract

Hydrogen decrepitation (HD) is an effective and environmental friendly technique for recycling of neodymium-iron-boron (NdFeB) magnets. During the HD process, the NdFeB breaks down into a matrix phase ($Nd_2Fe_{14}BH_x$) and a grain boundary phase ($NdH_{2.7}$) that are 10-20 μm and <2 μm in size respectively. Recycled NdFeB material has a higher oxygen content compared to primary source material. This additional oxygen mainly occurs at the Nd-rich grain boundary phase (GBP) because rare earth elements oxidise rapidly when exposed to air. This higher oxygen level in the material results in a drop in density, coercivity, and remanence of sintered NdFeB magnets. The particle size of the GBP is too small to separate by sieving or conventional screening technology. In this work, an attempt has been made to separate the GBP from the matrix phase using a hydrocyclone. HD powder, obtained from hard disk drive (HDD) scrap NdFeB sintered magnets, was used as a starting material and passed through a hydrocyclone for a total number of six times. The X-ray fluorescence (XRF) analysis showed the matrix phase had been directed to the underflow while the GBP was directed to the overflow. The overflow streams have significantly higher amounts of oxygen than the starting material and underflow streams. The optimum separation was achieved with three passes.

Keywords: Hydrocyclone, Centrifugal separation, Fine particle separation, NdFeB, Recycling, Rare earth elements

Introduction

The growing need for sustainable technologies is resulting in an increasing emphasis on the recycling of materials. This development is particularly important in the case of NdFeB sintered magnets where there is an appreciable growth in demand and a high degree of wastage [2]. NdFeB magnets possess the highest energy product of all permanent magnets which makes them highly efficient and suitable for lightweight mobile applications [3]. Therefore, they are widely used in computer hard drives (HDDs), load speakers, medical imaging, household electrical appliances, hybrid and electric vehicles (HEVs and EVs), wind turbines, and many other small consumer electronic devices. The amount being used varies between a few grams (e.g. loudspeakers) to tonnes of materials (e.g. wind turbines) [4].

Previous studies at the University of Birmingham have shown that hydrogen can efficiently be used to separate NdFeB magnets from HDD scrap [5]. NdFeB magnets become demagnetised when reacted with hydrogen, thus allowing the powder to be separated much more readily. During this hydrogen decrepitation process, the NdFeB magnets absorb hydrogen and break down into an interstitial matrix phase hydride ($Nd_2Fe_{14}BH_x$) and a grain boundary phase hydride ($NdH_{2.7}$) that are 10-20 μm and <2 μm in size respectively [6]. During sintering of NdFeB magnets, the Nd-rich GBP melts down, resulting in liquid phase sintering. When the recycled HD powder is used to re-manufacture sintered NdFeB magnets, the GBP has a higher oxygen content and therefore it does not melt during the re-sintering process which results in a lower density magnet and a reduction in coercivity, remanence, and maximum energy product. The aim of this work is, therefore, to separate the grain boundary phase (<2 μm) with higher oxygen content from the matrix phase (10-20 μm) using a hydrocyclone.

Hydrocyclones have been used in the chemical and mineral industries for many years. Their usage is very wide in the mineral, chemical and bio industries due to the simple design, operational flexibility and low operation and maintenance costs. The devices use centrifugal forces to separate two products of different densities or sizes [7, 8]. Despite their simplicity and low cost, they are very efficient for solid-liquid separations [9]. By using suitable materials and methods of construction, hydrocyclones can be operated at high temperature and/or pressure circumstances, where the development of high efficient devices could have a significant impact in the energy and processing industries [10].

1 Experimental

1.1 Material

The starting material used in this study was a hydrogen decrepitated (HD) powder obtained from hard disk drive (HDD) scrap. A batch of NdFeB magnets, obtained from hard disk drive scrap, was processed at 2 bar hydrogen and room temperature for 2-4 hours. The extraction of NdFeB magnets from hard disk drives is explained in detail by Walton et al., 2015 [5]. Hydrogen decrepitated material was then exposed to air (for controlled oxidation) before mixing it with water. Water acts as a medium for processing of the powder through the cyclone and it reduces the build-up of triboelectric charges between fine particles. This, therefore, allows for better separation efficiency in the case of very fine particles.

1.2 Working principle of hydrocyclone

A hydrocyclone consists of two main parts as shown in figure 1. The first is a cylindrical part with feed inlet. This part also includes an outlet, located at the top of the cylinder, extends into the cylinder and is known as the vortex finder. The second main part is conical and is connected to the cylindrical section at the top and to the underflow at the bottom end. The latter part is known as the spigot [11].

The feed slurry, under pressure, enters at the tangential inlet at the top of the hydrocyclone. As the feed enters the chamber, a rotation of the slurry inside of the cyclone begins to accelerate the movement of the particles. This circular acceleration of the fluid directs the heavier particles towards the outer wall under the action of a centrifugal force. The particles migrate in a spiral pattern through the cylindrical section into the conical section. Radial movement is hindered by the drag force as the particles move through the carrying fluid. At this point, the smaller particles migrate toward the centre of cyclone and spiral upward through the vortex finder. This product, which contains the finer particles and the majority of the water, is termed the overflow. As the flow descends in the hydrocyclone, the layer adjacent

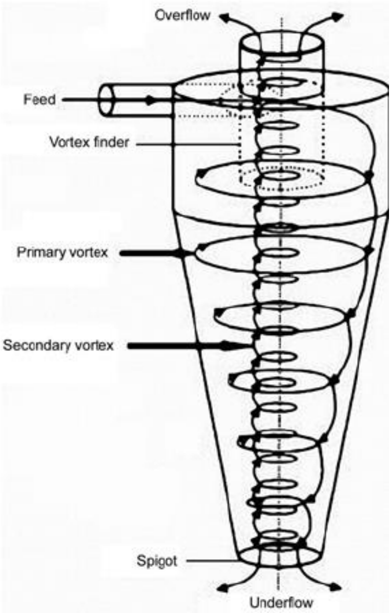

Figure 1. Schematic diagram of a hydrocyclone [1]

to the hydrocyclone wall becomes loaded with heavy particles, which exit the device through the spigot orifice of the cyclone, is termed as underflow [7, 8].

1.3 Methods

The experiments were carried out with a Mozley C700 rig, using a 50 mm hydrocyclone (vortex finder size was 14.3 mm and diameter of the spigot was 4.5 mm which is illustrated in figure1).

The system was operated at a constant feed pressure of 0.8 bar. 1 kg of HD material was mixed with 10 liter water in the tank and circulated through the cyclone. At equilibrium, the hydrocyclone underflow and overflow were sampled for a set period, normally 2 s. Underflow and overflow sample volumes were measured. The underflow of the previous pass was mixed with 10 liter water and used as the feed for the next pass. The experiment was repeated for six passes and samples were collected after each pass.

Samples from each pass were then filtered and dried at 80 °C in an oven in air. These samples were then analysed using an XRF spectrometer (S8 Tiger, Haworth 503A) as pressed pellets. 0.5 g of sample material was mixed with 0.1 g wax and then pressed in the form of a pellet. Full detection mode (8 minutes) was selected for XRF analysis. Oxygen analysis was performed by Less Common Metals (LCM), Cheshire, UK using an LECO instrument. Particle size distribution was performed using a Malvern Mastersizer 2000S.

2 Results and Discussion

Results obtained from hydrocyclone separation are presented in table 1. It can be seen that there is a sudden increase, from 1.62% to 2.45%, in the material going into the overflow between the third and fourth pass, which shows a yield loss. It means that most of the smaller particles are separated into the overflow in first three passes, and there is not much GBP left to separate in the feed as we go for the fourth hydrocyclone pass.

Table 1: Hydrocyclone data for hydrogen decrepitated NdFeB

Hydro-cyclone passes	Feed (g)	Underflow (g)	Overflow (g)	Overflow/ Reject (%)	Underflow/ Yield (%)
Pass 1	1000.00	987.07	12.93	1.29	98.71
Pass 2	793.67	779.93	13.74	1.73	98.27
Pass 3	653.32	642.76	10.57	1.62	98.38
Pass 4	563.27	549.45	13.82	2.45	97.55
Pass 5	486.06	474.39	11.67	2.40	97.60
Pass 6	428.64	418.30	10.34	2.41	97.59

Table 2 presents the results obtained from XRF and oxygen analysis of the feed and underflow fractions. The term ∑REE represents the sum of rare earth elements in the material (Nd + Dy). The hydrided matrix phase ($Nd_2Fe_{14}BH_x$) being large size particles should report to the underflow and grain boundary phase ($NdH_{2.7}$) being smaller should report to the overflow.

As both of these particles have Nd in them, we have to optimise the number of passes required for this separation based on total rare earth elements.

Table 2: Chemical analysis of underflow streams in weight %

Samples	Fe	Nd	Dy	∑REE	Oxygen (%)
Starting Material	47.45	33.48	1.21	34.69	1.1
Underflow 1	54.55	33.38	1.07	34.45	1.9
Underflow 2	54.02	33.05	1.23	34.28	2.1
Underflow 3	56.53	32.38	1.19	33.57	1.9
Underflow 4	55.78	32.48	1.23	33.71	2.0
Underflow 5	57.17	32.70	1.24	33.94	2.1
Underflow 6	54.55	33.38	1.07	34.45	1.9

It can be seen from the table 2 that the concentration of iron is increasing with each hydrocyclone pass whereas the amount of neodymium (Nd) is decreasing as expected. On the other hand, total rare earth content decreases until the third pass and then it starts increasing again. The reason for this could be that either all the small particles have separated at the end of the third pass or the remaining small particles adhere to the larger particles by triboelectric charges and cannot be further separated.

The XRF and oxygen analysis of the overflow fractions are presented in Table 3. The presence of Fe and Dy in the overflow means that some of the large particles (matrix phase) are also going to the overflow alongside small particles (GBP). Although Dy is also present in the GBP which means that some of the Dy detected in overflow comes from both matrix phase and GBP. From the fourth pass, the amount of Fe suddenly started to increase in the overflow, which means that after third pass there is more of the matrix phase in the overflow than grain boundary phase.

Table 3: Chemical analysis of overflow streams in weight %

Samples	Fe	Nd	Dy	∑REE	Oxygen (%)
Starting Material	52.80	33.03	1.03	34.06	1.1
Overflow 1	20.59	68.54	1.21	69.75	15.7
Overflow 2	21.74	65.19	1.24	66.43	15.9
Overflow 3	22.65	67.52	1.27	68.79	18.1
Overflow 4	31.16	61.00	1.33	62.33	15.6
Overflow 5	32.63	59.98	1.24	61.22	20.4
Overflow 6	35.32	54.41	1.19	55.60	17.4

The oxygen content in all underflow samples (Table 2) is slightly higher than the starting material (feed) which is likely to be due to the neodymium reacting with water to form neodymium hydroxide [6]. Formation of $Nd(OH)_3$ increases the amount of oxygen present in the starting material. On the other hand, there is a considerably higher amount of oxygen is present in overflow streams (Table 3). This means that the smaller particles, which are mainly neodymium oxide and hydroxide, are successfully separated from the matrix phase by this separation process.

The particle size distribution results from the Mastersizer are shown in figure 2 below. The figure shows that the particles in the starting material (feed) were distributed between 1.5 microns and 120 microns. It can also be seen that in the feed, 10% of total particles are less than 14 microns.

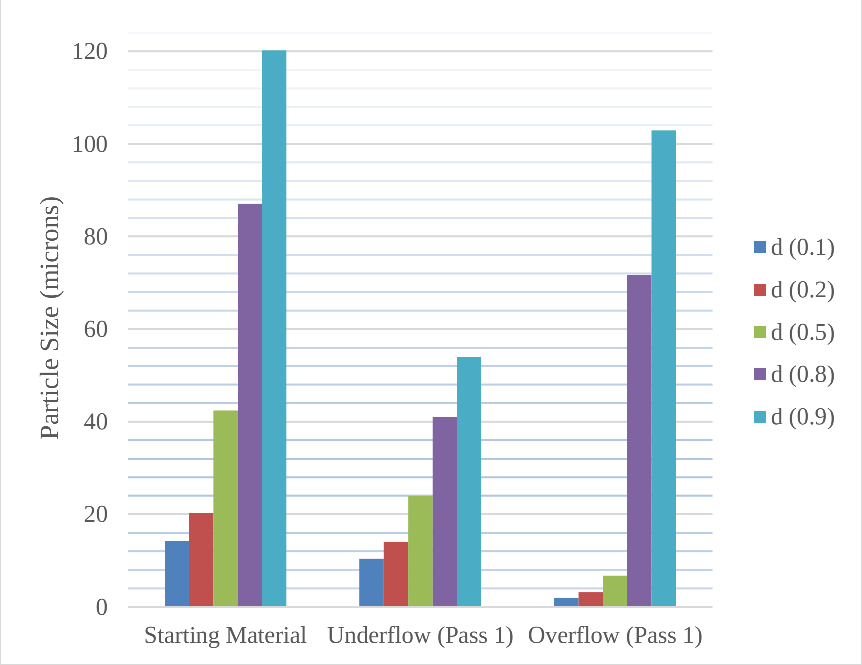

Figure 2. Particle size distribution in feed, underflow and overflow after first pass

It can be seen that the particle size has considerably reduced in the underflow obtained after the first pass. It seems that larger particles initially present in the feed kept breaking down into smaller particles as they pass through the hydrocyclone system. The reason for this size reduction is that the powder produced by the HD process is extremely friable. The friability in the material is due to the presence of microcracks along the grains produced during absorption of hydrogen [12].

The overflow stream has mainly smaller particles (GBP) along with some larger particles (matrix phase). Evidence of the presence of matrix phase in overflow was also found in the chemical analysis (Table 3). Figure 2 also shows that particle size distribution (D50) is reduced from 42 microns (starting material) to 24 microns in underflow and further to 7 microns in overflow just after the first pass.

3 Conclusion

Hydrocyclone separation has been shown to be an effective method to separate the matrix $Nd_2Fe_{14}BH_x$ from the oxygen rich grain boundary phase for HD processed NdFeB. The oxygen-rich grain boundary phase was successfully separated in the overflow along with some matrix phase particles. The overflow streams were found to have a higher quantity of oxygen than the starting material and underflow streams. This hydrocyclone separation process was also optimised and the total number of passes required for optimum separation was found to be three. After the third pass, a considerable amount of matrix phase was found in the overflow resulting in a yield loss.

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