2. WARM COMPACTION OF AN Fe-Si/Fe POWDER MIXTURE

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Abstract. In this study, we present high-density nanostructured Fe-Si/Fe-based soft/soft composite, prepared by the warm compaction method. Ball-milled Fe-33Si (a/o) powder, pure Fe powder, and polyvinyl pyrrolidone (PVP) were mixed in an acetone solvent to create a powder mixture and then compacted at various temperatures and pressures. Sample densities were obtained up to a 90% theoretical value. The warm-compacted sample prepared at 650 MPa and 240 °C for 1h had the highest compaction properties in comparison with other samples prepared at pressures of either 300 or 400 MPa, and at room temperature or 120 °C.

1. INTRODUCTION

Soft magnetic composite materials, such as an Fe-based alloy, are characterized by magnetic and thermal isotropy, a very low eddy current loss, a relatively low total core loss at low and medium frequencies, large anisotropy constants, low coercivity, and high Curie temperatures [1-7]. Additionally, it has been reported that the presence of silicon in Fe-Si alloy powders plays a key role in not only decreasing magnetic anisotropy, but also increasing electrical resistivity, resulting in a further decreased coercivity and eddy current loss [1, 2].

Due to a rapid increase in the demand for soft magnetic composite parts, recent attempts have been made to improve their properties, such as an increase in the silicon content, the addition of insulator materials, and various other processing methods. However, conventional compaction methods make it very difficult to obtain a high density, because the high-silicon content causes the Fe-based material to be too brittle. Moreover, sintering processes cannot be used to make high density, due to the presence of polymer materials, such as epoxy insulators.

High pressures should be applied to produce a compressed powder magnetic core without sintering. For this reason, the high density of the compacted sample, and the elimination of residual stresses in the compaction step, are very important. Various approaches have been proposed to increase the density without using the sintering process. Among these approaches, a warm compaction method has been used for high and uniform densities, a reduction of residual stresses, and the improvement of magnetic properties [3-5,8]. This method has been established with the knowledge that metallic powders have better plasticity and compressibility at elevated temperatures and are, therefore, easier to be deformed to form high-density bulks, compared to cold compaction. In the warm compaction method, the material powder is preheated and compacted in a heated die. The warm compaction process has been developed to manufacture cost-effective and highly dense sintered products, with densities equivalent to those attained by double pressing and double sintering processes [8]. In this study, powder mixtures with Fe-33Si (a/o) and Fe were used to increase the compaction density by having the soft Fe phase act as a buffer for the high pressure during the compaction process.
Fig. 1. (a) SEM images of Fe-33Si powder ball-milled for 20 h and (b) ball-milled Fe-Si/Fe powder mixture.

Fig. 2. The density changes for different compaction pressures and temperatures.

2. EXPERIMENTAL PROCEDURES

Commercial Fe-33Si (a/o) alloy powder, and iron powder (Hoeganaes Co.) produced by gas atomization were used as starting materials. The purity of the Fe-33Si powder was above 99%, while the purity of the iron powder was above 99.9%. Initially, the particles were smaller than 20 nm. The Fe-33Si powder was sealed in a grinding unit made with stainless steel balls and a stainless steel jar under an argon atmosphere. A high-energy ball-milling process was then performed by an attritor (Zoz Gmbh, CM101) at 400 rpm for 20 hours, with a ball-to-powder weight ratio of 10:1. The ball-milled Fe-33Si powder was then mixed with the pure Fe powder for 5 hours by a 3-D mixer. Polyvinyl pyrrolidone (PVP) was added to the powder mixture continuously with an acetone solvent. After the mixed powder was dried in an oven, the composition of the powder mixture was Fe-10Si (a/o). The powder mixture was then cold and warm compacted, using pressures of 300, 400, 500, and 650 Mpa, in a cylindrical die, having a diameter of 10 mm. To investigate the effect of compaction temperature, the powders were compacted at room temperature, 120 °C, 180 °C, and 240 °C for 1h.

Phase identification of the fabricated powders was performed using an X-ray diffractometer (XRD, Cu K$_\alpha$; $\lambda = 1.540562$ Å), and electron probe microanalysis (EPMA). The density of the samples was determined by the Archimedeans principle. The magnetic properties of the samples were evaluated with a vibration sample magnetometer (VSM) at an applied magnetic field of up to ± 1.0 MA/m at room temperature and a B-H curve analyzer and a impedance analyzer in the frequency range of 10~1000 kHz at 10 mT.

3. RESULTS AND DISCUSSION

Microstructures of the pure Fe and ball-milled Fe-33Si alloy powders were obtained over a period of 20 hours (Fig. 1). The starting powder had an average particle size of 3 μm. In the case of the ball-milled Fe-Si powder, the average crystalline size was calculated by the Williamson-Hall equation [9] from the X-ray diffraction pattern, and the result was nearly 16 nm. Furthermore, X-ray diffraction analysis confirmed that the Fe-33Si alloy powder was composed of fcc-Fe$_3$Si and hcp-Fe$_5$Si$_3$. It is known that the density of a randomly packed particle is in the range of 70% when no deformation is involved. To obtain a higher density, it is necessary to perform plastic deformation of the particles. However, because the Fe$_3$Si and Fe$_5$Si$_3$ are hard phase materials, it is difficult to compact by conventional compaction method. Moreover, the deformation of a...
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Fig. 3. XRD analysis of cold and warm compacts of Fe-Si/Fe mixed powder.

nanostructured powder cannot easily be compared with a micron-sized powder, due to the reduced dislocations in the powder, which explains the low density obtained in the nanostructured powder compacts.

In this study, the change in green densities for different cold compaction pressures increased linearly with the increasing compaction pressure from 300 to 650 MPa. The green densities of 300 and 650 MPa were 4.75 and 5.31 g/cm³, respectively. Additionally, the green density of the cold compaction with polyvinyl pyrrolidone (PVP) was nearly 0.8 g/cm³ higher than the sample without PVP. In fact, the main functions of PVP in the powder mixture were the reduction of friction between powder particles during compaction and the encapsulation of the powder. This caused a higher density through an increased effective pressure on the powder, resulting in improved mechanical properties.

The warm compaction densities of the Fe-Si/Fe powder mixture at different pressures under various temperatures were also determined (Fig. 2). It was clear that the compacted powder density increased with an increase in the compaction temperature, under constant pressure. There was a significant influence of temperature on the deformation behavior. The deformation of metals was considered to be a thermally activated process, where both elastic and plastic deformation was affected. Additionally, soft phase iron powder can achieve high density due to an increase in its plastic deformation capability, as opposed to the hard phase Fe-Si in a Fe-Si/Fe soft magnet powder mixture. A monotonous increase of the density was observed for the Fe-Si/Fe powders over the entire studied temperature range (Fig. 2). This may be explained by the fact that the yield strength of metallic Fe-Si and Fe decreased linearly with temperature in the region between 20 and 240 °C. To investigate the effect of PVP contents on green density, warm compaction was performed at 650 MPa and 240 °C. The green densities of 0, 0.2, 0.6, and 1.2 w/o PVP content were 6.31, 6.49, 6.51, and 6.51 g/cm³, respectively. As previously mentioned, PVP led to a higher green density by reducing friction between powder particles during compaction. Alternatively, due to the low density of the polymer, the compacted powder density was lowered at higher amounts of PVP. Therefore, the limit of the polymer contents was 1.2 w/o PVP in this study. However, the green density did not increase above a PVP content of 0.6 w/o.

Fig. 3 shows the XRD patterns of cold and warm compacts of Fe-Si/Fe mixed powder. The α-Fe (Fe powder) and Fe₃Si (Fe-33Si powder) peaks were confirmed from cold and warm compacted samples. In the cold compacted sample, the Fe and Fe₃Si peaks were broadened, whereas peaks of warm compacted sample were more sharpened and narrowed. It was considered because stresses induced by compaction pressure were relieved during warm compaction.

An EPMA mapping image of the warm compacted sample at 650 MPa and 240 °C was obtained (Fig. 4). The image confirmed that while the Fe-Si powders (Si mapping image) were maintained spherical shape, the iron powders (Fe mapping image) were deformed among the Fe-Si powders. It was evidenced that the density of the Fe-33Si/Fe composite sample was higher than the density of the gas atomized Fe-10Si alloy sample, because the soft phase iron powder was filled at the unoccupied site by deformation.

It is generally known that higher density results in improved magnetic properties. The magnetic properties also were sensitive to stress, and were deteriorated by compaction. However, these stresses could be relieved by heat treatment at an appropriate temperature [1,4]. A hysteresis loop was obtained after warm compaction in Fig. 5. From the result of the warm compacted sample at 650 MPa and 240 °C, the coercivity $H_c$ was 876 A/m, and the saturation magnetization $M_s$ was 144 A·m²/kg. It is known that saturation magnetization depends on the composition of materials; however, the coercivity is affected by various factors, such as density, strain, grain size, and magnetic anisotropy. Generally, the hysteresis loop was behaved discontinuous when there existed two phases, while the VSM result seemed to behave like single phase magnetic material. This result was considered that the Fe-Si
and Fe powders used in this work have similar magnetic properties, $H_c$ and permeability, except $M_s$.

Fig. 6 shows the magnetic losses measured in the frequency of up to 1000 kHz at 10 mT with various compaction pressures. It was confirmed the core losses logarithmically increased with a frequency increase. It is known that core loss can be presented by the sum of hysteresis loss, eddy current loss and residual loss. Among those, eddy current loss is proportionate to flux density and frequency; on the other hand, it is inverse proportion to electrical resistant. Also, hysteresis loss is affected by crystalline anisotropy, magnetostiction, saturation magnetization, pores, impurities and defects. Especially, core loss was significantly increased as green density decrease. It was considered that the effect of pore filling was dominantly affected to decrease hysteresis loss than residual stresses increased that during warm compaction process. The result of core loss in the range over 100 kHz was considered the increase of hysteresis loss by residual pores.

For this reason, temperature plays a very important role in higher densities, and in the elimination of residual stress. The core loss could be enhanced,
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if compaction temperature increases until decomposition of binder which acts as an insulator.

4. CONCLUSION

The warm compaction method was successfully fabricated high-density parts using premixed Fe-Si/Fe powder containing a small amount of PVP binder. Higher density under warm compaction was achieved due to the plastic deformation capability of Fe-Si/Fe powder by particle rearrangement. Results were shown that the warm compacted sample at 650 MPa and 240 °C had the lowest coercivity of 876 A/m and the lowest core loss. It was considered that this result was related to the higher density, and lower residual stresses of the warm compacted sample.

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REFERENCES