



In-operando high-speed holography and pyrometry of composition-controlled micro-explosions and agglomerate breakup in Al-Mg energetic fuels

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ABSTRACT

The combustion behavior of Al-Mg alloy particles with varied Mg contents in CH₄/O₂/Ar post-flame was investigated in combination with high-speed imaging, three-color pyrometry, and microscopic digital inline holography (DIH). Al-Mg alloy particles with precisely controllable composition and tunable reactivity were first synthesized using a scalable ultrasonic atomization method. A notable transition in combustion behavior occurs when the Mg content approaches ~ 33 wt%. Particles with ≥ 33 wt% Mg exhibit significantly higher temperatures, frequent micro-explosions, and dramatically shortened burn times, whereas pure Al and Mg-lean particles burn slowly with negligible explosive events. High-speed DIH imaging reveals that the rapid micro-explosion events can occur on microsecond timescales, with particle expansion velocities reaching ~ 20 m·s⁻¹ and fragmentation producing submicron-to-micron-scale combustion products. DIH also reveals that particle agglomerates undergo disaggregation during explosion events, breaking apart into secondary particles. The observations support a mechanism in which preferential Mg vaporization within molten particles generates internal pressure that ruptures the oxide shell and triggers micro-explosive fragmentation. Among the compositions investigated, Mg content of ~ 33 wt% provides a favorable balance between enhanced combustion reactivity and high energy density. These findings highlight the role of alloy composition in controlling particle combustion performance in high-temperature reacting gas flows.

1. Introduction

Metal additives such as aluminum (Al) powders are widely used to enhance energy density in propellants and pyrotechnics due to their high enthalpy release and high combustion temperatures (Dreizin, 2009; Tian et al., 2025). However, micron-sized Al particles often suffer from long ignition delays, particle agglomeration, and slow oxidation rates, which limit their effectiveness (Sundaram et al., 2017; Yang et al., 2025). Alloying Al with magnesium (Mg) has emerged as a promising strategy: Al-Mg alloys ignite at significantly lower temperatures and burn faster than pure Al, yielding higher combustion efficiency (Dreizin and Schoenitz, 2015; Gao et al., 2025). For example, Al-Mg powders have been shown to exhibit shorter burn times and elevated flame

temperatures in combustion experiments, making them attractive for advanced energetic formulations (Gao et al., 2025; Yao et al., 2024).

Notably, Al-Mg alloy particles often display rapid micro-explosion behavior under energetic heating. Previous studies in solid-propellant combustion revealed that when Al-Mg alloys are rapidly heated, they can undergo violent fragmentation, ejecting molten droplets into the flame (Ao et al., 2020; Liu et al., 2022). The propensity to micro-explode depends on particle size and alloy composition (Liu et al., 2022; Feng et al., 2020), though the detailed mechanism in a flame environment remains unclear. Understanding these transient combustion behaviors at the single-particle scale is crucial for optimizing metal additive performance. Resolving these short-lived breakup processes requires diagnostics capable of tracking freely moving particles with sufficiently

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high spatial and temporal resolution.

Advances in high-speed diagnostics now allow *in situ* probing of individual burning metal particles. Digital inline holography (DIH) has been demonstrated as a powerful method to capture three-dimensional trajectories and size distributions of burning metal particles with a long depth of field beyond the capability of conventional imaging (Mazumdar et al., 2020; Yang et al., 2026; Huang et al., 2021). Simultaneously, high-speed three-color imaging pyrometry can measure spatially-resolved flame luminosity and particle surface temperatures (Wang et al., 2019; Yang et al., 2026; Yang et al., 2027).

The combination of controlled alloy synthesis and multiscale diagnostics provides a direct framework for isolating the effect of Mg content on particle combustion dynamics. Ultrasonic atomization (UA) has emerged as an effective method for producing reactive metal powders with controlled composition and morphology (Singh et al., 2026; Priyadarshi et al., 2024). UA can generate Al-based alloy particles with tunable oxidation behavior by adjusting alloy composition and solidification pathways (Singh et al., 2026). This method enables high-yield production of uniform spherical particles while allowing precise control of alloy composition and microstructure, which strongly influence particle ignition, oxidation pathways, and micro-explosion behavior during combustion. In this study, UA was used to systematically synthesize powders of Al-Mg alloys spanning multiple phase regimes of the binary system. While increasing Mg content enhances reactivity, it also reduces the overall energy density of the fuel (Gao et al., 2025; Yao et al., 2024). Therefore, varying Mg content is essential to identify compositions that provide a favorable balance between combustion reactivity and energy density.

In this work, we focus on how Al-Mg composition controls single-particle combustion and micro-explosion dynamics in a premixed $\text{CH}_4/\text{O}_2/\text{Ar}$ flame. Al-Mg powders spanning 0–100 wt% Mg were synthesized by ultrasonic atomization and sieved to a common 32–53 μm size fraction to enable direct comparison across compositions. We combine macro-scale color imaging and shadowgraphy for population-level combustion statistics with micro-scale DIH for resolving transient particle morphology and breakup, together with time-resolved three-color pyrometry for particle temperature measurements. TG-DSC analysis and thermochemical calculations provide complementary information on oxidation behavior and energetics. Our objective is to quantify how Mg content influences micro-explosion frequency, burn time, particle temperature, and transient breakup dynamics, and to relate these observations to the underlying combustion mechanism. These insights should inform the design of high-energy metal fuels and propellants.

2. Method

2.1. Synthesis and characterization

Al-Mg alloy powders were synthesized from high-purity elemental Al and Mg feedstocks (ThermoFisher Scientific, $\geq 99.9\%$) using an ultrasonic atomization (UA) system (Amazemet) (Singh et al., 2026). The weighed elemental mixtures corresponding to the target compositions listed in Table 1 were loaded into a boron-nitride-coated graphite crucible housed within an induction-heated chamber. Before heating, the chamber was evacuated and backfilled with argon (Ar) twice to minimize residual oxygen and moisture. The feedstock was then subjected to stepwise heating with dwell steps at 473 and 773 K, while the chamber was continuously flushed with Ar to remove adsorbed species. The charge was subsequently heated under Ar to ~ 1123 K for pure Al and to 973 K for the Al-Mg alloys, followed by a 5 min dwell to ensure complete melting and compositional homogenization. Pure Al was atomized directly after this hold. For the Mg-containing alloys, the melt was then cooled to 923 K prior to atomization to reduce Mg evaporation while maintaining sufficient fluidity. Atomization was initiated by removing the graphite sealing rod, allowing the melt to flow into the spray chamber and impinge on a carbon-fiber composite plate mounted on a 60 kHz sonotrode. Ultrasonic vibrations generated capillary waves on the melt surface that fragmented the molten stream into droplets, which rapidly solidified into spherical alloy particles while falling through the inert atmosphere (Priyadarshi et al., 2024). A controlled pressure differential between the heated melt chamber and the spray chamber enabled continuous melt feeding and stable atomization (Singh et al., 2026). During atomization, the spray chamber pressure was maintained between 100 and 200 mbar using active pressure control, with automatic Ar replenishment whenever the pressure fell below the lower limit and venting whenever it increased above the 200 mbar threshold. During the first few seconds of atomization, transient spattering was occasionally observed due to incomplete wetting of the sonotrode surface; however, once wetting was established, a steady atomization regime was obtained. The atomization process typically produced an 80–90 wt% yield of powders below 300 μm (Singh et al., 2026). After atomization, the powders were sieved, and the 32–53 μm size fraction was used for all combustion diagnostics to ensure consistent particle size and enable direct comparison of thermal response across compositions.

The sample set and their chemical compositions determined from X-ray diffraction (XRD) are summarized in Table 1. Five powder compositions were selected using CALPHAD-guided phase diagram design with Thermo-Calc software (2024b version) using TCBIN: TC Binary Solutions v1.1 database (Spencer, 2008; Andersson et al., 2002) to span the major condensed phases in the Al-Mg binary and guide composition selection while systematically increasing Mg content and lowering equilibrium phase-transition temperatures (Table 1; Fig. 1 (a) and

Table 1

Chemical composition of five powders investigated in this work, including pure Al, Al-Mg alloys, and pure Mg. Also listed are dominant phases identified by XRD, the melting and boiling points calculated using Thermo-Calc (Fig. S1), the gravimetric and volumetric energy densities estimated for oxidation with O_2 , and our measured heat of combustion from calorimetry.

Powder name	Elemental composition	Major phases (by XRD)	Melting point (K)	Boiling point (K)	Theoretical gravimetric energy density ($\text{kJ}\cdot\text{g}^{-1}$ metal)	Theoretical volumetric energy density ($\text{kJ}\cdot\text{cm}^{-3}$ metal)	Measured heat of combustion by calorimetry ($\text{kJ}\cdot\text{g}^{-1}$ metal)
Al	Al 100 wt%	α -Al	933	2790	31	84	29.1 ± 0.3
Al-15% Mg	Al 85 wt%, Mg 15 wt%	α -Al and β - Al_3Mg_2 (mass ratio $\sim 1:1$)	860	1680	30	74	27.1 ± 0.1
Al-33% Mg	Al 67 wt%, Mg 33 wt%	β - Al_3Mg_2	723	1540	29	65	23.7 ± 0.4
Al-50% Mg	Al 50 wt%, Mg 50 wt%	γ - $\text{Al}_{12}\text{Mg}_{17}$	735	1480	28	58	20.0 ± 1.3
Mg	Mg 100 wt%	α -Mg	923	1385	25	43	21.5 ± 1.8

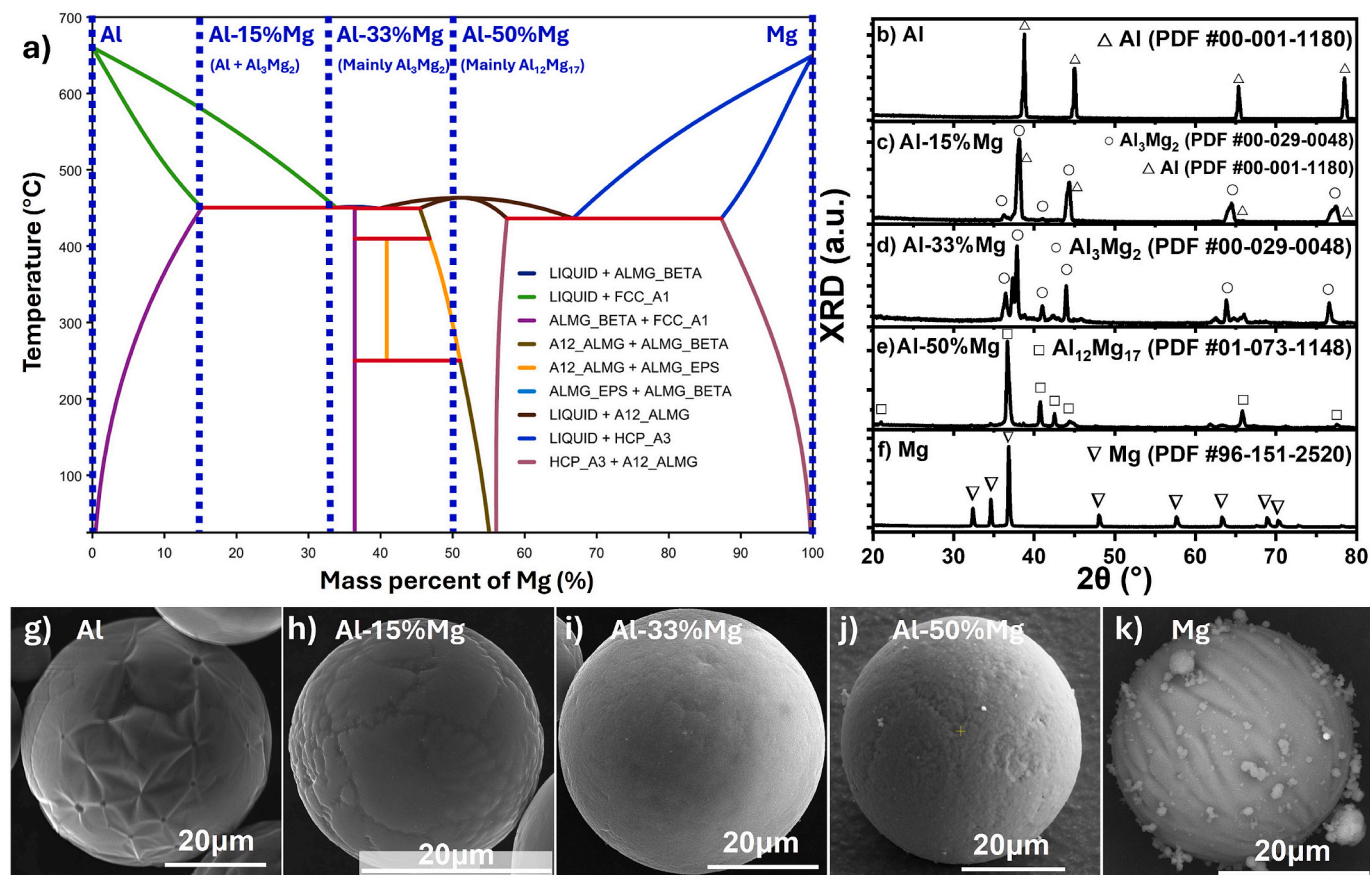


Fig. 1. (a) Al-Mg binary condensed phase diagram calculated using Thermo-Calc software showing the variation in melting temperature with Mg content, where blue dashed lines mark the composition used in this work (0, 15, 33, 50, and 100 wt% Mg); XRD patterns for the as-synthesized metallic powders with (b) 0 wt%, (c) 15 wt%, (d) 33 wt%, (e) 50 wt%, and (f) 100 wt% Mg, respectively; SEM characterization of surface morphologies of the as-synthesized metallic powders with (g) 0 wt%, (h) 15 wt%, (i) 33 wt%, (j) 50 wt%, and (k) 100 wt% Mg.

Fig. S1). To characterize the as-synthesized particles, XRD was performed using PANalytical Empyrean Series 2 on a silicon zero-background plate at a scan rate of 7°/min in the 2θ range of 10–80°, as shown in Fig. 1 (b–f). The target Mg mass fractions were 0, 15, 33, 50, and 100 wt% Mg, which represent pure α-Al, an Al-rich alloy (α-Al + β-Al₃Mg₂), β-intermetallic-dominant compositions (Al₃Mg₂), γ-intermetallic-dominant compositions (Al₁₂Mg₁₇), and pure α-Mg, respectively. The five selected powders are hereafter denoted Al, Al-15%Mg, Al-33%Mg, Al-50%Mg, and Mg, where the numbers denote nominal Mg content in wt%. The phase diagrams in Fig. 1 (a) and Fig. S1 show that increasing Mg content substantially lowers the melting and boiling temperatures of the alloys, which is expected to influence particle ignition, vaporization, and explosion behavior in combustion environments.

In addition to lowering phase-transition temperatures, increasing Mg loading also affects the thermochemical energy content of the alloys (Table 1). The theoretical gravimetric energy density was calculated by assuming complete oxidation of the metal components in O₂ based on the reactions $4\text{Al} + 3\text{O}_2 = 2\text{Al}_2\text{O}_3$ and $2\text{Mg} + \text{O}_2 = 2\text{MgO}$, using standard formation enthalpies of the oxide products. The corresponding volumetric energy densities were estimated using the bulk densities of the dominant phases in each alloy composition. Although packed spherical particles contain interparticle voids, the bulk density provides a reasonable estimate of the volumetric energy density of the alloy itself, while in practical energetic formulations these voids are typically filled by polymer binders. As shown in Table 1, the calculated energy density decreases gradually from 31 kJ g^{−1} and 84 kJ cm^{−3} for pure Al to 25 kJ g^{−1} and 43 kJ cm^{−3} for pure Mg, reflecting the lower heat of oxidation for Mg compared with Al on both mass and volume bases. Therefore,

increasing Mg content introduces a trade-off: higher Mg fractions are expected to facilitate melting, vaporization, and more vigorous combustion, while simultaneously reducing the overall energy density. By varying the Mg loading, this study systematically examined this balance to identify the compositions that provide both favorable combustion reactivity and high energy density. The heats of combustion were also measured using a Parr 6725 Semimicro Calorimeter equipped with a Parr 6772 Calorimetric Thermometer. Each metal powder was mixed with benzoic acid as a combustion aid and pressed into a cylindrical pellet before electrical ignition in an oxygen-filled bomb. The heat contribution from benzoic acid was subtracted, and the resulting heat release was normalized by the metal mass, as reported in Table 1. The measured heats of combustion show the same general decrease with increasing Mg content, consistent with theoretical trends.

The as-synthesized powders were placed onto aluminum stubs using conductive carbon tape and examined using a Tescan MIRA 3 GM field-emission scanning electron microscope (FE-SEM) to characterize powder size, sphericity, and surface morphology. Representative SEM images of all five compositions are shown in Fig. 1(g–k). The powders in the selected 32–53 μm fraction are predominantly spherical, while their surface morphology varies with alloy composition. For cross-sectional imaging, powders were mounted in epoxy and polished to assess internal microstructure and the possible presence of internal porosity using a backscattered-electron detector with enhanced Z-contrast and sensitivity (Flickinger et al., 2026). As shown in Fig. 2 (a), the synthesized Al-33%Mg alloy (β-Al₃Mg₂ dominant) exhibits a predominantly spherical morphology with relatively smooth surfaces, consistent with effective droplet formation and solidification during ultrasonic atomization. The cross-sectional SEM image reveals a dense internal structure without

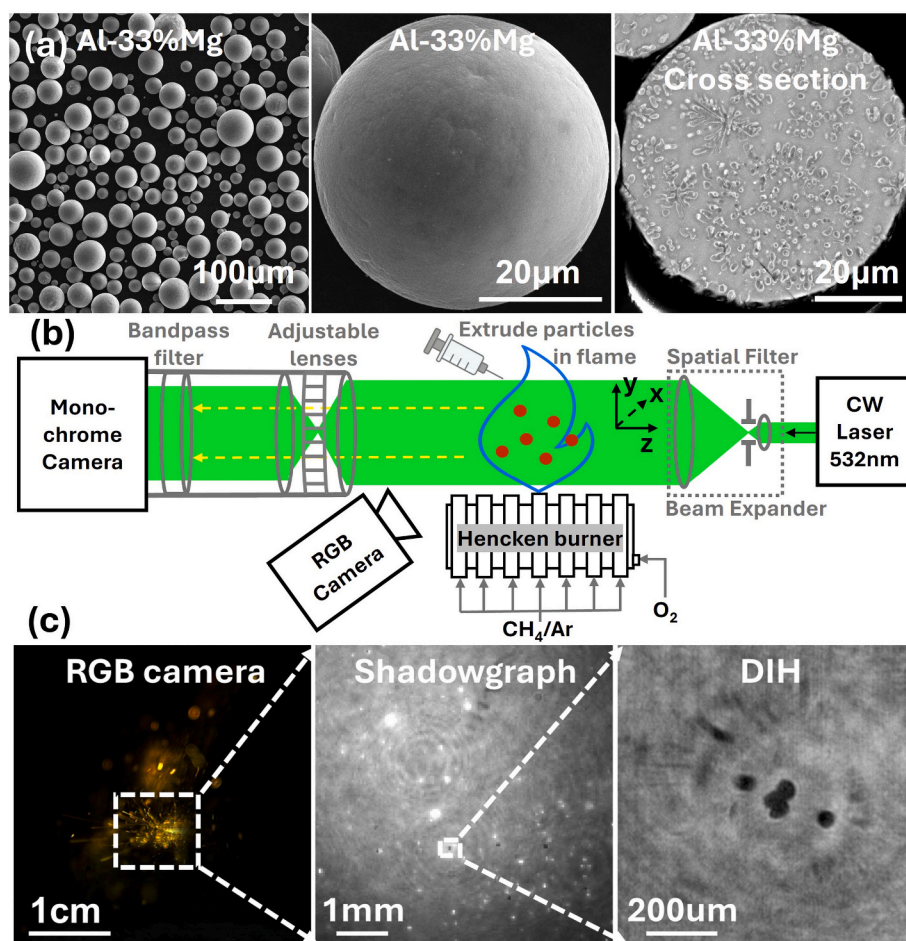


Fig. 2. (a) Representative SEM images showing the particle sizes of the 32–53 μm Al-33%Mg metallic powders with their cross section; (b) Schematic experimental setup of combustion diagnostics, including the Hencken burner and DIH. (c) Representative snapshots of adjustable high-speed imaging system, including macro-scale RGB imaging, shadowgraphy, and micro-scale DIH.

visible voids, confirming good consolidation during solidification and structural integrity of the alloy powder. In addition, numerous fine microstructural features are distributed throughout the particle, likely arising from the solidification microstructure of the Al₃Mg₂ intermetallic phase. Further SEM characterizations of atomized Al alloys can also be found in our previous work (Singh et al., 2026).

2.2. Hencken burner

As illustrated in Fig. 2 (b) and Fig. S2, a CH₄-flame burner was used to generate a spatially uniform high-temperature gas environment for particle combustion; essentially a wall-less plug flow reactor. The Hencken burner is packed with stainless-steel spheres through which multiple tubes are inserted. A mixture of CH₄ and Ar is supplied through the tubes, while O₂ is introduced through the spaces between the tubes and stainless-steel balls, producing a uniform laminar outflow that forms a quasi-premixed flame above the burner surface (Jacob et al., 2016). The Al-Mg particles were loaded into a syringe equipped with a gauge-16 needle positioned ~ 3 cm above the burner with the injection direction oriented toward the burner center. Upon injection, the aerosolized particles were sufficiently diluted so that the combustion behavior of individual particles or small agglomerates could be considered as isolated events. This assumption is supported by the microscopic observations discussed in Section 3. The post-flame environment employed in this work corresponds to a CH₄/O₂/Ar mixture with a molar ratio of 1:2:2, producing a H₂O/CO₂-rich and O₂-lean region above the flame. Gas flow rates of CH₄, O₂, and Ar were maintained

by MKS mass flow controllers at 1, 2, and 2 standard L·min⁻¹, respectively. The gas-phase composition and temperature distribution were further calculated with a freely propagating flame model using Chemkin-Pro and USC Mech II (Wang, et al., 2026), as shown in Fig. S2. The adiabatic temperature of the flame is computed to be ~ 2890 K. The simulation also indicates that at the position where particles burn (>1 cm above the flame sheet), the gas environment becomes mainly H₂O/CO₂-rich with limited residual O₂.

2.3. High-speed optical diagnostics with DIH and pyrometry

The combustion behavior of the Al-Mg particles ejected into the CH₄ flame was visualized using high-speed DIH, as illustrated in Fig. 2 (b). This technique, with prolonged depth of field compared to conventional microscale imaging, enables three-dimensional tracking and sizing of moving particles with high spatial and temporal resolution, making it well suited for capturing rapid fragmentation and micro-explosion events in high-speed flow conditions. A collimated continuous-wave 532 nm laser (Coherent Verdi V-6) was used for illumination. The beam was spatially filtered through a 5-μm pinhole and expanded to a 25 mm diameter using a pair of plano-convex lenses with focal lengths of 10 mm and 100 mm (Thorlabs, models P5W, C060TMD-A, and LA1509-A, respectively). The resulting in-line holograms were recorded using a high-speed CMOS camera (Phantom VEO 1310S, binning mode) operating at 40,000 to 100,000 fps with an exposure time of 1 μs. Infinity K2 Distamax long-distance microscope lenses (NTX tube with CF-4 objective) were used to achieve a spatial resolution of 2.8 μm·pixel⁻¹ across a

0.9×0.7 mm field of view at 100,000 fps (or 1.8×1.4 mm at 40,000 fps). Additional DIH measurements were also performed with camera binning disabled, providing a finer spatial sampling of $\sim 1.4 \mu\text{m}\cdot\text{pixel}^{-1}$ at 40,000 fps to confirm rapid particle disappearance in micro-explosion under higher-spatial-resolution configuration. The optical focal plane was aligned at the center of the Hencken burner. This configuration enabled high-contrast diffraction patterns generated by particles larger than several μm to be captured. The recorded holograms were numerically reconstructed using Fourier-domain angular-spectrum back-propagation under scalar diffraction theory (Yang et al., 2027). For each frame, the hologram was propagated through a series of axial positions, and the reconstructed plane providing the clearest particle image was selected for subsequent analysis (Yang et al., 2026; Vasudevan et al., 2026). Particle trajectories and transient morphological changes were then tracked frame-by-frame. The reconstruction procedure, equations, representative raw/refocused holograms, and Python implementation are provided in Fig. S3–S4 and Supplementary Code S1 (Yang et al., 2027; Wu et al., 2021; Chen et al., 2017). To suppress broadband light emissions from the combustion zone, a narrow-band-pass optical filter (Thorlabs FLH532-4, 532 ± 2 nm) was installed in front of the detector when viewing the microscopic events. For shadowgraphy, the same setup was used except that the band-pass filter was removed, and the lens was replaced with a Nikon AF Micro Nikkor lens to achieve a 12×8.7 mm field of view at $36 \mu\text{m}\cdot\text{pixel}^{-1}$ at 100,000 fps.

High-speed color imaging was performed to investigate macroscopic combustion events and record pyrometry data using a Phantom Miro 110 camera equipped with a Nikon AF Micro Nikkor lens (field of view $\sim 6 \times 2.6$ cm, aperture f/16, spatial resolution $\sim 47 \mu\text{m}\cdot\text{pixel}^{-1}$). Three-color imaging pyrometry was applied to the color-camera data to map surface temperatures. The raw intensities of the three-color channels (red, green, and blue; RGB) were extracted by debayering the Bayer pattern images (Yang et al., 2026; Wang et al., 2019; Yang et al., 2026). Temperatures were then estimated at each pixel by taking ratios of the three-color channels (G/R, B/G, and B/R) and comparing them with theoretical temperatures assuming graybody emission. The pyrometry was calibrated with blackbody sources (ThorLabs SLS201L and Mikron M390) and has an expected uncertainty of ~ 200 – 300 K. Detailed comparisons of the window sizes, pixel sizes, camera, and lens used in DIH, shadowgraphy, and pyrometry are provided in Fig. 2 (c) and Fig. S5.

2.4. TG-DSC analysis

Thermogravimetric analysis (TGA) and Differential scanning calorimetry (DSC) of the particles was performed using a NETZSCH STA 449 F3 Jupiter thermogravimetric analyzer. As particle combustion primarily occurs in a post-flame region dominated by stable oxidizing species, such as CO_2 and H_2O , with relatively low O_2 concentration according to the kinetic simulation shown in Fig. S2, simultaneous TG–DSC measurements were used to compare the relative oxidation behavior of the powders under simplified H_2O –, CO_2 –, and O_2 –containing environments relevant to the post-flame chemistry. These slow-heating measurements are intended to probe composition- and oxidizer-dependent thermal reactivity (not to reproduce the conditions of the microsecond-scale micro-explosion in flame tests). All samples (~ 1 mg per run) were heated to 1470 K at a ramp rate of $10 \text{ K}\cdot\text{min}^{-1}$, held at 1470 K for 120 min, to characterize the mass changes under different flow conditions (total flow rate kept at $50 \text{ mL}\cdot\text{min}^{-1}$) and investigate fundamental reactions of Al–Mg alloys with O_2 , CO_2 , and H_2O vapor. Due to the limited vapor pressure of H_2O at room temperature (24 Torr), 3% H_2O vapor in Ar was used to probe the reactivity of the powders under humid oxidizing conditions. Because the O_2 concentration in the CH_4 flame decreases to $\leq 5\%$ according to the kinetic simulation, experiments with 5% O_2 in Ar were also performed. These results are presented in Section 3.3 and Fig. S6–S9. The post-heating products were collected and characterized by XRD to identify the reaction products, as

discussed in Section 3.3 and Fig. S10–S11. The DSC heat-flow signals were recorded simultaneously with the TG measurements and normalized by sample mass. These measurements provide complementary information on phase transitions and oxidation-associated thermal events.

3. Results and discussion

3.1. High-speed pyrometry, shadowgraphy, and statistical analysis

The combustion behavior of atomized Al–Mg alloy particles (32 – $53 \mu\text{m}$) in the CH_4 flame was characterized using color imaging, three-color pyrometry, and shadowgraphy. Fig. 3 shows the observed particle dynamics for alloys with increasing Mg content: Panels (a1) to (a5) show representative color images of particles traversing the flame, while panels (b1) to (b5) present the corresponding three-color pyrometry and average particle temperature. Parallel shadowgraph images are shown in panels (c1)–(c10) to visualize individual particle explosion events. These snapshots are from supplementary Video S1. Statistical analysis of the observed particles is summarized in Fig. 3 (d–e). Based on the shadowgraph images, particle trajectories within the field of view were classified into three categories according to their optical appearance and evolution: (i) particles that remain dim throughout the observation window and exit the field of view without visible change; (ii) particles that appear bright immediately upon entering the field of view and remain incandescent without a clear onset of oxidation; and (iii) particles that initially appear dark but subsequently turn bright during flight, indicating the onset of rapid oxidation reactions. The explosion frequency was determined statistically from the entire particle population as the ratio of particles undergoing micro-explosions to the total number of particles observed. This analysis therefore reflects the fractions of dim particles, continuously incandescent particles, and exploding particles within the observation window, as shown in Fig. 3 (d). A total of 290 particle trajectories were analyzed by shadowgraphy, including 50, 55, 55, 76, and 54 particles for Al, Al–15%Mg, Al–33%Mg, Al–50%Mg, and Mg, respectively. Among these populations, 0, 1, 29, 44, and 30 particles underwent visible micro-explosion, corresponding to explosion fractions of 0, 2, 53, 58, and 56%, respectively, as detailed in Table S1. For particles in category (iii), the burn time is defined as the time interval between the frame in which the particle first transitions from dark to bright and the frame where the particle either undergoes micro-explosion or cools and returns to a dark appearance. Particles belonging to categories (i) and (ii) were excluded from burn time determination because the start or end of the oxidation process cannot be unambiguously identified within the observation window. Statistical trends of the measured burn time, particle temperature, and alloy boiling temperature are shown in Fig. 3 (e).

For pure Al particles in Fig. 3 (a1), no micro-explosions were observed within the field of view. Most particles appeared dim in the color images. Because three-color pyrometry relies on thermal emission, the weak incandescence resulted in insufficient signal for reliable temperature retrieval, as shown in Fig. 3 (b1). The absence of micro-explosions and low radiative intensity are also evident in the corresponding shadowgraph images in Fig. 3 (c1) and (c2), where particles remain mostly dark and intact. These observations indicate that pure Al particles undergo rather slow oxidation in the flame and rarely reach temperatures sufficient to induce vaporization. A small portion of the Al particles has sufficient incandescence for pyrometry, showing an average particle temperature of 2050 K. However, they represent less than 3% of all the particles, as shown in Fig. 3 (d). Therefore, the average temperature of pure Al particles is estimated to be well below 2050 K, much lower than its boiling point (2790 K). In Fig. 3 (e), the measured burn time for pure Al particles exceeds the 40 ms observation window of the color imaging, indicating that their combustion process occurs on a longer timescale than the residence time within the field of view.

Introducing 15% Mg significantly enhances particle luminosity. As shown in Fig. 3 (a2), most particles exhibit stronger incandescence

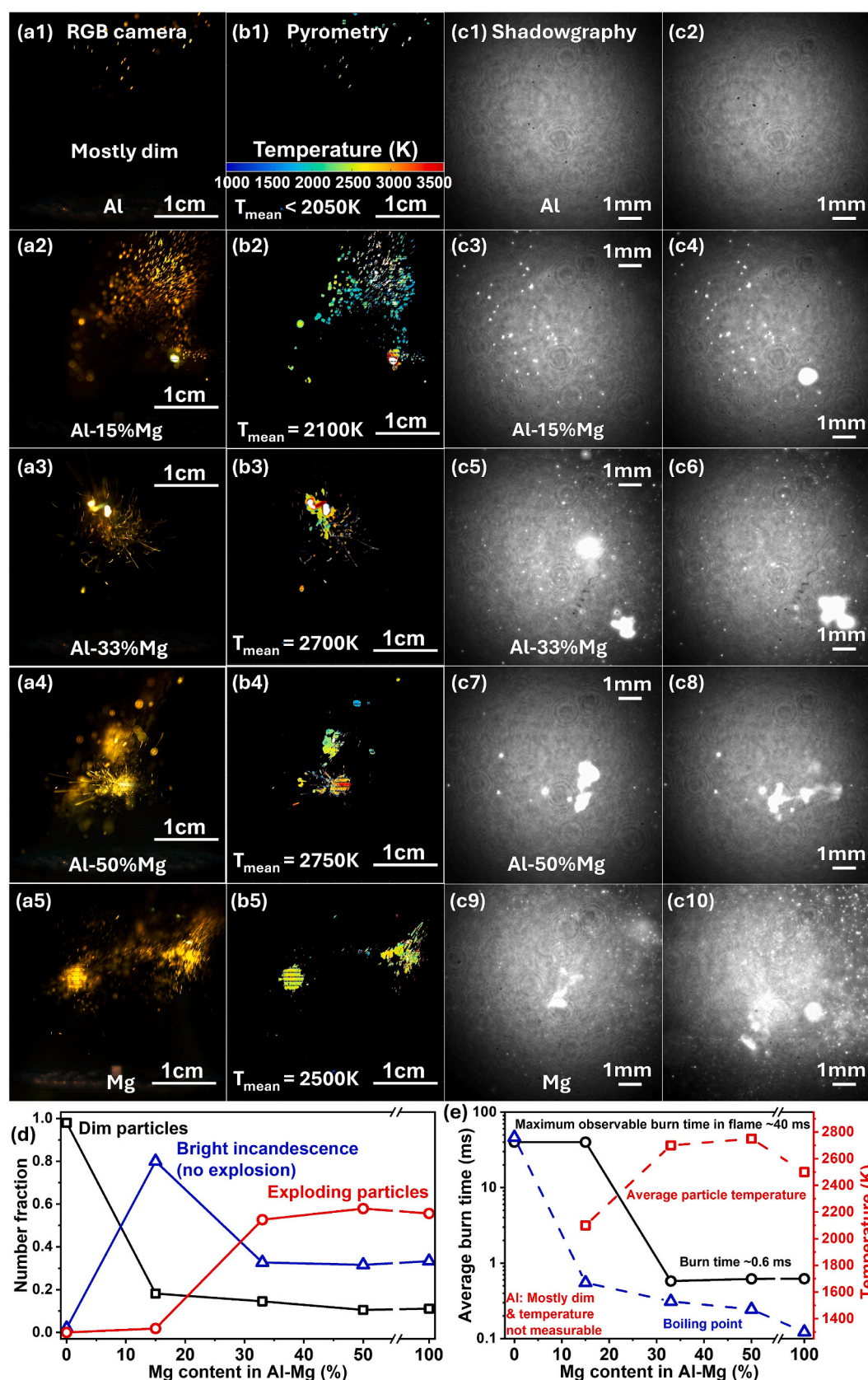


Fig. 3. Combustion imaging in $\text{CH}_4/\text{O}_2/\text{Ar}$ post-flame, including (a) color imaging, (b) pyrometry and (c) shadowgraphy of five metal powders with Mg = 0, 15, 33, 50, and 100 wt%, respectively; (d) Statistical analysis of explosion frequency; (e) Burn time, boiling point, and average particle temperature. Details can be viewed in the supplementary video S1. The statistical analysis includes $N = 50, 55, 55, 76,$ and 54 particles for Al, Al-15%Mg, Al-33%Mg, Al-50%Mg, and Mg, respectively; complete classification counts are provided in Table S1.

compared with pure Al. The corresponding pyrometry in Fig. 3 (b2) indicates particle temperatures approach ~ 2100 K. As shown in Fig. 3 (c3) and (c4), occasional explosion events are observed in shadowgraph images, but the fraction of such events remains relatively small. Enhanced luminosity suggests increased heat release and more vigorous oxidation compared with pure Al particles. Nevertheless, the statistical analysis in Fig. 3 (d) shows that most particles at this composition still burn without explosive fragmentation.

A significant change in combustion behavior occurs when the Mg content reaches 33% or higher. As shown in Fig. 3 (a3) – (a5), particles with Mg contents of 33%, 50%, and 100% exhibit intense incandescence and rather frequent micro-explosions. The shadowgraph images in Fig. 3 (c5)–(c10) clearly capture particle explosion events, where these molten droplets ignite, fragment, eject secondary particles, and generate smokes. As shown in Fig. 3 (d), the fraction of micro-explosion events increases markedly with Mg content, reaching $\sim 60\%$ for alloys containing 33 wt% Mg or more. At the same time, the fraction of dim particles decreases to $\sim 10\%$ within the observation window. These results indicate that the 33 wt% Mg and higher compositions tested are significantly more reactive than Mg-lean particles. Three-color pyrometry measurements show that particle temperatures for these alloys are substantially higher than those observed for lower Mg compositions.

Fig. 3 (e) also shows the measured burn times. The pure Al and the 15 wt% Mg particles exhibit burn times exceeding the 40 ms observation limit of the imaging system. In contrast, when the Mg content reaches 33 wt% or higher, the burn time decreases dramatically and stabilizes near ~ 0.6 ms. Previous studies have reported similar trends for Al-Mg alloy particles, where increasing Mg content significantly shortens ignition delay (Gao et al., 2025) and increases explosion frequency (Yao et al., 2024). Moreover, Gao et al., 2025 have shown that the introduction of Mg can significantly improve the combustion efficiency of Al-Mg fuels, with measured heats of combustion approaching theoretical values and overall efficiencies exceeding 99%. Consistent with this trend, our results further show that, among the compositions investigated, the 33 wt% Mg alloy provides a favorable balance between enhanced combustion reactivity and the high energy density of Al. Three main factors likely contribute to this transition: (1) First, the addition of Mg substantially lowers both the melting and boiling temperatures of the alloy phases, which promotes earlier melting and increased vapor generation during particle heating. Thermodynamic calculations of the Al-Mg phase diagram indicate that the boiling temperatures, T_b , decrease dramatically from ~ 2790 K for Al to ~ 1540 – 1385 K for the 33–100 wt % Mg compositions. Meanwhile, average particle temperatures, T_p , increase to ~ 2500 – 2700 K for Mg-rich compositions. The resulting vapor formation inside the particles can generate internal pressure and trigger fragmentation events, dramatically accelerating the reaction by dispersing molten material and increasing surface area for oxidation (Feng et al., 2020). (2) In addition, Mg oxidation releases heat rapidly and can promote the subsequent oxidation of Al within the alloy particle, because Mg has a stronger tendency to oxidize and loses electrons more readily than Al. Experimental observations have shown that the heat released from Mg combustion can effectively initiate and sustain Al combustion, leading to more intense reactions and shorter combustion durations (Gao et al., 2025). (3) Moreover, the presence of Mg also modifies the structure of the surface oxide layer. Compared with the dense and protective Al_2O_3 layer that forms on pure Al particles, oxide shells formed on Al-Mg alloys tend to be more porous and less protective due to the formation of MgO and mixed oxides (Gao et al., 2025; Yao et al., 2024). MgO has a Pilling-Bedworth ratio of ~ 0.81 , which favors defect-rich oxide layers (Kim et al., 2011). This structural change can facilitate oxidizer transport and reduce the kinetic barrier for ignition and oxidation.

The observed micro-explosions are consistent with the superheating-driven fragmentation mechanism reported for multi-component droplets containing components with different volatilities (Guerrieri et al., 2017; Lyu et al., 2021). In such systems, the more volatile component

can become trapped within a surrounding shell of the less volatile component and become superheated as the droplet temperature increases. Once the local vapor pressure exceeds a critical threshold, vapor bubbles nucleate and grow rapidly inside the liquid, eventually causing the droplet to fragment in a micro-explosion. For Al-Mg alloys, Mg acts as the volatile component because its boiling point is far lower than that of Al. The buildup of Mg vapor within the molten particle can generate internal bubbles that grow until reaching a critical size, leading to rapid fragmentation of the particle. Such bubble nucleation and growth processes have been analyzed by Belal et al., 2018 using classical nucleation theory and are expected to occur readily for Al-Mg particles at temperatures above ~ 2300 K. This mechanism is consistent with the sharp increase in explosion frequency observed when the Mg content reaches 33 wt% and particle temperatures T_p of 2500–2750 K correspond to superheat ratios, $S = T_p/T_b \approx 1.7$ – 1.9 . At such superheat levels, rapid internal Mg vapor generation can drive bubble growth, jetting, and rupture of the mixed oxide shell.

3.2. High-speed holographic imaging by DIH

Although conventional imaging provides statistical information on particle explosion frequency and burn time, it cannot easily track the shape changes of individual particles due to the limited depth of field and resolution. DIH solves this limitation. The DIH sequences in Fig. 4 are representative individual particle/agglomerate events selected to resolve the transient breakup dynamics. Fig. 4 (a) shows the disaggregation process of a collection of Al-Mg particles, obtained from DIH with a bandpass filter to block strong incandescence emissions. At 125–225 μs , the agglomerate undergoes an abrupt breakup event, separating the agglomerate into multiple particles that fly in different directions. Some particles, as shown in the 300–350 μs snapshots, further undergo secondary micro-explosions and disappear. Fig. 4 (b) further confirms that rapid secondary micro-explosions occur within tens of μs , as detailed in Fig. S3–S4. Therefore, to visualize such fast processes, it is necessary to increase the frame rate to 100,000 fps and remove the bandpass filter to increase light sensitivity, as shown in Fig. 4 (c). Upon micro-explosion, we can observe the size of the visualized particle rapidly expanding from ~ 40 μm to ~ 250 μm within 10 μs , corresponding to an expansion radial speed of $10 \text{ m}\cdot\text{s}^{-1}$. Such rapid radial expansion indicates a violent internal gas-generation process during particle breakup, consistent with the superheating mechanism discussed in Section 3.1. Strong incandescence appears in the subsequent frame, and the original micron-sized particle rapidly disappears within ~ 20 μs of the explosion event. The same disappearance behavior was also observed using the higher-spatial-resolution DIH configuration ($\sim 1.4 \mu\text{m pixel}^{-1}$, without binning), indicating fragmentation into products below the reliable detection limit of the present DIH system. These observations are consistent with rapid refinement into micron-to-submicron products, although the final fragment-size distribution cannot be quantitatively resolved by the present measurements. Such rapid micro-explosions coexist with the other slower oxidation processes.

It is important to note that while Mg vaporization and expansion are endothermic processes, the required thermal energy is supplied by the rapid exothermic oxidation of Mg, which occurs near the particle surface and through defects in the oxide shell. Evaluating the timescale of internal heat conduction is therefore essential, as it determines how efficiently heat generated at the surface is transported into the particle interior to drive vaporization. Rapid heat transfer would allow this locally generated heat to penetrate into the molten core, enabling transient superheating of Mg-rich regions and promoting vapor generation prior to rupture. The characteristic heat conduction time t_{cond} was estimated from $t_{\text{cond}} \sim L^2/\alpha$, where L is particle radius ($\sim 20 \mu\text{m}$) and $\alpha = k/(\rho c_p)$ is thermal diffusivity ($\sim 1 \times 10^{-4} \text{ m}^2\cdot\text{s}^{-1}$ for Al/Mg), as detailed in Table S2. For pure Al and Mg, the calculated conduction

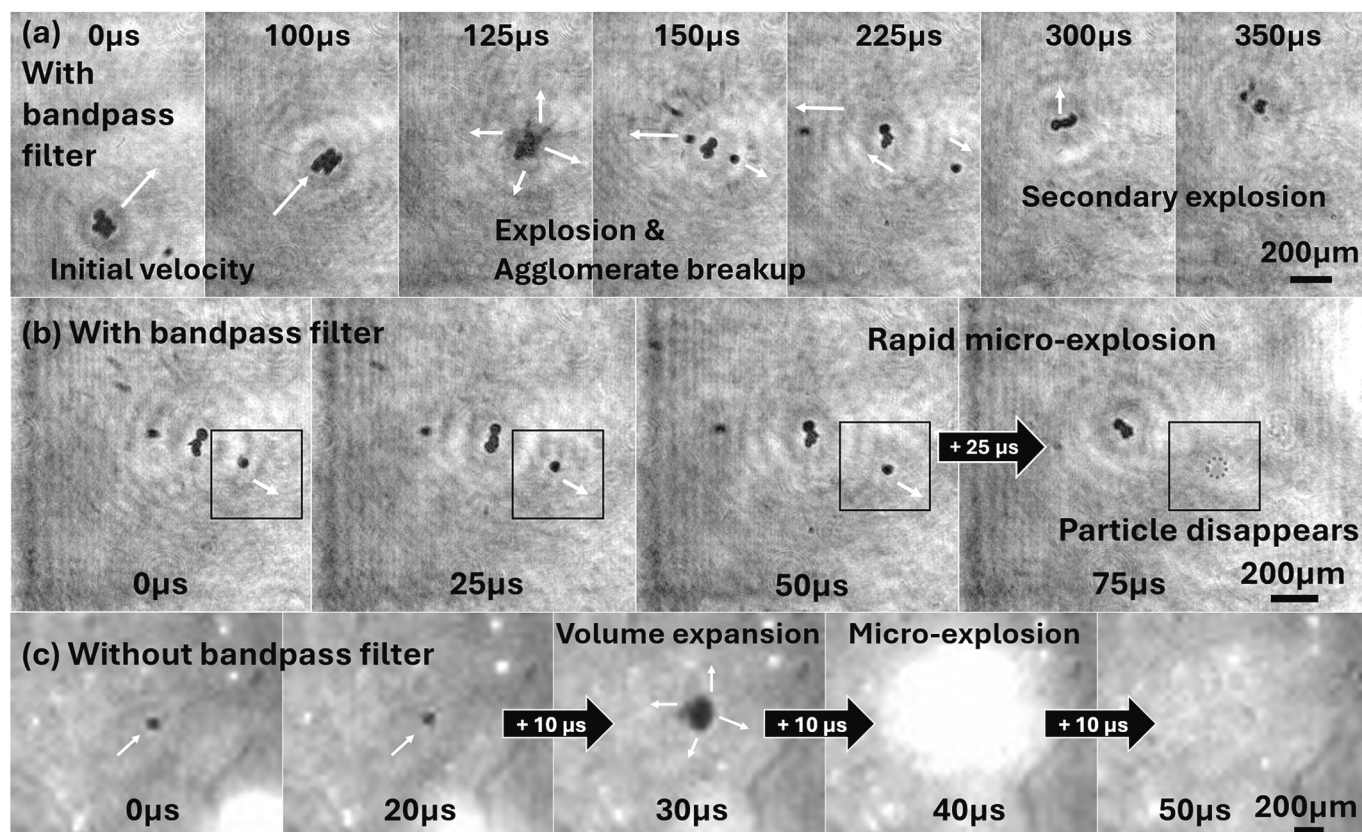


Fig. 4. DIH imaging the explosion process of Al-Mg powders in $\text{CH}_4/\text{O}_2/\text{Ar}$ post-flame, including (a) agglomerate explosion and breakup into secondary particles and (b-c) rapid micro-explosions of individual particles. Raw hologram stacks are in Fig. S3-S4.

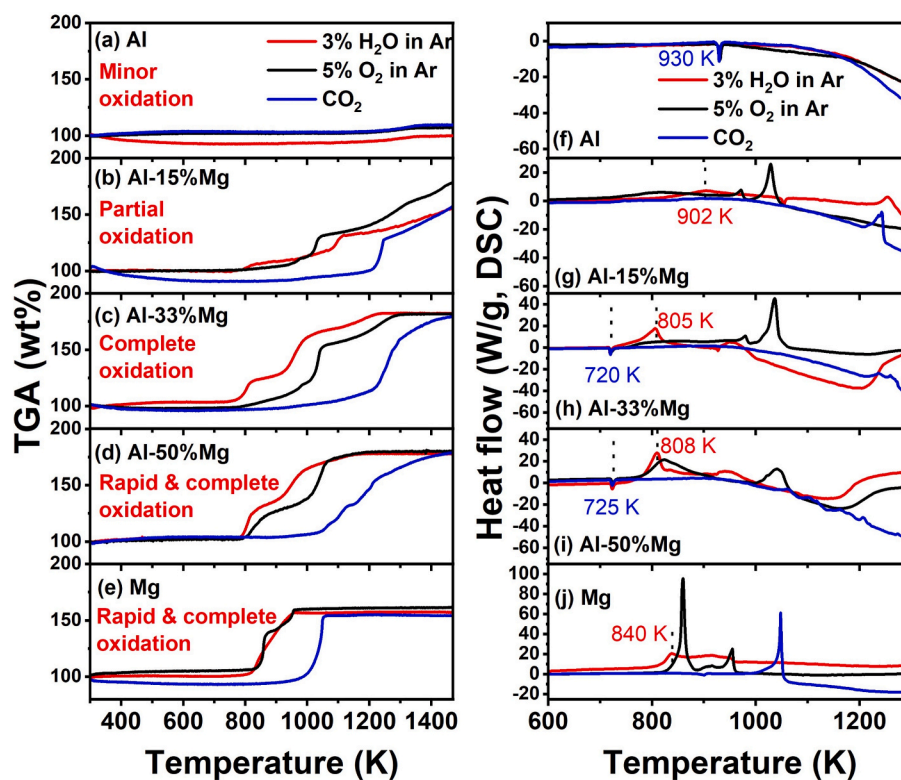


Fig. 5. TG and DSC of five metal powders spanning pure Al, Al-Mg alloys (Mg = 15, 33, and 50 wt%), and pure Mg in CO_2 (pure), H_2O (3% in Ar), and O_2 (5% in Ar) at a ramp rate of $10 \text{ K} \cdot \text{min}^{-1}$, as detailed in Fig. S6-S9. In the DSC figures (f-j), blue labels indicate endothermic melting features, whereas red labels indicate the first prominent exothermic features in the H_2O -containing environment.

times are $\sim 5 \mu\text{s}$, and thus comparable to the rapid volume expansion observed during the prompt micro-explosion events in Fig. 4 (c), which finish within $\sim 20 \mu\text{s}$. This comparison suggests that thermal energy within the particle can redistribute on a timescale equivalent to the explosive breakup process. Therefore, internal heat conduction is sufficiently fast to allow rapid superheating driven by surface oxidation, facilitating vapor generation within the molten particle prior to the observed micro-explosive fragmentation.

3.3. TG-DSC analysis and XRD of post-heating products

Thermogravimetric (TG) analysis of the five Al-Mg powders was conducted up to 1470 K at a heating rate of $10 \text{ K}\cdot\text{min}^{-1}$ under different oxidizing environments. According to the kinetic simulation shown in Fig. S2, the flame temperature under the present flow conditions can reach $\sim 2890 \text{ K}$. In the post-flame region where the particles burn, the O_2 concentration decreases to below $\sim 5\%$, while hot H_2O and CO_2 become the dominant oxidizing species. Therefore, Fig. 5 compares the TG and DSC responses of the five powders in H_2O vapor, CO_2 , and O_2 -containing environments. Because of the limited vapor pressure of H_2O at room temperature, Ar saturated with H_2O was used, corresponding to $\sim 3\%$ H_2O vapor in the total 50-sccm flow. Additional TG analysis, including onset temperature, leveling-off temperature, and oxidation extent, is provided in Fig. S6–S9.

For pure Al powders ($\sim 40 \mu\text{m}$), the sample mass varies within $\pm 5\%$ during heating in H_2O , CO_2 , and O_2 environments, indicating that oxidation is minimal within the temperature range studied. This behavior is consistent with the well-known protective character of the Al_2O_3 surface layer and the extremely high ignition temperatures reported for Al particles of similar sizes (approaching $\sim 2000 \text{ K}$) (Sundaram et al., 2016). When 15 wt% Mg is introduced, the alloy becomes noticeably more reactive, and partial oxidation occurs. When the Mg content reaches 33 wt% or higher, the powders can be completely oxidized below 1470 K. In addition, Mg-rich samples (33–100 wt% Mg) exhibit significantly faster oxidation rates in H_2O compared with CO_2 , indicating that steam is the most effective oxidizer among the combustion products of a CH_4 flame.

The corresponding DSC profiles provide complementary information on both phase transitions and oxidation-associated heat release (Fig. 5f–j). Endothermic features are observed at $\sim 930 \text{ K}$ for Al, 720 K for Al-33%Mg, and 725 K for Al-50%Mg. These temperatures agree closely with the calculated melting temperatures (Table 1 and Fig. 1), providing an experimental consistency check on the CALPHAD calculations. Distinct melting endotherms are not clearly resolved for Al-15% Mg and Mg, likely because oxidation-related exothermic heat release overlaps with or precedes the corresponding phase-transition signals. More importantly, the DSC measurements reveal a strong composition dependence in the onset of oxidation-associated heat release. In H_2O -containing flow, the first prominent exothermic feature occurs at $\sim 902 \text{ K}$ for Al-15%Mg, whereas Al-33%Mg, Al-50%Mg, and Mg exhibit early exothermic events at approximately 805, 808, and 840 K, respectively. Thus, the compositions containing $\geq 33 \text{ wt}\%$ Mg exhibit oxidation-associated heat release below $\sim 850 \text{ K}$, consistent with their substantially earlier and more rapid mass gain in TG. The thermal response is also strongly oxidizer-dependent. O_2 generally produces stronger exothermic features than H_2O , consistent with its larger oxidation enthalpy (Table 4), whereas oxidation by H_2O releases less heat because H_2 is formed as a reduced coproduct. In contrast, the major CO_2 -associated reaction features occur at substantially higher temperatures, consistent with the delayed oxidation observed in the corresponding TG curves. Together, the TG and DSC measurements show that increasing Mg content not only increases the ultimate oxidation extent but also substantially lowers the characteristic temperature for rapid reaction.

The oxidation products were further characterized using XRD after TG heating. The diffraction patterns are shown in Fig. S10 and Fig. S11,

and the quantitative phase fractions obtained from Rietveld refinement are summarized in Tables 2 and 3. For pure Al, the products consist primarily of Al and Al_2O_3 , consistent with the limited oxidation observed in TG. As Mg content increases, the dominant oxidation products gradually shift to MgO and MgAl_2O_4 spinel. In particular, MgAl_2O_4 becomes the major product phase for intermediate compositions (33–50 wt% Mg). The relative fractions of MgO and MgAl_2O_4 also depend on the oxidizing environment: the H_2O vapor promotes the formation of MgAl_2O_4 , whereas CO_2 oxidation produces a mixture of MgO and MgAl_2O_4 . It should be noted that directly collecting combustion products from particles burning in a fast-flow, high-temperature CH_4 flame is experimentally challenging. As a result, the product ratios obtained from TG experiments may differ from those formed under the flame conditions used in this study. Ruan et al., 2024 performed combustion of Al-Mg wire (200- μm diameter) in an H_2 flame and analyzed the burnt wire tips using SEM-EDS, showing that Mg is preferentially oxidized while Al can remain partially unreacted. Their observation supports the interpretation that Mg plays a dominant role in initiating oxidation reactions in Al-Mg alloys, while Al oxidation proceeds subsequently as the reaction progresses (Dreizin and Schoenitz, 2015; Ruan et al., 2024; Aly and Dreizin, 2015). Other studies suggest that Mg oxidation occurs preferentially in Al-Mg alloys, forming MgO and Al_2O_3 as the initial oxidation products (Wang et al., 2022) with subsequent MgAl_2O_4 spinel formation at higher temperatures through a solid-state reaction between MgO and Al_2O_3 (Wang et al., 2022). Because spinel formation involves solid-state diffusion between oxide phases, it may be more readily observed under slower heating conditions, such as TG experiments, than during the rapid combustion processes in the CH_4 flame.

Table 4 summarizes representative global reactions between Al-Mg alloys and the oxidizers O_2 , CO_2 , and H_2O , together with their thermodynamic properties. The calculated Gibbs free energies indicate that oxidation reactions with all three oxidizers remain thermodynamically favorable at both room temperature and elevated temperatures approaching the flame temperature ($\sim 2890 \text{ K}$). Among the three oxidizers, reactions with O_2 exhibit the largest negative enthalpy changes, indicating that O_2 oxidation is thermodynamically the most exothermic pathway for both Al and Mg. In contrast, oxidation reactions with CO_2 and H_2O are less exothermic because part of the reaction energy is consumed in reducing these oxidizers to CO or H_2 . This thermodynamic trend is also consistent with the DSC measurements in Fig. 5, where the O_2 -containing environment generally produces more intense exothermic features than H_2O . Despite this thermodynamic trend, the TG results show that oxidation reactions of Mg-rich alloys are kinetically more favorable towards H_2O . This is because steam oxidation can proceed through rapid heterogeneous reactions with lower activation barriers and may promote the formation of porous oxide structures that facilitate oxidizer transport to the metal surface (Dreizin and Schoenitz, 2015; Nie et al., 2016; Wang et al., 2026). Table 4 also provides additional support for Mg vaporization as the source of the rapid internal gas-volume increase observed during micro-explosion: Our DIH in Fig. 4 (c) captured the rapid volume expansion associated with internal pressure buildup.

Table 2

XRD refinement results of TG post-heating products of Al, Al-15%Mg, Al-33%Mg, Al-50%Mg and Mg in H_2O , as detailed in Fig. S10.

Powder composition	Al (wt%)	Al_2O_3 (wt%)	MgAl_2O_4 (wt%)	MgO (wt%)
Al 100 wt%	80%	20%	0%	0%
Al 85 wt%, Mg 15 wt%	0%	65%	35%	0%
Al 67 wt%, Mg 33 wt%	0%	0%	100%	0%
Al 50 wt%, Mg 50 wt%	0%	0%	80%	20%
Mg 100 wt%	0%	0%	0%	100%

Table 3

XRD refinement results of TG post-heating products of Al, Al-15%Mg, Al-33% Mg, Al-50%Mg and Mg in CO₂, as detailed in Fig. S11.

Powder composition	Al (wt%)	Al ₂ O ₃ (wt%)	MgAl ₂ O ₄ (wt%)	MgO (wt%)
Al 100 wt%	80%	20%	0%	0%
Al 85 wt%, Mg 15 wt%	0%	56%	31%	13%
Al 67 wt%, Mg 33 wt%	0%	16%	67%	17%
Al 50 wt%, Mg 50 wt%	0%	28%	39%	33%
Mg 100 wt%	0%	0%	0%	100%

Table 4

Global reactions of Al-Mg alloy particles with O₂, CO₂, and H₂O, and corresponding thermochemical properties (ΔH° , ΔS° , ΔG°).

Chemical reactions	ΔH° (kJ/g metal)	ΔS° (J·K ⁻¹ /g metal)	ΔG° at 298 K (kJ/g metal)	ΔG° at 2890 K (kJ/g metal)
2Al + 3 CO ₂ → Al ₂ O ₃ + 3CO	-15.3	-1.0	-15.0	-12.4
2 Al + 3 H ₂ O (g) → Al ₂ O ₃ + 3 H ₂	-17.6	-3.3	-16.6	-8.0
4Al + 3O ₂ → 2 Al ₂ O ₃	-31.0	-5.8	-29.3	-14.3
2Al + Mg + 4CO ₂ → MgAl ₂ O ₄ + 4CO	-14.9	-0.9	-14.7	-12.4
2 Al + Mg + 4 H ₂ O (g) → MgAl ₂ O ₄ + 4 H ₂	-17.0	-3.0	-16.1	-8.3
2Al + Mg + 2O ₂ (g) → MgAl ₂ O ₄	-29.4	-5.3	-27.8	-14.1
Mg + CO ₂ → MgO + CO	-13.1	-1.0	-12.8	-10.3
Mg + H ₂ O (g) → MgO + H ₂	-14.8	-2.7	-14.0	-7.0
2 Mg + O ₂ → 2MgO	-24.8	-4.5	-23.4	-11.7

However, none of the considered global reactions of Al or Mg with H₂O, CO₂, or O₂ produces a positive net change in gas-phase moles: reactions with H₂O and CO₂ convert the gaseous oxidizer to an equal molar amount of H₂ or CO ($\Delta n_{\text{gas}} = 0$), whereas reactions with O₂ consume gas ($\Delta n_{\text{gas}} < 0$). Thus, together with the measured particle temperatures

substantially exceeding the boiling temperatures of the Mg-rich alloys and the rapid volume expansion observed by DIH, Mg vaporization is the most plausible source of the internal gas-volume increase that drives micro-explosion.

3.4. Mechanism

Fig. 6 summarizes the proposed mechanisms governing the combustion and fragmentation behavior of Al-Mg particles. For particle agglomerates, as illustrated in Fig. 6 (a), as heating proceeds, Mg within the alloy is oxidized first due to its higher reactivity and more porous oxide shell than Al. The condensed-phase oxidation of Mg releases substantial heat locally, which promotes the vaporization of Mg. The resulting vapor generation is likely responsible for the observed disaggregation. Such breakup processes reduce particle agglomeration and increase the effective reactive surface area of the fuel.

For individual particles, as shown in Fig. 6 (b), the micro-explosions mechanism is likely driven by superheating-induced internal vaporization. During rapid heating in the flame, the Al-Mg particle melts and forms a liquid alloy core surrounded by an oxide shell. Because Mg has a much lower boiling temperature than Al, Mg preferentially vaporizes within the molten particle once the particle temperature exceeds the alloy boiling temperature. The buildup of Mg vapor generates internal pressure within the particle. When this pressure exceeds the mechanical strength of the surrounding oxide shell, the shell ruptures, producing a rapid micro-explosion that ejects molten fragments and vapor products.

The presence of Mg, therefore, plays three roles: (1) it promotes ignition and heat release through rapid, condensed-phase oxidation; (2) it breaks up agglomerates through both rapid, external evaporation and through internal evaporation that induces micro-explosive fragmentation; and (3) lastly it refines individual powders into much finer particles through micro-explosions, thus accelerating combustion.

4. Conclusion

The combustion behavior of Al-Mg alloy particles was investigated in a CH₄/O₂/Ar post-flame using high-speed imaging, three-color pyrometry, and DIH. The Al-Mg alloy particles were synthesized using a

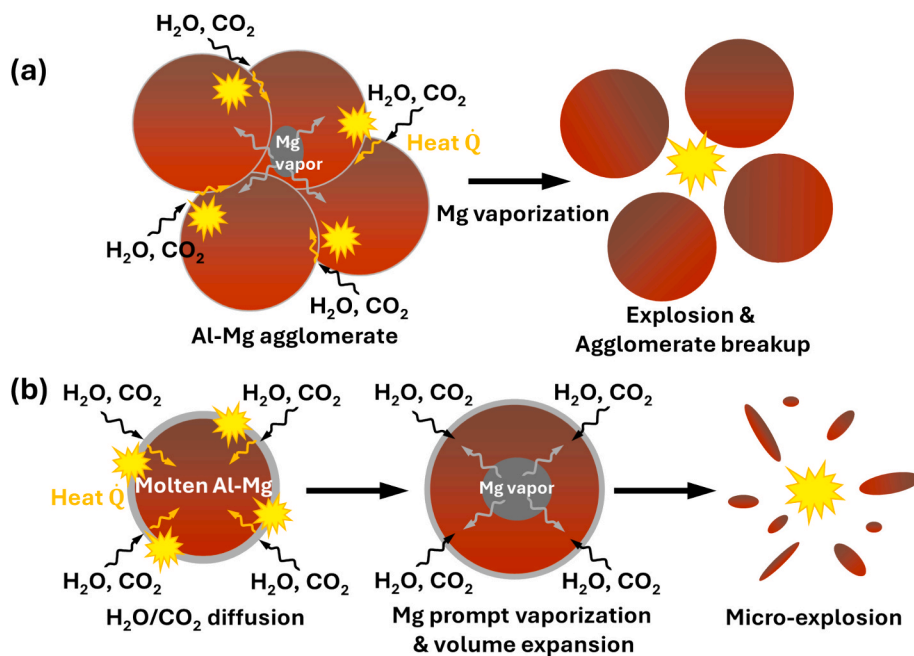


Fig. 6. Schematic illustration of the proposed combustion mechanisms of Al-Mg particles: (a) agglomerate breakup due to external evaporation and internal evaporation that leads to micro explosion and (b) single-particle micro-explosion that refines particle size and speeds combustion.

scalable ultrasonic atomization method, which enables controlled alloy composition and produces spherical particles suitable for combustion studies. A clear transition in combustion behavior occurs when the Mg content reaches ~ 33 wt%. Particles containing ≥ 33 wt% Mg exhibit significantly higher temperatures, frequent micro-explosions, and dramatically shorter burn times compared with Mg-lean particles. DIH imaging directly visualizes individual particle breakup and reveals that micro-explosion events can occur on microsecond timescales, with rapid particle expansion and fragmentation producing submicron-to-micron-scale combustion products below the detection limit. DIH also reveals that particle agglomerates undergo disaggregation during explosions, breaking apart into smaller fragments. These results support a mechanism in which preferential Mg vaporization within the molten alloy core generates internal pressure that ruptures the oxide shell and triggers micro-explosive fragmentation. Among the compositions studied, the 33 wt% Mg alloy provides a favorable balance between enhanced combustion reactivity and the high energy density of Al. The results also demonstrate that high-speed holography is a powerful tool for directly visualizing rapid explosion processes of micron-scale energetic materials.

CRedit authorship contribution statement

Lei Yang: Writing – original draft, Methodology, Formal analysis, Data curation. **Chetan Singh:** Writing – original draft, Data curation. **William V. Dam:** Data curation. **Keren Shi:** Data curation. **Yuxin Zhou:** Data curation. **Timothy P. Weihs:** Writing – review & editing, Resources, Methodology, Investigation, Conceptualization. **Michael R. Zachariah:** Writing – review & editing, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supporting material is free online. Supplementary material includes Al–Mg phase diagrams incorporating gas-phase equilibria, flame temperature and species profiles, raw and reconstructed holographic images, optical diagnostic configurations and spatial resolutions, thermogravimetric analysis under H₂O, CO₂, and O₂ environments, XRD characterization of post-heating products, particle combustion statistics, internal heat conduction timescale calculations, DIH reconstruction code, and high-speed combustion video (Figures S1–S11, Tables S1–S2, Supplementary Code S1, and Video S1). The videos for Fig. 3 are in the supplemental video S1. Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ces.2026.125332>.

Data availability

Data will be made available on request.

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