



Development of Ba-hexaferrite coatings for electromagnetic wave absorption applications

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ABSTRACT

In this research, Ba-hexaferrite coatings for electromagnetic wave absorption applications were deposited by thermal spraying. A suitable powder feedstock was manufactured by blending a $\text{BaCO}_3 + \text{Fe}_2\text{O}_3$ mixture, which was then agglomerated by spray-drying. The agglomerates were processed by air plasma spraying (APS) without any further treatment or were heat-treated and reactively-sintered to stoichiometric Ba-hexaferrite prior to spraying. As-sprayed coatings were analyzed by Raman spectroscopy and X-ray diffraction (XRD). Whereas the deposition of untreated agglomerates did not result in adequate amounts of crystalline Ba-hexaferrite in the coatings, the APS processing of reactively-sintered agglomerates led to a high content of Ba-hexaferrite and similar magnetic properties to those of Ba-hexaferrite bulk materials.

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1. Introduction

The requirements to suppress electromagnetic noise and to avoid electromagnetic interference in electronic devices have made the development of electromagnetic wave absorbers a very important subject [1,2]. For these applications a ferromagnetic barium hexaferrite ($\text{BaFe}_{12}\text{O}_{19}$) is often applied. These ferrites are based on a complex magnetoplumbite structure, which results in a saturation magnetization of 72 emu/g ($\text{A} \cdot \text{m}^2/\text{kg}$) and a remarkable magnetocrystalline anisotropy field of 17 kOe [3]. The latter leads to a ferromagnetic resonance frequency of about 50 GHz, which is suitable for mm-wave applications [3]. State-of-the-art commercial hexaferrite-based absorbers are bulk ceramics, which are fitted to components by bonding or brazing, or paints and polymer composites filled with absorbing powder, which need a very high thickness in order to be effective [4]. These kinds of absorbers have limited possibility to be adjusted to the geometry of the devices. A further disadvantage is the cost intensive procedure to fix the small sintered tile onto large-scale devices. Such limitations can be overcome by the application of thermal spraying technologies for the preparation of absorbing coatings.

Although there is much interest in the application of ferrites in the form of thick films and planar magnetic devices, little work has apparently been published on the use of thermal spraying to produce

such deposits. Some of the published studies concern the deposition of absorbing coatings based on lossy conductors, dielectrics and spinel ferrites [5], the preparation of spinel ferrite coatings [6–9] or the production of $\text{BaFe}_{12}\text{O}_{19}$ from Ba- and Fe-nitrate solutions by inductively-coupled plasma [10]. In most cases, the interaction of the powder particles with the spray jet results in a phase transformation of the feedstock and/or in compositional alterations, which lead to an instability of the ferrite phase. In the case of amorphous as-sprayed coatings subsequent annealing is necessary for the crystallization of the ferrite phases, but this restricts the applications of thermal sprayed coatings significantly [7,8,11]. If secondary phases appear beside the ferrite phase, the magnetic properties of the coating are negatively affected [9]. By using “cold” spraying parameters for the APS process it is possible to reduce the secondary phases within the coating significantly and the coatings achieve nearly the same properties as bulk ferrites [9,14].

Previous experiments by the authors have shown that, when processing $\text{BaFe}_{12}\text{O}_{19}$ and $\text{BaCoTiFe}_{10}\text{O}_{19}$ hexaferrites by APS or HVOF spraying, excessively “hot” spray parameters result in the formation of undesirable secondary phases (e.g. spinel and amorphous phases), although it is possible to restore single-phase hexaferrite structure by heat-treatments at 1100–1300 °C [12,13]. Large amounts of crystalline hexaferrite can however be retained in the as-deposited coatings by adjusting the parameters in order to embed a controlled fraction of unmelted material in the sprayed layer [14].

Previously [14], the feedstock powder consisted of spray-dried agglomerates of micrometric Ba-hexaferrite particles. In this study, by

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contrast, the spray trials with APS are performed using spray-dried mixtures of micrometric reagent particles ($\text{BaCO}_3 + \text{Fe}_2\text{O}_3$). This agglomerated mixture is either sprayed without any further pre-treatment or heat-treated before spraying, in order to produce reactive sintering. All coatings are analyzed with regard to phase composition, microstructure and magnetic properties.

2. Experimental method

The feedstock powders were based on a blend of BaCO_3 (98.5%, Merck, Darmstadt, Germany) and Fe_2O_3 (99.65%, NK- α SEW, IROX-NKK Co. Ltd., Tokyo, Japan). The powders were homogenized in a water/ethanol slurry for 1 h using a Turbula homogenizer (System Schatz Type T2F, Willy A. Bachofen AG, Maschinenfabrik, Basel, Switzerland). This solution was spray-dried to achieve spherical agglomerations by the spray dryer Niro pilot P6.3 (GEA Niro Inc., Columbia, USA). This process is explained in detail in [15]. The spray-dried powder was classified within the spray-drying system into a fraction of $-65 + 20 \mu\text{m}$. The particle size distribution was measured with a Lecotract-LT100 (St. Joseph, Michigan, USA) particle size analyzer. After classification and before spraying, batches of the powder were heat-treated at 1150 °C for 4 h to achieve reactive sintering with the formation of Ba-hexaferrite phase within the powder. After heat-treatment the powder possessed a particle size distribution of $-70 + 0.1 \mu\text{m}$. Due to the fine particles the powder was sieved to separate particles smaller than 20 μm . So the used powder had a particle size distribution of $-70 + 20 \mu\text{m}$ with a suitable flowability for APS.

The blended and spray-dried powder (not sintered) as well as the spray-dried and sintered powder was processed by the APS system F4-HBS from Sulzer Metco (Hatterheim, Germany). The spray-dried (non-sintered) powder was sprayed in order to achieve a phase transformation to Ba-hexaferrite phase during spraying using the high particle temperatures within the process. To achieve such in situ reaction, the spraying parameters (listed in Table 1) were adjusted to high particle temperatures (high H_2 content in the plasma gas). While spraying the sintered powder, “cold” spraying parameters (no H_2) were chosen without hydrogen to avoid complete melting and hence extensive phase transformation of the powder during processing [14].

The synthesis of the powder and the structural evolution of the coatings were monitored by X-ray diffraction (XRD) using X'Pert PRO MPD Diffractometer (PANalytical, Almelo, The Netherlands). The microstructures of the coatings were observed on their polished cross-sections with a scanning electron microscope (FEI XL30, Eindhoven, The Netherlands) combined with energy dispersive X-ray spectroscopy (EDXS, ISIS Link System, Oxford Instruments, Oxford, UK).

Micro-Raman spectroscopy (LabRAM, Horiba Jobin-Yvon, Longjumeau, France; laser wavelength: 632.81 nm) was performed on the polished cross-sections, in order to assess the local phase composition of the samples. The spectra of bulk $\text{BaFe}_{12}\text{O}_{19}$ were also acquired for reference. Bulk ceramic samples were prepared from the stoichiometric powders (see the homogenization procedure above) calcined at 1100 °C for 6 h, followed by 1 h milling in an attritor mill and sintering at 1300 °C for 3 h. The XRD and SEM analysis confirmed that the ceramics were single phase. The magnetic properties of the coatings were measured

with a vibrating sample magnetometer (VSM, LakeShore 7312, Westerville, OH, USA) at a maximum field of 1.35 T.

3. Results and discussion

3.1. Characterization of the powders

As expected the blended powder consists of a BaCO_3 and hematite (Fe_2O_3), whereas the heat-treated powder is single phase Ba-hexaferrite (Fig. 1). So it can be concluded that the heat-treatment process leads to a complete phase transformation of the blended powder. After sintering the powder morphology is still spherical which has a positive effect on the flowability during processing. After sintering the powder becomes a bit finer, due to breaking of some agglomerates during sintering. These fine particles were subsequently sieved out with a 20 μm sieve.

3.2. Characterization of the coatings

The blended powder was sprayed aiming to obtain an in situ reaction of the powder mixture $\text{BaCO}_3 + \text{Fe}_2\text{O}_3$ (hematite) into the Ba-hexaferrite phase. The coating APS 1 (Fig. 2A), produced from the blended powder, possesses a brittle structure with a lot of cracks and no clear typical lamellas. It consists of several areas with different contrasts reflecting local variations of the chemical and/or phase composition. It would seem that the blended mixture was largely melted and formed a liquid phase, but the recrystallization of this melt was not completed during spraying. The XRD patterns of these APS coatings (Fig. 3) show a clear multiphase structure. The spinel peaks have the highest intensity, suggesting a high spinel content within the coating. Due to the high cooling rates (10^6 K/s) of molten impacting particles [17], the cooling time is not long enough for the recrystallization to the Ba-hexaferrite phase, because this phase can only be formed from the melt through various peritectic reactions, and the spinel phase crystallizes as first [16]. In this case also hematite can be detected, which comes from the feedstock.

The coatings sprayed from the sintered powder (example is shown in Fig. 2B) exhibit a significantly different structure than the previously mentioned ones. They consist of dense regions and also highly porous regions. A reactively-sintered agglomerate is porous but possesses high cohesive strength, so that it does not fall apart during spraying. Because of its low thermal conductivity, the outer shell melts, whereas the core remains partly unmelted. The liquid material from the outer shell can infiltrate and close the porosity of the agglomerate. Such densified agglomerates give rise to dense regions within the sprayed coating. The unmelted particles built up the high porosity of about 15%. This high content of unmelted material, which is embedded within the coating, suggests that a high amount of Ba-hexaferrite must be present. Accordingly, the XRD patterns of three APS sprayed coatings (Fig. 4) show very strong Ba-hexaferrite peaks. Additional minor peaks corresponding to hematite can be observed for the coatings APS 3 and 4 while only Ba-hexaferrite peaks can be observed for the coating APS 5. It has to be noted that the main spinel peaks overlap with those of the Ba-hexaferrite (marked with M in Fig. 4). Therefore a minor amount of the spinel phase cannot be undoubtedly excluded based on the XRD results. By comparing the three patterns it can be observed that the Ba-hexaferrite vs. hematite fraction varies and this is consistent with

Table 1
APS spraying parameters of the blended/agglomerated powder and the blended/agglomerated/sintered powder.

Sample	Current [A]	Gas:Ar [SLPM]	Gas:H ₂ [SLPM]	Carrier gas [SLPM]	Spray distance [mm]	Particle size [μm]	Injection angle [°]	Coating thickness [μm]	Powder type
APS 1	500	50	6	3.5	130	20–65	90	107	Blended
APS 2	300	50	6	3.5	130	20–65	90	98	Blended
APS 3	500	50	0	3.5	130	20–70	90	73	Blend./sinter.
APS 4	500	50	0	3.5	180	20–70	90	75	Blend./sinter.
APS 5	300	50	0	3.5	100	20–70	90	24	Blend./sinter.

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