Study on the electromagnetic properties and microwave absorbing mechanism of flaky FeSiAl alloy based on annealing and phosphate coating

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Keywords: FeSiAl, low-frequency absorbing materials, annealing, phosphating, electromagnetic performance

Abstract
A low-frequency composite absorbing material with high permeability and low permittivity in low frequency was prepared after the flaky FeSiAl alloy powder was annealed in N₂ atmosphere and modified with zinc-based phosphate solution. The effects of annealing temperature and phosphating time on the phase composition, morphology and static magnetic properties of the flaky FeSiAl alloy were investigated with x-ray diffraction (XRD), scanning electron microscope (SEM) and vibrating sample magnetometer (VSM). With the aid of vector network analyzer (VNA), the electromagnetic properties and absorbing performance of the powder treated in two ways were studied. The results showed that after annealing in N₂ atmosphere, the crystallization degree of FeSiAl powder increased, the crystal type underwent a phase transition of A2-A2/B2-A2/B2/D03, the permittivity decreased, and the permeability was greatly improved. After phosphating, the phosphating reaction products were deposited on the surface of FeSiAl particles, the permittivity was significantly reduced, the low-frequency impedance matching and low-frequency absorption performance were improved. The causes and frequency changes of electromagnetic wave absorption peaks were analyzed with the λ/4 interference cancellation theory, the interface reflection model and the impedance matching principle.

1. Introduction

With the development of radar detection technology, the low-frequency (0.5 ~ 4 GHz) radars, especially decimeter-wave and meter-wave radars are playing an increasingly important role. The application of early warning radars and beyond-horizon radars makes the development of low-frequency absorbing materials urgent [1, 2]. In addition, in civilian areas, low-frequency electromagnetic waves can also cause biological damages to human health through thermal effects [3], so the development of ‘thin, wide, light and strong’ low-frequency absorbing materials [4] is of great significance.

Soft magnetic alloy powders, represented typically by FeSiAl, has high saturation magnetization and a high Snoek limit in the low-frequency range [5]. The shape effect produced by the flattening increases its permeability in the low-frequency range. Also due to the presence of solid solution elements, the eddy current effect is small, so the flaky soft magnetic alloy powder has a good application prospect as a low-frequency radar wave absorber. However, due to the large size of FeSiAl particles, they are more likely to overlap adjacent particles after flattening, forming a conductive network, resulting in an abnormal increase in permittivity, and thus lower absorption strength [6]. The current treatment of flaky soft magnetic alloys is mainly reducing their permittivity to improve their impedance matching performance. Wang et al [7] used magnesium-based phosphating solution to modify the flaky FeCoZr soft magnetic alloy powder. After phosphating, the permittivity decreased, the absorption peak moved toward the high-frequency range, and the reflectivity reached −11.4 dB at 6.2 GHz.
Zhang et al [8] heat-treated the flaky FeSiAl alloy powder under the protection of N₂/H₂, selectively oxidized elements Si and Al, and protected element Fe. At 700 °C in heat treatment, the permeability increased slightly while the permittivity decreased by 50%, and the reflectivity reached −8.9 dB at 1 GHz. Sun et al [9] prepared FeSiAl/graphite composite material via the high-energy ball milling process. The graphite layer suppressed the generation of eddy currents between contact particles, lowered the permittivity, and had −21 dB absorption at 6.7 GHz.

In view of the current high permittivity, poor low-frequency absorbing performance, and lack of in-depth researches on the absorption mechanism of the flaky FeSiAl alloy, this paper adopted two methods—annealing with N₂ protection and subsequent surface phosphating modification—to improve the permeability and reduce the permittivity of the flaky FeSiAl alloy, realized its effective absorption of the electromagnetic waves in the L band (1 ~ 2 GHz), studied the influence of annealing and phosphating on the absorbing performance of flaky FeSiAl, and analyzed the absorbing mechanism.

2. Experimental procedure

2.1. Experimental materials
Flaky FeSiAl alloy powder (mass fraction: 85% for Fe, 10% for Si, and 5% for Al) prepared with the high-energy ball milling method in the early stage; acetone, zinc oxide, nitric acid and phosphoric acid purchased from Tianjin Damao Chemical Reagents Factory; high-purity nitrogen purchased from Shijiazhuang Chemical Reagent Factory; and self-made additives for phosphating and experimental water.

2.2. Heat treatment process
A tube furnace was used for annealing in an N₂ atmosphere. The heating rate was set to be 5 °C min⁻¹, and the temperature was kept at 400 ~ 800 °C for 2 h. Furnace cooling was adopted for cooling purpose.

2.3. Phosphating process
The reagents were weighed and mixed at the mass ratio of (ZnO: HNO₃: H₃PO₄)=1:0.5:2.5. 300 ml deionized water and a small amount of homemade additives were added to the solution. The solution was stirred till it became transparent. FeSiAl alloy powder which was annealed at 700 °C and had the best magnetic properties was chosen for phosphating treatment. 2 g of the annealed sample was weighed and put into acetone for ultrasonic cleaning for 10 min, and then placed in 200 ml phosphating solution after magnetic separation. The mixed liquid was heated to 80 °C with water bath using medium temperature phosphating, heating, and mechanically stirred. Samples were taken separately after the liquid was phosphated for 2 min, 4 min and 6 min, cleaned and dried before surface phosphated flaky FeSiAl soft magnetic alloy powder was obtained.
Hitachi S-4800 scanning electron microscope was used to observe the morphology of the samples; X-ray diffractometer was applied to analyze the crystal structure of the samples (Cu Kα radiation, scanning angle 10° ~ 90°, scanning rate 8°/min); Model6000 PPMS-VSM was used to test the static magnetic properties of the samples at room temperature; Agilent-N5242A vector network analyzer was used to measure the electromagnetic parameters of the samples on the 0.5 ~ 6 GHz frequency band, and Matlab was used to calculate the reflection loss of the samples based on the transmission line theory.

3. Results and discussion

3.1. Morphology
Figure 1 shows the morphology of flaky FeSiAl alloy powder and the annealed samples. After annealed, FeSiAl still maintained flaky particles. The flakes were about 20 ~ 40 μm in diameter, and less than 1 μm in thickness. At high resolution, it can be seen that the surface of the unannealed particles was relatively smooth, while there were some fine particles on the surface of samples annealed at 600 °C or above. As the annealing temperature rose, the number of such fine particles about 10 ~ 20 nm big in diameter increased gradually on the surface. The main reason for this was that at a high temperature and under great pressure, N₂ reacted with FeSiAl to form nitride on the surface, and the reaction process became more adequate as temperature rose [10].
Figure 2 shows the morphology of the samples phosphated for different lengths of time after they were annealed at 700 °C, and the EDS of the samples phosphated for 2 min. It can be seen that flake particles had good dispersibility after phosphating, and the phosphating precipitates were obviously adsorbed on the surface of the samples after the samples were phosphated for 2 min and 4 min, and the surface precipitates increased gradually as the time of phosphating lengthened. After 6 min of phosphating, the sample contained a lot of agglomerated precipitate impurities, showed poor purity, and the phosphating effect was poor. Figure 2(d) shows the energy spectrum of the sample phosphated for 2 min whose main elements were still Fe, Si, Al, and which contained small amounts of Zn, P, O and C, in which C came from the additive of auxiliary film forming.

3.2. Structural analysis

Figure 3(a) shows the X-ray pattern of flaky FeSiAl alloy powder and annealed samples. It can be seen that FeSiAl alloy had only three diffraction peaks (110) (200) (211) in the crystal form, with weaker diffraction peaks and larger half-heights, indicating that the grains were small and there were a lot of defects and lattice distortions. After annealed at 500 °C, a (200) diffraction peak appeared at 31.5°, indicating that the crystal structure changed from A2 phase to a superlattice structure of B2 phase. When the annealing temperature reached 600 °C, a (111) diffraction peak appeared at 27.5°, indicating the appearance of the D03 phase structure in FeSiAl. After that, as the annealing temperature rose, the samples’ diffraction peak height became larger and the half-height width became narrower, the degree of crystallinity was improved and internal defects were further reduced. It can be seen that the crystal structure of FeSiAl powder, after annealed at different temperatures, followed such phase transition as A2-A2/B2-A2/B2/D03. In addition, because it was difficult for N2 to react with FeSiAl and there were few nitrided products, the characteristic diffraction peak of nitrided did not appear after annealing. The XRD pattern of each sample after phosphating was shown in figure 3(b). The precipitation products were mainly Zn3(PO4) · 4H2O and Zn2Fe(PO4)2 · 4H2O. The phosphate precipitation diffraction peak of the sample after 2 min of phosphating was not obvious, due to the small amount of phosphate precipitation, only significant the FeSiAl diffraction peak. There are obvious diffraction peaks of FeSiAl and phosphate in the sample after phosphating for 4 min. After phosphated for 6 min, FeSiAl didn’t show an obvious diffraction peak due to the formation of a large number of phosphating products and the reaction consumption the flaky particles.

3.3. Static magnetic properties

Figure 4 shows the hysteresis loops of FeSiAl alloy powder annealed at different temperatures. The specific magnetostatic parameters of each sample are listed in table 1. It can be seen that, as the annealing temperature rose, the saturation magnetization (Ms) of FeSiAl alloy didn’t show obvious regular changes, which is similar to that of unannealed flaky FeSiAl alloy. This is because Ms is mainly determined by material composition, but less affected by structural changes. As the annealing temperature rose, the coercivity (Hc) of the samples decreased gradually, showing superparamagnetic properties. The reason is that Hc is affected by grain size, internal stress, grain boundary and internal defects. However, annealing can reduce the internal defects and internal stress, resulting in the decrease of Hc [11]. The paramagnetic characteristics of the powder after annealed helped the movement of magnetic domains and free electrons. Therefore, the magnetostatic properties of the flaky FeSiAl alloy can be improved by annealing.
3.4 Electromagnetic parameters

Figure 5 shows the permittivity of flaky FeSiAl alloy powder, annealed FeSiAl alloy powder at 700 °C and samples with different phosphating duration. It can be seen that the real part \(\varepsilon'\) and imaginary part \(\varepsilon''\) of permittivity of the annealed FeSiAl were significantly lower than those of unannealed FeSiAl. The maximum value of real part dropped from 140 to 58, and the maximum value of imaginary part from 180 to 17. This is because the nitrides on the surface of FeSiAl alloy after annealing formed a high resistance layer, which blocked electron transfer among different particles and reduces the conductivity of the powder, resulting in permittivity decrease. As the phosphating duration lengthened, the real and imaginary parts of the permittivity of the phosphated samples were further reduced. The sample had the lowest permittivity after it was phosphated for 6 min, with the maximum value of real part and that of the imaginary part being 16 and 6, respectively. The real part of the permittivity is mainly related to the polarization of the FeSiAl particles, while the dielectric polarization of the flaky particles is mainly its intrinsic electron displacement polarization [12]. In this study, due to the absorption of phosphate particles, the contact between adjacent flaky particles was blocked, and the electronic displacement was limited, leading to the decrease of polarization degree, and then the decrease of the real part of the permittivity. The imaginary part of the permittivity is mainly related to the conductivity of the material. According to free electron theory [13], electrical loss of magnetic materials:

![Figure 5. Permittivity of each sample (a) real part and (b) imaginary part.](image1)

![Figure 6. Permeability of each sample (a) real part and (b) imaginary part.](image2)

Table 1. Magnetostatic parameters of FeSiAl alloy powders annealed at different temperatures.

| Samples          | Origin sample | 400 °C | 500 °C | 600 °C | 700 °C | 800 °C |
|------------------|---------------|--------|--------|--------|--------|--------|
| \(H_C/\text{Oe}\) | 13.6          | 9.9    | 5.4    | 4.6    | 3.6    | 2.9    |
| \(M_s/\text{(emu g}^{-1}\) | 128           | 121    | 136    | 126    | 129    | 137    |

![Table 1](image3)
Where $\varepsilon_0$ is the vacuum permittivity, $\rho$ is the metal resistance and $f$ is the frequency. After phosphating on the surface of FeSiAl alloy, the alloy particles are insulated from each other, and it is difficult to form a conductive network, which decreases its conductivity decreases. According to the above formula, the imaginary part of the permittivity of the material will decrease obviously after phosphating.

Figure 6 shows the permeability of flaky FeSiAl alloy powder, samples annealed at 700 °C and samples later obtained with different phosphating duration. With the increase of frequency, the real part ($\mu'$) of permeability decreased gradually, and the imaginary part ($\mu''$) increased first and then decreased, showing a single peak. After annealed at 700 °C, the real part and imaginary part of permeability of FeSiAl alloy powder increased, especially in the low-frequency range of less than 2.5 GHz. The permeability of magnetic material can be expressed by formula (2) [14]:

$$\varepsilon'' = \frac{1}{2\pi \varepsilon_0 \rho f}$$

(1)

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$$\mu_i = \frac{M_i^2}{a k_H M_s + b \lambda \xi}$$

(2)

Where $\mu_i$ is the initial permeability, $a$ and $b$ are constants, $k$ is the proportional coefficient, $\varepsilon$ is the lattice elastic strain coefficient, and $\lambda$ is the magnetostrictive coefficient. In the formula, the permeability depends on $M_i$ and $H_c$. Just as in the analysis of static magnetic properties of the annealed samples, when $M_i$ is basically the same, $H_c$ gradually decreases with the increase of annealing temperature. Therefore, according to formula (2), the permeability of FeSiAl alloy powder increased after annealing. After annealing, the internal stress was eliminated, the soft magnetic properties were improved, and the magnetic anisotropy was reduced, therefore the resonance frequency of the imaginary part of the permeability was reduced. As the phosphating duration lengthened, the real part and the imaginary part of the permeability of the samples phosphated for 2 min and 4 min were slightly lower than those of the annealed FeSiAl alloy, while the permeability of the samples phosphated for 6 min was significantly lower. Firstly, the phosphate particles on the surface hindered the domain wall displacement of FeSiAl alloy in the process of magnetization; secondly, in the phosphating process, the electrochemical reaction of Fe dissolving into Fe$^{2+}$ occurred in the anode area, which led to the decrease of Fe content in the alloy, and then permeability reduction. After 6 min of phosphating, the permeability of the sample decreased significantly, mainly because the phosphating lasted too long, the precipitation products increased and agglomerated in large numbers, which seriously reduced the proportion of FeSiAl alloy.

Through the theoretical model of transmission line, the electromagnetic wave reflection loss intensity (RL) of the material can be calculated [15]:

### Table 2. The absorption characteristic parameters of each sample.

| Samples          | $f_m$/GHz | $RL_{min}$/dB |
|------------------|-----------|---------------|
| Origin sample    | 1.4       | −4.0          |
| 700 °C          | 1.6       | −8.6          |
| 2 min          | 1.8       | −10.7         |
| 4 min          | 2.6       | −10.4         |
| 6 min          | 4.4       | −9.2          |

Figure 7. (a) RL-$f$ curves of each sample and (b) 3D RL-$f$ curves of the sample after 2 munites of phosphating.
Where $Z_{in}$ is the normalized input impedance, $c$ is the speed of vacuum light, $f$ is the microwave frequency, $t$ is the thickness of the absorber, and $\mu_r$ and $\varepsilon_r$ are relative permittivity and permeability.

The $RL$-f curves of each sample calculated with the transmission line theory were shown in figure 7(a). The thickness of each sample was set to be 2.5 mm, and the detailed data of reflectance can be found in table 2. After annealed at 700 °C, the absorption strength of FeSiAl alloy powder increased compared with that of unannealed FeSiAl alloy powder, the reflectivity decreased from $-4.0$ dB to $-8.6$ dB, and the absorption peak frequency shifted to high frequency, from 1.4 GHz to 1.6 GHz. The permittivity of the phosphated FeSiAl alloy samples was further reduced and the absorption strength was improved compared with that of the FeSiAl alloy annealed at 700 °C. The absorption peak frequency ($f_{m}$) increased with the increase of phosphating time. Among them, the sample phosphated for 2 min had the lowest reflectivity, low absorption peak frequency and excellent low-frequency absorption effect, with $-10.7$ dB absorption at 1.8 GHz, the frequency band when the reflectivity was

![Table 3. Performance comparison with other soft magnetic alloys.](image)

| Sample | $f_{m}$/GHz | Absorption band below $-8$ dB GHz$^{-1}$ | Relative Bandwidth | Thickness mm$^{-1}$ |
|--------|-------------|-----------------------------------------|--------------------|-------------------|
| [16]   | 2.1         | 1.3–2.5                                 | 1.9                | 3                 |
| [17]   | 1.6         | 1.3–1.9                                 | 1.5                | 2.5               |
| [18]   | 1.3         | 1–1.7                                   | 1.7                | 2                 |
| [19]   | 2.5         | 2–4.5                                   | 2.25               | 2.25              |
| [20]   | 2.2         | 1.5–2.7                                 | 1.8                | 3                 |
| This work | 1.8         | 1.1–2.6                                 | 2.4                | 2.5               |

![Figure 8. S11 and S11-OPEN of annealed at 700 °C(a) and phosphate for 2 min (b), 4 min (c), 6 min (d).](image)

\[
RL = 20 \log \left| \frac{Z_{in} - 1}{Z_{in} + 1} \right| 
\]  

(3)

\[
Z_{in} = \sqrt{\frac{\mu_r}{\varepsilon_r}} \tan h \left[ j \left( \frac{2\pi}{c} \right) \sqrt{|\mu_r| |\varepsilon_r| |f|} \right] 
\]  

(4)
lower than $-8\text{dB}$ (84% echo energy attenuation) was $1.1 \sim 2.6$ GHz. Figure 7(b) shows the reflectance curves of samples of different thickness which were phosphated for two minutes. It can be seen that the absorption peak frequency and reflectance decreased gradually as thickness increased. When the thickness was 5.7 mm, it reached the matching thickness and had the lowest reflectance of $-29\text{ dB}$.

In this paper, the microwave absorbing properties of FeSiAl alloy modified by annealing and phosphating solution were compared with those of other soft magnetic alloys, as shown in table 3. It can be seen from the table that, compared with other soft magnetic alloy materials in the literature, the FeSiAl composite prepared in this paper had lower absorption frequency and a higher relative bandwidth level ($\text{relative bandwidth} = \text{high frequency cut-off frequency}/\text{low frequency cut-off frequency}$ in the absorption band below $-8\text{ dB}$). It can be seen from the comparison that the FeSiAl composite microwave absorbing material designed and prepared in this paper had such advantages as requiring low experimental conditions, being easy to be prepared and massively produced, and having a good low-frequency effect and a wide frequency band.

### 3.5. Absorbing mechanism analysis

There are two ways for the absorbing materials to attenuate electromagnetic waves, that is, internal electromagnetic loss and interference cancellation of reflected waves at front and back interfaces. In order to explore the reason behind the improvement of microwave absorbing properties of flaky FeSiAl alloy powder after phosphating, it is necessary to distinguish the ways of electromagnetic wave energy loss. The front and back interface reflection model \cite{21} is introduced here. When the electromagnetic wave vertically irradiates the surface of the absorbing material, one part of the electromagnetic wave is reflected by the front interface of the material ($S_{11\text{-OPEN}}$); the other part enters the interior of the material and exits from the front interface of the material after being reflected by the metal back plate, and the exit electromagnetic wave is vector superimposed with the surface reflection wave to form the total reflection wave ($S_{11}$). When the total energy of incident wave is 1, the equation is as follows:

$$2S_{11\text{-OPEN}} \pm S_{11} + E_{\text{loss}} = 1 \quad (5)$$

Where $E_{\text{loss}}$ is the inner energy loss of the absorbing material, and the scattering parameters $S_{11\text{-OPEN}}$ and $S_{11}$ can be measured by vector network analyzer. Figure 8 shows the variation curve of $S_{11}$ and $S_{11\text{-OPEN}}$ with frequency. At the same time, when the total energy of incident electromagnetic wave is set as 1, the energy distribution of electromagnetic wave in each part of the absorbing material calculated according to equation (5) is listed in table 4.
It can be seen from figure 8 and table 4, after surface phosphating, the front interface electromagnetic wave reflection energy of flaky FeSiAl alloy decreased, and the internal attenuation of the electromagnetic wave energy increased, indicating improvement of the impedance matching performance. The reason is that in the process of phosphating, phosphate is formed by phosphating reaction and deposited on the surface of FeSiAl particles, which obstructs the transition and transfer of free electrons in FeSiAl alloy and reduced the permittivity. Secondly, the decrease of permittivity further weakens the skin effect of FeSiAl alloy and improves the impedance matching performance. With the increase of phosphating reaction time, the higher the phosphate content, the better the impedance matching performance of FeSiAl alloy. The results showed that the $S_{11}$-open of sample phosphated for 4 min was $-5.5$ dB, meaning that the surface direct reflection energy was 0.28, a figure significantly lower than that of the unphosphorized FeSiAl alloy (0.36). The total reflectance of the samples phosphated for 2 min and 4 min, respectively, was basically the same, but the surface reflection wave energy of the samples phosphated for 2 min was higher, showing more interference loss characteristics than that of the samples phosphated for 4 min. Therefore, in figure 7, the absorption peak of the samples phosphated for 2 min was narrower than that of the samples phosphated for 4 min. In addition, due to the aggregation of phosphating products and the decrease of effective magnetic components, the sample phosphated for 6 min or more showed lower matching performance.

According to the transmission line theory, the characteristic impedance of each sample can be calculated by equation (4). Figure 9 shows the curves of frequency and the characteristic impedance of each sample. The normalized characteristic impedance of free space was known (real part being 1, imaginary part being 0) [22], so the closer the real part of characteristic impedance of each sample was to 1, the closer the imaginary part to 0, and the better the impedance matching performance. It is difficult to directly distinguish the distance between each curve and the characteristic impedance of free space with figure 9 alone. Therefore, the surrounding area of each curve and the target curve (real part being 1, imaginary part being 0) was calculated in table 5. The larger the area was, the worse the matching was. As shown in table 5, the characteristic impedance of each sample after phosphating was closer to the free space impedance, and the impedance matching performance was improved. The sample phosphated for 4 min had the best matching performance, which was consistent with the calculation results of the front and back interface reflection model. The matching performance of the sample phosphated for 6 min was different from that in table 4. The main reason is that the matching performance of absorption peak frequency was calculated in the front and back interface reflection model, while the matching performance in the range of $0.5 \sim 6 \text{ GHz}$ was calculated in table 5.

| Samples  | $S_{\text{real part}}$ | $S_{\text{imaginary part}}$ | $S_{\text{total}}$ |
|----------|------------------------|-----------------------------|------------------|
| 700 °C   | 42.63                  | 7.36                        | 49.99            |
| 2 min    | 41.52                  | 8.13                        | 49.65            |
| 4 min    | 36.10                  | 8.27                        | 44.37            |
| 6 min    | 39.07                  | 6.77                        | 46.84            |

Figure 10. (a) Dielectric loss tangent and (b) magnetic loss tangent of each samples.
Figure 10 showed the electrical loss tangent (a) and magnetic loss tangent (b) of each sample. In figure 10(a), the imaginary part of the phosphated samples’ permittivity was greatly reduced due to the existence of phosphating film, resulting in a gradual decrease of the electrical loss performance of the material as phosphating went on. In figure 10(b), the magnetic loss of phosphated samples was basically the same as that of non-phosphated samples when the frequency was lower than 3 GHz. In the range of 3 ~ 6 GHz, the magnetic loss of the sample phosphated for 2 min was slightly higher than that of FeSiAl alloy annealed at 700 °C, while that of the sample phosphated for 4 min and that for 6 min were lower than that of the annealed samples. In addition, each sample showed much higher magnetic loss than electrical loss, indicating that magnetic loss plays a leading role in the internal loss of each phosphated flaky FeSiAl alloy.

Figure 11. Relationship between permittivity real part and imaginary part (a) and curve of $C_0$-f (b) of 2 min phosphating.

Figure 12. The $\lambda/4$ thickness and $RL$-f curves of annealed at 700 °C (a) and phosphate for 2 min (b), 4 min (c), 6 min (d).
Cole-Cole semicircle and theory of $C_0 = \mu'^n (\mu'')^{-2} f^{-1}$ are often introduced to study the specific loss mechanism of electric loss and magnetic loss [23]. In figure 11(a), there were several approximate Cole-Cole semicircle curves between the real part’s $25 \sim 35$ and the imaginary part’s $5 \sim 8$ of the permittivity, indicating that there was dielectric relaxation near the frequency of 6 GHz, which was mainly caused by interface polarization and dipole polarization. In the strong electromagnetic field in the range of $0.5 \sim 6$ GHz, the magnetic loss modes of materials mainly included natural resonance and eddy current loss [24], in which the eddy current loss can be characterized by the value of $\mu'^n (\mu'')^{-2} f^{-1}$. When the magnetic loss of materials was all from eddy current loss, the value of $C_0$ changing with frequency was constant. Therefore the magnetic loss type of materials can be analyzed by observing the change trend of $C_0$ curve. In figure 11(b), the $C_0$ value tended to be constant in the range of $0.5 \sim 2$ GHz, indicating that the magnetic loss was mainly eddy current loss, while in the range of $2 \sim 6$ GHz, the curve rose gradually, indicating that the magnetic loss was mainly natural resonance.

When the reflected wave frequency of the front interface and the back interface was the same, if the wave path difference ($\Delta$) was an odd multiple of the half wavelength, the interference phenomenon would appear, so as to counteract the reflected echo and form an absorption peak, then the matching thickness $t_m$ at the absorption peak of the absorbing material had the following relationship with the frequency $f_m$ [25]:

$$t_m = \frac{n \pi c}{4 f_m \sqrt{\varepsilon_r |\mu_r|}} \quad (n = 1, 3, 5 \cdots)$$

Where: $n$ means that when the thickness of the absorber was an odd multiple of $\lambda/4$, the material met the $\lambda/4$ interference cancellation model. Figure 12 shows the reflectivity and $\lambda/4$ medium thickness curves of each sample. It can be seen that the absorption peak frequency did not coincide with $\lambda/4$ wavelength interference thickness, and there was a deviation of about 1 GHz. This is because the $\lambda/4$ interference model represents the thickness of the absorber corresponding to zero reflection at different frequencies, indicating that the absorption peak frequency of flaky FeSiAl and FeSiAl composites after surface phosphating was not completely determined by the thickness interference, but the overall deviation was small, and when $t_m = 2.5$ mm, the change trend of the absorption peak frequency determined by the interference was the same as that of the actual absorption peak frequency. Therefore, the reason for the change of $f_m$ after phosphating can still be explained qualitatively by formula (6). When $t_m$ was determined, $f_m$ is determined by $\sqrt{|\varepsilon_r||\mu_r|}$ in inverse proportion. The $\sqrt{|\varepsilon_r||\mu_r|}$ of each sample is shown in figure 13. It can be seen that after phosphating, $\sqrt{|\varepsilon_r||\mu_r|}$ of each sample was lower than FeSiAl alloy after heat treatment. Therefore, phosphating made the absorption peak shift to high frequency.

4. Conclusion

(1) Firstly, the internal defects and residual stress of flaky FeSiAl alloy were eliminated by annealing in the $N_2$ atmosphere, the permeability was improved and the permittivity was reduced. Secondly, the permittivity was further reduced and the impedance matching performance was improved by surface modification by phosphating of flaky FeSiAl alloy powder which was annealed at $700 \degree C$. When the phosphating time was 2 min, the sample reflectivity reached $-10.7$ dB at 1.8 GHz and the reflectivity in the $1.1 \sim 2.6$ GHz was less than $-8$ dB, which can effectively absorb the L-band electromagnetic wave.
After annealing, the FeSiAl particles still maintained a sheet-shaped structure, and the nitriding products of FeSiAl formed on the surface of the particles blocked the movement of free electrons and promoted the decrease of permittivity. As annealing temperature increased, the crystal structure of the flaky FeSiAl alloy underwent a phase transformation of A2-A2/B2-A2/B2/D03, the coercivity gradually decreased, the magnetic properties were released, and the permeability was improved.

Through surface phosphating treatment, a phosphating layer can be formed on the surface of FeSiAl particles, which obstructs the transition and transfer of free electrons, promoted further permittivity reduction, and improved the absorption strength. According to the front and back interface reflection model and impedance matching theory, the reflected energy of electromagnetic waves on the surface of the alloy powder after phosphating was lower, so that more electromagnetic waves can enter the absorber and be lost. This is the internal reason for the increase in the absorption strength of FeSiAl alloy after phosphating. According to the \( \lambda/4 \) interference cancellation theory, the frequency of the absorption peak was affected by \( \sqrt{\varepsilon_r \mu_r} \). After phosphating, the absorption peaks moved to high frequency due to permittivity reduction of the FeSiAl alloy powder.

**Acknowledgments**

This paper is supported by the Natural Science Foundation of Hebei Province, China (No. E2015506011) and Innovation and Development Science Foundation of the PLA Army Engineering University Shijiazhuang Campus (No. KYSZQZL1910).

**Data availability statement**

All data that support the findings of this study are included within the article (and any supplementary files).

**Appendices**

\[
e'' = \frac{1}{2\pi \varepsilon_0 pf}
\]

\[
\mu_i = \frac{M_i^f}{akH_s M_p + b\lambda \xi
\]

\[
RL = 20 \log \left| \frac{Z_m - 1}{Z_m + 1} \right|
\]

\[
Z_{im} = \sqrt{\frac{\mu_r}{\varepsilon_r}} \tan h \left\{ f \left( \frac{2\pi}{c} \right) \sqrt{[\mu_r] \varepsilon_r} \left[ \frac{1}{ft} \right] \right\}
\]

\[
2S_{11 - OPEN} \pm S_{11} + E_{loss} = 1
\]

\[
t_m = \frac{nc}{4f_m \sqrt{[\varepsilon_r][\mu_r]}} (n = 1, 3, 5 \cdots)
\]

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