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Effect of heat treatment in air on surface composition of iron-phosphate based soft magnetic composite components

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ABSTRACT

Soft magnetic composite materials (SMC) manufactured by conventional powder metallurgical techniques for electromagnetic applications constitute individually encapsulated ferromagnetic powder particles with an insulating surface layer, bonded together into 3D finished structures. The production procedure includes compaction of the SMC base powder mixed with a lubricant substance and a post-annealing treatment that aims to relieve the stresses induced in the component during pressing. In the present study, the effect of the heat treatment process to the nature of the insulating layer was investigated under different temperature regimes using analytical techniques. Its surface chemistry was determined based on the XPS depth profiling technique, and its morphology and structure were evaluated using HR-SEM and XRD. Differences between interior and exterior areas of the samples suggested the development of an oxide scale in the outer regions that prevents its further bulk oxidation at temperatures above 500 °C, while below that temperature incomplete de-lubrication takes place.

1. Introduction

Soft magnetic composite (SMC) materials for electromagnetic applications represent promising alternatives to traditionally used laminated ferrites and electrical steels in the sustainable energy market [1–3]. Emerging from well-established manufacturing techniques offered by the Powder Metallurgy (PM) technology, the SMC materials are an extremely appealing option to the end user due to the cost efficient production routes of net-shaped components, efficient material utilisation, high tolerances, high degree of reproducibility and property consistency for parts in large scale volume production [4–6]. Their true strength though lays in the design freedom possibilities available that originate from their isotropic magnetic behaviour. Owing to the latter, new functional structures can be realised with 3D-flux capabilities leading to applications of higher complexity. Continuous development of the materials connected to the SMC technology over the past two decades has enabled the manufacturing of parts covering low, medium and high frequency applications, such as inductor cores, rotating machines, fast switching actuators and sensors [1,3,7,8].

The concept of the SMC is based on encapsulating each iron particle with an electrically insulating coating and subsequently stacking the particles together through a conventional uniaxial compaction process into three-dimensional finished structures [1,6,9]. In order to preserve the initial state of the coated powder, the SMC powder is admixed with organic lubricants prior to the compaction to effectively reduce inter-particle and particle to die wall friction. The insulator coating serves a dual purpose: (i) reducing the part's core losses by substantially increasing its bulk resistivity; (ii) introducing mechanical strength by acting as a "binder" material between the particles. The former is accomplished by confining the deleterious effects of the produced eddy currents in each iron particle, while the latter is achieved with a tailored curing process through a post heat treatment. The latter consists also a necessary step of the production of SMC components that aims in relieving any stresses induced during the compaction of the powder [1,6]. The temperature range for this treatment should be such that the deterioration of the coating is prevented while at the same time sufficient stress relief takes place, improving the component's magnetic properties by lowering its hysteresis losses [9–11].

It is thus obvious that the insulating coating is the paramount feature of the SMC technology. Its physical, morphological, electrical and chemical properties such as thickness, surface coverage, porosity, structure, strength, cohesion to the matrix, resistivity and chemical durability after treatment are inextricably linked to the component's density, strength, permeability, induction saturation and core losses which are of utmost importance to its performance [9–13]. Both organic and inorganic compounds can be used as insulating substances, making it possible to tailor the

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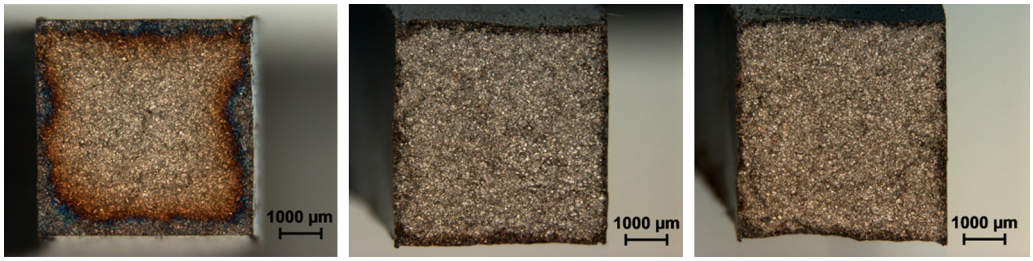

Fig. 1. Fractured surfaces of SMC parts heat treated in air at 400, 500 and 600 °C (from left to right).

finished part's properties so as to match the preferences of the desired application, thus adding great versatility and functionality to the technique [1,3,12–14]. In this context, inorganic phosphate coatings are of interest due to their thermostability, good electrical insulating properties, mechanical and chemical durability, effective functionalisation of their surface to more advanced and complex compounds and ease of application to mass production [13–17]. Phosphatising is a well-known metal conversion treatment, largely used in automotive industry and for radio equipment applications. Metal surfaces are usually treated either by spraying or immersion baths, which consist of phosphoric acid solutions in aqueous or volatile organic solvents, producing crystalline or amorphous-like phosphate coatings [15,16].

An extensive volume of research publications is associated with the study of phosphatised surfaces, providing information on their properties, quality, chemistry, growth kinetics, mechanisms and parameters of formation [15–21]. The usage of analytical techniques for analysis of coatings on flat substrates is a common practice in material science nowadays. However, the number of similar investigations on spherical or irregular powder particles is significantly lower [13,14,22–25]. Recently, a methodology to investigate the chemical composition, surface characteristics and thickness of the phosphatised iron powder was developed involving the use of surface analytical techniques and electron microscopy. For these matters, the effect of the specific geometry and topography of the samples along with the charging nature of their insulating surface in the analyses and evaluation process were taken into account based on modelling of the XPS composition depth profiling on powder samples [26]. Here, the ultimate aim is to better investigate the complex surfaces of finished SMC parts after compaction and heat treatment at different temperature regimes, providing insight into the influence of the process treatment on the characteristics of the coating. For this purpose, heat-treated components at temperatures of 400, 500 and 600 °C, chosen from previous analyses of magnetic and mechanical properties [11] critical to the performance of a product, are studied using complementary analytical techniques such as high-resolution scanning electron microscopy (HR SEM) coupled with energy dispersive X-ray microanalysis (EDX), X-ray photoelectron spectroscopy (XPS) and X-ray diffraction (XRD). The surface morphology, coverage, chemical composition analysis, chemical state depth profiling and thickness determination of the insulating layer are determined based on this approach. The results of these investigations are of the greatest importance in modelling and tailoring an optimum treatment process for such components.

2. Experimental procedure

2.1. Material

In the current study, the final SMC components were prepared based on commercially available soft magnetic powder, Somaloy® 500 by Höganäs AB, Sweden. The SMC powder grade is based on water-atomised high purity iron powder of supreme compressibility that is typically used for manufacturing parts of high density. The iron particles are further insulated with an inorganic phosphate coating based on a patented process [2]. The thickness of the insulating surface coating formed is in the nanometre range in order to minimise its effect on the compressive behaviour of the grade and in that manner to improve the magnetic performance of the finished part [26]. Components were produced under traditional uniaxial die compaction at 800 MPa using a powder mix, which contained 0.5 wt.% of organic lubricant. Subsequent heat treatment was performed according to the process described by Zhou et al. [11] in a belt furnace in air for 30 min at the temperatures of 400, 500 and 600 °C. Parts were pressed into standardised magnetic rings (toroids), normally used in iron core loss measurements, of 5 mm × 5 mm cross section and outer/internal diameter dimensions of 55 mm/45 mm, respectively [27]. The samples were fractured in air and the produced fracture surfaces of their cross sections were used for surface analysis as shown in Fig. 1.

2.2. Characterisation techniques

2.2.1. Scanning electron microscope (SEM)

Surface morphology, characteristics and analysis of the surface coverage were conducted by means of a LEO Gemini 1550 (CARL ZEISS – LEO electron microscopy, GmbH, Germany) scanning electron microscope equipped with a field emission gun. For the best topographical information from the surface of the samples, secondary electron imaging was performed using an InLens detector (CARL ZEISS – LEO electron microscopy, GmbH, Germany) at a low acceleration voltage of 5 kV. Further chemical microanalysis was carried out using energy dispersive X-ray spectroscopy utilising an X-MAX 20 mm^2 detector (Oxford Instruments, High Wycombe, England) calibrated at 5 kV acceleration voltage. Silicon standard was used for calibration of EDX setup before each measurement, which was optimised according to our previous investigations [26]. Due to the surface roughness of the samples, the results from the compositional analysis from this technique are only considered qualitatively as complementary information to the chemical analysis performed by XPS.

2.2.2. X-ray diffraction (XRD)

Phase identification analysis of the exterior surface regions of the heat-treated components was carried out using a Bruker D8 ADVANCE diffractometer equipped with a Cr-Kα source. The measurements were performed under grazing angle mode with an incident angle of 3° over a range of $40° < 2\theta < 140°$ with 0.05° 2θ/step and 5 s/step.

2.2.3. X-ray photoelectron spectroscopy (XPS)

Chemical state analysis, compositional depth profiling and coating thickness determination were carried out using XPS for the cross sections of all heat-treated components. Analyses were performed for both interior and exterior regions in order to determine