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Application of Taguchi method for optimization of process parameters in decalcification of samarium–cobalt intermetallic powder

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ABSTRACT

Decalcification studies on samarium–cobalt intermetallic alloy powder produced by reduction–diffusion process have been carried out under wide range of experimental conditions based on Taguchi method. This approach has been adopted to arrive at optimum conditions for the leaching operation for calcium removal without affecting the yield of Sm_2Co_{17}. The percentage effect of number of water wash, acid wash, pH and digestion time on decalcification was investigated. Conditions predicted by Taguchi method successfully brought down the residual calcium value to about 1750 ppm in Sm_2Co_{17} suitable for further operation related to making permanent magnet. The Sm_2Co_{17} phase was found to be stable even after long period of leaching operations as characterized by scanning electron microscopy and X-ray diffraction. Leaching of calcium from large batch product samples have been demonstrated successfully with 99.5% yield and residual calcium in the range of 1740–1750 ppm. Acetic acid and propionic acid showed comparable results in decalcification.

1. Introduction

Permanent magnets based on rare earth-transition metal intermetallics are the basis of ever increasing number of commercial and scientific applications, including electric motors, NMR scanners, wind-mills, compact discs, actuators for robotics and flight control, etc. [1–3]. The advantage of permanent magnets in these applications is their ability to exhibit high level, constant magnetic fluxes without applying an external magnetic field or electrical current. Samarium–cobalt based magnets with high energy product and excellent coercive force, are ideally compact and suitable to highly efficient machine and components in which higher operation temperature, higher corrosion and oxidation resistance are crucial [4]. Iron–neodymium–boron (Nd–Fe–B) and other rare earth-iron/cobalt-based intermetallics have been investigated due to their superior magnetic properties [5–7]. However, corrosion resistance is a primary concern with Nd–Fe–B type of magnet and it has limitations to be used at higher temperature. In view of growing indigenous demand of samarium–cobalt based permanent magnet in particular Sm_2Co_{17} for specific applications, attention has been paid to prepare this magnetic material. The Sm_2Co_{17} powder can either be prepared by melting individual elements, samarium (Sm) and cobalt (Co), with required composition and casting the resultant melt [8,9] or by reduction–diffusion (R–D) process [5–10]. The former process requires higher temperature (>1600 °C) and thus resulting in significant loss of Sm. The R–D process involves mixing the required amount of samarium oxide (Sm_2O_3) with Co powder and heating the mixture at higher temperature (~1100–1200 °C) in the presence of calcium (Ca) under inert atmosphere to form the intermetallic. The advantage of the R–D process over the co-melting process is that it starts with rare earth oxides instead of the costly rare earth metals. Further, the energy input for crushing the material to bring into powder form before compaction and sintering is less for R–D process. Thus this method is economical for large scale production. The resulting alloy powder in the R–D process contains significant amount of Ca (10–15%) in the form of calcium oxide (CaO) or unreacted Ca, which need to be removed before using the powder for making permanent magnets. If not removed to the maximum permissible limits, it interferes in the subsequent stages of magnetization by decreasing the energy product value. Chen et al. [11] have reported that with increase in Ca content in the rare earth-transition metal based intermetallics from 2100 ppm to 3200 ppm, the maximum energy product $(BH)_{max}$ decreased from 24.7 MGOe to 21.0 MGOe. It clearly indicates that the Ca content in the alloy powder should be <2000 ppm in order to obtain $(BH)_{Max}$ value more than 25 MGOe. In general Ca is removed by water washing (leaching) to a greater extent by taking the advantage of partial solubility of calcium hydroxide in aqueous solution [12]. However to bring the value to the desired level of <2000 ppm use of organic acid such as, acetic acid in presence of ammonium hydroxide is reported [13]. Use of formic acid, NH_4Cl, EDTA, propionic acid, neodecanoic acid for the complexation of Ca to yield corresponding

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soluble compound has also been reported [13–15]. Literature survey revealed that the reports on decalcification of rare earth based intermetallics alloy powder are available mostly in patents [11–16], where the optimization of process parameters is seldom described. Additionally the conditions reported vary considerably from one report to another. The leaching is carried out under agitated condition in order to enhance the mass transfer. Experimental variables including solid to liquid ratio (S/L), number of water and acid wash cycle, concentration of acid, digestion time, agitator speed etc., play important roles in decalcification. It is therefore desirable to investigate the role of these process variables to arrive at optimum conditions for effective leaching of Ca without distorting the required phase suitable for magnetization. With this objective present work has been initiated to study the effect of different parameters on decalcification and tried to optimize them to get minimum residual Ca with maximum yield of Sm_2Co_{17} powder. In order to optimize the process variables, Taguchi method [17] has been adopted and applied successfully in the present work to select the most influential parameters and their effect on decalcification. Four parameters namely, number of water wash cycle, number of acid wash cycle, pH of the solution and digestion time in each cycle have been selected and varied in three different levels. Other parameters, such as, agitation speed, solid to liquid ratio and temperature were kept fixed at levels decided based on our experience. Apparently this is a four factor three level system. If one uses full factorial design [18], 3^4 or 81 numbers of experiments are required to be carried out to get necessary information whereas Taguchi method allows us to carry out only nine experiments in order to get same information. With the help of these nine experiments the leaching parameters have been optimized and discussed in this paper. A comparative evaluation on leaching of Sm–Co alloy powder by acetic and propionic acid has also been carried out in this investigation.

2. Taguchi method

Taguchi method [17] is a multi-parameter optimization procedure, which is very useful in identifying and optimizing dominant process parameters with a minimum number of experiments. The method is based on an orthogonal array [19] of experiments. An orthogonal array is a minimal set of experiments with various combinations of parameter levels. Output of the orthogonal array, which indicates the relative influences of various parameters on the formation of the desired product, is used to optimize an objective function. There are three types of objective functions: larger-the-better, smaller-the-better and nominal-the-best. The influences are commonly referred in terms of S/N (signal to noise) ratio.

For optimization of residual calcium, smaller-the-better type of objective function has been used. In this case the exact relation between S/N ratio and the signal is given by

$$\mathrm{S/N} = -10\log\left(\frac{1}{n}\right)\sum_{i}^{n} y_i^2 \quad (1)$$

whereas for optimization of yield of Sm_2Co_{17} powder, larger-the-better type of objective function has been used. Here, the exact relation between S/N ratio and the signal is given by

$$\mathrm{S/N} = -10\log\left(\frac{1}{n}\right)\sum_{i}^{n} 1/y_i^2 \quad (2)$$

where y_i is the signal (residual calcium or yield of Sm_2Co_{17} powder) measured in each experiment averaged over n repetitions. The effect of a parameter level on the S/N ratio, i.e., the deviation it causes from the overall mean of signal, is obtained by analysis of mean (ANOM). The relative effect of process parameters can be obtained from analysis of variance (ANOVA) of S/N ratios. Computation of ANOM and ANOVA are done by using following relations.

$$m_i = \left(\frac{1}{N_l}\right)\sum S/N \quad (3)$$

and

$$Sum\,of\,squares\,(SoS) = \sum_{i=i}^{i=j} N_l(m_i - \langle m_i \rangle)^2 \quad (4)$$

where m_i represents the contribution of each parameter level to S/N ratio, $\langle m_i \rangle$ is the average of m_i's for a given parameter and the coefficient and N_l represents the number of times the experiment is conducted with the same factor level in the entire experimental region. SoS is obtained by using ANOVA. This term is divided by corresponding degrees of freedom (DoF = number of parameter level minus 1) to derive relative importance of various experimental parameters by utilizing Eq. (5)

$$Factor\,effect = \frac{SoS}{\{DoF\,X\,\sum(SoS/DoF)\}} \quad (5)$$

Several researchers [20–23] have extensively used this technique in optimization of parameters in various processes. To the best of our knowledge for the first time this technique is being used to optimize the leaching parameters to get the minimum residual calcium with maximum yield of desired intermetallic phase in reduction–diffusion product.

3. Experimental

3.1. Synthesis of samarium–cobalt intermetallics

Calculated amount of Sm_2O_3 (>99% pure, supplied by Indian Rare Earth Limited) was mixed with Co powder (99.6% pure,

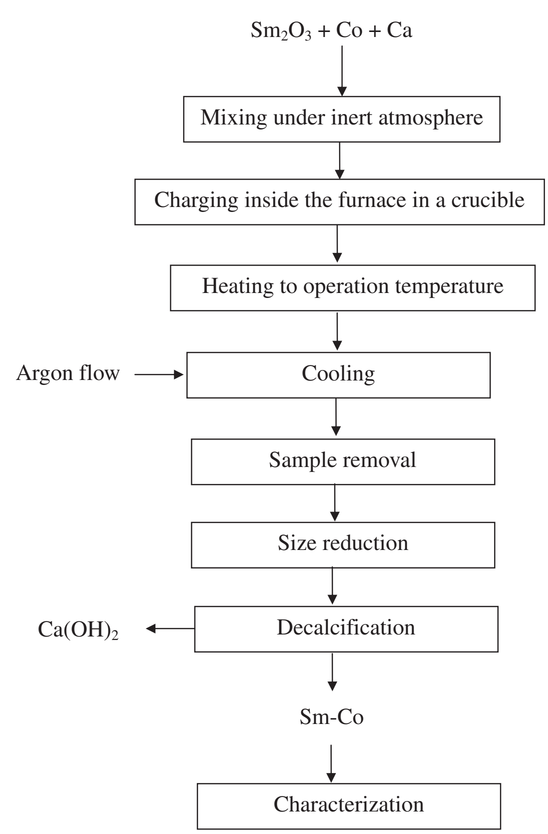

Fig. 1. Schematic diagram of R–D process for producing Sm_2Co_{17} intermetallic.