

## Phase Behaviour and Physical Properties of (CO<sub>2</sub> + Diluents) Mixtures in the Context of CCS

Lorena F. S. Souza<sup>S</sup>

*Department of Chemical Engineering, Imperial College London, London, United Kingdom*

Saif Z. S. Al Ghafri

*School of Mechanical and Chemical Engineering, University of Western Australia, Perth, Australia*

J. P. Martin Trusler<sup>C</sup>

*Department of Chemical Engineering, Imperial College London, London, United Kingdom*

*m.trusler@imperial.ac.uk*

Knowledge of the phase behaviour of (CO<sub>2</sub> + diluent) mixtures is crucial in carbon capture, transport and storage (CCS) operations. For the design, optimisation and safe operation of these processes, accurate equations of state (EOSs) are required to describe thermodynamic properties of the various mixtures of CO<sub>2</sub> with relevant substances, including H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub>, CO, Ar, O<sub>2</sub>, and H<sub>2</sub>S. A popular and potentially very-accurate approach is to use empirical multi-fluid multi-parameter EOS models. In order to adjust the interaction parameters in those models, reliable and accurate experimental thermophysical data are indispensable. In the present study, new VLE measurements have been made on two relevant binary systems: (CO<sub>2</sub> + CH<sub>4</sub>) and (CO<sub>2</sub> + CO), and also of the distribution of trace levels of H<sub>2</sub>S between the two phases. The measurements were carried out at temperatures ranging from just above the triple-point to just below the critical point of CO<sub>2</sub> and at pressures from the vapor pressure of pure CO<sub>2</sub> up to approximately 15 MPa or, if lower, the mixture critical pressure. Density measurements on (CO<sub>2</sub> + CO) were carried out with a vibrating-tube densimeter (VTD) at a temperature range of (283.15 to 373.15) K and pressures up to 48 MPa. The new VLE and density data, together with earlier results from our laboratory on (CO<sub>2</sub> + H<sub>2</sub>) and (CO<sub>2</sub> + N<sub>2</sub>), have been compared with the predictions of available thermodynamic models, including GERG-2008 and EOS-CG. Additionally, the data have been correlated with the Peng-Robinson EOS, generally incorporating a single temperature-dependent binary interaction parameter. EOS-CG showed significantly better agreement, especially close to the critical point, than found with the Peng-Robinson model. The new experimental VLE data help to fill important knowledge gaps relating to the thermophysical properties of mixtures of CO<sub>2</sub> with substances relevant in CCS.