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Formation Process of the Integrated Core(Fe-6.5wt.%Si)@Shell(SiO₂) Structure Obtained via Fluidized Bed Chemical Vapor Deposition

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Received: 11 March 2020; Accepted: 14 April 2020; Published: 17 April 2020

Abstract: As electromagnetic functional materials, soft magnetic composites (SMCs) have great potential for applications in high-energy electromagnetic conversion devices. The most effective way to optimize the performance of an SMC is to incorporate it into insulated ferromagnetic core-shell particles with high structural uniformity and integrity. Fluidized bed chemical vapor deposition (FBCVD) is a facile and efficient technique for the synthesis of ferromagnetic/SiO₂ core-shell particles. However, the formation mechanism and conditions of integrated ferromagnetic/SiO₂ core-shell structures during the FBCVD process are not fully understood. On this basis, the formation process and the deposition time required for transformation of the Fe-6.5wt.%Si substrate into the Fe-6.5wt.%Si/SiO₂ composite, and finally into the Fe-6.5wt.%Si/SiO₂ core-shell structure, were investigated. Deposition of the insulative SiO₂ coating onto the Fe-6.5wt.%Si particles was described by the three-dimensional island nucleation theory. The SiO₂ islands were initially concentrated in rough areas on the Fe-6.5wt.%Si particle substrates owing to the lower heterogeneous nucleation energy. Deposition for at least 960 s was necessary to obtain the integrated ferromagnetic/SiO₂ core-shell structure. The uniformity, integrity, and thickness of the insulative SiO₂ coating increased with the increasing deposition time. The results in this study may provide a foundation for future kinetics investigations and the application of FBCVD technology.

Keywords: Fe-6.5wt.%Si/SiO₂ core-shell structure; formation process; deposition time; fluidized bed chemical vapor deposition

1. Introduction

Soft magnetic composites (SMCs) have become increasingly important in materials research, because they are magnetically isotropic, highly permeable, and inhibit eddy current loss [1–3]. The most effective way to optimize the features of SMCs is to incorporate them into core-shell (C-S) structures by coating the ferromagnetic core particles with an insulative shell. The C-S structure can significantly enhance the surface resistance of ferromagnetic particles and reduce the effective diameters of eddy currents [4,5] for the realization of high-energy electromagnetic conversion devices.

Over the past decade, many researchers have focused on the design and controlled synthesis of insulated ferromagnetic C-S structures and the relationships between their structures and activities [6,7]. The design and preparation of SMCs and ferromagnetic/inorganic C-S structures using diverse insulative oxide coatings including Al_2O_3 [8,9], SiO_2 [10], Fe_3O_4 [11], MgO [12], and ZnO [13] were reported. Yan and coworkers investigated the influence of processing parameters on the magnetic properties of SMCs [14–16]. Shokrollahi and coworkers studied the mechanisms by which C-S structures influenced the magnetic properties of SMCs [17,18]. These research achievements promoted the development of basic theory and expanded the range of applications for SMCs. As basic and applied research on SMCs has progressed, it has been widely accepted that increasing the structural integrity and uniformity of C-S structures will reduce iron loss at extremely high-energy densities. Furthermore, the thickness of the insulative shell coating affects carrier migration and the coupling efficiency of the ferromagnetic particles [5,19–21]. Therefore, the synthesis of C-S composite particles with few defects seems particularly important for obtaining SMCs with excellent properties. While many studies have focused on SMC characterization and measurement, few have investigated the formation mechanisms and optimal conditions for the production of C-S structures with ferromagnetic cores. For example, fluidized bed chemical vapor deposition (FBCVD) has been recognised as an efficient technique for the synthesis of ferromagnetic/ SiO_2 core-shell particles [22,23]. However, FBCVD is a complex two-phase gas and solid flow system, and the controlled synthesis of ferromagnetic/ SiO_2 C-S particles is influenced by many factors. The formation mechanism and conditions of integrated ferromagnetic/ SiO_2 core-shell structures during the FBCVD process are not fully understood.

Aiming at these problems above, a more in-depth study of the formation mechanism of ferromagnetic/ SiO_2 C-S structural evolution during FBCVD was carried out in this paper. The influence of deposition time on the microscopic characteristics of as-deposited particles was systematically investigated at a constant precursor concentration using ferromagnetic particles with the same temperature and morphology. We studied the time needed for the transition from Fe-6.5wt.%Si particles to an Fe-6.5wt.%Si/ SiO_2 composite to obtain Fe-6.5wt.%Si/ SiO_2 C-S particles. The relationship between the deposition time and the static magnetic properties of the Fe-6.5wt.%Si/ SiO_2 C-S particles was also investigated. The results could promote the application and development of FBCVD technology.

2. Materials and Methods

Spherical atomized Fe-6.5wt.%Si particles (>99 wt.%) with an average size of $\sim 70\ \mu\text{m}$ were used as substrates. The tetraethyl orthosilicate ($\text{C}_8\text{H}_{20}\text{O}_4\text{Si}$) precursor was supplied by Sinopharm Chemical Reagent Co., Ltd. High purity argon (Ar, 99.99 wt.%) was used as carrier gas to transport tetraethyl orthosilicate vapor.

In a typical process, 10 g of Fe-6.5wt.%Si particles was first heated to 920 K in a quartz tube furnace of the fluidized bed reactor. Simultaneously, 80 mL/min of high purity Ar was introduced into the reactor to suspend and fluidize the Fe-6.5wt.%Si particles. Then, the gaseous $\text{C}_8\text{H}_{20}\text{O}_4\text{Si}$ precursor was introduced into the fluidized bed reactor at atmospheric pressure using 80 mL/min of high purity Ar as a carrier gas. The gasification reaction of the $\text{C}_8\text{H}_{20}\text{O}_4\text{Si}$ precursor occurred in a liquid evaporator at 423 K. The deposition time was set to 60, 120, 240, 480, 960, 1920, or 3840 s before cooling the furnace to room temperature.

The phases obtained after FBCVD were analyzed using a D8 Advance X-ray diffractometer (XRD, Bruker, Billerica, MA, USA) equipped with a $\text{CuK}\alpha$ radiation source from (2θ) 10° to 90° . The surface and cross-sectional morphologies of all particle samples were examined using a JSM7610F field-emission scanning electron microscope (FE-SEM, JEOL, Tokyo, Japan). The cladding ratio of SiO_2 insulation on the surface of the Fe-6.5wt.%Si substrate was statistical analyzed by an Image J software after skeletonize treatment. The chemical states and electronic structures of the original Fe-6.5wt.%Si and Fe-6.5wt.%Si/ SiO_2 C-S particle surfaces were determined via X-ray photoelectron spectroscopy (XPS) on a 250Xi spectrometer (ThermoFisher, Waltham, MA, USA) equipped with an $\text{AlK}\alpha$ radiation source. The pass energy, energy step size, and scan time for XPS were 30.0 eV, 0.050 eV, and 120.3 s,

respectively. The electrical resistivity under a pressure of 2 MPa was measured on a ST2722 powder resistivity tester (Jingge Electronic Co., LTD, Suzhou, China). Hysteresis loops were recorded on a 7404-S vibrating sample magnetometer (VSM, Lake Shore Cryotronics, OH, USA) under an applied magnetic field ranging from $-20,000$ to $20,000$ Oe. The saturated magnetizations of all particle samples are expressed in emu/g, which is the unit in the centimeter–gram–second (CGS) system of units.

3. Results and Discussion

3.1. Synthesis Process of Fe-6.5wt.%Si/SiO₂ C-S Structure

XRD was employed to analyze the structures of the original Fe-6.5wt.%Si particles and particle samples after deposition over various intervals. As shown in Figure 1a, three sharp crystalline peaks were observed at (2θ) 44.94° , 65.37° , and 82.78° in the pattern of the original Fe-6.5wt.%Si particle sample, which corresponded to the (110), (200), and (211) crystal planes of α -Fe(Si), respectively. The pattern was consistent with that of the International Centre for Diffraction Data powder diffraction file 09-065-9130. These results indicated that the Fe-6.5wt.%Si particle substrate had a body-centered cubic (bcc) lattice structure. The enlarged image in Figure 1b revealed another broad peak centered at 23° in the XRD patterns of the Fe-6.5wt.%Si particles after FBCVD treatment, which indicated that the SiO₂ coating on the particles was an amorphous phase. The diffraction peak intensity of the amorphous SiO₂ phase increased gradually as the deposition time increased, likely because more SiO₂ was deposited onto the surfaces of the particles. The average crystal sizes, micro-strains, and lattice constants of the α -Fe(Si) phase with their corresponding deposition times are analyzed after XRD structural refinement using a Jade 6.0 software (Materials Data, USA). The results are shown in Table 1. The microstructure of the α -Fe(Si) phase did not undergo any obvious changes during FBCVD. The average crystal size, micro-strain, and lattice constant were approximately 56.1 nm, 0.126%, and 2.8510 Å, respectively. These results indicated that the deposition of amorphous SiO₂ did not affect the crystal structure of the Fe-6.5wt.%Si particle substrate.

Figure 1. (a) X-ray diffractometer (XRD) patterns and (b) partially enlarged XRD patterns of the original Fe-6.5wt.%Si particles and the particles obtained after fluidized bed chemical vapor deposition (FBCVD).

Table 1. The average crystal sizes, micro-strains, and lattice constants of α -Fe(Si) in the original Fe-6.5wt.%Si particles and the particles obtained after fluidized bed chemical vapor deposition (FBCVD).

Samples	Crystal Size (nm)	Micro-Strain (%)	Lattice Constant (Å)
Original Fe-6.5wt.%Si particles	55.1 ± 0.10	0.126 ± 0.0016	2.8508 ± 0.006
Particle samples after deposition for 60 to 3840 s	60 s	53.7 ± 0.06	0.131 ± 0.0013
	120 s	57.3 ± 0.08	0.122 ± 0.0017
	240 s	56.1 ± 0.09	0.128 ± 0.0017
	480 s	54.6 ± 0.09	0.123 ± 0.0019
	960 s	58.7 ± 0.09	0.128 ± 0.0018
	1920 s	58.8 ± 0.11	0.127 ± 0.0020
3840 s	54.1 ± 0.13	0.126 ± 0.0021	2.8513 ± 0.006

The morphologies of the particles were investigated via FE-SEM. The typical morphology of the atomized Fe-6.5wt.%Si particle substrate can be seen in Figure 2a. Wrinkles and pits (outlined in black on the Figure 2a) randomly distributed over the surfaces of the particles resulted from the combined effects of superheating, flow pressure, and collision during solidification. The surface stress would be increased owing to the appearance of those wrinkles and pits, and the stress centralizes around the wrinkles and in the pits [24]. The successful deposition of insulative SiO₂ nanoparticles onto the Fe-6.5wt.%Si particles is indicated in Figure 2b–h. The surface roughness clearly increased as the quantity of deposited SiO₂ increased owing to longer deposition times. The wrinkles and pits became less defined as the quantity of deposited SiO₂ increased with the increasing deposition time. It was possible that the FBCVD SiO₂ deposition process was similar to that of traditional chemical vapor deposition [25]. It could then be divided into four major evolutionary stages. When deposition was performed for 60 s (Figure 2b), the SiO₂ insulation nucleated on the surface of the Fe-6.5wt.%Si particles by absorbing SiO₂ or merging. When the deposition time was extended to 120 s (Figure 2c), the growing SiO₂ nuclei formed three-dimensional islands once their concentration reached a certain threshold. As the FBCVD reaction progressed, the SiO₂ islands continuously came into contact by gathering and through the secondary nucleation of young SiO₂ nuclei. Particles obtained after deposition for <960 s are shown in Figure 2d–f. Most of the adjacent SiO₂ islands merged, leaving only a small number of stripe-shaped gaps. Fresh SiO₂ continued to nucleate and grow within the gaps to form an integrated and continuous SiO₂ insulative coating in the final stage of deposition. The particles obtained after deposition for 1920 s and 3840 s are shown in Figure 2g,h, respectively. The vast majority of the initial SiO₂ islands was located in the wrinkled and pitted areas of the Fe-6.5wt.%Si substrate (tagged in red in Figure 2c). According to the model of heterogeneous nucleation on rough surfaces [26], the nucleation energy can be calculated using Equation (1).

$$\Delta G_{Het} = \left(\frac{16\pi r_{ns}^3}{3\Delta g_v^2} \right) \left[\frac{(1 - \cos \theta^*)^2 (2 + \cos \theta^*)}{4} \right] \quad (1)$$

where ΔG_{Het} is the heterogeneous nucleation energy on a rough surface; r_{ns} is the ratio of the interfacial energies of a SiO₂ nucleus and an Fe-6.5wt.%Si particle; and Δg_v is the difference between the free energies of the SiO₂ nucleus and the Fe-6.5wt.%Si particle. θ^* is the epigenetic contact angle between the SiO₂ nucleus and the Fe-6.5wt.%Si particle, which can be calculated using Equation (2) according to the Wenzel model [27].

$$\cos \theta^* = f \cos \theta \quad (2)$$

where f is the roughness factor of a rough surface, and θ is the intrinsic contact angle between a SiO₂ nucleus and an Fe-6.5wt.%Si particle. The schematic illustration of the intrinsic contact angle (θ) and epigenetic contact angle (θ^*) between the SiO₂ nucleus and the Fe-6.5wt.%Si particle was shown in Supplementary Figure S1. As the SiO₂ nucleus is present on an island structure, the value of θ is less than 90° [28,29]. Many more nucleation sites were thus present in the wrinkled and pitted areas, because ΔG_{Het} decreased as the value of f increased. This indicated that the Fe-6.5wt.% Si/SiO₂ composite structure could form quickly during FBCVD. However, the time required to form the integrated Fe-6.5wt.% Si/SiO₂ C-S structure is not known with certainty, and further investigation is needed.

Figure 2. (a) Field-emission scanning electron microscope (FE-SEM) images of the original Fe-6.5wt.%Si particles; particle samples following FBCVD for (b) 60 s, (c) 120 s, (d) 240 s, (e) 480 s, (f) 960 s, (g) 1920 s, and (h) 3840 s.

To investigate the formation and transformation processes of the Fe-6.5wt.%Si/SiO₂ C-S structure, electron backscatter diffraction analysis was performed on sections of the Fe-6.5wt.%Si/SiO₂ composite particles after deposition for >120 s. The cross-sectional FE-SEM images are shown in Figure 3a–f. The thickness and uniformity of the SiO₂ insulation and the cladding ratio increased dramatically with the increasing deposition time. The Stranski–Krastanovs mode of nucleation and growth was observed [30], which was in good agreement with the results of previous analyses. Within a relatively short period of 120 s, roughly 61.8% of the Fe-6.5wt.%Si substrate surface in Figure 3a was covered with insulative SiO₂ cladding, with an average thickness of 93 nm. As the deposition time increased to 960 s (Figure 3d), the Fe-6.5wt.%Si substrate surface became completely covered with layers of insulative SiO₂ cladding, with thicknesses ranging from 107 nm to 262 nm. Moreover, the average value of the SiO₂ insulation thickness was 184.5 nm. The results indicated that the evolution of the Fe-6.5wt.%Si/SiO₂ composite structure into the integrated Fe-6.5wt.%Si/SiO₂ C-S structure might take 960 s or longer. With deposition times of 1920 s and 3840 s (Figure 3e–f), the Fe-6.5wt.%Si/SiO₂ C-S structure was much more recognizable. The cladding ratios and thicknesses of the SiO₂ layers after deposition for various amounts of time are shown in Figure 3g. The experimental data were fitted using the classic Kolmogorov–Johnson–Mehl–Avrami equation [31] to investigate the relationship between the cladding ratio and the deposition time.

$$C_{\text{SiO}_2} = 1 - \exp(-0.0274t^{0.735}) \quad (3)$$

where C_{SiO_2} is the cladding ratio of SiO_2 insulation on the surface of the Fe-6.5wt.%Si substrate, and t is the FBCVD reaction time. This fitting equation demonstrated here can provide basic information to precisely control the morphology characteristics of Fe-6.5wt.%Si/ SiO_2 C-S structure particles during the FBCVD process.

Figure 3. Cross-sectional images of Fe-6.5wt.%Si/ SiO_2 composite particles after deposition for (a) 120 s, (b) 240 s, (c) 480 s, (d) 960 s, (e) 1920 s, and (f) 3840 s. (g) The relationships between the cladding ratio, SiO_2 insulation thickness, and deposition time.

The chemical states of the Fe-6.5wt.%Si/ SiO_2 composite particles after deposition for > 480 s and their variation with deposition time were studied via XPS. The results are shown in Figure 4 and Table 2. The XPS survey spectrum of the Fe-6.5wt.%Si/ SiO_2 composite particles after deposition for 480 s contained characteristic C, Si, O, and Fe peaks. For calibration purposes, the C_{1s} electron binding energy corresponding to adsorbed carbon was set at 284.8 eV. The intensities of the Si and O signals increased with deposition time, while the Fe signal decreased in intensity. The Fe_{2p} peak existing in the Fe-6.5wt.%Si/ SiO_2 composite particles after deposition for 480 s was because the SiO_2 is not fully coated on the Fe-6.5wt.%Si particle surface to form the Fe-6.5wt.%Si/ SiO_2 C-S structure. When the deposition time increased above 960 s, the Fe_{2p} peak was nearly undetectable, indicating that formation of the Fe-6.5wt.%Si/ SiO_2 C-S structure was complete after deposition for 960 s and the thickness of SiO_2 insulation is higher than 10 nm. Moreover, the deconvoluted Fe_{3p} spectra for electrons on the surfaces of Fe-6.5wt.%Si/ SiO_2 C-S particles after deposition for 1920 s is shown in Supplementary Figure S2. Four Si_{2p} peaks at electron binding energies of 102.54 eV, 103.31 eV, 103.84 eV, and 104.07 eV [32,33] are visible in the deconvoluted Si spectrum of Fe-6.5wt.%Si/ SiO_2 C-S particles after deposition for 1920 s, as shown in Figure 4e. According to the ^{29}Si magic angle spinning nuclear magnetic resonance (MAS NMR) and chemical kinetics results [34–36], there are four categories of Si groups with different oxidation states during thermal decomposition of the $C_8H_{20}O_4Si$ precursor. Moreover, the concentrations of four categories of Si groups decrease in the order $Si(OSi)_4$ (group Q1) > $Si(OSi)_3OH$ (group Q2) > $Si(OSi)_2(OH)_2$ (group Q3) > $Si(OSi)(OH)_3$ (group Q4). In accordance with the results of the quantitative analysis (Table 2), the Si_{2p} electron structure on the Fe-6.5wt.%Si/ SiO_2 C-S surface after deposition for 1920 s indicated the presence of Si in Q3 groups (102.54 eV, 12.81 at.%), Q1 groups (103.31 eV, 42.56 at.%), Q2 groups (103.84 eV, 31.08 at.%), and Q4 groups (104.07 eV, 8.35 at.%). Similarly, the O_{1s} electrons on the surfaces of the Fe-6.5wt.%Si/ SiO_2 C-S particles after deposition for 1920 s were assigned to Q3 groups (531.24 eV, 14.07 at.%), Q1 groups (532.62 eV, 43.87 at.%), Q4 groups (532.81 eV, 11.33 at.%), and Q2 groups (532.97 eV, 29.24 at.%) [33]. The peak at 534.03 eV was ascribed to O atoms bound to the Fe-6.5wt.%Si alloy surface (1.99 at.%). It was noted that the sum of the O atoms in Q2 groups and O atoms bound to the Fe-6.5wt.%Si alloy surface (31.23 at.%) was in close proximity to the at.% of Si in Q2 groups (31.08 at.%). This indicated that the O atoms interacting with the Fe-6.5wt.%Si alloy surface were also in Q2 groups. Although the binding energies of Si_{2p} and O_{1s} electrons did not change as the deposition time increased to 4000 s (Figure 4h), their relative proportions in Q3 and Q4 groups decreased slightly. This meant that Si groups with a high degree of polymerization were more thermally stable [37,38]. The Fe-6.5wt.%Si/ SiO_2 C-S structural formation

process during FBCVD is shown schematically in Figure 5, which is based on the combined XRD, SEM, and XPS results.

Figure 4. X-ray photoelectron spectroscopy (XPS) survey spectra of Fe-6.5wt.%Si/SiO₂ composite particles after deposition for (a) 480 s, (b) 960 s, (c) 1920 s, and (d) 3840 s. The deconvoluted Si 2p and O 1s spectra for electrons on the surfaces of Fe-6.5wt.%Si/SiO₂ C-S particles after deposition for 1920 s (e,f) and 3840 s (g,h).

Table 2. Quantitative elemental analysis of Si_{2p} and O_{1s} on the surfaces of Fe-6.5wt.%Si/SiO₂ composite particles after deposition for 1920 s and 3840 s.

Deposition Time	Peaks	Relative Sensitivity Factors	Position	Full Width Half Maximum	Area	Atomic Ratio
1920 s	Si _{2p}	1	102.54 eV	1.40	4610.1	12.81%
	Si _{2p}	1	103.31 eV	1.31	10,896.3	42.56%
	Si _{2p}	1	103.84 eV	1.53	7956.0	31.08%
	Si _{2p}	1	104.07 eV	1.16	2138.9	8.35%
	O _{1s}	1	531.24 eV	1.98	21,095.4	14.07%
	O _{1s}	1	532.62 eV	1.53	65,018.5	43.87%
	O _{1s}	1	532.81 eV	1.80	16,911.4	11.33%
	O _{1s}	1	532.97 eV	1.87	43,835.3	29.24%
	O _{1s}	1	534.03 eV	2.61	2978.7	1.99%
3840 s	Si _{2p}	1	102.54 eV	1.70	4343.8	15.95%
	Si _{2p}	1	103.31 eV	1.39	11,862.6	43.55%
	Si _{2p}	1	103.87 eV	1.55	8693.9	31.92%
	Si _{2p}	1	104.07 eV	1.80	2337.9	8.58%
	O _{1s}	1	531.24 eV	1.87	20,603.5	13.28%
	O _{1s}	1	532.62 eV	1.45	69,023.3	44.49%
	O _{1s}	1	532.81 eV	1.33	13,883.4	8.95%
	O _{1s}	1	532.97 eV	1.91	46,698.8	30.10%
	O _{1s}	1	534.03 eV	1.16	4922.8	3.17%

Figure 5. Schematic illustration of the Fe-6.5wt.%Si/SiO₂ C-S structural formation process during FBCVD.

3.2. Static Magnetic Properties

The static magnetic properties of the original Fe-6.5wt.%Si particles and Fe-6.5wt.%Si/SiO₂ C-S particles were analyzed using a VSM. The results are shown in Figure 6. On the basis of their hysteresis loops, all of the particle samples exhibited excellent soft magnetic properties. The saturated magnetization (M_s) values of the Fe-6.5wt.%Si/SiO₂ C-S particles were obtained in the range from 176.9 to 182.0 emu/g as the deposition time increased from 960 s to 3840 s. The M_s values of the Fe-6.5wt.%Si/SiO₂ C-S particles were slightly lower than those of the original Fe-6.5wt.%Si particles (188.1 emu/g). The decrease in the M_s values with the increasing deposition time might have been the result of the growing thickness and quality of the non-magnetic amorphous SiO₂ shells on the core Fe-6.5wt.%Si particles. This also had the effect of reducing the magnetic moment per unit

volume. The theoretical value of M_s according to the core-surface model [39] could be calculated using Equation (4).

$$M_s = \frac{M_{s(Fe-6.5wt.\%Si)} \times v_{(Fe-6.5wt.\%Si)} + M_{s(SiO_2)} \times v_{(SiO_2)}}{v_{(Fe-Si)} + v_{(SiO_2)}} \quad (4)$$

$$= \frac{M_{s(Fe-6.5wt.\%Si)} \times d^3 + M_{s(SiO_2)} \times ((d+r)^3 - d^3)}{(d+r)^3}$$

where $M_{s(Fe-6.5wt.\%Si)}$ is the M_s of the pure Fe-6.5wt.%Si particles (188.1 emu/g); $M_{s(SiO_2)}$ is the M_s of the non-magnetic amorphous SiO_2 shell (0 emu/g); d is the average radius of the original Fe-6.5wt.%Si particles (35 μm); and r is the thickness of the non-magnetic amorphous SiO_2 shell. The theoretical and measured M_s values of all the particle samples are summarized in Table 3. It was clear that the measured M_s values agree with the theoretical results approximately, which verified the correctness of the experimental data in this work and indicated that both the microstructural characteristics and the saturated magnetization values of the particles could be controlled with the deposition time. However, the value of H_c changed little, which was thought to be because the test conditions of vibrating sample magnetometer (step-size of 50 Oe and testing precision of 1%) cannot satisfy the demands of H_c of the soft magnetic materials [40].

Figure 6. Hysteresis loops and details about the applied magnetic fields and saturated magnetization of the original Fe-6.5wt.%Si particles and Fe-6.5wt.%Si/ SiO_2 C-S particles after deposition over various intervals.

Table 3. Theoretical and measured saturated magnetization values of the original Fe-6.5wt.%Si particles and Fe-6.5wt.%Si/ SiO_2 C-S particles following deposition.

Samples	Theoretical Value (emu/g)	Measured Value (emu/g)
original Fe-6.5wt.%Si particles	188.1	188.1
Fe-6.5wt.%Si/ SiO_2 C-S particles 960 s	182.0	182.9
1920 s	180.3	181.0
3840 s	176.9	178.4

The electrical resistivities of the original Fe-6.5wt.%Si particles and the Fe-6.5wt.%Si/ SiO_2 C-S particles are shown in Figure 7. According to the percolation theory, an increase in the electrical resistivity of a conducting/insulating C-S structure is the result of disruption of the electrical conductivity network [41]. Conduction in the original Fe-6.5wt.%Si particles and the Fe-6.5wt.%Si/ SiO_2 C-S particles could be described using a simple circuit model. On one hand, the original Fe-6.5wt.%Si particles were in continuous contact to form chain return, which was equivalent to the current passed through series-connected resistors. On the other, the conductive Fe-6.5wt.%Si cores of the Fe-6.5wt.%Si/ SiO_2 C-S particles were covered with insulative amorphous SiO_2 shells, which corresponded to capacitance.

Therefore, the electrical resistivities of Fe-6.5wt.%Si/SiO₂ C-S particles after deposition for 480 s (2054 $\mu\Omega\cdot\text{m}$) were higher than those of the original Fe-6.5wt.%Si particles (0.91 $\mu\Omega\cdot\text{m}$) by four orders of magnitude. The increase in the SiO₂ shell thickness with the increasing deposition time led to more obvious capacitance effects. This increased the electrical resistivities of the Fe-6.5wt.%Si/SiO₂ C-S particles to 2347 $\mu\Omega\cdot\text{m}$ (1920 s) and 2660 $\mu\Omega\cdot\text{m}$ (3840 s).

Figure 7. Electrical resistivities of the original Fe-6.5wt.%Si particles and Fe-6.5wt.%Si/SiO₂ C-S particles following deposition.

4. Conclusions

We studied the formation of an integrated Fe-6.5wt.%Si/SiO₂ C-S structure during fluidized bed chemical vapor deposition. The Fe-6.5wt.%Si/SiO₂ C-S particles were synthesized using Fe-6.5wt.%Si particle substrates and a gaseous C₈H₂₀O₄Si precursor via FBCVD for different amounts of time. The Fe-6.5wt.%Si/SiO₂ composite particles could be obtained in 60 s. These were converted into integrated ferromagnetic/SiO₂ C-S structures after deposition for ≥ 960 s. The process of integrated ferromagnetic/SiO₂ C-S structure formation was described using the three-dimensional island nucleation theory. Islands began to form in wrinkled and pitted areas of the Fe-6.5wt.%Si particle surfaces. The deposited SiO₂ was an amorphous phase with four Si groups and five O groups. As the deposition time increased from 120 s to 960 s, the cladding ratio increased from 61.8% to 100%, and the average thickness of the SiO₂ layer increased from 93 nm to 184.5 nm. The average thickness of the SiO₂ layer increased as the FBCVD reaction progressed, and it became more uniform. The saturated magnetization values of the Fe-6.5wt.%Si/SiO₂ C-S particles decreased from 182.0 emu/g to 176.9 emu/g as the deposition time increased from 960 s to 3840 s, while their electrical resistivity increased from 2054 $\mu\Omega\cdot\text{m}$ to 2660 $\mu\Omega\cdot\text{m}$. We believe this work will help promote the use of FBCVD for the synthesis of high-performance Fe-6.5wt.%Si/SiO₂ and other ferromagnetic-based SMCs for future applications.

Supplementary Materials: The following are available online at <http://www.mdpi.com/2075-4701/10/4/520/s1>, Figure S1: Schematic illustration of intrinsic contact angle (θ) and the epigenetic contact angle (θ^*) between the SiO₂ nucleus and the Fe-6.5wt.%Si particle; Figure S2: The deconvoluted Fe3p spectra for electrons on the surfaces of Fe-6.5wt.%Si/SiO₂C-S particles after deposition for 1920 s.

Author Contributions: Conceptualization, writing—original draft preparation, Z.W. and J.J.; methodology, C.X.; investigation, X.L.; writing—review and editing, K.X.; visualization, project administration, H.K. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Natural Science Foundation of Anhui Province (1908085QE190), Chinese National Science Foundation (51904002, 51674181), and Key Program of Scientific Research Fund of Anhui Provincial Education Department (KJ2019A0077).

Conflicts of Interest: The authors declare no conflict of interest.

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