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Poster Session Day 1

Mass Law in Thermal Diffusion: Insights from Noble Gases

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Thermal diffusion leads to a partial separation of molecules from initial homogeneous chemical or isotopic mixture, with the extent of separation governed by the magnitude of a thermal gradient and the nature of intermolecular interactions (e.g. [1][2]). For given isotope mixture, the thermal diffusion separation has been typically assumed to be mass proportional (e.g. [3][4]). A decade ago, we reported an unexpected violation of the mass law for thermal diffusion of two polyatomic molecules (O_2 and SF_6) within their triple or quadruple isotope matrix, respectively ([5][6]). The underlying mechanisms of this deviation remain not fully understood. Expanding the types of molecules tested could significantly enhance our understanding and application of such anomalous gas behavior.

Monoatomic molecules represent the simplest case and serve as a valuable benchmark for thermal diffusion studies due to their lack of intramolecular structure, such as symmetry, and their rotational/vibrational energy states. Despite numerous studies on this topic, high-precision investigations focusing on mass dependency within multiple isotope systems are lacking.

In this study, we report measurements of thermal diffusion separation of neon and argon in their triple isotope systems (Neon: ^{20}Ne, ^{21}Ne, ^{22}Ne; Argon: ^{36}Ar, ^{38}Ar, ^{40}Ar). Our preliminary results indicate that the self-diffusion of these pure gases adheres well to the expected mass law. However, when mixed with a polyatomic molecule (e.g., an Ar-O_2 admixture), the argon isotope fractionation is observed to deviate subtly, but significantly, from the expected mass dependency law. This deviation increases with a higher proportion of polyatomic molecules (e.g., a higher O_2/Ar ratio). Our observations introduce complexity into the theoretical understanding of thermal diffusion separation, suggesting that additional factors beyond, e.g., symmetry and inelastic collisions, might influence the mass relationships. Potential mechanistic interpretations will be discussed.

References

[1] Grew and Ibbs, 1952; [2] Mason et al, 1966;[3] Holleran, 1955; [4] Grachev and Severinghaus, 2003; [5] Sun and Bao 2011a; [6] Sun and Bao 2011b