Supporting Information

Pushing the limits of energy performance in micron-sized thermite: core-shell assembled liquid metal modified-Al@Fe_2O_3 thermites

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A. TG experiment of GLM-Al and ref-Al.

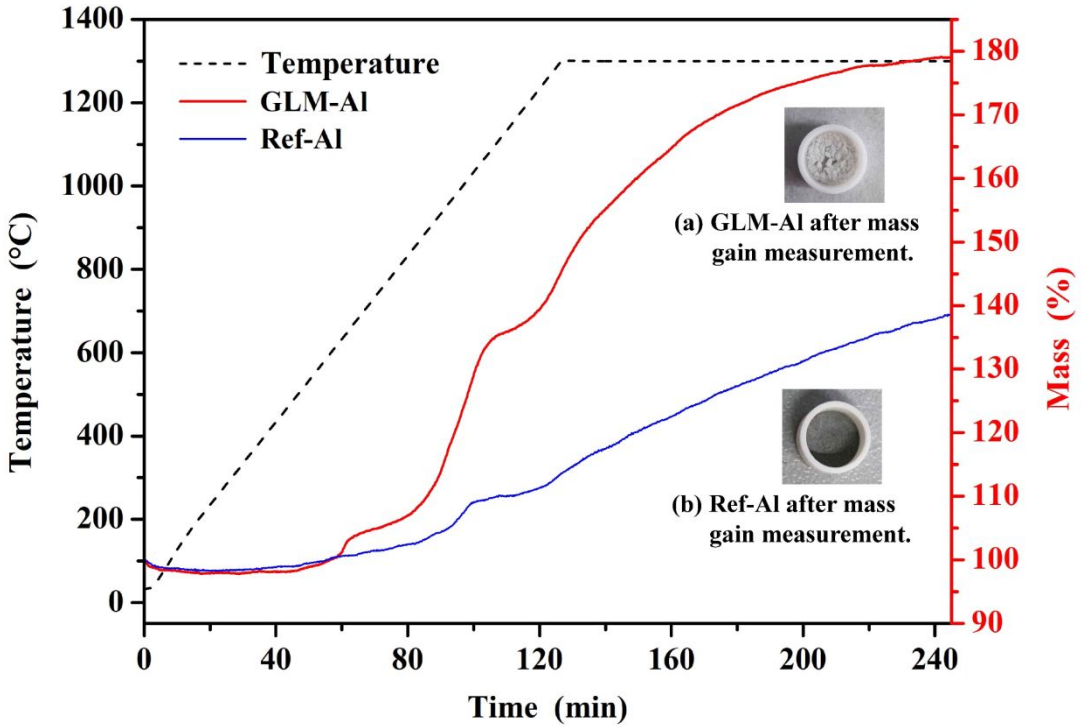

Figure S1. TG curves of GLM-Al and ref-Al under oxygen atmosphere.

B. Schematic of the ignition system.

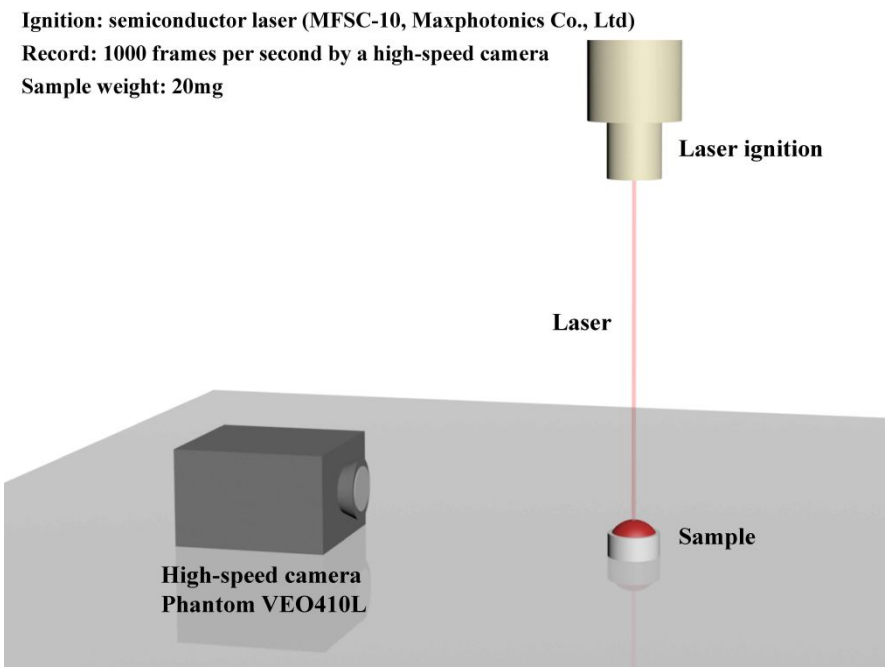

Figure S2. Schematic of the laser ignition system.

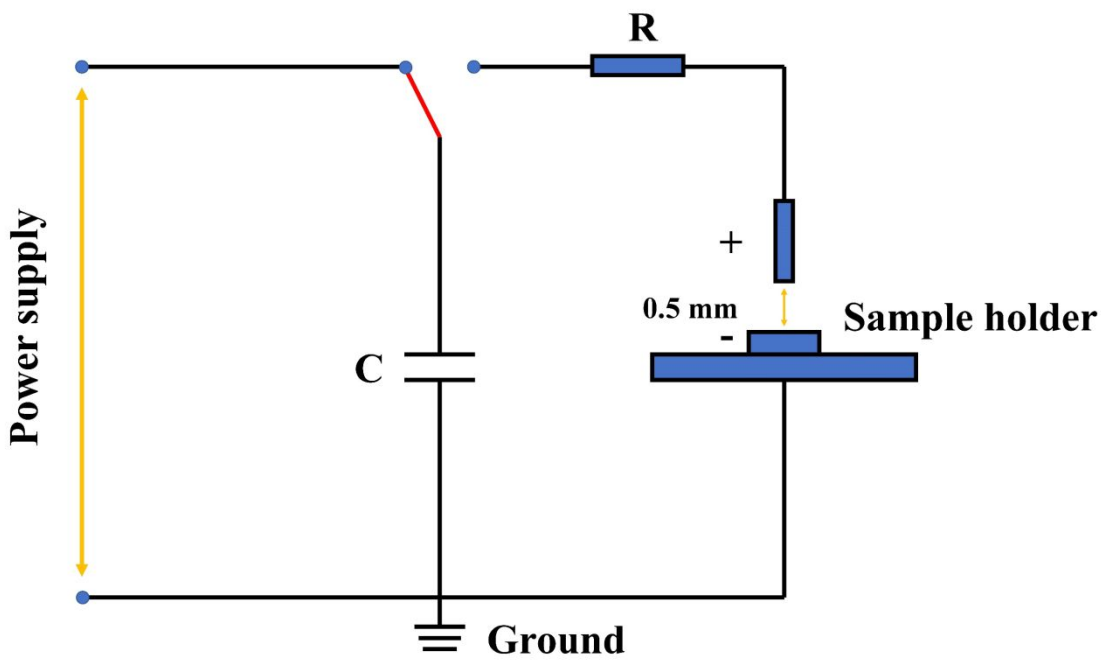

Figure S3. Schematic of the ESD ignition system.

C. Microphotograph of raw-Al, GLM-Al and ref-Al.

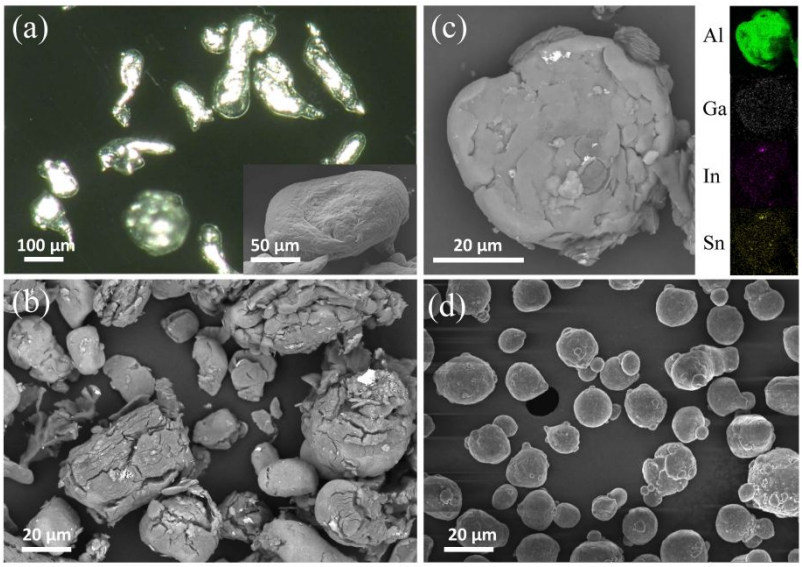

Figure S4. (a) Optical micrograph and SEM image of the raw-Al; (b) SEM image of the GLM-Al; (c) EDS mapping of GLM-Al; (d) SEM image of the ref-Al.

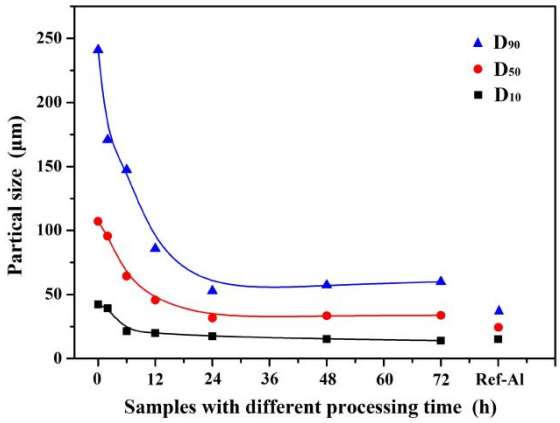

Figure S5. Particle size reduction trend of GLM-Al.

Table S1. Particle size (μm) of GLM-Al with different processing time.

Sample	GLM-Al							Ref-Al
	0 h	2 h	6 h	12 h	24 h	36 h	72 h	
D_{10}	42.3	39.4	21.5	20.0	17.5	15.4	14.1	15.8
D_{50}	107.0	95.6	64.3	45.7	31.6	33.5	33.8	24.4
D_{90}	241.0	171.0	147.5	85.8	52.8	57.3	60.0	36.8

D. Internal strain of raw-Al, GLM-Al and ref-Al.

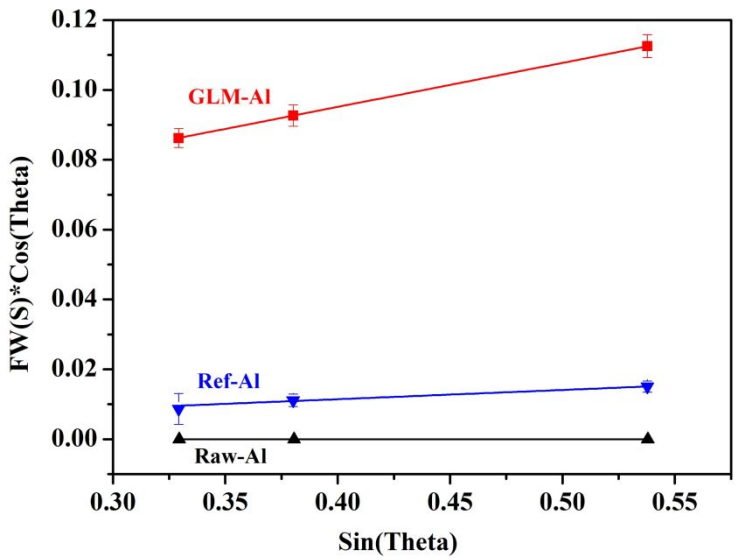

Figure S6. Internal strain of raw-Al, GLM-Al and ref-Al were fitted by Williamson-Hall analysis.

E. SEM images of GLM-Al@Fe_2O_3 and ref-Al@Fe_2O_3.

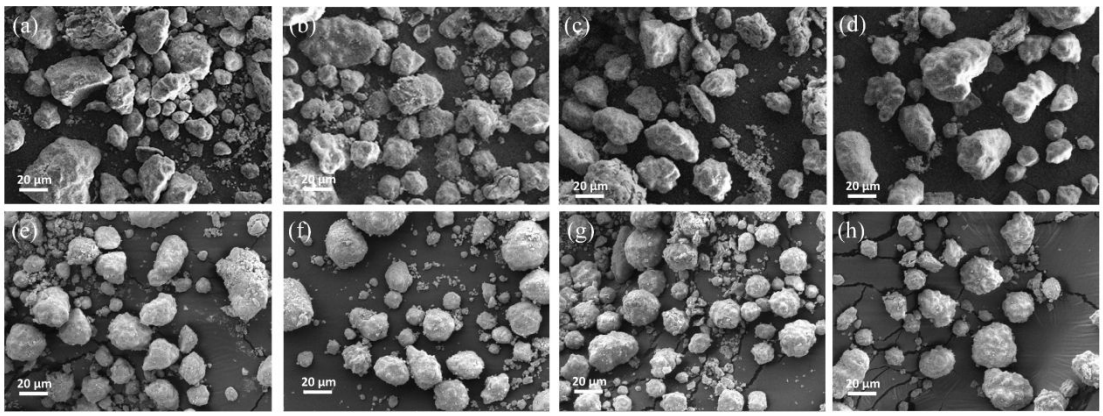

Figure S7. SEM images of GLM-Al@Fe_2O_3 φ=1.1 (a); φ=1.3 (b); φ=1.5 (c); φ=1.7 (d); SEM images of ref-Al@Fe_2O_3 φ=1.1 (e); φ=1.3 (f); φ=1.5 (g); φ=1.7 (h).

F. XPS spectra of GLM-Al.

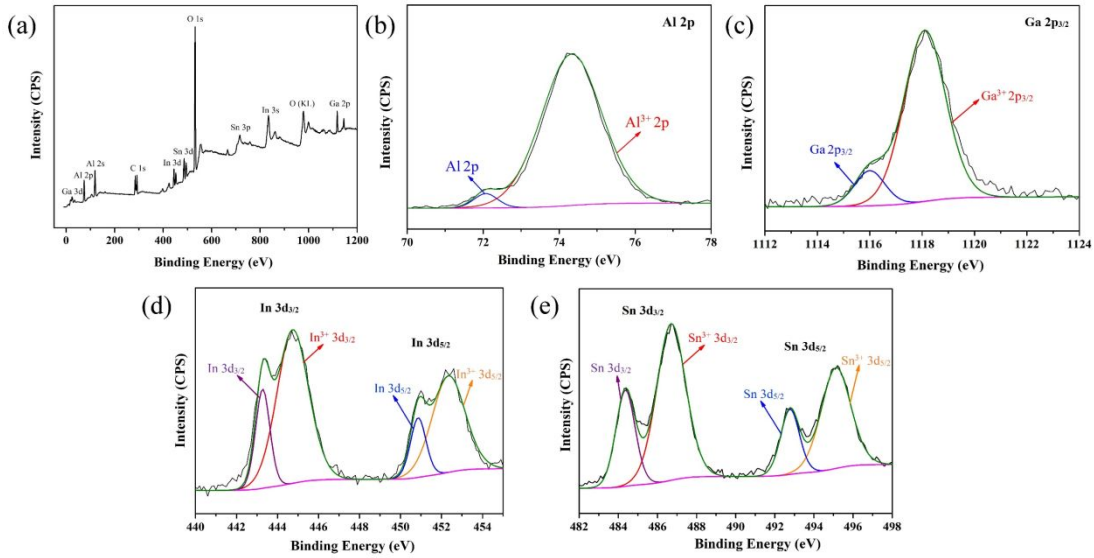

Figure S8. XPS spectra of GLM-Al. (a) Survey scan; (b) Al 2p; (c) Ga 2p; (d) In 3d; (e) Sn 3d.

XPS was used to figure out the chemical states of the elements in the surface of GLM-Al. The survey scan in **Figure S8a** confirms the anticipated existence of Al, O, Ga, In and Sn in GLM-Al. **Figure S8b** shows that the Al 2p spectra of the GLM-Al split into two peaks located at 72.1 and 74.3 eV, revealing the presence of Al and Al_2O_3. **Figure S8c** shows the corresponding Ga 2p demonstrating two distinct peaks (Ga $2p_{3/2}$ and Ga^{3+} $2p_{3/2}$) consistent with Ga and Ga_2O_3. **Figure S8d** shows the In 3d spectra of the GLM-Al. Four peaks (In $3d_{3/2}$, In^{3+} $d_{3/2}$, In $3d_{5/2}$, and In^{3+} $3d_{5/2}$) are observed because of spin-orbit coupling and In oxidation. **Figure S8e** shows the Sn 3d spectra of the GLM-Al, which is similar to above In 3d.[1-2] According to the XPS spectra of GLM-Al, the surface of GLM-Al is coated by Galinstan and its oxides, which is owning to Al surface wetted by Galinstan and Galinstan was oxidized by air.

G. DSC curves of different materials.

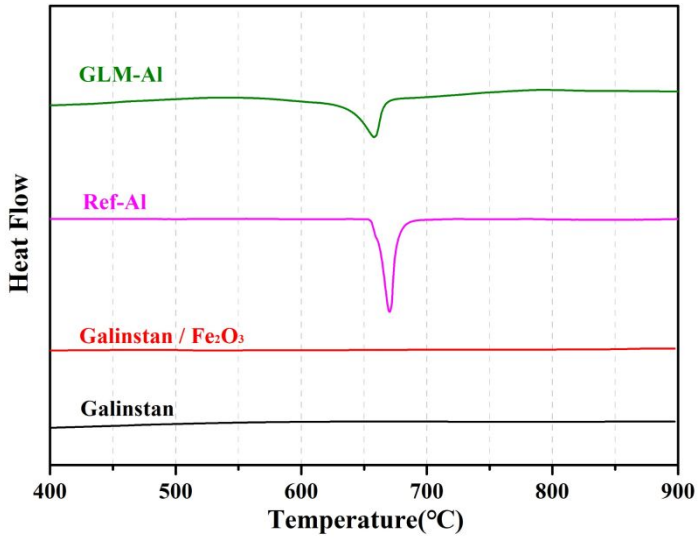

Figure S9. DSC curve of GLM-Al, ref-Al, physical mixture of Galinstan with Fe_2O_3 and Galinstan.

H. XRD of GLM-Al@Fe_2O_3.

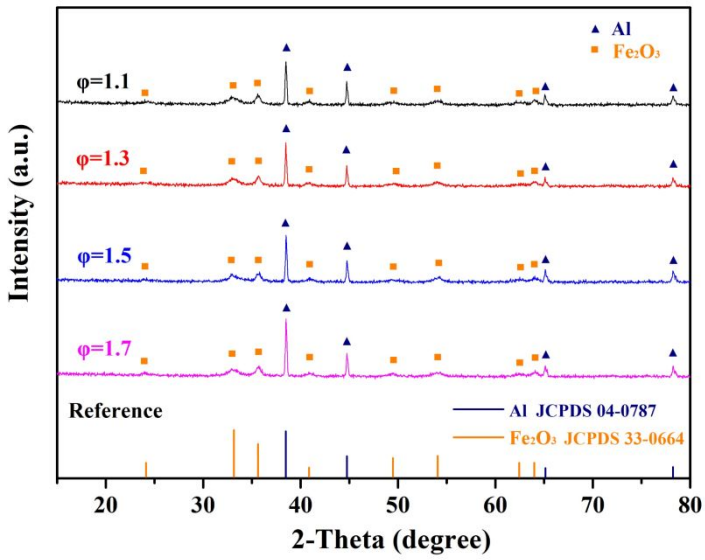

Figure S10. XRD patterns of GLM-Al@Fe_2O_3 thermites.

I. High-speed images of laser ignition.

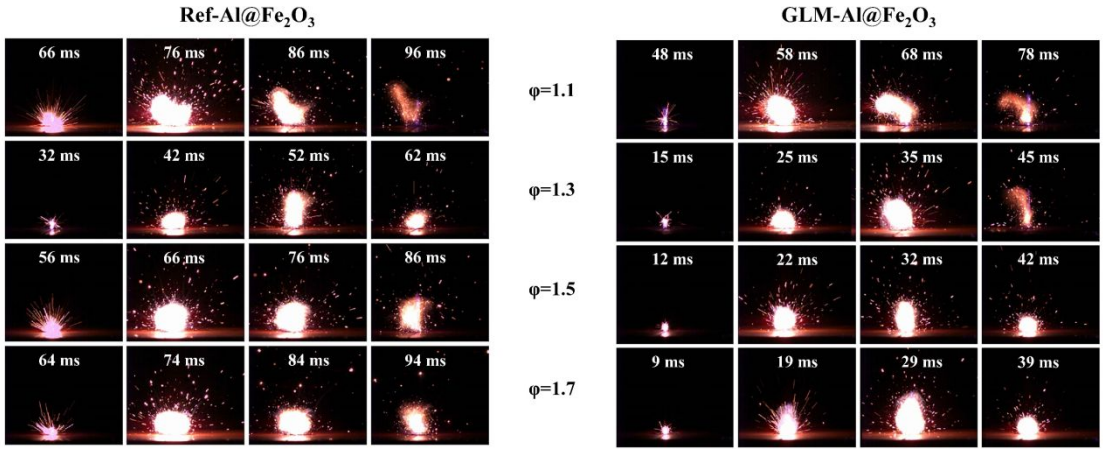

Figure S11. Images from a high-speed camera during laser ignition tests of the thermites.

J. Storage stability.

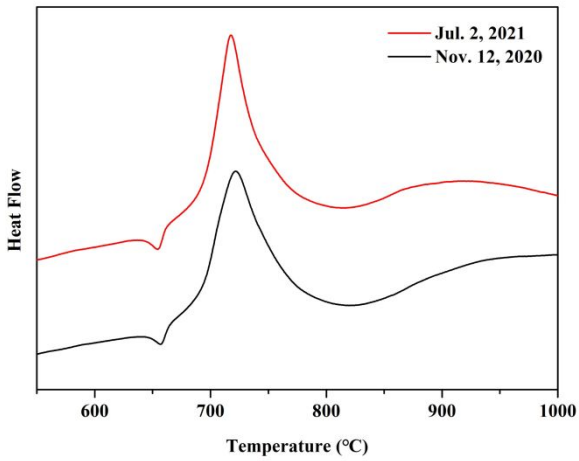

Figure S12. DSC curves of GLM-Al@Fe_2O_3 for different test time.

We performed the DSC test again on GLM-Al@Fe_2O_3, which was tested on November 12, 2020. Comparing the two DSC curves, we found no significant change in the DSC curve after 232 days of storage.

K. Video and images of the reaction of GLM-Al with water at room temperature.

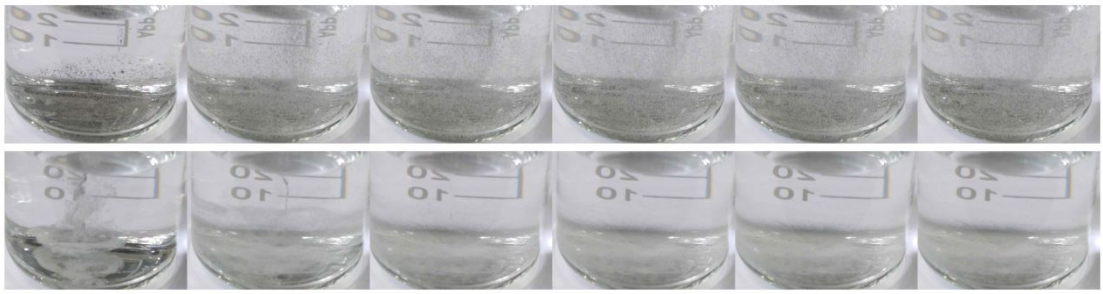

Figure S13. Images of the reaction of GLM-Al with water at room temperature (above); images of adding ref-Al to water at room temperature (below).

Movie S1. The reaction of GLM-Al with water was recorded by high-speed camera (Phantom VEO410L, AMETEK Inc.) at 1000 frames per second. Note: video playback speed 30 frames per second.

RERERENCES

(1) Guan, X.; Zhou, Z.; Luo, P.; Wu, F.; Dong, S. Hydrogen generation from the reaction of Al-based composites activated by low-melting-point metals/oxides/salts with water. *Energy* **2019,** *188*, 116107.

(2) Jacob, A. R.; Parekh, D. P.; Dickey, M. D.; Hsiao, L. C. Interfacial Rheology of Gallium-Based Liquid Metals. *Langmuir* **2019,** *35* (36), 11774-11783.