**Supporting Information**

**Recycling and utilization of coal gasification residues for fabricating Fe/C composites as novel microwave absorbents**

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Table S1.  Industrial analysis of CGR

|  |  |  |  |
| --- | --- | --- | --- |
| Mad (wt%) | Aad (wt%) | Vad (wt%) | FCad (wt%) |
| 2.06 | 7.76 | 9.70 | 80.48 |

Table S2.Chemical constituent analysis of CGR

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| Cad (wt%) | Had (wt%) | Nad (wt%) | St,ad (wt%) | Oad (wt%) |
| 81.19 | 2.67 | 1.15 | 0.80 | 4.37 |

**Characterization**

The X-ray diffraction (XRD, X’PertPRO, Philips Co., Ltd., the Netherlands) and Cu-K*α* radiation (*λ*=1.5406 Å) were applied to characterize the phase composition and crystalline state of the Fe/CGR composites. The compositional and chemical states analysis was performed on X-ray photoelectron spectroscopy (XPS, ESCALAB 250Xi spectrometer, Thermo Fisher), equipped with an Al Ka monochromator X-ray source. Raman spectra were recorded on a micro-Raman spectrometer (Renishaw) with the laser wavelength of 532 nm. Under thermogravimetry (TG) and Differential scanning calorimetry (DSC) (TGA/DSC 3+, METTLER TOLEDO, Switzerland), the thermal performance of Fe/CGR was examined under argon atmosphere with a heating rate of 10 °C/min. The field emission scanning electron microscopy (FESEM, S-4800) was used to observe the morphology of the sample, and the scanning electron microscopy (SEM) images were acquired. The crystal structure and microstructure of samples were recorded by a transmission electron microscope (TEM, JEOL JEM-2100). The chemical constituents of the Fe/CGR composites were measured by X Ray Fluorescence (XRF, PANalytical B. V., Holland). The magnetic properties were also measured by the Vibrating Sample Magnetometer (VSM, Lakeshore Model 7400) at room temperature. Meanwhile, the samples were uniformly mixed with 30 wt.% paraffin wax, which were molded into annular-shaped specimens with outer diameters of 7.00 mm and inner diameters of 3.04 mm. Electromagnetic parameters, including the complex permittivity and the complex permeability, were characterized by the vector network analyzer (VNA, Agilent 85050D) in the 2-18 GHz.

Fig. S1 shows the typical Raman spectra of Fe/CGR-0.5M, Fe/CGR-1.0M, and Fe/CGR-1.5M composites. It is well known that the D band (around1350 cm−1) and G band (around 1580 cm−1) are related to the lattice disorders in the sp2-hybridized carbon atoms and stretching vibrations of sp2 at carbon atoms, respectively. The former is usually forbidden in perfect graphite, whereas the latter corresponds to the highly oriented sp2 hexagonal graphitic lattice [1, 2]. Consequently, the degree of graphitization is generally assessed by the intensity ratio of ID and IG (ID/IG), and the graphitization degree is always strengthened with the decrease in ID/IG. As shown in Fig. S1, the values of ID/IG are slightly increased with the increase in concentration of Fe3+, due to the carbothermal reduced reaction will consume more carbon in the matrix, and then lead to the increase in disordered degree and defects in carbon [3]. Besides, the carbothermic reduction process not only consumes carbon but also significantly affects the state of carbon [4]. Apparently, the Raman spectrum of three samples has little difference on the phase structure, which is attributed to the fact that the solid-phase reaction only occurs at the interface. Therefore, the significant change in the degree of carbon graphitization is also mainly concentrated at the interface.



Fig. S1. Raman spectra of three samples with different concentration of Fe3+.

For further confirm the elemental chemical constituents of Fe/CGR composites. The XPS spectra are given in Fig. S2. From Fig. S2a, it can confirm the existence of C, Fe, Si, N, O elements on the surface of Fe/CGR composite. In Fig. S2b, there are three peaks at 284.8 eV, 286.0 eV and 288.6 eV, which can be attributed to C-C/C=C, C-O-C and O-C=O, respectively [5], further confirming that the reducing agent-carbon source is sufficient.



Fig. S2. XPS spectra for the Fe/CGR-800: full spectrum (a) and narrow spectrum of C (b).



Fig. S3. Hysteresis loops of the Fe/CGR composites.

Fig. S3 shows hysteresis loops of the Fe/CGR composites, it is obvious that the Fe/CGR composites display ferromagnetic behavior. Moreover, the saturation magnetization (*M*s) is dependent on the content of Fe. And the *M*s value of Fe/CGR-0.5M, Fe/CGR-1.0M, Fe/CGR-1.5M are 22.5 A∙m2/kg, 33.4 A∙m2/kgand 55.7 A∙m2/kg, which is caused by the different grain size and anisotropy constant of the loaded Fe phase [6-8].



Fig. S4. The $ε^{''}$-$ε^{'}$ plot of Fe/CGR-0.5M (a), Fe/CGR- 1.0M (b), Fe/CGR-1.5M (c).

Fig. S4 shows the Cole-Cole diagram of different samples. According to Debye relaxation theory, the relative complex permittivity $ε\_{r}$ can be expressed as [9-10]:

 $ε\_{r}=ε\_{\infty }+\frac{ε\_{s}-ε\_{\infty }}{1+j2πfτ}=ε^{'}-jε^{''}$ (1)

And the $ε^{'}$, $ε^{''}$can be expressed as:

$ε^{'}=ε\_{\infty }+\frac{ε\_{s}-ε\_{\infty }}{1+(2πf)^{2}τ^{2}}$ (2)

$ε^{''}=\frac{2πfτ(ε\_{s}-ε\_{\infty })}{1+(2πf)^{2}τ^{2}}$ (3)

Where $ε\_{s}$ is the static dielectric constant, $ε\_{\infty }$ is the optical dielectric constant, *f* is the frequency and $τ$ is polarization relaxation time. So according to equation (2) and equation (3), the relationship between $ε^{'}$ and $ε^{''}$ can be explained by the following equation:

$(ε^{'}-\frac{ε\_{s}-ε\_{\infty }}{2})^{2}+(ε^{''})^{2}=(\frac{ε\_{s}-ε\_{\infty }}{2})^{2}$ (4)

Based on this equation, one Cole-Cole semicircle represents one polarization relaxation process, and a semicircle between the curves of $ε^{'}$and $ε^{''}$ represents one Debye relaxation process. Fig. S4 shows the Cole-Cole semicircle of Fe/CGR composites with different Fe loading. Obviously, Fe/CGR composites show multiple Debye relaxation processes [11]. With the increase of Fe content, the radius of the semicircle gradually increases, and the polarization ability of the sample improves. Combined with SEM analysis (Fig. 5), the unique morphologies of CGR matrix provide abundant active sites for Fe/CGR composites, when electromagnetic waves enter into the microwave absorbents, there are a large number of positive and negative charges gathering in the Fe-CGR interface, resulting in strong interfacial polarization, which is conducive to the attenuation of electromagnetic waves, and thus improves the microwave absorption performance of the composite.



Fig. S5. The C0 value curves of Fe/CGR-800 composites.

It can be seen that the C0 values of the three samples fluctuate greatly in the whole testing frequency (Fig. S5). This shows that the main source of magnetic loss is not eddy current loss, so it can be inferred that the magnetic loss of the Fe/CGR-800 composites comes from natural resonance and exchange resonance [12-13].



Fig. S6. Frequency dependence of the loss tangents of Fe/CGR-800 composites.

Fig. S6 shows the frequency variation curves of the dielectric and magnetic loss factor for Fe/CGR-800 composites. As can be seen, the $tan δ\_{e} $value of each sample is larger than that of $tan δ\_{m}$ in the whole frequency range, indicating that the microwave attenuation is mainly from dielectric loss [14-15].

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