Supplemental Materials

Mass Production of Graphene Materials from Solid Carbon Sources using A Molecular Cracking and Welding Method

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Fig.S1 shows TGA curves of the raw and the purified graphene samples. As-obtained samples presented an onset degradation temperature around 430 °C and a final degradation temperature around 660 °C. Graphene samples usually present thermal oxidation ranging 450–700 °C. Amorphous carbon presents a lower oxidation temperatures (<400 °C). Based on TGA results, the purity of the graphene products in this work is between 92%-96%. Beside the graphene structures, there are 3-5% amorphous carbon and 0.3%-3% catalyst metals.

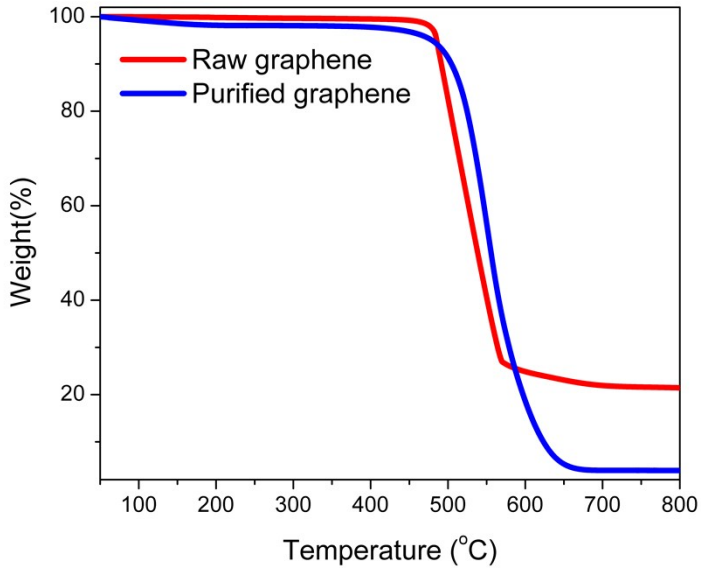

Figure 1S TGA of graphene samples

Fig.2S showed XRD patterns of the 10 % Fe/ lignin mixtures after thermal carbonization at different temperatures. Based on the XRD pattern, iron was reduced into FeO at 500 °C, iron was reduced to α-Fe at 600 °C, however, these iron nanoparticles were too active to stay in the air, and they were burnt to iron oxides when the sample bottle was open. When the temperature was increased to 700 °C, the α-Fe was partly transformed to γ-Fe, and these nano particles were stable in the air (XRD pattern was not shown here), this might be due to iron particles were encapsulated in carbon shells. Further increasing the temperature to 750 °C, most of the iron was converted to Fe_3C. As the temperature was up to 900 °C, the material mainly showed the strong Fe_3C diffraction peaks. When the temperature increased to 950 °C, the presence of the diffraction peak at around 26.5° for the product materials, besides of the Fe_3C peaks, indicated that graphene material is formed. A stronger graphene diffraction peak was observed for the 1000 °C sample.

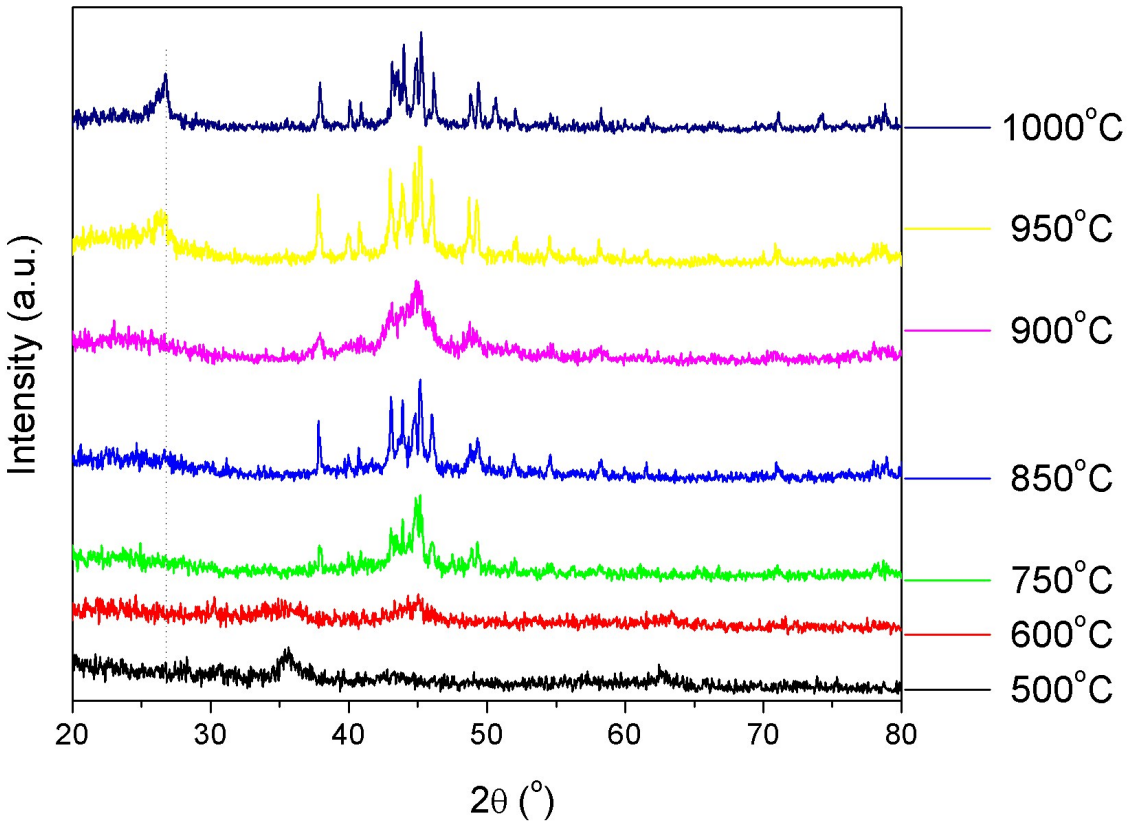

Figure 2S XRD patterns for carbonized 10 % Fe- lignin mixtures at different temperatures

Fig.3S illustrated the SEM images of Fe-lignin samples thermally treated at different temperatures. SEM images showed that nanoparticles were the main products for samples in the range of 500 to 800 ºC, while nanoplates composed of the sample at 1000 ºC.

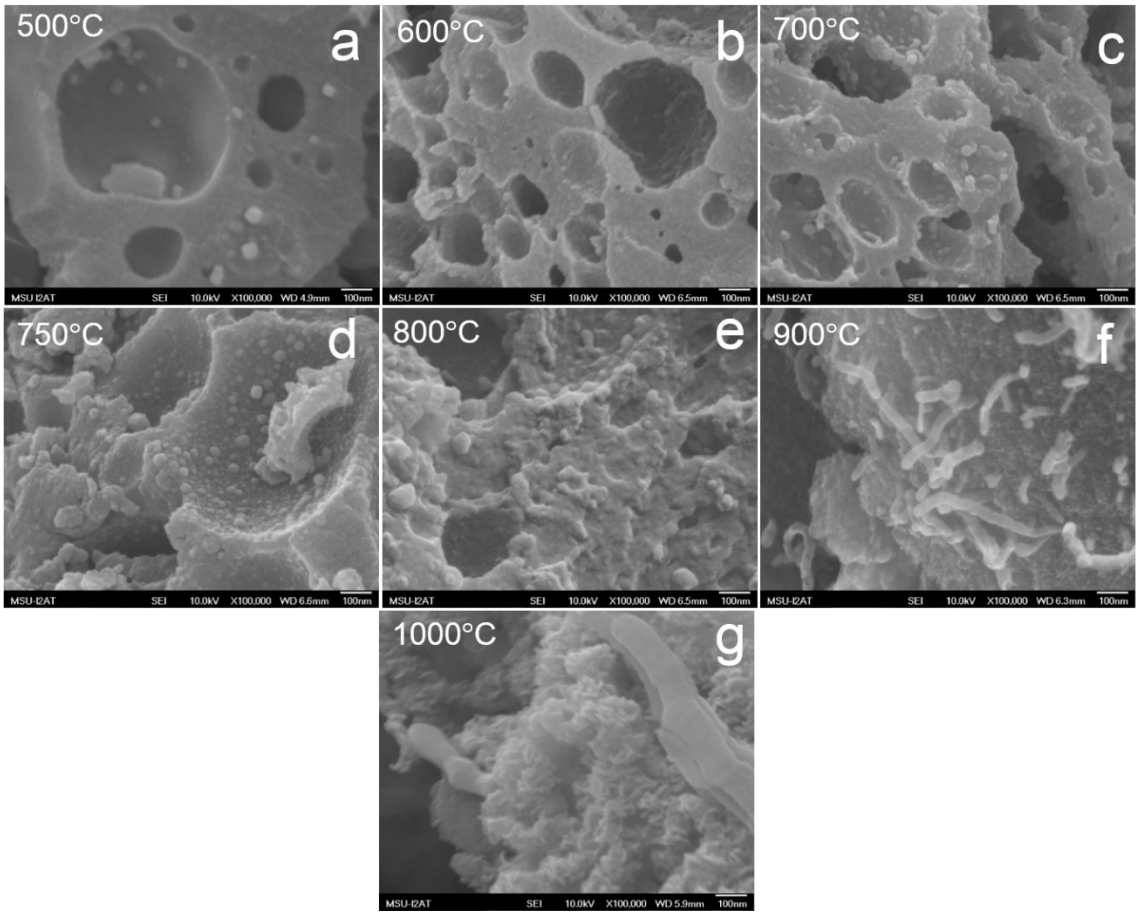

Figure 3S SEM images of the samples treated at different temperatures.

HRTEM images of the samples treated at different temperatures were showed in Fig. 4S. Fig.4Sa indicated that the γ-Fe_2O_3 nanoparticles were tightly encapsulated in amorphous carbon for the sample produced at 500 ºC. Fig.4Sb illustrated the carbon encapsulated α-Fe nanoparticles at 600 ºC. Fig.4Sc showed iron nanoparticles were embedded in 2-3 layers of graphene at 700 ºC. As the temperature reached to 750 ºC, iron nanoparticles

became to multilayer graphene-encapsulated iron nanoparticles (MLGEINs) with a light color shell (corresponding to graphene layers) and dark core (corresponding to the iron core) as shown in Fig.4Sd. The size of iron particles increased significantly to 30-80 nm (Fig.4Sf) when the temperature increased to 900 ºC, and the core-shell structures were cracked, graphene shells were skinned off the iron cores. Fig.4Sg demonstrated the formation of graphene materials and the separation of iron particles from graphene structures.

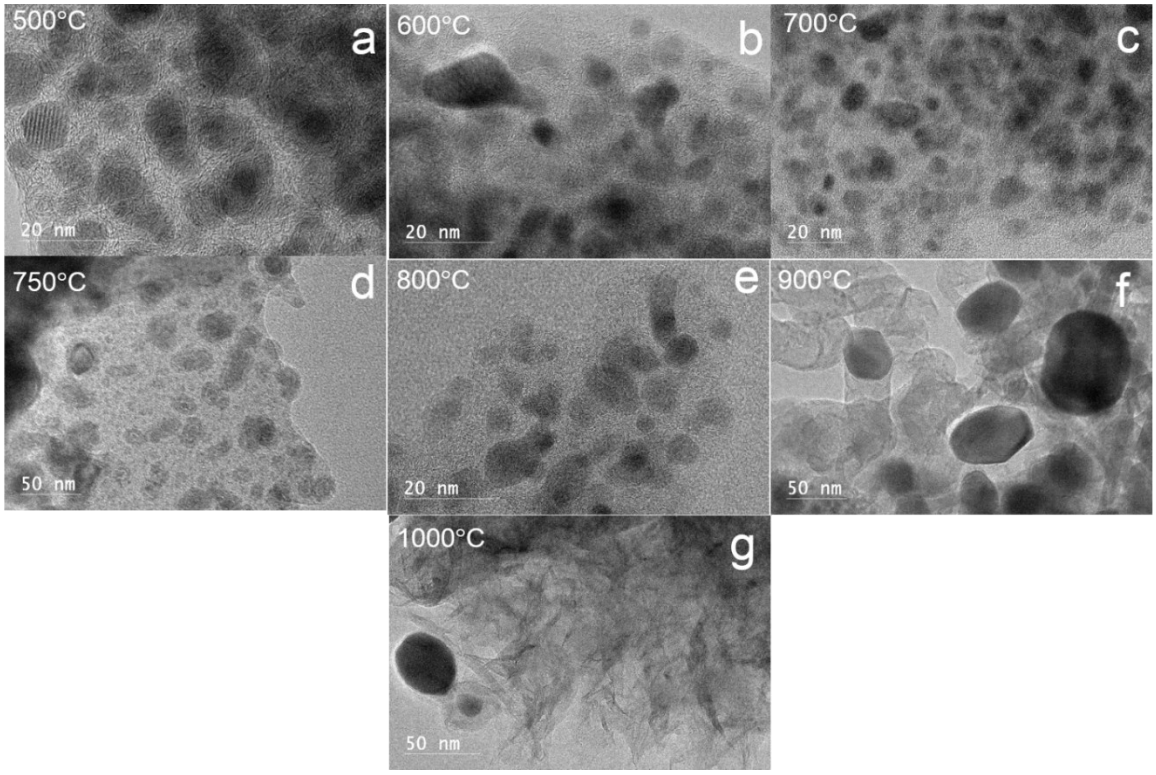

Figure 4STEM images of the samples treated at different temperatures.

The effects of different thermal treatment times, including 0, 0.5,1, 3, and 5 hours, on graphene products were evaluated. XRD, SEM, and TEM data was obtained for the resulting graphene-based materials (Figs.5S-7S). The results show that different morphologies of graphene-based materials may be formed by tuning thermal treatment time, including those comprising graphene chains, fluffy graphene, curved graphene sheets and flat graphene sheets.

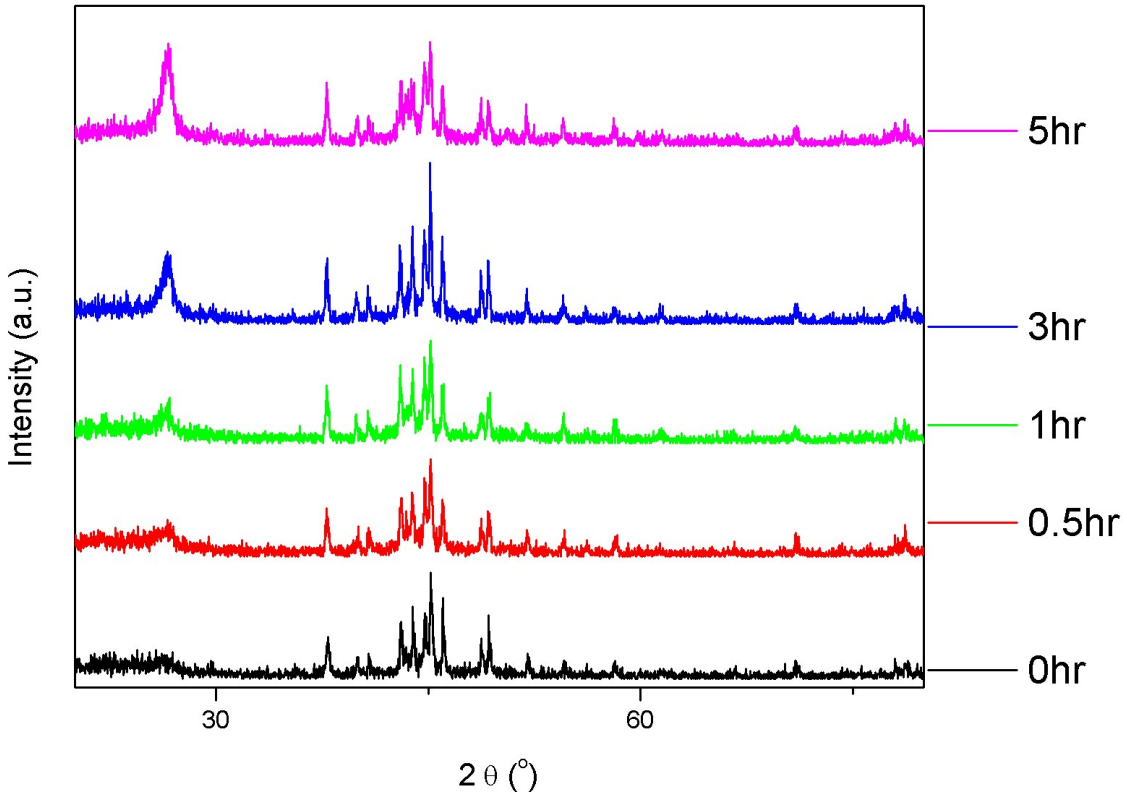

Figure 5S XRD patterns of the products with different heating times.

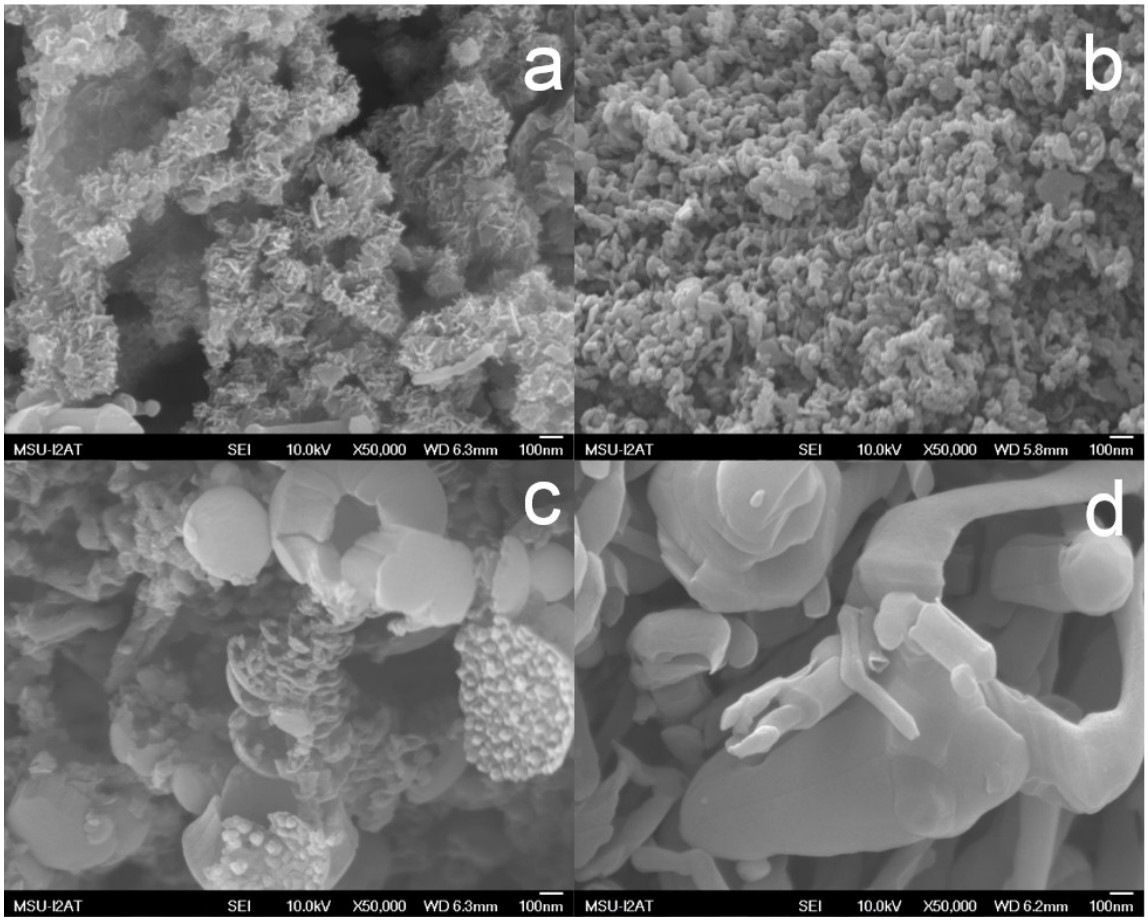

Figure 6S SEM images of the products with different heating times.

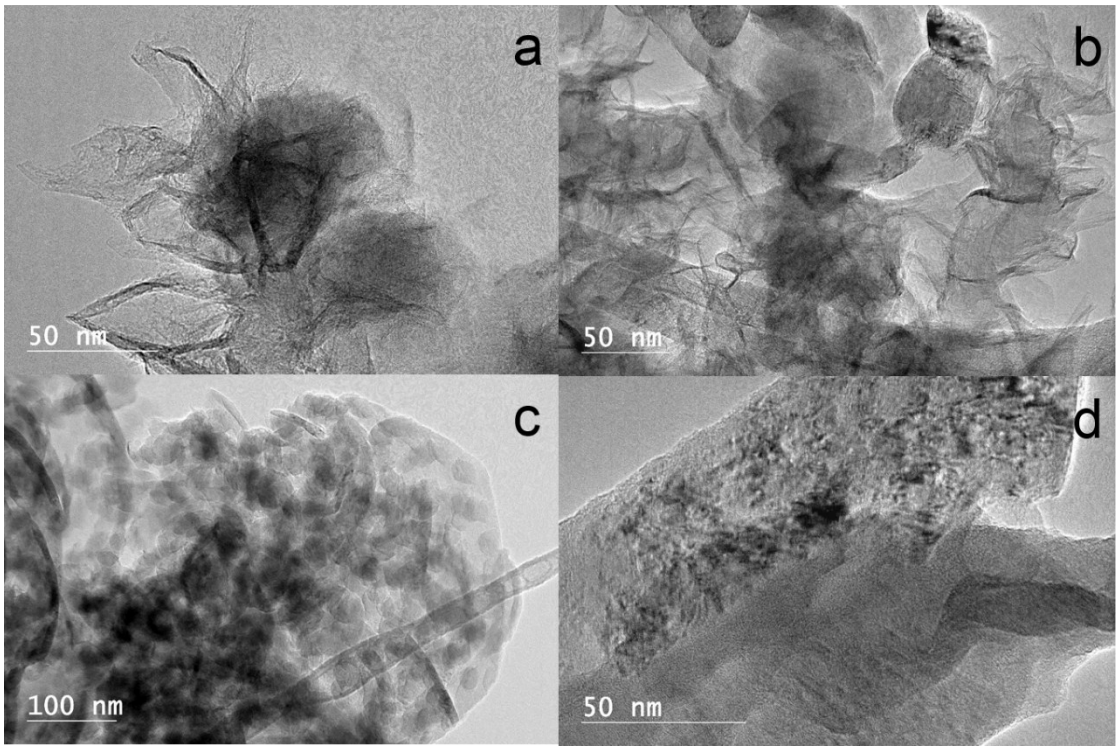

Figure 7S TEM images of the products with different heating times.

Fe-lignin precursor sample was separated to different size: ≤ 44, 44-125, 125-177, 177-250, 250-420 μm. Figs.8S-9S demonstrated XRD, SEM, and TEM results of the products with different precursor particle size. The results showed the geometry size and thickness of the graphene materials decreased with increasing of precursor particle size. XRD, SEM, and TEM data was obtained for the resulting graphene-based materials. The SEM data is shown in Fig. 10S, along with SEM data showing the effects of heating time. The results show that different morphologies of graphene-based materials may be formed by tuning precursor particle size and heating time, including those comprising graphene chains, fluffy graphene, curved graphene sheets and flat graphene sheets. In general, longer heating times and smaller precursor particle size can produce larger sized graphene sheets.

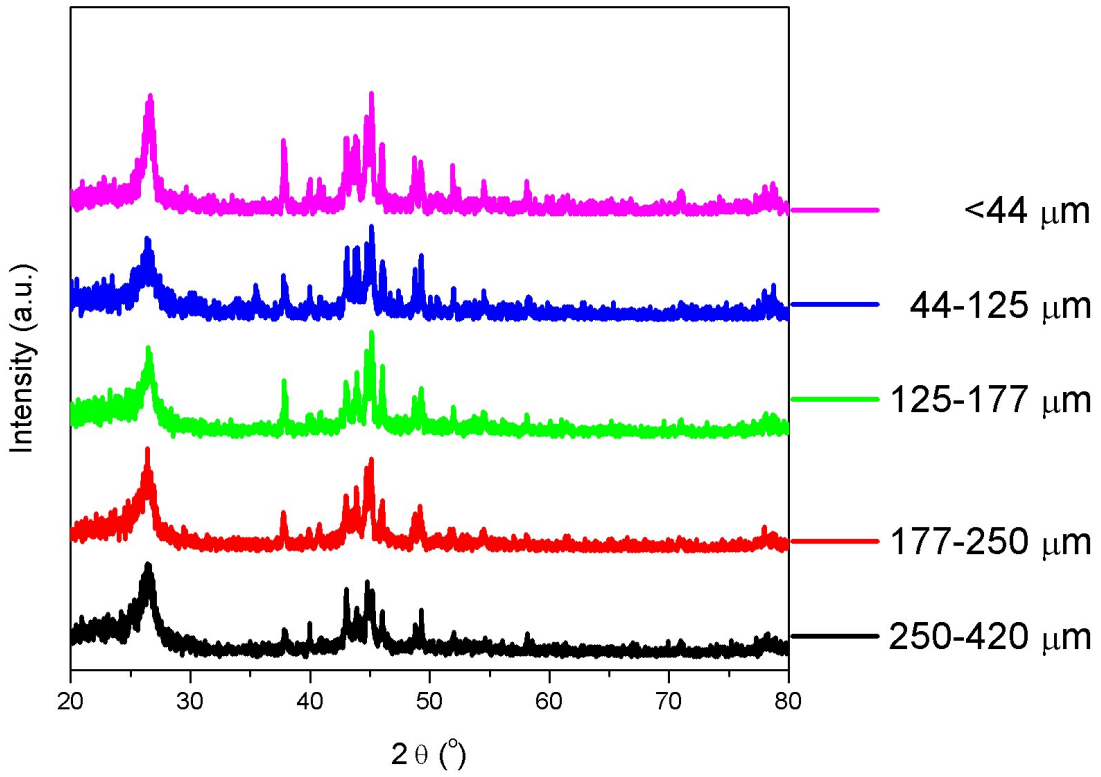

Figure 8S XRD results of the products with different precursor particle size.

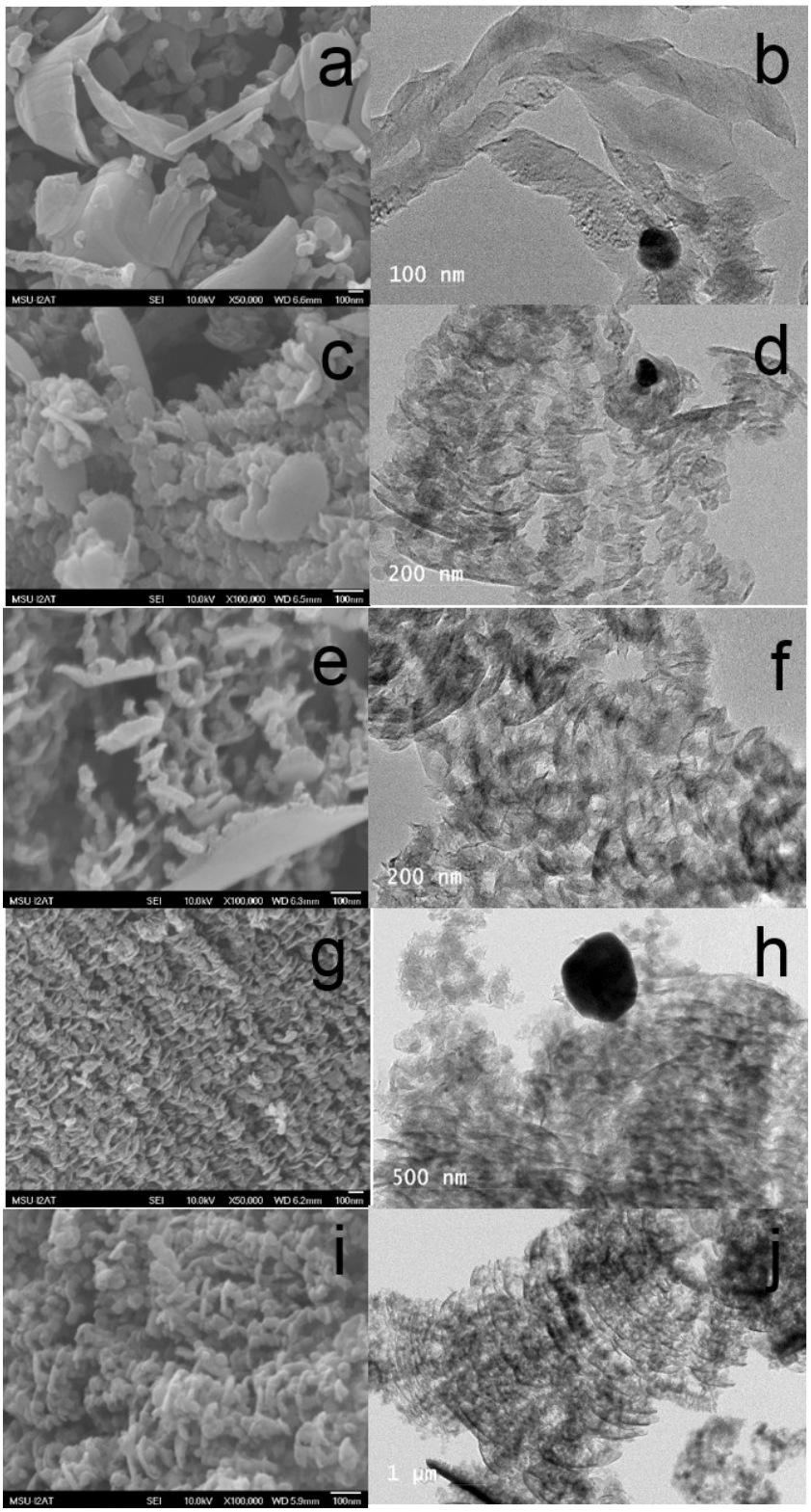

Figure 9S SEM and HRTEM images of the products with different precursor particle size.

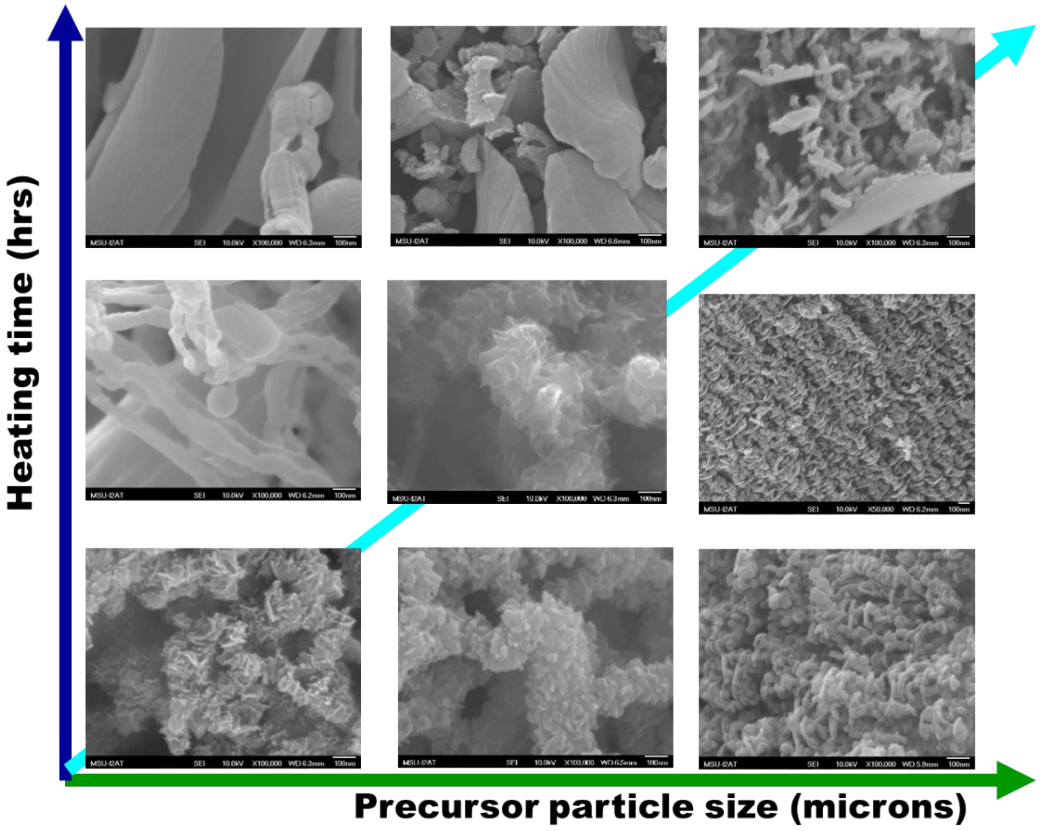

Figure 10S Effect of precursor particle size and heating time on the product structures and morphology (SEM images).

The idea welding temperatures were found at least 1,000°C, with heating time ranged from 0.5-1 hour. The selected welding gases were methane (CH4), and natural gas (NG). The optimized Fe-lignin precursor particle size was examined between 125 to 250 microns (μm).