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DEVELOPMENT OF Fe-Si-B POWDERS FOR SOFT-MAGNETIC APPLICATION

RAZVOJ PRAHOV NA OSNOVI Fe-Si-B ZA MEHKOMAGNETNE MATERIALE

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Soft magnetic composite (SMC) materials based on Fe-Si-B powder have been developed. The most important property of SMC materials, especially those used in a high-frequency range, is that the soft-magnetic powder particles are well insulated from each other. The Fe-Si-B powders were prepared by water-atomisation and were compared with some commercially available powders. The surfaces of microcrystalline powders were investigated using high-resolution Auger-electron spectroscopy (HRAES) and X-ray photoelectron spectroscopy (XPS). The electrical break-down voltages, permeability and electrical losses of SMC materials were measured.

Key words: SMC, Fe-Si-B, insulating layer, XPS, AES

Na osnovi prahu Fe-Si-B smo razvili mehkomagnetne kompozitne materiale. Če so namenjeni za uporabo v visokofrekvenčnem področju, morajo imeti zelo dobro izolacijo med posameznimi delci-prahovi. Prahove Fe-Si-B smo pripravili po postopku vodne atomizacije in jih primerjali z nekaterimi komercialnimi. Površine mikrokristaliničnih prahov smo preiskali z visoko ločljivim Augerjevim elektronskim spektrometrom (HR AES) in z rentgenskim fotoelektronskim spektrometrom (XPS). Izmerili smo tudi nekatere električne lastnosti mehkomagnetnih kompozitov, kot so: električna prebojna napetost, permeabilnost in električne izgube.

Ključne besede: SMC, Fe-Si-B, izolacijska plast, XPS, AES

1 INTRODUCTION

SMC (soft magnetic composite) materials are compressed soft-magnetic powder particles insulated from each other with epoxy resin or some other type of dielectric[1,2]. The magnetic properties of these composites depend mainly on the magnetic properties of the powder particles, their shape and their size distribution in the dielectric matrix. One of the most interesting properties of these materials is the possibility to tailor their composition and processing to specifically meet the requirements of an application[3]. For higher frequency applications an excellent insulation between the particles and a smaller particle size are required to reduce eddy-current losses. Technological processing of SMC materials demands high pressures of compaction for the powder mixture, and because of the use of liquid binders at this stage of the process, direct contact between the particles cannot be avoided. The binder only fills the voids between the particles so the insulation of the particles must be achieved at an earlier stage of the process. This insulation is designed to surround the powder particles with a uniform, thin, electrically insulating layer, and as a result, the eddy currents remain restricted to the individual particles.

The experimental Fe-Si-B powders were prepared by water-atomisation. The surfaces of the micro-crystalline powders were studied. In addition to the study of the newly developed Fe-Si-B powders a commercially available Höganäs SMC powder called Somaloy 500 and Basf's EW carbonyl iron powder were analysed and compared with the Fe-Si-B powders. Using high-resolution Auger-electron spectroscopy (HRAES)[4] and X-ray photoelectron spectroscopy (XPS) analyses, the powder insulation layers were investigated. Some electromagnetic parameters such as permeability, electrical losses and U-I characteristics were also measured.

2 EXPERIMENTAL PROCEDURE

The experimental Fe-Si-B powders were prepared by water-atomisation using a David Mckee type D5/2 pilot water-atomiser this resulted in an almost spherical particle shape morphology and a relatively broad particle size distribution. The chemical composition of the powders was analysed and is as follows: Fe 88,1 %, Si 6,84 %, B, 3,14 %, Ni 1,45 %, C 0,025 % and Al 0,12 %. The particle size distribution was determined by using a laser granulometer (Cilas HR 850B) and sieve separation. The sieve analyses were performed using a Granulometer HR 850-B.

The Fe-Si-B powder was annealed in the air from 300 to 900 °C for 15 minutes in order to study the microstructural changes during thermal treatment. Powders were hot mounted and the microstructure was examined using standard metallographic methods.

Microstructural changes during heating were monitored using Nikon optical microscope Microphot FXA and a JSM-35 scanning electron microscope at a 25 keV accelerating voltage. The microhardness was measured using a Vickers micro hardness tster (Shimadzu, Japan) with an applied load of 100 g.

The surfaces of the Fe-Si-B powder samples were analysed by HRAES and XPS using a VG Microlab 310 F instrument. Auger analyses were carried out on the cross-sections of powders, which were mechanically compressed into a soft Bi-Pb-In-Sn alloy and metallographically prepared (polished) using a standard procedure. For XPS analysis, powders were fixed on a silver tape and ion etched with an ion current of 1,7 μA over the whole area (approximately 50 mm^2). The spectra were first measured over the non-sputtered surfaces and then over the surfaces after several sequential sputtering cycles.

The quality of the electrical insulation layer was measured by an Iskra NL 036 0-4000V isolation controller. During the measurement the powder was compressed with a pressure of 100 kPa. The controller generated on AC voltage, if the current reached 100 μA the voltage was automatically shut off.

3 RESULTS AND DISCUSSION

Somaloy 500 is a plain iron powder with particles of an irregular shape and high surface area. The particles are surface coated and very soft (85 ± 5 HV0.1) with a particle size from 20 to 100 μm. The microstructure is polycrystalline ferrite with a hardness of 85 HV0.1. Somaloy 500 was developed for soft-magnetic applications with 3-dimensional flux, such as electrical machines, transformers, ignition system and sensors. Basf's EW consists of carbonyl iron particles regularly shaped and with an onion-like shell structure which are mechanically very hard (≈900 HV0.1). Due to its very small size (from 3 to 8 μm) it is mostly used for high-frequency applications.

The Fe-Si-B powders that were experimentally produced using water-atomisation are regularly shaped because of alloying elements such as Si and B, which increase the surface energy. By using different sieves the powders were separated in terms of their size (**Figure 1**). For subsequent experiments only powders of size below 45 μm were selected. In **Figure 2** the size distributions of the Fe-Si-B powders and Basf's EW powders analysed usiung a laser diffraction technique are shown. The mean particle size of the smallest Fe-Si-B and EW fraction is 28.7 μm and 3.5 μm, respectively. **Figure 3** shows a cross-section of the Fe-Si-B powders. It is clear that some particles fuse during the rapid solidification and some of them are hollow. In contrast the Somaloy 500 is irregularly shaped and has larger average size (**Figure 4**).

In **Figure 5** the microstructures of Fe-Si-B powders annealed in air are shown. In the as-water-atomised state the microstructure is amorphous or microcrystalline. Above 400 °C and 500 °C the complete microcrystalline structure appeared, while at 600 °C a single phase still exists but with a grain structure that exhibits larger grain boundary surfaces. Above 700 °C an α-(Fe, Si, Ni) solid solution and the Fe_2B phase form[5]. A scanning electron microscopy (SEM) cross-section of water-atomised particles shows the microcrystalline structure with a very

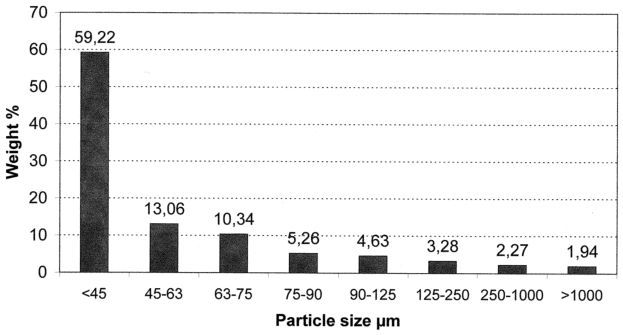

Figure 1: The particle size distribution of Fe-Si-B water-atomised powder
Slika 1: Velikostna porazdelitev vodno atomiziranega prahu Fe-Si-B

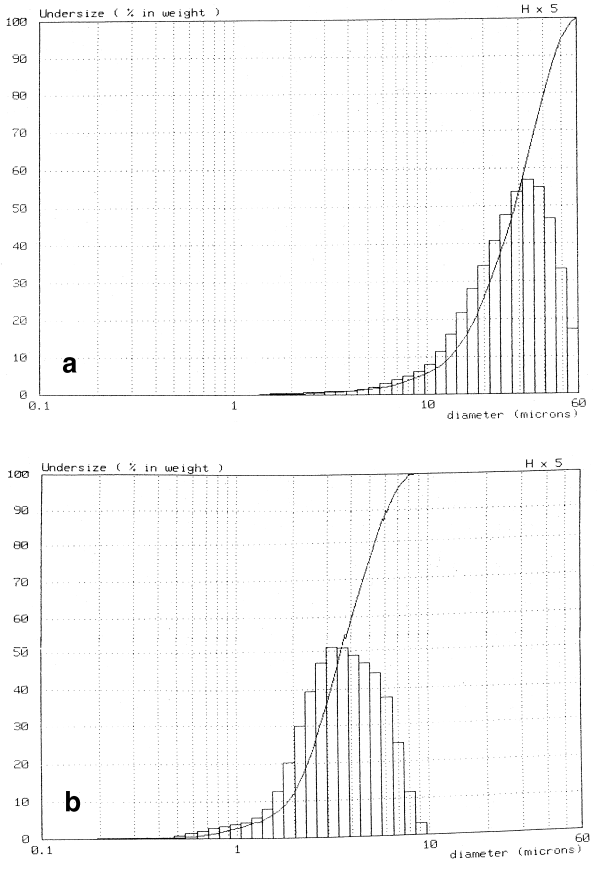

Figure 2: (a) The particle size distribution of a sieved Fe-Si-B powder fraction < 63 μm, **(b)** particle size distribution of EW powder
Slika 2: (a) Velikostna porazdelitev sejalne frakcije < 63 μm, **(b)** velikostna porazdelitev EW-prahu

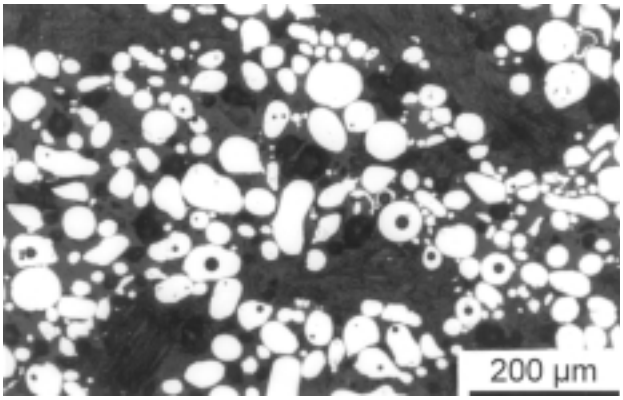

Figure 3: OM image of cross-section of Fe-Si-B powder particles (as polished)
Slika 3: Optičnomikroskopski posnetek prereza prahu Fe-Si-B (polirano)

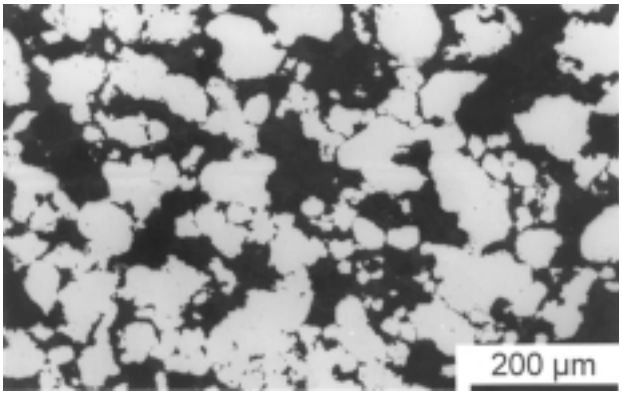

Figure 4: OM image of cross-section of Somaloy 500 powder particles (as polished)
Slika 4: Optičnomikroskopski posnetek prereza prahu Somaloy 500 (polirano)

smooth surface (**Figure 6**). The micro-hardness was measured for particles of different sizes from 1 mm down to 0.045 mm, and annealed at temperatures from 300 to 900 °C. It was found that there was no size-dependent particle hardness and also that during annealing in air the hardness begins to change slightly when annealing above 800 °C. The hardness was 950 ± 50 $HV_{0.1}$ and dropped when annealing above 800 °C and 900 °C to 750 ± 50 $HV_{0.1}$ and 720 ± 50 $HV_{0.1}$, respectively.

Figure 7a shows an SEM image of an Fe-Si-B powder particle on which an AES line scan was performed and in **Figure 7b** the AES profile is shown. The AES line scan was made from the matrix material into the powder particle. Therefore, In and Sn were detected first. On the border there was some C, which remains on the sample even after ion etching in a small gap between the matrix and the analysed powder particle. The spectra show the depletion of Fe from the surface and, in addition intensive peaks of O and Si. The

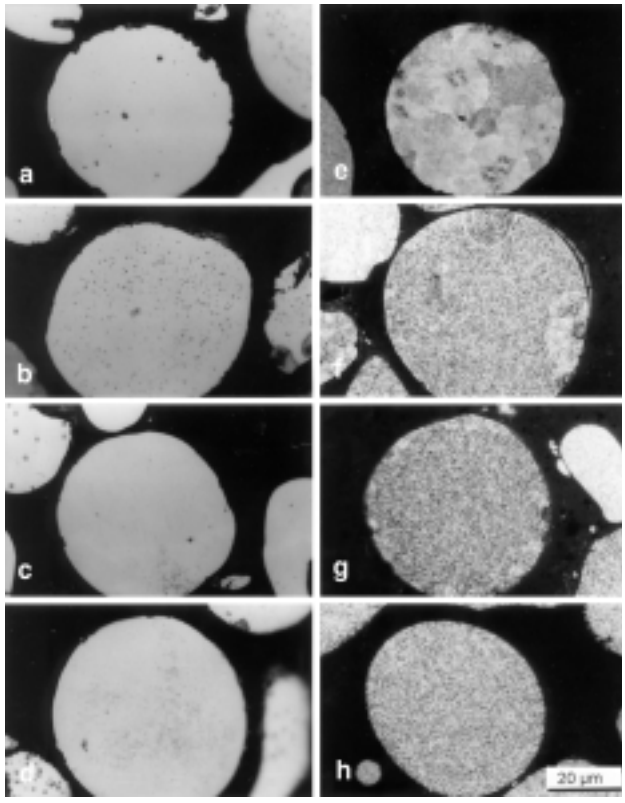

Figure 5: Microstructure of Fe-Si-B powder in (**a**) as-atomised state and annealed for 15 minutes in air at temperature (**b**) 300 °C, (**c**) 400 °C, (**d**) 500 °C (**e**) 600 °C, (**f**) 700 °C, (**g**) 800 °C (**h**) 900 °C
Slika 5: Mikrostruktura prahu Fe-Si-B po (**a**) vodni atomizaciji in žarjenju 15 minut na zraku pri temperaturi (**b**) 300 °C, (**c**) 400 °C, (**d**) 500 °C (**e**) 600 °C, (**f**) 700 °C, (**g**) 800 °C (**h**) 900 °C

profiles suggest that on the particle surface an Fe-O-Si compound no thicker than 1 µm was formed.

Figures 8 shows XPS spectra of as-atomised powder particles with a size below 45 µm. In **Figure 8a** binding-energy values (uncorrected) for Fe $2p_{3/2}$ of 707.3

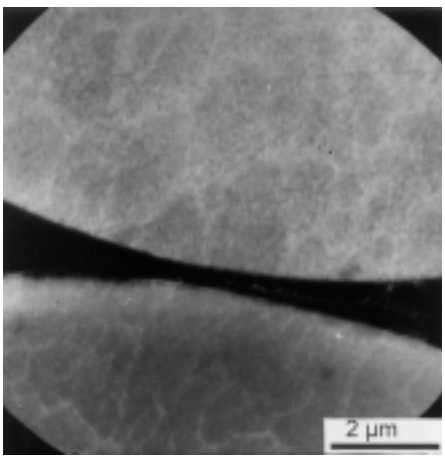

Figure 6: Microstructure of water-atomised Fe-Si-B powder shows a microcrystalline appearance (SEM image)
Slika 6: Vodno atomiziran prah Fe-Si-B ima mikrokristalinično mikrostrukturo (SEM-posnetek)

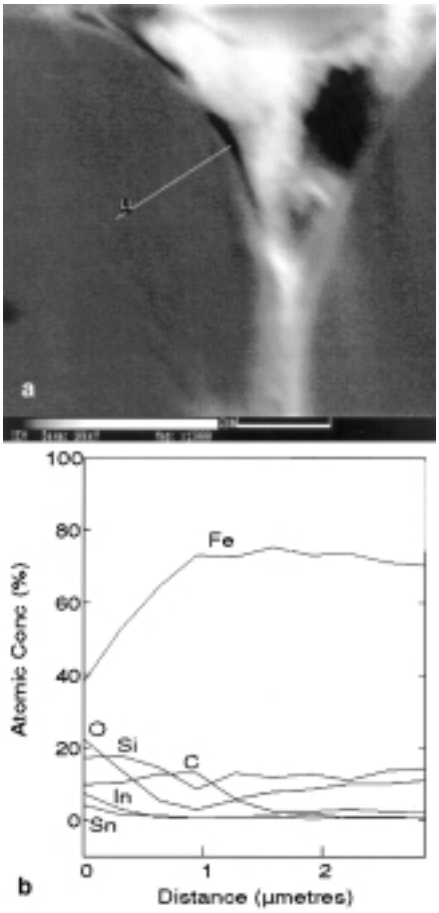

Figure 7: **(a)** SEM image of Fe-Si-B powder particle, **(b)** the AES line-scan profile
Slika 7: **(a)** SEM-posnetek prašnega delca Fe-Si-B, **(b)** linijska profilna analiza AES

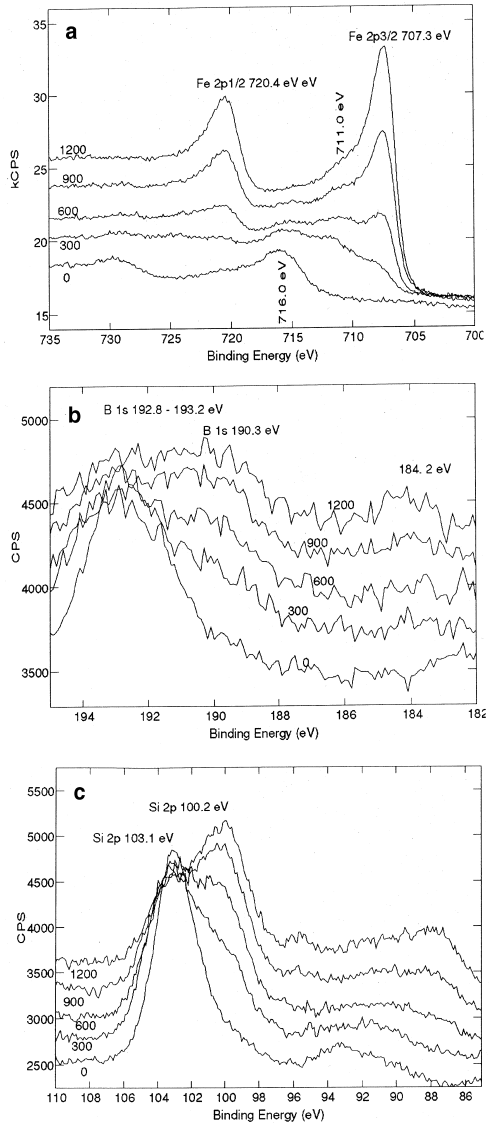

Figure 8: XPS spectra of water-atomised Fe-Si-B powders sputtered from 0 to 1200 s: (**a**) Fe 2p, (**b**) B 1s, (**c**) Si 2p; spectra in (**a**) are not corrected for charge shift
Slika 8: XPS-spektri vodno atomiziranih prahov Fe-Si-B ionsko jedkanih od 0 to 1200 s: (**a**) Fe 2p, (**b**) B 1s, (**c**) Si 2p; spektri v (**a**) niso korigirani glede na energijski pomik spektra zaradi nabijanja

eV, 710.7 - 710.9 eV and 712.6 eV can be observed in the spectra sputtered for 0, 300, 600 and 900 s. Using C 1s peaks from beneficial carbon contamination, the values of approx. 707 eV, 710.4 - 710.6 eV and 712.0 eV are obtained after a charge-shift correction. The first two can be interpreted as metallic Fe or Fe boride and Fe_2SiO_4[6,7], while the last (measured on the non-sputtered surface) may belong to a thin Fe_2O_3 layer on the Fe_2SiO_4 substrate. The supposition that, at least partially the intensity of the Fe $2p_{3/2}$ at 707 eV in **Figure 8a** is due to Fe boride is corroborated by the B 1s at 187.8 eV in **Figure 8b,** which also corresponds to Fe boride[6,7]. The B 1s at 192.5 eV in Figure 4b corresponds to B_2O_3[6,7]. **Figure 8c** shows the spectra of the Si 2p after different sputtering times where the initial peak at 102.5 eV corresponds to Fe_2 SiO_4[6,7], thus corroborating the identification of the Fe $2p_{3/2}$ peak at approximately 710.5 eV in **Figure 8a** and the additional peak at approximately 99.4 eV develops with sputtering. This additional peak corresponds to metallic silicon[6,7].

In order to find out the composition of the surface of the commercially available powders the AES analyses of both Somaloy 500 and EW were carried out. In both cases P and O were found to be on the surface. P 2p with two components at 132.2 eV and at 2p at 134.9 eV was found by XPS. Since most references[6,7] quote single binding-energy values for P 2p, unresolved into its closely spaced ($\Delta E_B \approx 0.8$ eV) 2 $p_{3/2}$ and 2 $p_{1/2}$ components, further deconvolution was not performed. The peaks at 132.2 eV and 134.9 correspond to P_2O_5 and some phosphate, respectively.

Following a procedure that is most probably similar to the one performed with the commercially available powders, our Fe-Si-B powder was also additionally chemically treated in 1 wt.% of 60 % H_3PO_4 diluted in

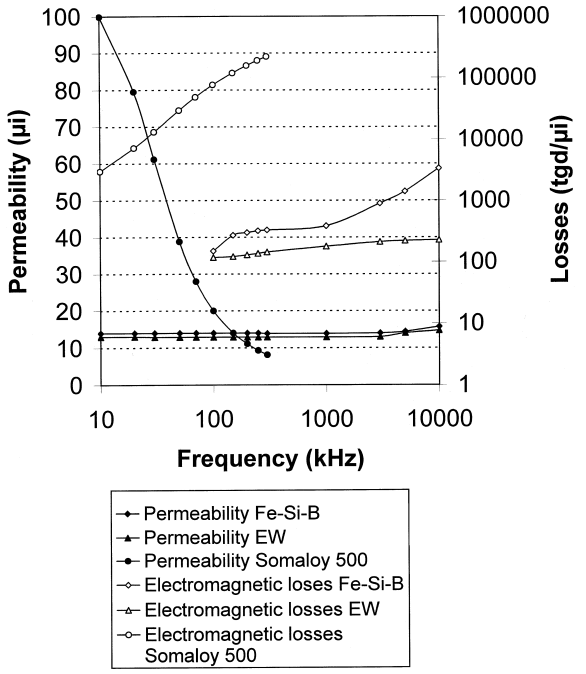

Figure 9: Permeability and electromagnetic losses measured on toroids compressed from Fe-Si-B, EW and Somaloy 500 powders.
Slika 9: Permeabilnost in elektromagnetne izgube, merjene na toroidih, stisnjenih iz prahu Fe-Si-B, EW in Somaloy 500

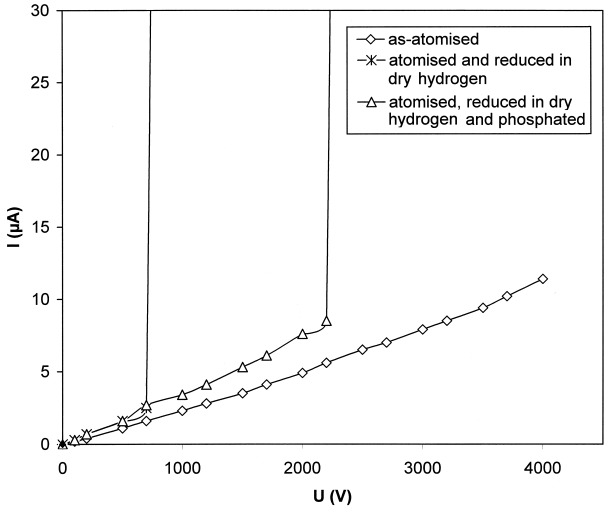

Figure 10: U-I measurements of Fe-Si-B powders
Slika 10: meritve U-I prahov Fe-Si-B

acetone in order to build an insulation layer on each of the particles.

In **Figure 9** the permeability and the magnetic losses were measured for the compressed powder particles with epoxy resin. Toroids of size Φ24/Φ10 × 15 mm were manufactured. The surface of the Fe-Si-B powders was not additionally treated. The permeability is similar for the Fe-Si-B powder and the EW. In the low-frequency range the Somaloy 500 shows the highest permeability, which diminishes rapidly at higher frequencies due to the coarse powder particles. The EW is used in the high-frequency range so the losses are small even at 10 kHz. The Fe-Si-B powders seem to be very good even though the size of the particles is around 35 μm.

The results of U-I characteristic measurements for the Fe-Si-B powders are presented in **Figure 10**. It was shown that the best insulation layer was achieved for the as-atomised powders. But after mixing the powders with epoxy resin and compressing, the insulation performance of the powder particles drops significantly, most probably due to the high internal stresses imposed during compaction and due to the brittle nature of the insulating layer, which does not remain stable during compression. For this reason the powders were reduced and oxidised, and by phosphating, it was intended to achieve mechanically stable insulation layer. The work is still in progress. At the moment, it is clear that water-atomised particles have an excellent insulation layer with a poor mechanical performance. By reducing the surface of the powder particles in dry hydrogen and by additional chemical treatment in phosphoric acid a stable insulation layer might be obtained. This insulation layer should be built with iron atoms of the same valence. The good insulation characteristics are also obtained if the layer is free of oxygen or metal vacancies due to a resonance-tunnelling effect, and if the layer is very thin, otherwise the electron is exchanged with the layer itself[8].

4 CONCLUSIONS

Laboratory water-atomised Fe-Si-B powder particles for soft-magnetic applications were developed. By using XPS spectroscopy and by monitoring the electrical characteristics of the powder particles an explanation for the insulation performance can be proposed. Water-atomised Fe-Si-B powders seem to have a thin Fe_2SiO_4 layer on the surface, which results in a good insulating performance, however, unfortunately, also in poor mechanical characteristics. On the basis of AES and XPS studies of the surface of the commercially available powders Somaloy 500 and EW, it was found that a phosphorus-acid-treated surface of the Fe-Si-B powder gives a very good insulation layer. The development P-containing powder material is in progress. It is expected that during rapid solidification a P-based insulation layer will be built up in-situ.

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