



Effect of gliding arc plasma on the combustion characteristics of co-firing of NH_3 and metal powders

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ABSTRACT

This study investigates the potential synergistic effects of iron powders and gliding arc plasma (GAP) in ammonia (NH_3)-air flame. Combustion behaviour was analysed using optical diagnostics, and the resulting solid particles were characterised through advanced material analysis techniques. For the first time, the combustion behaviours of iron powders in an NH_3 swirl flame were examined, and the effects of GAP on the resulting metal powder combustion products were evaluated. The results demonstrate that the addition of iron powders significantly increases flame zone and, like GAP, elevates the temperature at the bottom of the combustion chamber, with their combined effect leading to an increase of 88 K. Iron powders also produce a distinct emission peak around 600 nm, corresponding to iron oxide molecular bands. Furthermore, the high-temperature, high-energy regions generated by GAP discharges induce partial burning of iron particles, reducing the visible solid particle content. Post-combustion analysis reveals that initially dispersed Fe particles aggregate into block-like structures primarily composed of Fe_2O_3 and Fe_3O_4 , with sintering and agglomeration effects being more pronounced in GAP samples. These findings provide new insights into the thermal behaviour and morphology evolution of metal powders during combustion and may inform novel strategies for their processing and treatment.

Novelty and significance statement

In this work, the combustion behaviours of different metal powders (iron and electric arc furnaces) in an ammonia swirl flame were analysed for the first time, and the effects of a gliding arc plasma on the combustion products of the metal powders were investigated. Through the combination of combustion diagnostics and materials characterisation techniques, this work offers a novel perspective on the co-firing of ammonia with metal powders under plasma-assisted conditions.

1. Introduction

Ammonia (NH_3), as a carbon-free fuel with well-established production and transportation infrastructures, has recently attracted significant research interest [1]. However, NH_3 exhibits relatively low laminar flame speed, leading to poor flame stability that requires improvement [2]. Meanwhile, iron (Fe) powder, as another carbon-free

energy carrier, can address the intermittency issues of renewable energy sources by offering a stable, transportable, and safe solution, while enabling circularity through its regenerative redox cycle [3]. Its abundance and high volumetric energy density make it a promising candidate for sustainable energy storage [4]. After combustion, the resulting Fe oxides can be reduced back to metallic Fe using green hydrogen, creating a fully carbon-free cycle [4]. A safe transportation and storage system provides great potential for the large-scale application of both NH_3 and Fe powder combustion [5]. Furthermore, the stable combustion of Fe powder is constrained by particle size and feed rate; excessively large particle sizes or excessively low feed rates can prevent the flame from being stably anchored and self-sustained [4]. Prasad et al. [6] reported that, to ensure flame self-sustainability in the applied burner, the maximum allowable particle-to-particle distance should be approximately 0.5 mm, or the total Fe particle surface area per unit burner volume should be at least $0.02 \text{ mm}^2/\text{mm}^3$, for oxidizer oxygen concentrations ranging from 13.5 % to 21 %. This highlights the inherent limitations of achieving self-sustained combustion with pure Fe powder. However, in the presence of an existing flame environment, the issues of instability associated with metal particle combustion can be effectively

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resolved. The co-combustion of Fe powder and NH_3 aims to meet the fundamental requirement of zero carbon emissions [7] while addressing the low combustion reactivity of NH_3 fuel, thereby enhancing overall combustion efficiency. To explore the feasibility of achieving a fully zero-carbon Fe fuel cycle, Chen et al. [8] first investigated the combustion behaviour of single Fe particles under NH_3 co-combustion conditions. The results show that the probability of micro-explosions of Fe particles increases significantly in a high-temperature NH_3 atmosphere, while it decreases with increasing oxygen concentration in the surrounding gas mixture.

Active control of NH_3 flames has got considerable attention due to its potential significance in improving combustion efficiency and stability. Among various control strategies, the use of plasma-assisted techniques has recently emerged as a promising approach for regulating flame behaviour [9]. Plasma has been demonstrated to effectively enhance ignition performance and flame stabilisation across a wide range of combustion systems [10]. These findings indicate that plasma offers distinctive advantages in promoting the combustion characteristics of NH_3 flames. Specifically, Choe et al. [11] reported that nanosecond repetitively pulsed (NRP) discharges can substantially reduce NO_x emissions. To elucidate the promoting mechanisms of NRP on NH_3 -air flames, Sun et al. [12] investigated flame development under NRP-induced conditions on the nanosecond timescale. Gliding arc plasma (GAP), has also been explored for its potential to enhance NH_3 combustion [13]. GAP has demonstrated notable capability in reducing NO_x emissions through the application of an external electric field [14, 15]. This approach offers a structurally simple and cost-effective configuration, as it requires only the installation of an electrode at the gas inlet, making it particularly attractive for industrial applications. Regarding emission characteristics, Aravind et al. [14] observed substantial NO reduction in non-premixed NH_3 -air flames at low plasma power, where the plasma interacted exclusively with the NH_3 stream. Furthermore, Wang et al. [15] examined GAP effects on air-stratified NH_3 flames, and through measurements of NH_2^* and OH^* radicals, they provided preliminary interpretations of NO_x variation under plasma activation. Tang et al. [16] utilized NH-PLIF technology to demonstrate that plasma further extends the peak NH concentration to higher values. With plasma activation, the average peak NH concentration in the near-field region increased by $>30\%$. Apart from the aforementioned experimental studies on plasma-assisted combustion (PAC) of NH_3 , several numerical investigations have also been conducted to better understand the underlying emission mechanisms. Moreover, GAP significantly enhances the decomposition of NH_3 , particularly in the presence of metals, which act as catalysts. In particular, the presence of a Fe catalyst enables GAP to achieve a decomposition rate of up to 90% [17], it is expected that NH_2 and H would be formed from the gliding arc effect, which could also be affecting the reactivity of Fe. Wang et al. [17] also indicate that metallic Fe serves as the true active phase; however, it is readily deactivated through interaction with adsorbed nitrogen atoms, leading to the formation of the less active Fe_4N_{10} phase. Notably, plasma effectively accelerates N_2 desorption from the catalyst surface, confirming its strong role in regenerating active sites. Kinetic analyses have revealed that the three-step dehydrogenation reactions occurring on the Fe catalyst dominate the NH_3 decomposition process assisted by GAP [18]. Nevertheless, plasma-particle interactions are widely applied in the reduction of Fe oxides, aiming to promote decarbonization in the steel industry [19]. This effect could provide new insights into solving future challenges associated with particulate pollutants. For instance, in electric arc furnaces (EAF), the steelmaking industry produces several million tons of EAF dust each year. This dust contains valuable metallic elements (e. g., Fe) yet also poses environmental risks due to the presence of hazardous metals such as Zn, Pb, and others [20].

This study aims to clarify whether synergistic interaction exists between metal powder and GAP in NH_3 flame systems. To the knowledge of the authors, this is the first work of the kind combining NH_3 , Fe and

GAP. Comparative experiments were conducted for NH_3 -Fe/EAF flames under GAP activation. The evolution of reactive intermediates was investigated using optical emission spectroscopy and high-speed chemiluminescence imaging of NH_2^* , while post-combustion solid residues were characterized to elucidate the role of Fe in NH_3 co-combustion under GAP-assisted conditions, using scanning electron microscopy (SEM) and X-ray diffraction (XRD). A novel approach for the treatment of metal powders was proposed.

2. Experiment setup

2.1. Experimental apparatus

Experimental investigations were carried out using a newly developed GAP-assisted dual-swirl combustor at the Centre of Excellence on Ammonia Technologies (CEAT), Cardiff University. A thorough description of the combustor design, including flow configurations, swirl generation, and ignition system, has been provided in our previous works [14,15], the details can be found in Fig. 1. In the combustion unit, the flow of NH_3 and air was controlled using high precision Alicat mass flow controller achieving an accuracy of $\pm 0.5\%$ within 95% of the full scale as shown in Fig. 1(a). Two radial co-swirlers having swirl numbers of 1.0 each were positioned 125 mm and 40 mm from the dump plane for inner and outer swirlers as shown in Fig. 1(b). The NH_3 is introduced through the central lance via the inner swirler, while air flows through the outer swirler and exits at the dump plane. This configuration typically results in a non-premixed flame. Plasma is primarily generated within the annular gap between the central lance and a conical electrode, which also serves as a bluff body. A high-voltage AC power supply (CTP-6 2000KA, ACS Materials, USA) is used to generate the gliding arc plasma. The change in input plasma power was accomplished by adjusting the input voltage of the power supply. Voltage and current waveforms were captured using a Tektronix MSO64B oscilloscope connected to a Tektronix P6015 high-voltage probe and a Cybertek HCP 8030 current probe (5 A, 1 V/A). During the experiment, the input plasma power was calculated in real time based on the voltage and current curves to keep it constant. In this experiment, the input frequency was kept constant at 13 kHz, and the plasma input power was controlled to be between 100–110 W. At the central lance of 16 mm diameter, a conical electrode made of tungsten-steel alloy with a maximum diameter of 11 mm was installed. A minimum electrode-to-wall gap of 2.5 mm was observed at breakdown. The arc was stretched upward by the gas flow until reaching its maximum length, after which it was blown out and the next breakdown followed. The flame is stabilised inside a quartz tube with a diameter of 58 mm and a height of 320 mm. The experimental study employed metallic powders consisting of elemental Fe and iron oxide (Fe_2O_3) particles. The Fe powders had a particle size distribution ranging from 20 to 25 μm , with relatively uniform morphology. The elemental composition of EAF dust is mainly composed of iron (Fe) and zinc (Zn), which were identified as

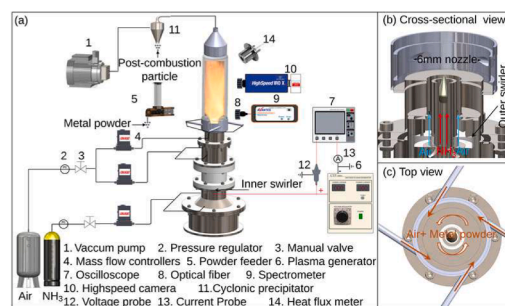


Fig. 1. Experiment setup of a plasma assisted NH_3 cofiring with metal powder. (a) the overall system, (b) cross-sectional view of combustor, (c) top view of combustor.

the dominant components, accounting for 26.0 wt.% and 27.6 wt.%, respectively. In addition, small amounts of other metallic elements, including calcium (Ca) (4.1 wt.%) and manganese (Mn) (1.9 wt.%), were also detected. Powder delivery was achieved using a small screw-type feeder, which allowed for precise control of the powder feed rate. After discharge from the feeder, the powders were transported into the combustor through a dedicated gas transport line, ensuring minimal agglomeration or sedimentation. Gas temperature (T) was measured with thermocouples (± 1.5 K accuracy) [21] at points along the axis inside the quartz tube, spaced every 80 mm. To further investigate the heat transfer characteristics, the total heat flux was measured at the same location as the thermocouple to ensure direct comparison with the flame temperature measurements. Heat flux measurements were performed using an SBG01-020 heat flux sensor (from Hukse Sensor Technology). The sensor has a nominal sensitivity of 0.3×10^{-6} V/(W·m⁻²), with a calibration uncertainty of $\pm 0.017 \times 10^{-6}$ V/(W·m⁻²). The measurement range is 0–20 kW/m². The exact measurement locations are indicated in Fig. 2. To enhance the residence time of the powder within the quartz tube, the air-powder mixture was introduced tangentially through four equally spaced inlets, which were distributed around a stainless-steel ring located between the combustor outlet and the bottom of the quartz tube. This configuration induced a swirling motion that promoted uniform suspension of particles and improved interaction with the reactive gas environment. Flame emission spectra were acquired using an AvaSpec-ULS spectrometer coupled with a UV–visible optical fibre. The optical probe was positioned 25 mm above the combustion chamber inlet at a working distance of 100 mm from the central axis, resulting in a sampling spot of approximately 2–3 mm in diameter within a 30×38 mm² measurement region. The combustor was ignited using a methane–air flame. Once ignition was achieved, the methane flow was gradually decreased while the NH₃ flow was increased until the methane was completely replaced and a stable NH₃ flame was established. Iron powder (or EAF dust) was then introduced into the combustor at a constant feed rate using the carrier gas. After the co-combustion flame became stable, the GAP system was switched on. All optical, temperature, and emission measurements were acquired only after the operating conditions had remained stable for at least 60 s. For solid product characterization, combustion was maintained under identical operating conditions, and solid products were collected after 5, 10, and 15 min of stable operation for subsequent analysis. Specifically, the carrier airflow was supplied independently via a mass flow controller and maintained at 4 SLPM, corresponding to a solid feed rate of approximately 0.3 g/min. Assuming complete oxidation of Fe-to-Fe₂O₃, the Fe particles consumed an additional oxygen amount of approximately 0.1 SLPM, corresponding to an additional air requirement of about 0.4 SLPM. Therefore, the overall stoichiometric air requirement was estimated to be 72 SLPM, resulting in an effective equivalence ratio of approximately ($\phi=1.01$). The addition of Fe particles therefore had a negligible influence on the global equivalence ratio, and the combustion condition can be considered nearly stoichiometric. A vacuum pump was used to draw the exhaust gases from the outlet of the combustion chamber through the probe, allowing for efficient collection of both particulate and gaseous combustion products for subsequent analysis. This setup ensured accurate measurement of emissions while minimizing particle loss along the sampling line. The combustion power of metal powder is around 20 W. The NH₃ flow rate was maintained at 20 SLPM, the combustion air flow rate was fixed at 71–102 SLPM (corresponding to an equivalence ratio of ($\phi_g = 0.7$ –1.0)), and the combustion power of NH₃ remained constant throughout all experiments.

2.2. Data processing method

The combustion products of the solid powders were collected using an emission sampling probe, which was connected to a cyclonic precipitator.

Post co-firing particulate image analysis was carried out using scanning electron microscopy coupled with energy dispersive spectroscopy (SEM-EDS), which included SEM: Zeiss Sigma HD field emission gun SEM, 20 kV beam energy, ~ 1.5 nA beam current using a 60 μ m diameter final aperture. For chemical analysis, samples were mounted on aluminium SEM stubs using carbon adhesive tabs, coated with a 15 nm thick layer of carbon prior to being loaded in the SEM. Two 150 mm² Oxford Instruments X-Max silicon drift detectors were used for semi-quantitative Energy Dispersive Spectroscopy (EDS) analysis. Backscattered electron images and EDS spectra were acquired and processed using Oxford Instruments Aztec software. EDS results were quantified using factory standards and data was normalised to 100 %.

NH₂* chemiluminescence images were captured using a LaVision high-speed camera equipped with a 630 nm filter at a frame rate of 4000 Hz. For each operating condition, 4000 consecutive images were recorded, corresponding to a total acquisition time of 1 s. The root-mean-square (RMS) intensity and the normalized RMS values are used to evaluate the stability of the reaction zone. A set of time-resolved images of NH₂* chemiluminescence are recorded as $I(x, y, t)$, the time-averaged intensity and the RMS fluctuation are computed by the following equations:

$$\bar{I} = \frac{1}{N} \sum_{t=1}^N I(x, y, t) \quad (1)$$

$$I_{RMS(x,y)} = \sqrt{\frac{1}{N} \sum_{t=1}^N (I(x, y, t) - \bar{I}(x, y))^2} \quad (2)$$

3. Result and discussion

3.1. Flame morphology

The flame morphology and temperature measurements of NH₃ and Fe powder cofiring under plasma-assisted conditions are shown in Fig. 2. The flame images of Fe and EAF powders in NH₃ exhibit no significant differences and EAF powders are therefore not shown.

The classic NH₃ flame is shown in Fig. 2(a). Under the current NH₃ and air flow conditions ($\phi_g = 1$), the visible flame height reaches only about 200 mm, roughly 2/3 of the quartz tube length. Meanwhile, the overall flame exhibits a dull orange colour. The addition of Fe powder significantly broadens the flame, with the flame covering the entire quartz tube, as shown in Fig. 2(b). The temperature exhibits a consistent trend across different operating conditions, rising monotonically with increasing height. Similarly, as shown in Fig. S1, the heat flux generally increases from the lower to the intermediate axial positions, reaching approximately 3.1 kW/m² at Test Point 3 for both cases. This increase

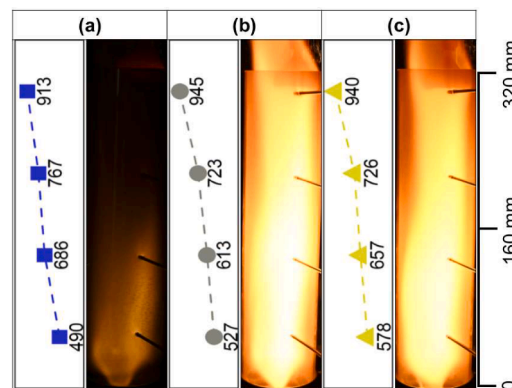


Fig. 2. Variation of flame morphology and near wall temperature measurement (K) with different conditions. (a) pure NH₃, (b) Fe powder cofiring with NH₃, (c) GAP assisted Fe powder cofiring with NH₃.

indicates enhanced heat transfer at the measurement location with increasing axial distance and is consistent with the experimentally observed increase in flame temperature. During its upward movement, the fuel gradually reacts with oxygen, releasing heat; together with the ascent of hot gases and convey effects, this causes the flame temperature to increase with height. The effect of GAP on flame morphology is more pronounced at the base of the flame. Temperature measurements also confirm this observation: as shown in Fig. 2(c), the flame base temperature increases by 88 K compared to that in Fig. 2(a). This temperature rise can be attributed to changes in power density caused by the thermal effects of electron collisions, as reported in previous studies [22], indicating that the influence of GAP on temperature cannot be neglected. Although the GAP increased the measured bottom flame temperature by approximately 51 K for the Fe powder cofiring with NH_3 case, its electrical power input (100–110 W) accounts for only about 2 % of the thermal power of the NH_3 flame (approximately 4.3 kW). Even if the entire plasma power were converted into sensible heat, the theoretical temperature rise would be 30–40 K [22]. Therefore, the observed temperature increase is not solely a thermal effect but is mainly attributed to plasma-induced enhancement of chemical reactions, including the generation of reactive radicals and accelerated chain-branching reactions, which increase local heat release and improve flame stabilization. In the authors' previous numerical study [22], the gliding arc plasma was found to increase the gas temperature of the NH_3 flame by approximately 50 K, as shown in Fig. S2 of the supplementary material. Similarly, experimental measurements by Alkhalifa et al. [23] also demonstrated a strong correlation between the plasma discharge power and the increase in gas temperature. The detailed calculation method for the gas temperature also has been included in the Supplementary Material. These observations indicate that GAP has a significant impact on both flame morphology and temperature distribution.

3.2. Optical diagnostics of combustion

Optical diagnostics were employed to analyse the cofiring of NH_3 and metal powders in GAP. The spectral measurements under different operating conditions are shown in Fig. 3, with a measurement range of 175–1100 nm.

The emission spectrum of a typical NH_3 flame exhibits several sharp peaks in the 600–800 nm region, associated with the NH_2 (α band, $^2\text{A}_1\text{--}^2\text{B}_1$) [15]. However, in contrast to the spectrum with Fe powder, the emission spectrum of pure NH_3 appears relatively featureless, showing an almost linear baseline in Fig. 3(a). This may also be attributed to the low combustion power of the flame. Molecular bands of FeO and spanning 550–650 nm contribute significantly to the red-orange colour

of the flame. According to the NIST Atomic Spectra Database [24], the dominant Fe I emission lines in the 580–620 nm wavelength range are located at approximately 593.2, 602.8, 606.6, and 613.8 nm, which may overlap with the broad FeO molecular emission band. Furthermore, comparison with the extracted Fe I emission spectra suggests that the potential contribution of atomic Fe emission to the measured spectra is relatively small compared with the dominant FeO molecular band. Therefore, the observed red-orange flame emission is primarily the result of the combined effects of the FeO molecular bands together with the superimposed sharp emission lines of Fe atoms [5], a similar spectral peak can also be observed in Fig. 3(b). Both molecular and atomic Fe species contribute significantly to the overall emission characteristics of the combustion process. The spectral characteristics of EAF powder with and without plasma is shown in Fig. 3(b). Similar to the Fe powder case, the introduction of plasma significantly enhances the overall spectral intensity. However, the spectral distribution becomes more complex, with additional emission features appearing in the 800–1000 nm region. This phenomenon is likely related to the complex composition and the presence of multiple metallic elements and impurities in the EAF powder. Overall, the introduction of plasma significantly enhances the radiative emission intensity during metal powder combustion and modifies the distribution of characteristic emission peaks, while the EAF powder exhibits richer spectral features due to its more complex composition.

In Fig. 4, the NH_2^* chemiluminescence images obtained by a high-speed camera during the co-combustion of Fe powder and NH_3 with and without plasma are compared. The bright spot counts in the images are further extracted for analysis.

In Fig. 4(a), under no plasma conditions, dense scattering spots appear at the bottom of the flame, indicating a high concentration of suspended Fe particles. When the GAP is applied, these scattering features are greatly reduced, showing fewer and more uniformly distributed particles in the flame, as it shown in Fig. 4(b). This difference mainly results from the multiple effects of the GAP on the Fe powder system. The high-temperature and high-energy region produced by the discharge increases the local gas and electron temperatures (up to 1–2 eV), probably inducing partial burning of iron particles [25], which reduces the number of solid particles. In addition, high-energy electrons and reactive species (N, H, O, OH, NH_2) rapidly react with particle surfaces via oxidation, nitridation, and reduction. The resulting volatile products promote particle fragmentation. The high-speed plasma jet also enhances mixing and turbulence, driving particles out of the main reaction zone and reducing their local concentration [26]. It is noteworthy that, under the influence of GAP, the number of luminous spots exhibits a quasi-periodic variation, as evidenced by the successive triangular peaks observed in Fig. 4(b). This behaviour is likely associated with the intrinsic periodicity of GAP generation, suggesting that temporal fluctuations in the input power density exert a significant influence on the combustion characteristics of the metal powders. Specifically, the periodic generation of GAP may lead to intermittent enhancements in the local energy supply [26], which in turn periodically intensifies the ignition and combustion processes of individual Fe particles. During intervals of increased power density, a larger number of particles can reach the ignition threshold, resulting in a rapid rise in the number of luminous spots and the formation of the triangular peaks observed in the temporal profile. Conversely, when the input power density decreases, the energy available for sustaining particle ignition and combustion diminishes, leading to a reduction in the number of visible luminous spots. This cyclic increase and decrease indicate that the combustion process is not purely steady but is dynamically modulated by the temporal characteristics of the energy input. Such modulation may influence not only the ignition frequency of particles but also the spatial distribution and lifetime of the luminous spots. Consequently, the observed quasi-periodic behaviour reflects a strong coupling between the plasma generation dynamics of GAP and the combustion response of the metal powder particles. Further investigation into this coupling

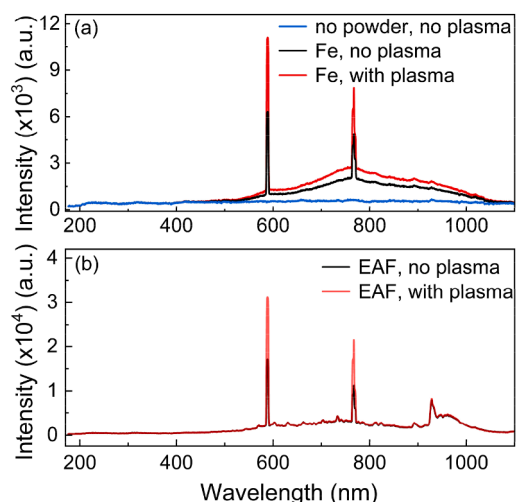


Fig. 3. Spectra of NH_3 -Fe/EAF cofiring in GAP.

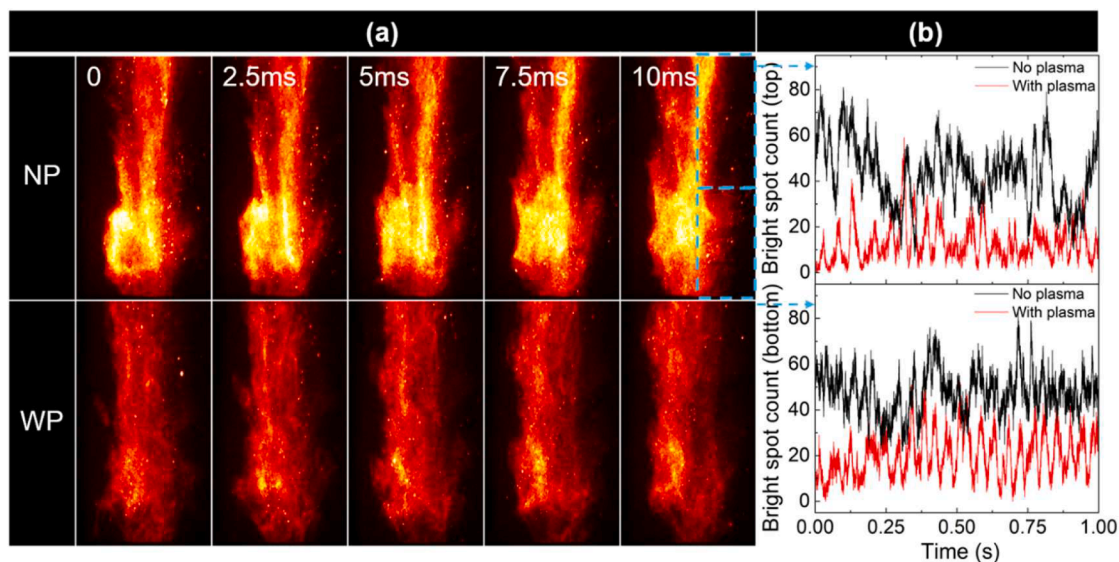


Fig. 4. NH_2^* images of NH_3 -Fe cofiring in GAP. (a) instantaneous images, (b) bright spot count of top and bottom part.

mechanism could provide deeper insights into the energy transfer processes and the stability of particle combustion under GAP-assisted conditions.

To clarify the coupling between the GAP discharge and flame dynamics, synchronized electrical measurements (voltage and current) have been included together with the bright spot count, as shown in Fig. 5. The voltage and current signals were recorded simultaneously with the high-speed imaging. Two representative discharge events are highlighted by the dashed lines. It can be observed that each discharge event corresponds to distinct electrical signatures characterized by simultaneous voltage fluctuations and current spikes. Following these discharge events, the number of bright spots increases gradually rather than instantaneously, indicating that the flame response is governed by the afterglow chemistry and transport of plasma-generated reactive species instead of the transient discharge pulse itself. This synchronized measurement therefore confirms the temporal coupling between the

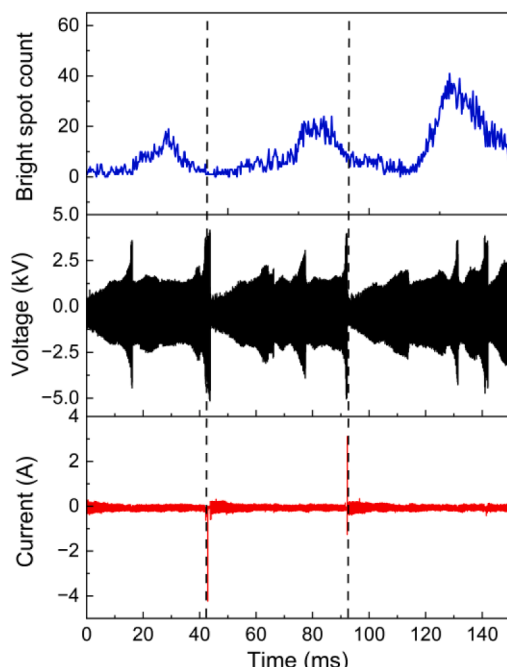


Fig. 5. Detailed V-I measurement with bright spot count.

GAP discharge and the evolution of flame bright spots while demonstrating that the flame dynamics occur on a longer timescale than the electrical discharge. It should be noted that the discharge operates at an AC frequency of 13 kHz, whereas the quasi-periodic variation of bright spot count occurs on a much longer timescale (tens of milliseconds). Therefore, the observed bright spot oscillation is not directly associated with the electrical excitation frequency but rather reflects the cumulative effects of plasma-induced radical production, transport, and subsequent flame response.

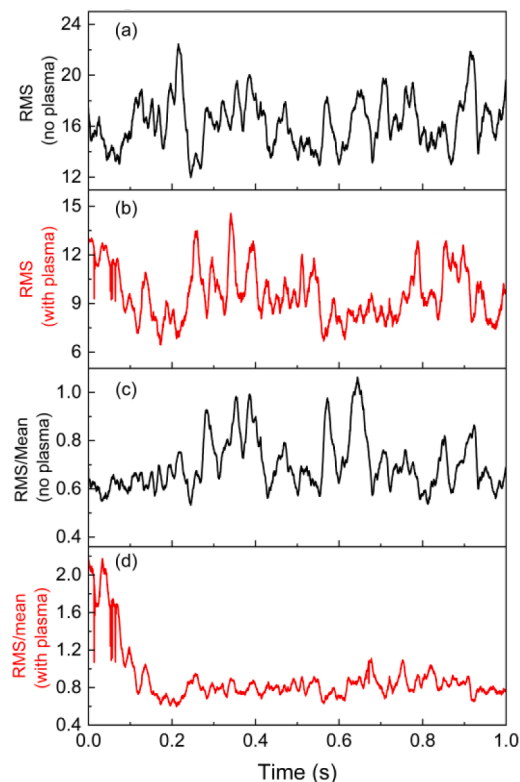


Fig. 6. Temporal evolution of NH_2^* chemiluminescence fluctuation during combustion. (a) RMS (no plasma), (b) RMS (with plasma), (c) normalized fluctuation intensity (RMS/Mean) (no plasma), (d) normalized fluctuation intensity (RMS/Mean) (with plasma).

Fig. 6 presents the temporal evolution of the NH_2^* chemiluminescence fluctuation during the combustion process.

As shown in Fig. 6(a), the RMS intensity of NH_2^* chemiluminescence exhibits significant temporal fluctuations, ranging approximately from 13 to 22 within the observation period. These large fluctuations indicate that the reaction zone is relatively unstable, which may be associated with the unsteady decomposition of NH_3 and the heterogeneous combustion behaviour of Fe particles.

In contrast, Fig. 6(b) shows that the RMS intensity is noticeably reduced, with values mainly distributed between 7 and 14. The decrease in RMS amplitude suggests that the fluctuation of the NH_2^* chemiluminescence becomes weaker, indicating an improvement in the stability of the reaction zone. This result implies that the combustion process becomes more stable under the modified operating conditions. The normalized fluctuation intensity, expressed as RMS/Mean, is illustrated in Fig. 6(c). The normalized RMS values fluctuate within the range of approximately 0.55–1.05. Compared with the absolute RMS intensity, the normalized parameter more clearly reflects the relative fluctuation of the chemiluminescence signal by eliminating the influence of overall intensity variation. The relatively moderate fluctuations indicate that the NH_2^* reaction zone maintains a quasi-stable structure during most of the combustion period. In Fig. 6(d), A rapid decrease is observed at the initial stage ($t < 0.2$ s), where the value drops sharply from about 2.1 to approximately 0.7. This behaviour corresponds to the ignition and transient establishment of the reaction zone, during which the decomposition of NH_3 and the ignition of Fe particles occur simultaneously. After this transient period, the value gradually stabilizes around 0.7–0.9, indicating that the combustion system enters a relatively steady state.

3.3. Characteristics of combustion products

The scanning electron microscopy (SEM) images of the original Fe powder, the combustion products of the Fe/EAF powder mixture in GAP are shown in Fig. 7.

The Fe powder shows a uniform dispersion of fine particles across the 500 μm scale. Individual particles are well-separated, mostly in their native micron-sized form, with no visible aggregation or coalescence. The surface texture appears relatively smooth, as it is shown in Fig. 7(a). By comparing Fig. 7(a) and (b), it can be observed that after combustion, the collected solid powder transforms from fine dispersed Fe particles into aggregated, block-like structures. These structures are irregular in shape but much more compact than the original particles. The increased particle size suggests sintering and oxide formation (Fe_2O_3 and Fe_3O_4), with interparticle gaps largely filled, leading to dense clusters. The observed phenomenon is mainly attributed to the sintering behaviour of micron-sized Fe particles during combustion [8]. Sintering of Fe initiates

at approximately 848 K and intensifies exponentially as the temperature increases. Another reason is that the metal oxides formed after combustion exhibit an increased molar volume which fills the interparticle gaps and results in the formation of a compact, block-like agglomerated oxide layer. This phenomenon is more pronounced in the GAP samples. As shown by the comparison between Fig. 7(b) and (e), the maximum size of the melting point (≈ 1811 K for Fe). Under such conditions, surface tension acts to minimize the surface energy of the molten droplets, resulting in the formation of stable, spherical particles [4]. Compared with pure Fe powder, EAF powder exhibits significantly less agglomeration behaviour after combustion in Fig. 7(d). This is mainly attributed to its heterogeneous initial particle morphology, complex composition (including Zn, impurities, and oxides), insufficient local temperature and melting, and the formation of pores due to surface chemical reactions during combustion. Fe powder particles are uniform and primarily consist of single-component Fe; during combustion, the local high temperature can partially melt the particles, and oxidation-induced expansion helps fill the interparticle voids, ultimately forming a dense agglomerated structure. In contrast, EAF powder has irregular particle morphology, high impurity content, and limited local melting, while Zn and other components volatilize or are reduced at high temperatures to form pores, hindering close particle bonding. As a result, EAF powder tends to form loosely aggregated structures rather than large, sintered blocks after combustion. The particle size distributions indicate a peak broadening on combustion of the iron dust consistent with sintering of the dust, and which appears to be greater with plasma than without plasma in Fig. 8. Specifically, plasma assistance promoted the sintering and agglomeration of the dust particles, resulting in a particle size distribution that was predominantly concentrated in the 92–170 μm range. In contrast, the combustion residues obtained without plasma assistance exhibited a much finer particle size distribution, with the majority of the particle volume fraction distributed in 30–92 μm . The ability to recover the metal oxide dust by reduction of the oxide after combustion will be important to the success of this approach; however, increases in the particle size distribution will affect the reuse of the dust and will need mitigation. Although the precise chemical pathways remain to be elucidated, this result highlights a promising route for in-situ reduction reactions in NH_3 flames.

The XRD patterns of the raw Fe powder and the corresponding combustion products obtained under different conditions are presented in Fig. 9.

As shown in Fig. 9(a), the pristine sample exhibits three prominent diffraction peaks located at 2θ angles of approximately 44° , 65° , and 82° , which can be indexed to the (110), (200), and (211) planes of body-centred cubic $\alpha\text{-Fe}$ [27]. The sharp and intense peaks indicate that the starting material mainly consists of highly crystalline metallic Fe without detectable secondary phases. After combustion in the NH_3 -air

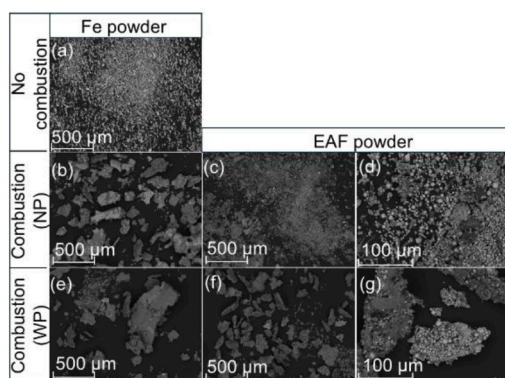


Fig. 7. SEM images of combustion product. (a) pure Fe powder, (b) Fe powder cofiring with NH_3 , (c-d) EAF powder cofiring with NH_3 , (e) GAP assisted Fe powder cofiring with NH_3 , (f-g) GAP assisted EAF powder cofiring with NH_3 .

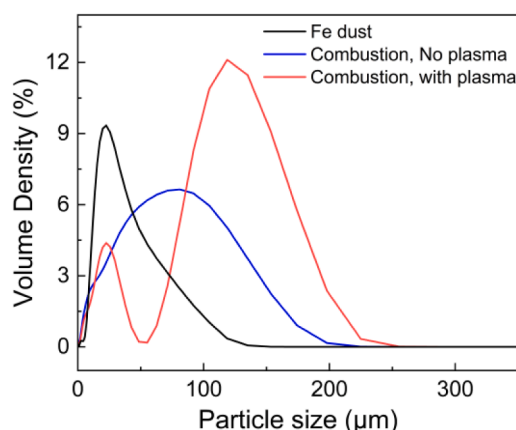


Fig. 8. Particle size distributions of combustion product.

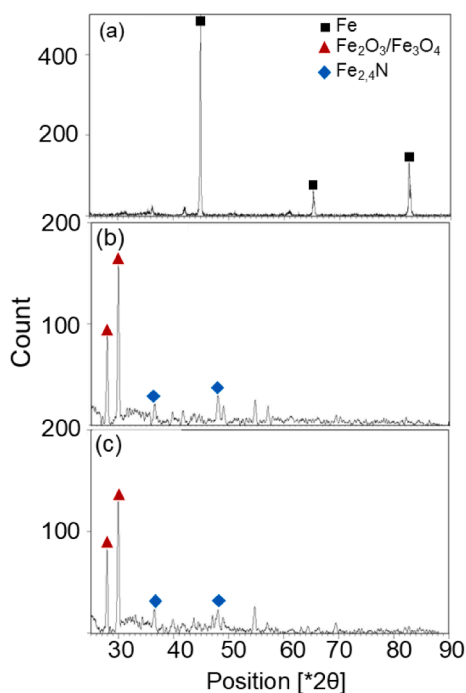


Fig. 9. XRD patterns of Fe powder and the combustion products under different conditions. (a) Raw Fe powder; (b) Combustion product in NH_3 -air atmosphere; (c) Combustion product under GAP-assisted NH_3 -air conditions.

atmosphere without plasma assistance in Fig. 9(b), the intensity of the α -Fe diffraction peaks decreases markedly, accompanied by the emergence of new peaks in the range of 29 – 31° these diffraction peaks can be assigned to Fe oxide phases, mainly Fe_3O_4 and Fe_2O_3 [28]. The appearance of these oxide phases indicates that oxidation reactions dominate during the combustion of Fe particles in the presence of oxygen and were also observed to correspond to a broadening amorphous peak/weakening crystal structure in the region below 25° . Under NH_3 -air combustion conditions, although NH_3 may provide a nitrogen source, the relatively high oxygen partial pressure promotes rapid oxidation of Fe, leading to the preferential formation of Fe oxides. In contrast, when GAP is introduced in Fig. 9(c), significant changes in the diffraction pattern are observed. The characteristic peaks corresponding to Fe oxides become weaker compared with those in Fig. 9(b), while additional diffraction peaks located around 37° and 49° show a slight increase in intensity. These peaks could possibly be attributed to Fe nitride phases, such as $\text{Fe}_{2.4}\text{N}$, noted in the literature [29]. However, the signal to noise ratio makes it difficult to distinguish the smaller diffraction features in this region with certainty, and although the Rietveld semi quantitative analysis of the trace was 3 % for these features, there is a low certainty attached to this. Meanwhile, a portion of the metallic Fe peaks remains detectable, suggesting that the oxidation degree of Fe is partially suppressed under plasma-assisted conditions. The formation of Fe nitrides under plasma conditions could be attributed to the enhanced activation of NH_3 molecules in GAP. The plasma environment promotes the dissociation of NH_3 into reactive nitrogen-containing species, including N, NH, and NH_2 radicals. These active species significantly facilitate nitridation reactions between Fe and nitrogen.

Moreover, hydrogen-containing radicals generated from NH_3 decomposition (H and H_2) may provide a locally reducing environment, which can partially inhibit the oxidation of Fe particles. Consequently, the introduction of GAP could alter the dominant reaction pathway from pure oxidation to a competitive oxidation-nitridation process, resulting in the formation of Fe nitride phases in the combustion products.

Future studies should aim to clarify the detailed synergistic

mechanisms between GAP and Fe/EAF powders through kinetic modelling. Further investigation into emission control and post-combustion particle recycling will help expand the practical applications of plasma-assisted metal- NH_3 combustion systems.

4. Conclusion

The objective of this study is to elucidate whether Fe/EAF powders and GAP exhibit synergistic effects in NH_3 flame systems. Optical diagnostic tools were applied to study the combustion behaviour, and the combustion product of particles were characterized using advanced material analysis techniques. The principal outcomes obtained from this work are listed below:

1. Incorporating Fe powders significantly enhanced the flame zone. Similar to GAP, this addition considerably elevated the bottom temperature of the combustion chamber, and the synergistic effect of both resulted in an 88 K increase.
2. In the presence of Fe/EAF powders, a distinct emission peak at approximately 600 nm was observed, attributable to FeO molecular bands, which imparts a red-orange appearance to the flame. Meanwhile, the high-temperature, high-energy regions generated by GAP discharges caused some Fe particles to melt, effectively reducing the visible solid particle content in the flame.
3. After combustion, dispersed metal particles transform into aggregated, block-like structures mainly composed of Fe_2O_3 and Fe_3O_4 . And the sintering and agglomeration effect is more pronounced in GAP. In contrast, EAF dust exhibited irregular particle morphology and limited local melting. After combustion, the residues showed porous, loosely aggregated structures rather than large, densely sintered blocks. Reactive nitrogen-containing species generated by plasma excitation may partially inhibit Fe oxidation, thereby favouring the formation of Fe nitride phases. This finding could offer novel perspectives for the processing of metal powders.

Based on the quasi-periodic variations of luminous spots observed in metal powders under GAP assistance in this study, future work could focus on the following aspects: a detailed investigation of the intrinsic periodicity of GAP generation and its energy release mechanisms to elucidate the coupling between plasma dynamics and particle ignition; and a systematic study of the effects of input power amplitude, frequency, and waveform on the combustion behaviour of individual particles and particle ensembles, combined with high-speed imaging and single-particle combustion analysis, to quantitatively assess the influence of plasma on particle ignition rate, combustion intensity, and luminous characteristics.

CRedit authorship contribution statement

Ziyu Wang: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. B. Aravind: Writing – review & editing, Supervision, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. Julian Steer: Writing – review & editing, Supervision, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. Syed Mashruk: Writing – review & editing, Resources. Agustin Valera-Medina: Writing – review & editing, Supervision, Resources, Funding acquisition.

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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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at [doi:10.1016/j.combustflame.2026.115378](https://doi.org/10.1016/j.combustflame.2026.115378).

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