Supporting Information

New strategy for Fabrication of High Performance Reactive Microspheres via Energetic Polyelectrolyte Assembly

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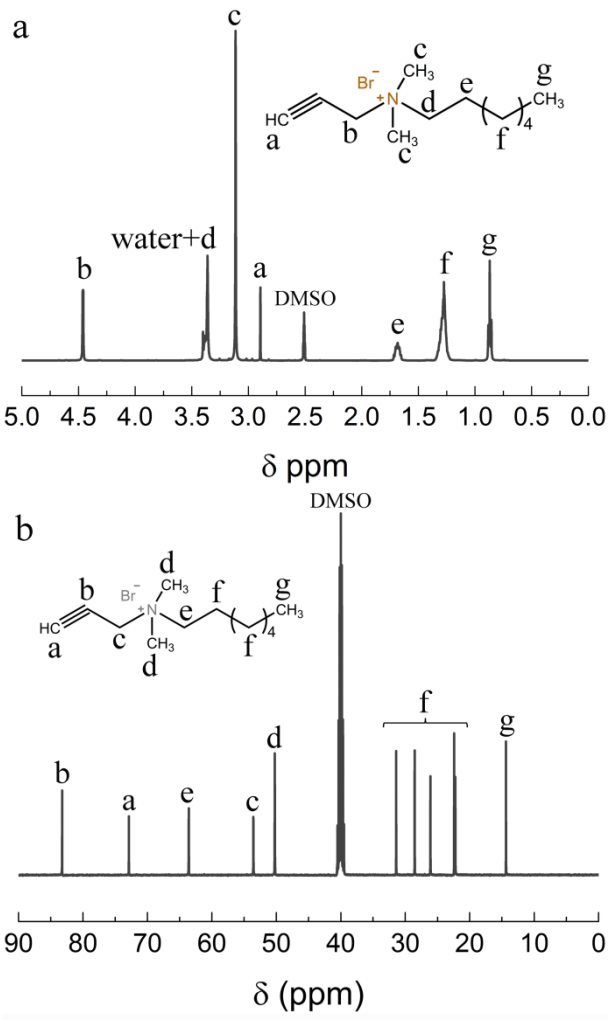

Fig. S1 ^{1}H-NMR(a) and ^{13}C-NMR(b) of

N,N-dimethyl-N-prop-2-yn-1-ylheptan-1-ammonium bromide

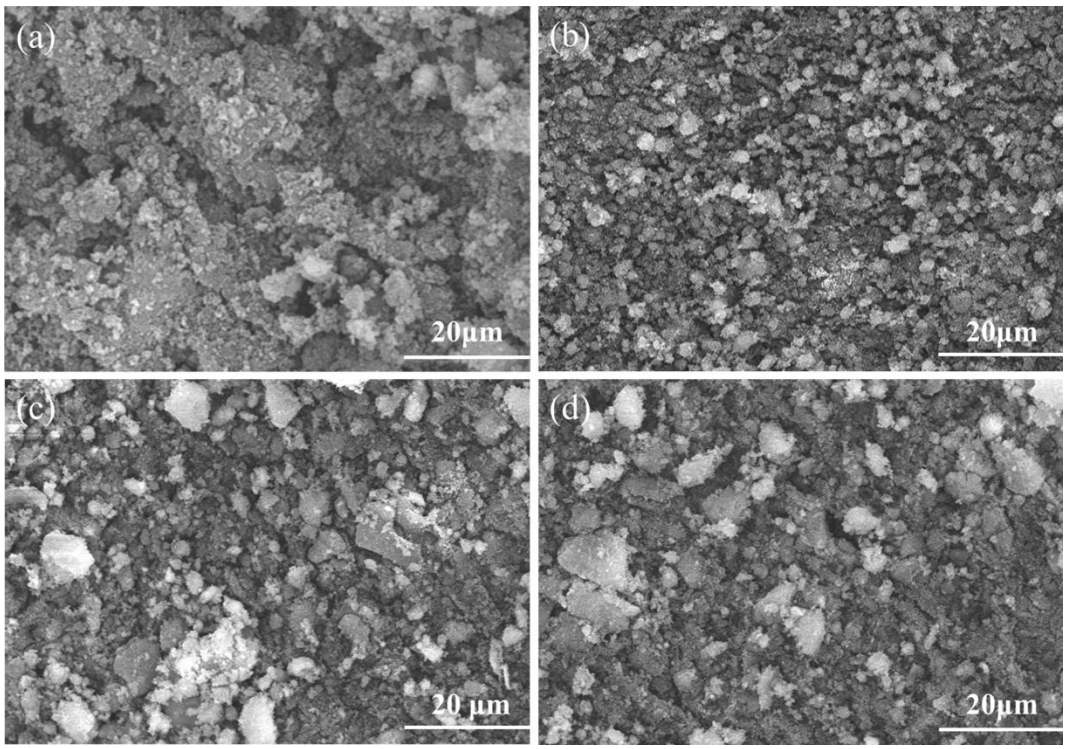

Fig. S2 SEM images (lower magnification) of assembled reactive Al/Fe_2O_3 microsphere with GEPEs content of 5 wt.%(a), 10 wt.%(b), 15 wt.%(c), and 20 wt.%(d).

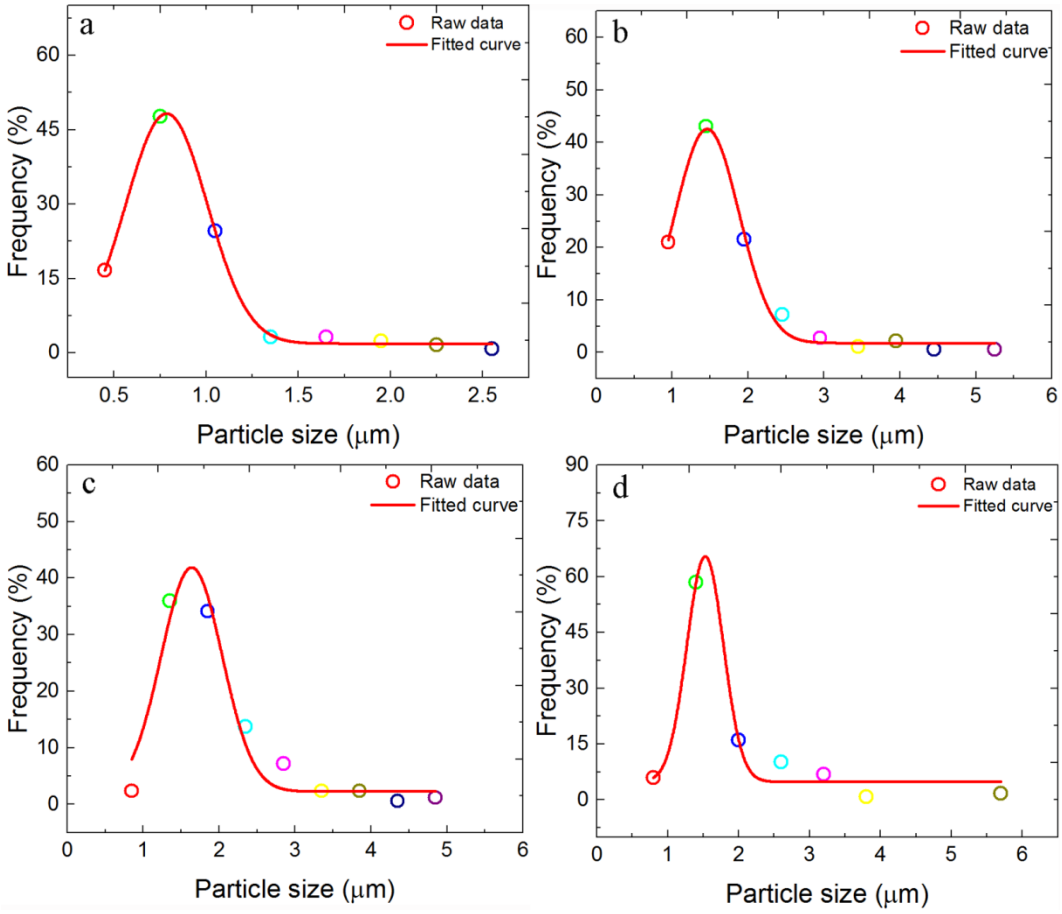

Fig. S3 The dimensions of five hundreds of assembled particles at different GEPEs-1.0 content of 5 wt.% (a), 10 wt.% (b), 15 wt.% (c), and 20 wt.%, are counted and the distribution are fitted by Gauss function.

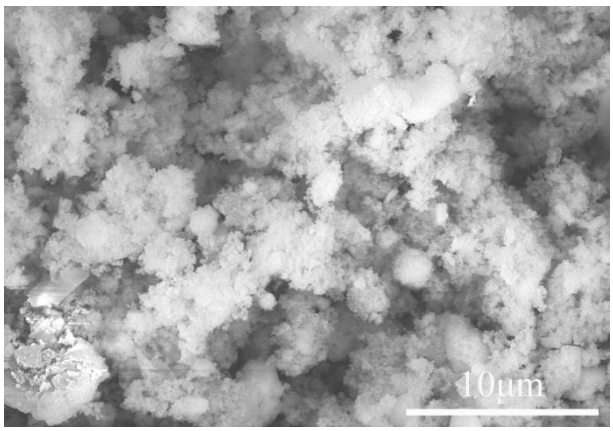

Fig. S4 SEM image of assembled NEMs using GEPEC-0.5 with content of 10 wt.%.

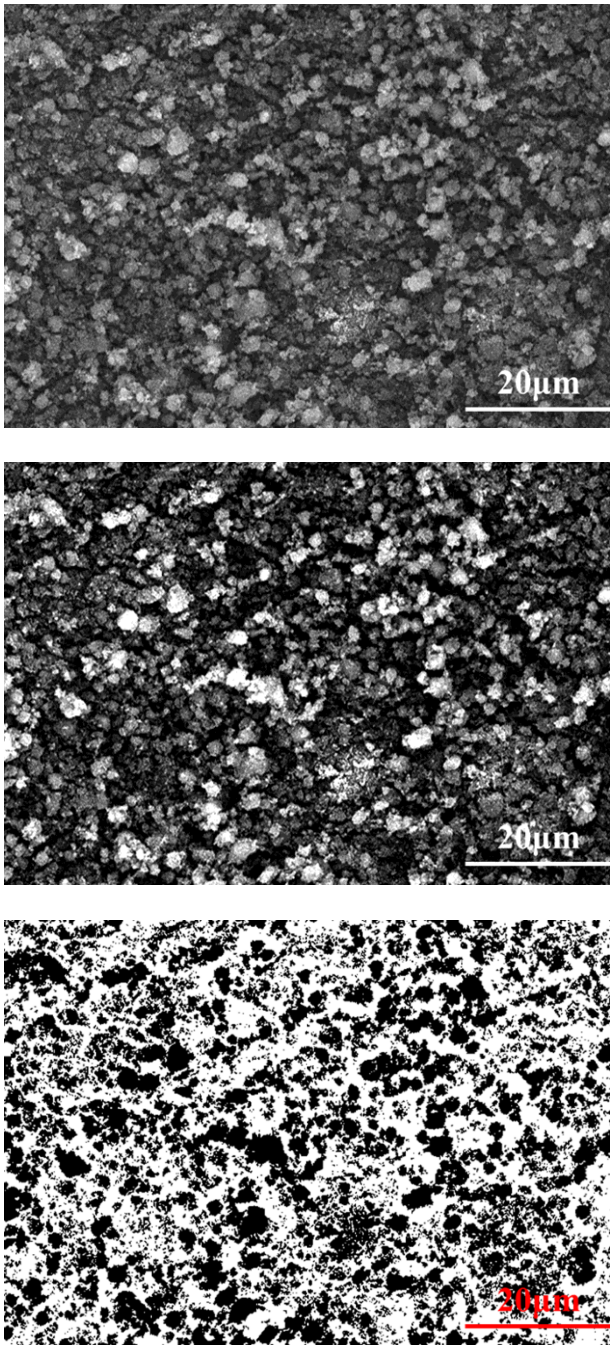

Fig. S5 The upper one is the original SEM image (Fig. S2b). The middle one is the image after adjusting brightness/contrast process. The lower one is the image analyzed by ImageJ.

By adjusting the image threshold, the boundaries of large sphere are sharpened against the background. The threshold is considered to be 0.9μm, a rough estimate, which is just below the smallest size in the statistical curve of the microsphere diameter (Fig. S3b). We assume that the white background is comprised of small particles with diameter below 0.9μm, which are not forming into microspheres during assembly process. The dark particles are the assembled microsphere with average diameter of 1.5 μm (Fig. S3b). The percentage of the dark area is 0.46. We define that the whole area of this image is A, so the white area is 0.54A and the dark area is 0.46A. It assumes that the area are comprised of cross-sectional area of spherical particles, then, the numbers of the assembled microspheres and unassembled particles could be calculated out. Next, the volume ratio can be obtained. At last the assembly ratio is acquired. The data are in the Table S1 below. Moreover, as the average dimension of the small particles (D_S) in the background area can't be known exactly, the estimated value, 0.9μm, is bigger than the true value, then, the assembly ratio is smaller than the actual value doubtlessly.

Table S1 The data of analysis of assembly ratio in detail

Average diameter of large particles	D_L	1.5 μm
Average diameter of small particles	D_S	0.9 μm
Total area composed of the dark particles	A_L	0.46A
Area covered by the white region	A_S	0.54A
Total number of 1.5 μm particles in the dark area	N_L	$4*0.46A/(\pi*1.5^2)$
Total number of 0.9 μm particles in the white area	Ns	$4*0.54A/(\pi*0.9^2)$
Volume Ratio: $V_L/V_S = (N_L*D_L^3) / (N_S*D_S^3)$		≈1.42
Assembly Ratio		58.6%

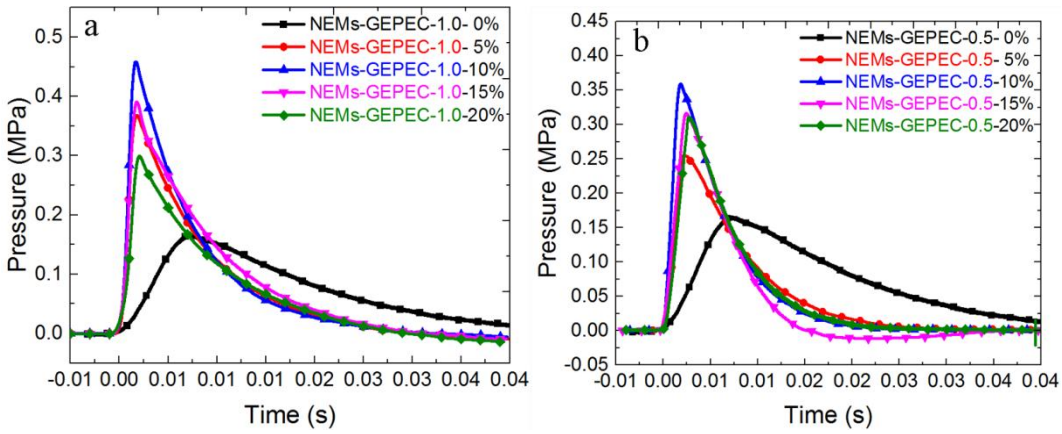

Fig. S6 Typical pressure-time profiles of assembled Al/Fe_2O_3 NEMs using GEPEC-1.0 (a) and GEPEC-0.5 (b) with different content respectively.

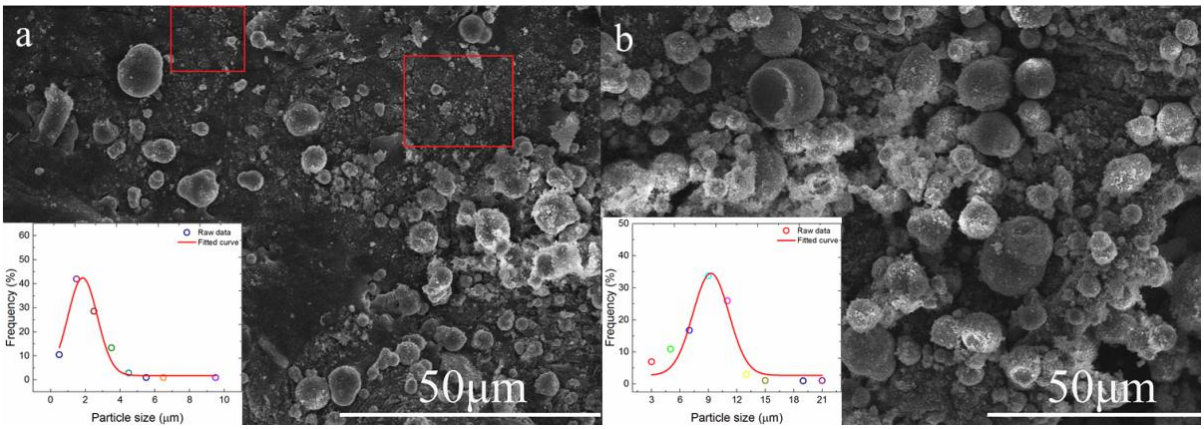

Fig. S7 SEM image of combustion products of assembled reactive Al/Fe_2O_3microsphere (a) and Al/Fe_2O_3 NEMs prepared by sonication (b).

It's important to note that the combustion products with larger diameters of 4~10 μm are also observed in the case of the assembled Al/Fe_2O_3 microsphere (a) but they are in small populations. A large amount of particles with smaller size are located in the area marked by red square (a).