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CHEMICAL THERMODYNAMICS AND THERMOCHEMISTRY

Emergence of Convective Flows during Diffusional Mass Transfer in Ternary Gas Systems: The Effect of Component Concentrations

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Abstract—Specific features of diffusional mass transfer in ternary gas mixtures and their relation to the concentration of the densest component in a mixture are studied experimentally under isothermic conditions. At certain compositions of the gas mixture, unique diffusion regimes develop in the system that are characterized by a considerable rate of mass transfer and are attributed to the presence of concentration convection. We show that transitions from the diffusional and convective regimes can be predicted using the stability theory extended to isothermal mixing in ternary gas systems. Theoretical predictions are compared to experimental data.

Keywords: diffusion, concentration, pressure, convection, stability theory

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INTRODUCTION

Theoretical descriptions of mixing by diffusion in gas systems at various technological settings has been dealt with in studies on mass transfer kinetics [1]. Diffusion in multicomponent systems is described by the Stefan–Maxwell equations [1] and, in comparison to binary systems, has a number of special attributes [2–5]. There may be no mass transfer of a species with a nonzero concentration gradient; conversely, a species can diffuse even if its concentration gradient is zero. Reverse diffusion can take place as well. Another feature of diffusion in multicomponent systems is the possibility of convection arising that play a part in creating a synergetic effect associated with considerable enhancement of component mixing in a given system [6]. Experimental studies and numerical modelling of the diffusion of binary solution vapors into inert gases [6, 7] suggest that convective instability, followed by the formation of structured flows, can arise in such systems. The effects reported in the cited works must depend on the composition of the gas mixtures and applied pressures, but the effect of these factors on the intensity of mixing were not addressed.

In this work, we present the results from an experimental study on the isothermal diffusion of a helium–argon binary gas system into nitrogen at different pressures and concentrations of the densest component. The experimental results are compared to the results from numerical modeling based on the theory of convective stability.

EXPERIMENTAL

Diffusion and convection mixing were studied using a two-bulb technique [8]. Since a detailed description of the experimental setup and the measuring cell was reported in [9], here we provide only a general description of the setup and its operating principles.

A schematic presentation of the setup is shown in Fig. 1. It consists of two parts. The first one is a gas flow control system that delivers gases from cylinders *A* and *B* to the measuring cell and contains set of needle valves *1–10*, pressure gauges *12* with membrane separators *11*, and buffer reservoir *13* to dump pressure fluctuations. The second part shows a cross section view of the measuring cell comprising two flasks of equal volume ($V_1 = V_2 = (76.2 \pm 0.5) \times 10^{-6}$ m^3) interconnected through a hollow cylinder channel with length $L = (70.00 \pm 0.05) \times 10^{-3}$ m and diameter $d = (4.00 \pm 0.02) \times 10^{-3}$ m. In our experimental work, we followed the protocol proposed in [9]. The upper *16* and lower *14* flasks were filled with gases from cylinders *A* and *B* to the pressure required in a given experiment. Fluoroplastic stopper *17* ensured gas-tight sealing of the flasks at the filling stage. Rod *18* was lifted by turning lever *19*, opening channel *15* that interconnected the two flasks; this marked the beginning of the mixing experiment. When the experiment was complete, the channel was closed and the gas mixtures from the flasks were analyzed via gas chromatography.