

Supplementary Information

Improvement of the anisotropic thermal conductivity of h-BN filled epoxy composites by changing the filler shape to spherical

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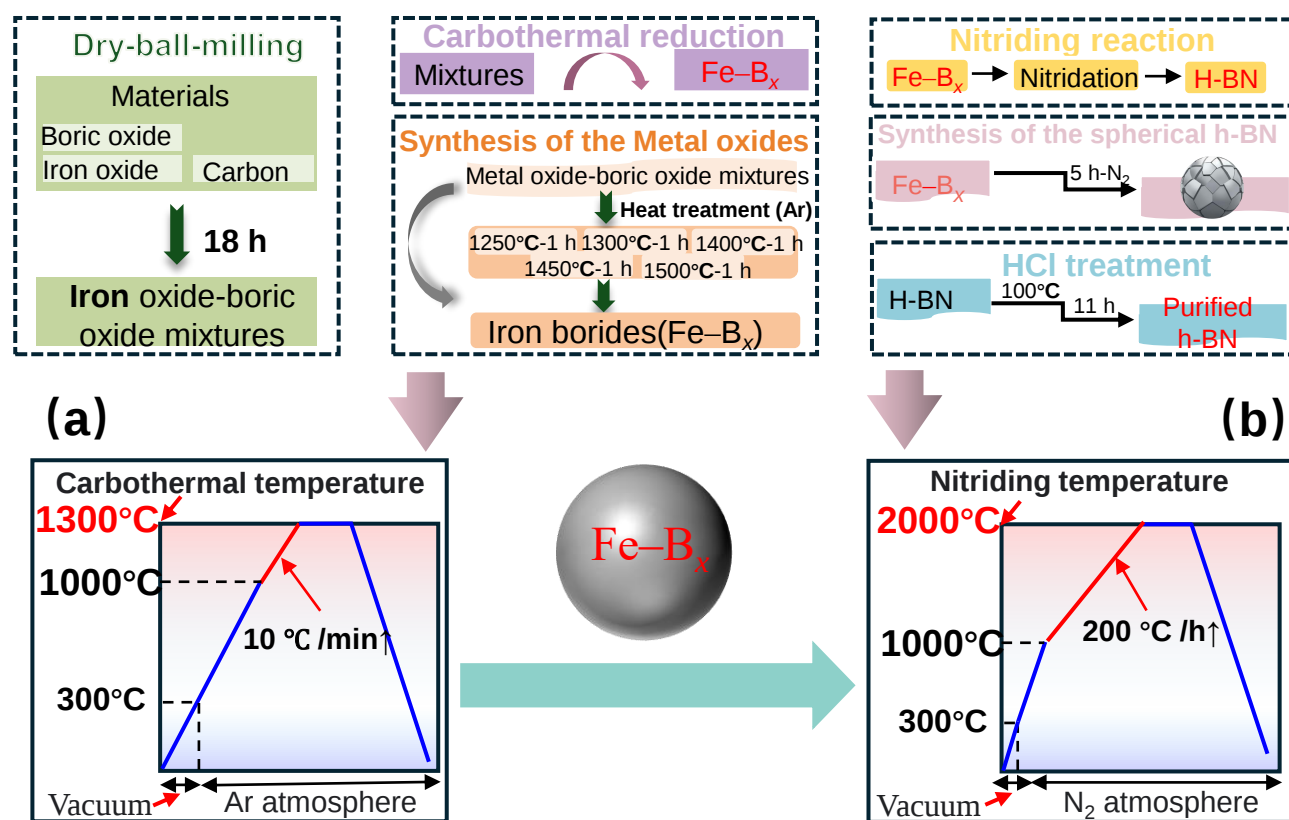


Figure S1. Synthesis of the spherical h-BN. **(a)** The heating rate of the carbothermal temperature and **(b)** nitriding temperature.

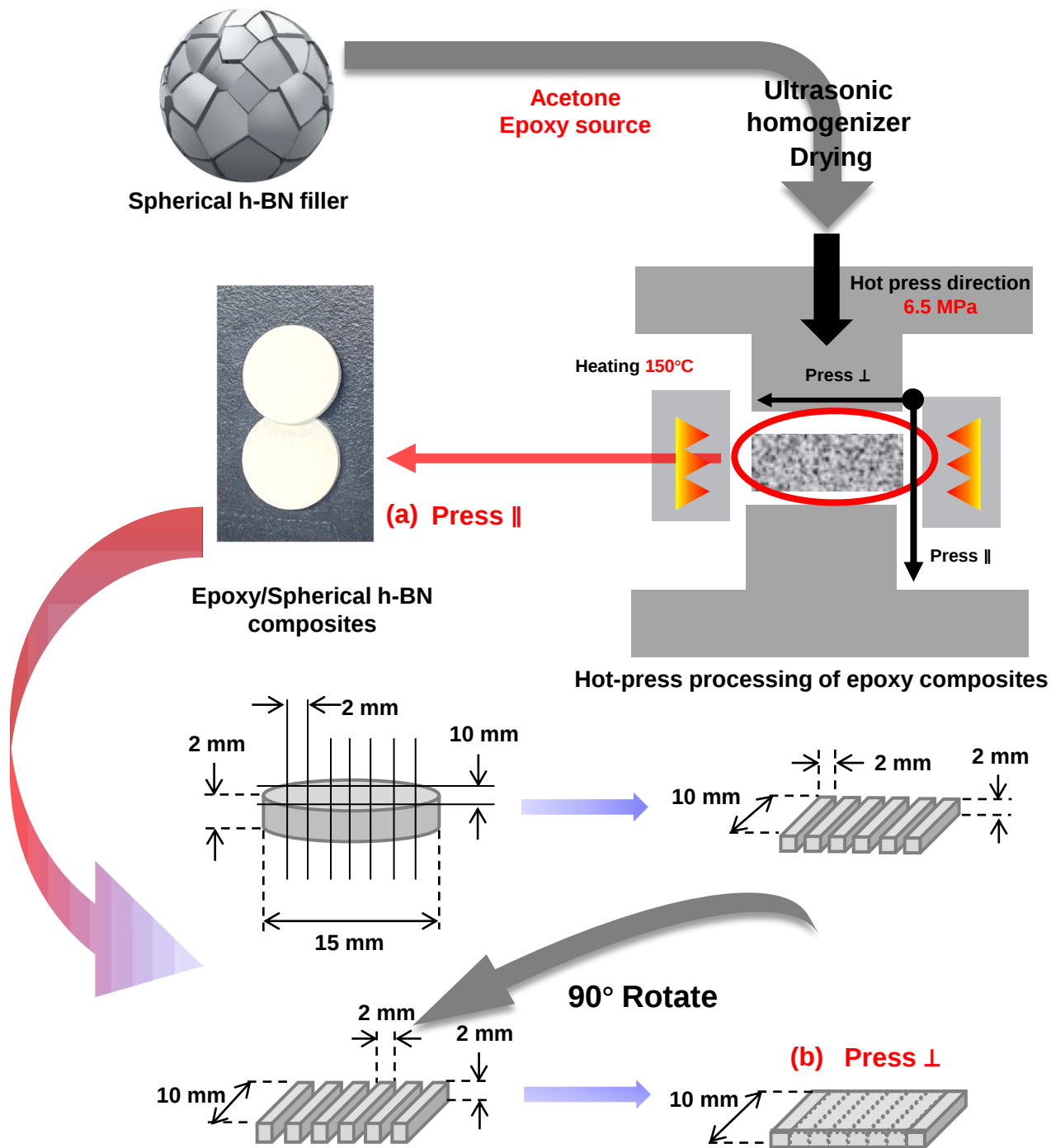


Figure S2. Prepare for thermal conductivity of the composite materials **(a)** parallel and **(b)** perpendicular to the hot-press direction

A sample of the hot-pressed composite material, with a diameter of 15 mm and thickness of approximately 2 mm, was prepared and ground into a disc-shaped sample with a thickness of 1 mm. This sample is used to measure the thermal diffusivity along the hot-pressing direction (denoted as press //), as shown in Figure S2(a), which is perpendicular to the preferred orientation of the platelet boron nitride particles. The thermal diffusivity perpendicular to the direction of hot pressing (denoted as press ⊥) was evaluated by measuring laminated samples.¹ This process involved cutting the disk samples, a 15 mm in diameter, into rectangular bars, each 2 mm wide, as shown in Figure S2(a), and these bars were then oriented vertically, to ensure that the cut side was positioned upwards and downwards. Subsequently, the cut rectangular bars were bonded for measurement (Figure S2(b)). These bonded samples, initially with 2 mm thick, were ground to form 1 mm thick squares measuring 10 mm × 10 mm. Finally, the tests were conducted at 25 °C. Moreover, the apparent densities of the composites were determined by measuring their volume and weight at 25 °C.

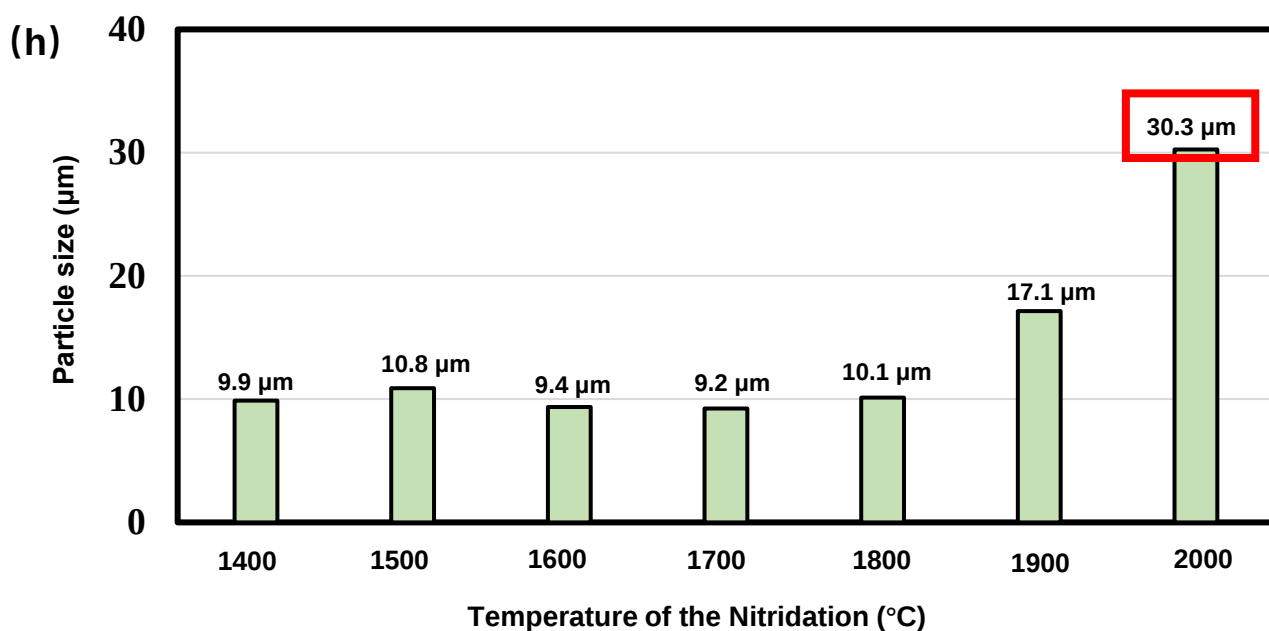
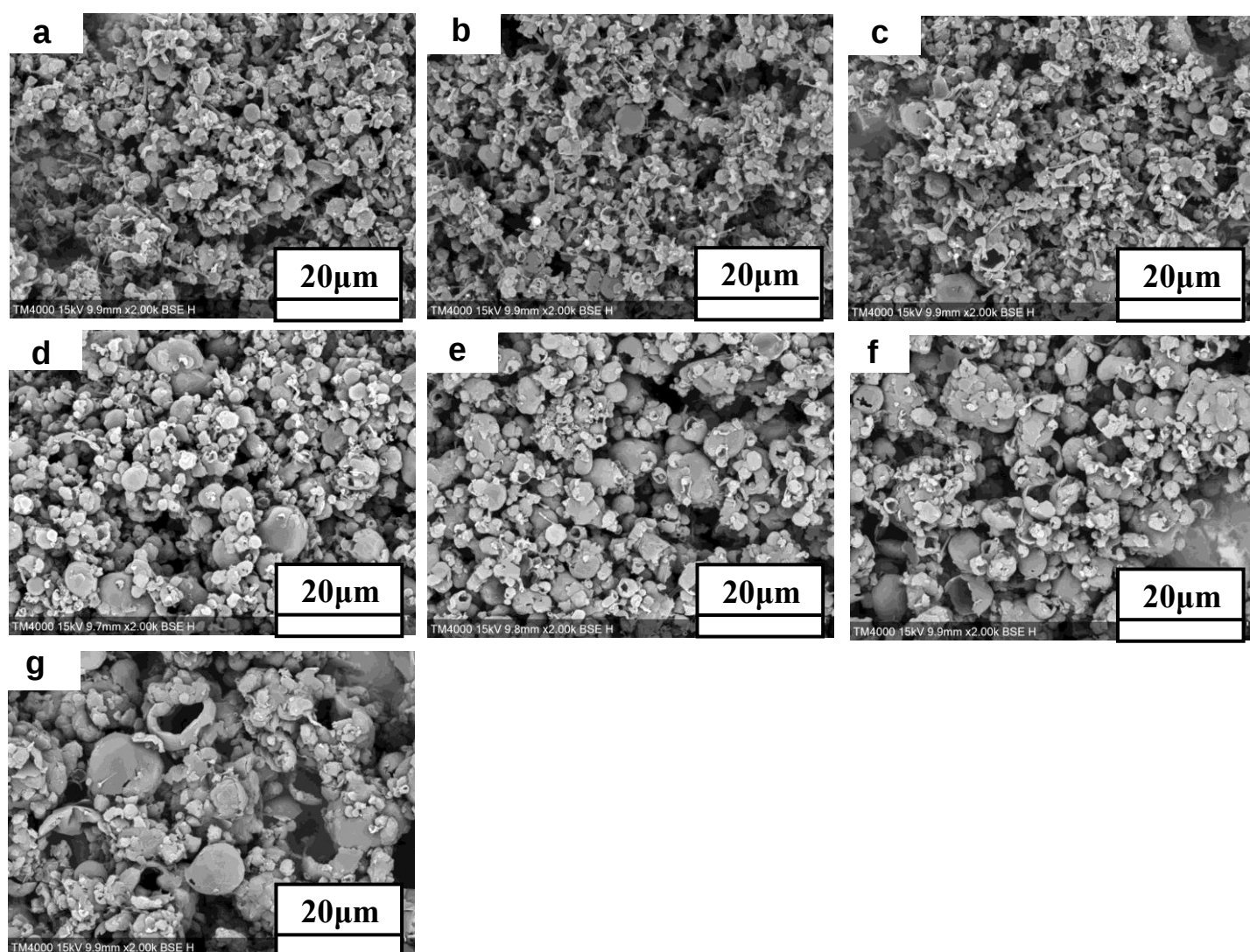


Figure S3. SEM images of the spherical h-BN through different nitriding temperature (a) 1400°C; (b) 1500°C; (c) 1600°C; (d) 1700°C; (e) 1800°C; (f) 1900°C; (g) 2000°C for 1 min based on the Fe-B_x which by carbothermal reduction of Fe₂O₃-B₂O₃ mixtures for at the 1300°C 1 h; (h) Effect of the temperature of nitridation on the particle size of obtained h-BN filler by the raw materials Fe-B_x which was synthesized by the carbothermal reduction at 1300°C for 1 h.

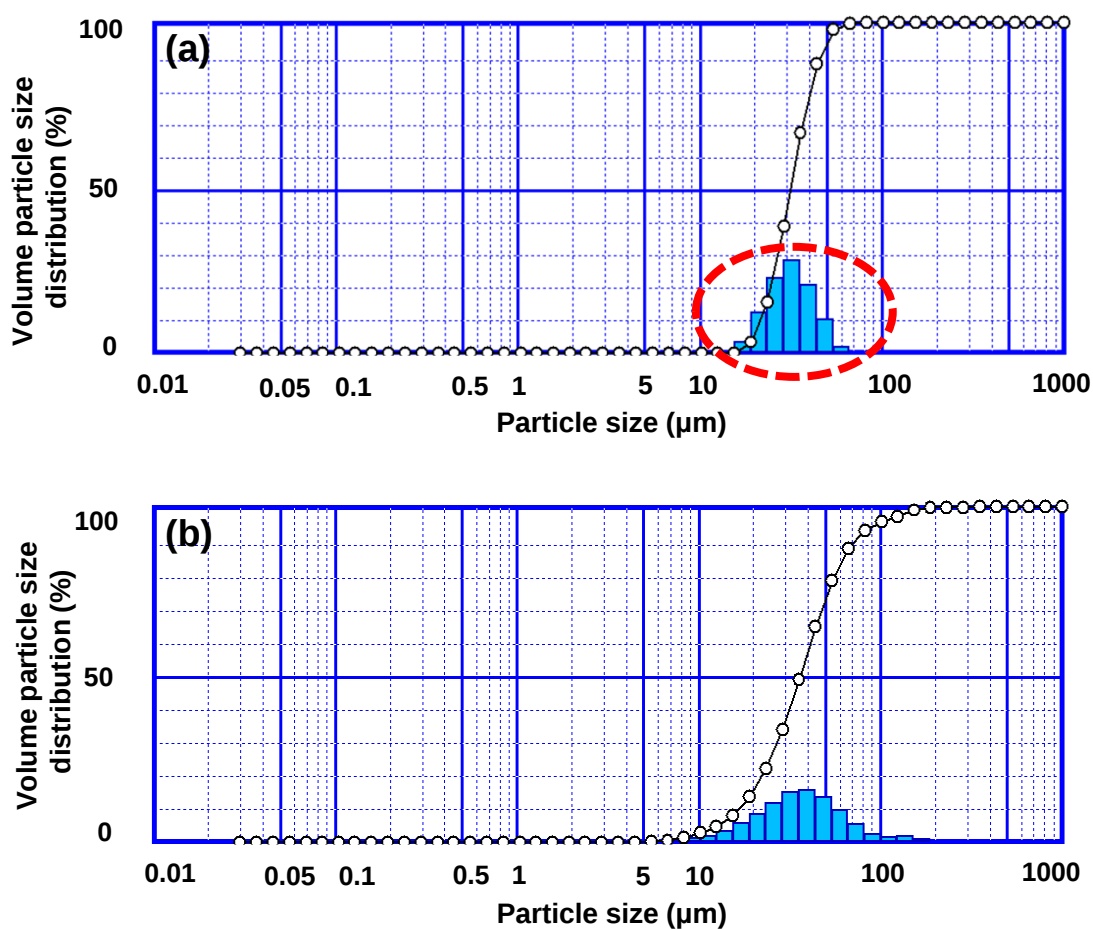


Figure S4. The particle size distribution of the synthesized spherical h-BN at 2000°C for 5 h based on Fe-B_x which was synthesized by the carbothermal temperature at **(a)**1300°C, **(b)**1400°C for 1 h.

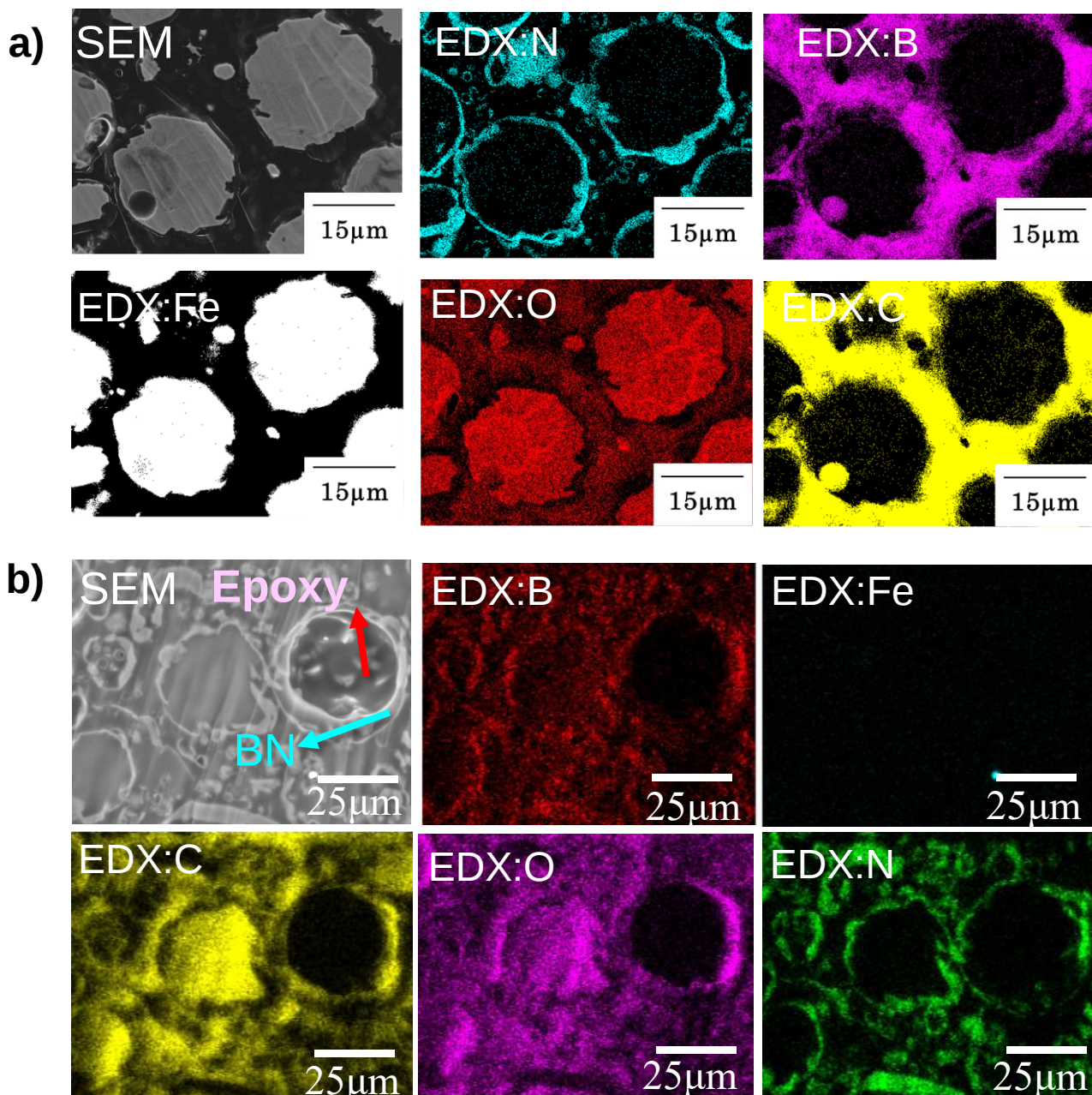


Figure S5. EDX of Epoxy/spherical h-BN composites with spherical h-BN fillers (a) before washed and (b) after washed with 6N-HCl solution at 100°C for 11 h based on the 1300°C of the carbothermal reduction of $\text{Fe}_2\text{O}_3\text{--B}_2\text{O}_3$ mixtures for 1 h indicate the existence of B, Fe, N, C and O, respectively.

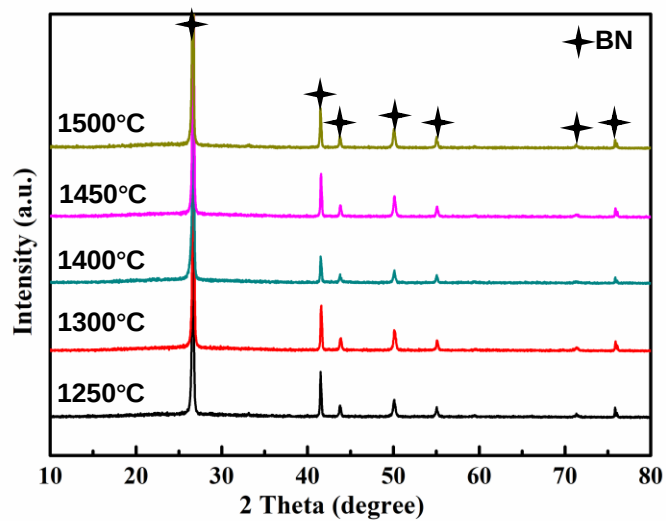


Figure S6. XRD patterns of the synthesized spherical BN based on the Fe-B_x before washed with 6N HCl solution at 100°C for 11 h on the different temperature of carbothermal reduction of $\text{Fe}_2\text{O}_3\text{-B}_2\text{O}_3$ mixtures for 1 h.

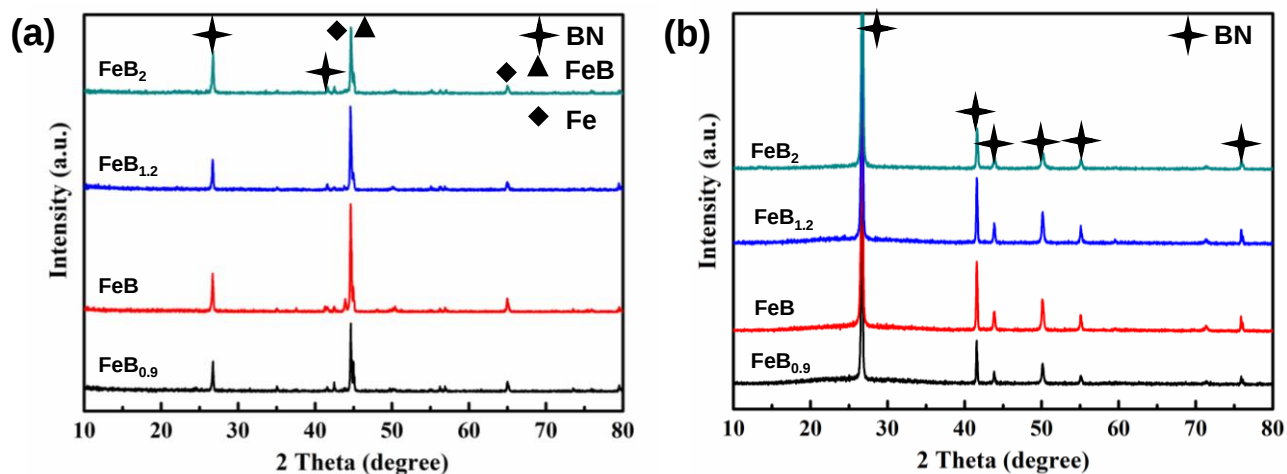


Figure S7. XRD patterns of the spherical h-BN fillers synthesized at 2000°C for 5 h by the spherical Fe-B_x which was synthesized at 1300°C for 1 h based on the different molar ratios between F₂O₃ and B₂O₃ **(a)** before washed with 6N-HCl solution, **(b)** after washed with 6N-HCl solution.

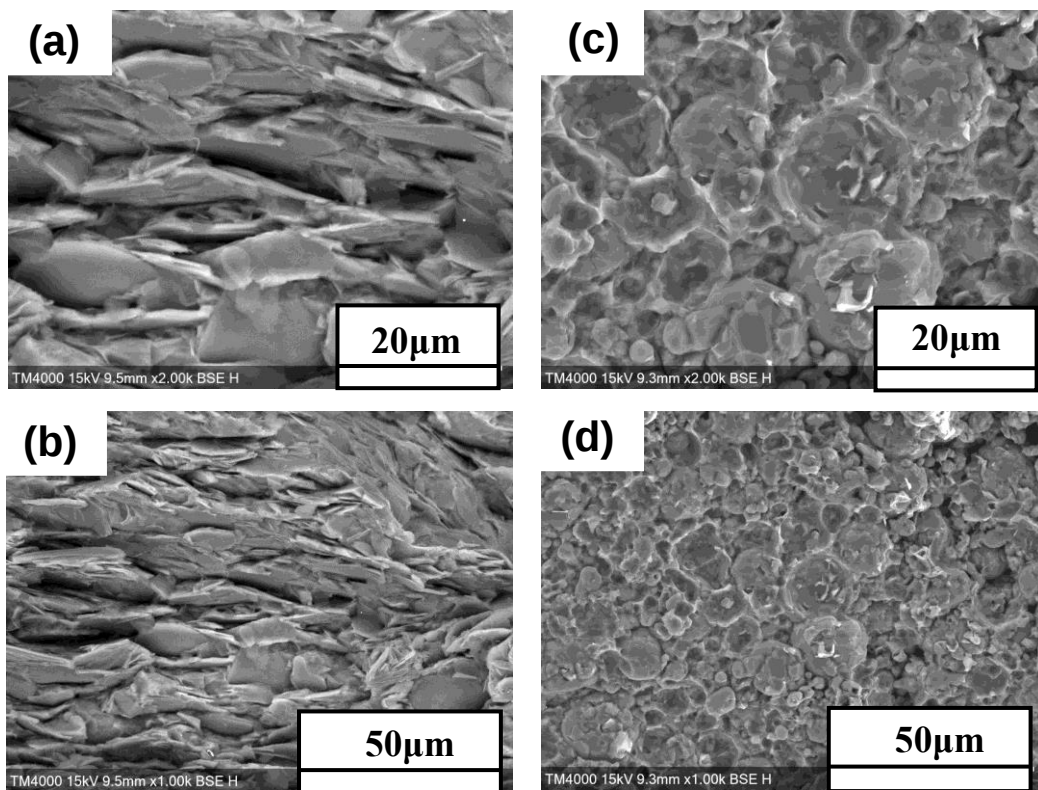


Fig. S8. Section views of fractured surfaces in epoxy composites containing 45vol% of the **(a, b)** the commercial platelet h-BN **(c, d)** the spherical h-BN filler.

Reference

1. Kusunose, T., Y. Uno, Y. Tanaka, T. Sekino. Compos Sci Technol. 2021, 208, 108770.