Abstract

Hydrogen is gaining recognition as an essential energy carrier in the worldwide endeavor to decarbonize the energy sector. Among the various methods for large-scale hydrogen storage, Underground Hydrogen Storage (UHS) in depleted gas reservoirs has emerged as a promising and cost-effective solution. This method involves injecting hydrogen, either as a pure component or as a mixture with methane, into porous rock formations that were previously used for natural gas storage. To optimize UHS and ensure its safe and efficient operation, it is essential to develop a comprehensive understanding of the reservoir dynamics. This requires a reliable 3D dynamic model capable of simulating the system's behavior under a range of operating conditions. One of the key challenges in accurately modeling these systems is the need for validated or calibrated Equations of State (EoS), which must be able to capture the complex interactions between the injected hydrogen or its mixtures and the existing reservoir fluids. In this context, laboratory experiments are critical for validating these models, particularly in terms of gas properties like compressibility and solubility under relevant pressure and temperature conditions.

This study provides results from a series of experiments that were carried out to characterize the behavior of H_2-CH_4 mixtures as well as CO_2 under conditions representative of those found in depleted gas reservoirs. The experiments were conducted using a Constant Mass Expansion (CME) experimental procedure in a PVT cell, which allows for precise measurement of gas volumetric behavior. The pressure range tested extended up to 300 bar, with temperatures spanning from 25°C to 55°C. Five different gases were analyzed: pure hydrogen (H_2), a 50% H_2-50% CH_4 mixture, a 10% H_2-90% CH_4 mixture, pure methane (CH_4), and pure CO_2. The experimental data were analyzed in terms of isothermal gas compressibility factor (z-factor) curves as a function of pressure for each considered gas composition. Experimental results were then compared to the GERG-2008 EoS, which provides an established reference for predicting the thermodynamic properties of gas mixtures.

In addition to compressibility studies, the solubility of hydrogen and its mixtures in brine under reservoir conditions is another critical aspect of UHS. Accurate estimation of hydrogen solubility is necessary to account for potential hydrogen losses to the aquifer. However, due to safety concerns, experimental data on hydrogen solubility in brine under high-pressure conditions are limited. To address this gap, the study, considered as first to date, also examined the solubility of H_2 and H_2-CH_4 mixtures in saline water (brine) under conditions that replicate a broad range of potential UHS reservoir scenarios. The experiments were conducted using the same PVT cell, with a pressure range from 1 bar up to 500 bar and temperatures of 45°C, 50°C, and 55°C. Two different brine samples, each representative of different reservoir conditions, were tested to assess the impact of brine composition on H_2 solubility. The gas mixtures considered were pure H_2, a 10% H_2-90% CH_4 mixture, and a 50% H_2-50% CH_4 mixture.

The results indicated that, while temperature and brine composition had minimal impact on hydrogen solubility within the tested range, the gas mixture composition significantly influenced the solubility behavior. Specifically, the solubility curve exhibited a steeper slope as the hydrogen percentage decreased in the gas mixture, indicating that methane content has a notable effect on hydrogen dissolution into brine. This finding underscores the importance of considering gas mixture composition when assessing potential hydrogen losses to the aquifer. A comparison of the

experimental data with existing literature models revealed some discrepancies, highlighting the need for further refinement of solubility models, particularly under the extreme pressures and temperatures typical of UHS operations.

The results from both the compressibility and solubility experiments are crucial for enhancing the understanding of UHS systems. By providing a more accurate representation of the thermodynamic properties of hydrogen and its mixtures, these findings contribute to the development of more reliable and efficient 3D reservoir models. These models will be essential for optimizing UHS design, ensuring the safety and long-term viability of hydrogen storage in depleted gas reservoirs, and mitigating risks associated with hydrogen loss and reservoir behavior. Ultimately, this work aims to advance the deployment of UHS as a key component of large-scale hydrogen storage solutions, helping to support the transition toward a sustainable and decarbonized energy future.