ABSTRACT

Carbon-free chemical energy carriers, such as hydrogen and ammonia, are likely to play a crucial role in decarbonizing future energy systems, ensuring clean, safe, efficient, and fuel-flexible combustion. Therefore, an understanding of fundamental flame properties, such as laminar flame speed, flammability limits, and inherent instabilities, namely Darrieus-Landau, diffusional-thermal, and Rayleigh-Taylor instabilities, is essential.

In the present work, intrinsic instabilities in laminar premixed hydrogen and ammonia flames have been studied computationally. Detailed numerical simulations of freely propagating flames are performed using classical inflow, outflow, and periodic boundary conditions, employing detailed chemistry and transport models to resolve the multi-scale nature of flame. The linear growth rate of flame-front perturbations is examined over a range of wavenumbers, followed by their long-term nonlinear evolution to quantify propagation characteristics. The Darrieus-Landau instability is expected to affect all premixed flames due to a density jump across the flame front. Diffusional-thermal instabilities are expected to develop due to the disparity between heat and mass diffusion, and the Lewis number characterizes this effect. An imbalance between convective and diffusive transport makes the flame weaker when diffusion is dominant and stronger when convection is dominant, as distinguished by the Peclet number. Under high-pressure conditions, an increase in the overall activation energy results in a thinner region where reactions dominate, making the flame weaker and more susceptible to instabilities.

The combustion properties of hydrogen and ammonia differ significantly from those of conventional fuels, and intrinsic instabilities in the flame front can affect overall reactant consumption rates under different Lewis, Zeldovich, and Peclet numbers. For mixtures whose controlling Lewis number (Le) is smaller than unity ($Le < 1$), cellular structures are pronounced. The dilution of nitrogen alters the flame structure of hydrogen-oxygen mixtures by reducing the Lewis and Peclet numbers. Linear analysis reveals that flames with low adiabatic temperatures, i.e., with Low Peclet numbers, are prone to high growth rates. The consumption speed also shows a similar enhancement. Similarly, CO_2-rich

hydrogen-air mixtures also show a reduction in Lewis and Peclet numbers, making the flame weaker and more susceptible to instabilities. Such mixtures are produced during gasification and steam reforming of coal, biomass, oil, natural gas, and methane, and could be used in combustion applications to reduce pollution in developing economies. The analysis of CO_2-rich hydrogen-air mixture in linear and nonlinear regimes showed enhanced growth and consumption rates as the equivalence ratio and temperature decreased, and CO_2 dilution and pressure increased, due to dominant mass diffusion. The Lewis number has a predominant effect in driving thermodiffusive instability. However, at low temperatures (around 100K), the flame density ratio increases by approximately a factor of 3 compared to room temperature (300K). It is expected to result in a strong Darrieus–Landau instability. Parametric analysis reveals higher growth rates and normalized consumption speeds for perturbed flames at 100K than at 300K in lean-to-rich hydrogen-air flames, due to synergistic interaction of Darrieus-Landau and thermodiffusive effects and the formation of long, finger-like structures. Understanding of flame dynamics at such low temperatures is essential from the safety and storage perspective of hydrogen.

Another regime of diffusional-thermal instability known as pulsating instability is expected for rich hydrogen-air flames with a Lewis number much larger than unity ($Le > 1$) and at sufficiently high Zeldovich number (Ze) such that $Ze(Le - 1) > 10.93$. Numerical simulations reveal longitudinal pulsation and transverse waves for such mixtures. Strikingly, ammonia/air flames propagate much more slowly (laminar flame speed < 10 cm/s) than hydrogen and are significantly affected by gravity due to comparable time scales of flame propagation and buoyancy-induced instabilities. Linear analysis reveals that an upward-propagating flame against gravity is more unstable and develops a deep cusp structure. Under elevated gravity conditions, a mushroom-type morphology is observed, and after symmetry-breaking, it further evolves into pulsations and shear-induced vortex shedding behind the flame cusp.