

Measurements of intra-diffusion coefficients for gaseous binary mixtures

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Introduction: Diffusion in gaseous mixtures is critical to many industrial and geological processes, yet experimental data are sparse, particularly for intra-diffusion coefficients at elevated pressures. Existing relationships between intra- and mutual-diffusion, well established in liquids, are less validated for gases. We address this gap by measuring intra-diffusion coefficients (D_i) in three binary gas mixtures across a range of compositions and industrially relevant pressures and temperatures.

Methods: Measurements were conducted using a custom-designed, NMR-compatible Sapphire High Pressure cell integrated into a 43 MHz benchtop NMR spectrometer (Figure 1). Intra-diffusion coefficients (D_i) were measured using the Pulsed Field Gradient Spin Echo (PGSE) technique across methane–nitrogen, methane–helium, and methane–hydrogen binary mixtures at temperatures from 28 to 40 °C and pressures up to 100 bar. The composition of each mixture was verified by NMR spectroscopy, and measurements were corrected for restricted diffusion effects using validated numerical simulations.

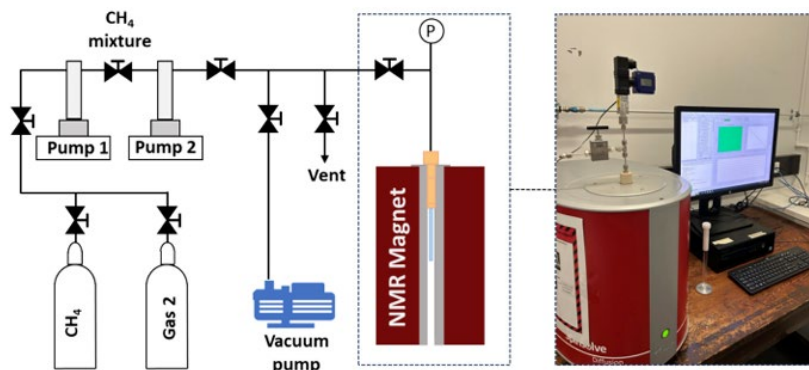


Figure 1: Experimental Setup

Results and Discussion: Intra-diffusion coefficients were obtained as a function of composition for all mixtures studied. Values of D_i were extrapolated to infinite dilution and compared to mutual diffusion coefficients (D_{12}) from both literature and Chapman-Enskog kinetic theory [1,2]. For methane–helium mixtures, agreement was within experimental uncertainty. For methane–nitrogen, D_i ($x_i=0$) exceeded D_{12} predictions by 8–10%. In the methane–hydrogen system, both CH_4 and H_2 intra-diffusion coefficients were measured, revealing good agreement for H_2 but a moderate deviation for CH_4 .

Conclusion: This study provides new experimental data on intra-diffusion in gaseous binary mixtures using an NMR-compatible high-pressure setup [3]. Results show generally good agreement with kinetic theory, but also reveal differences that suggest the need for refined models, particularly in mixtures with hydrogen. These data support the development of predictive models for high pressure and temperature gas diffusion as a function of composition.

References: [1] Berry Jr., AIChE J. (1960). [2] Chapman, S., The Mathematical Theory of Non-Uniform Gases (1990). [3] Kobeissi, S., Chem. Eng. Sci. (2025).