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Thermodynamic and transport properties of hydrogen containing streams

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The use of hydrogen (H₂) as a substitute for fossil fuel, which accounts for the majority of the world's energy, is environmentally the most benign option for the reduction of CO₂ emissions. This will require gigawatt-scale storage systems and as such, H₂ storage in porous rocks in the subsurface will be required. Accurate estimation of the thermodynamic and transport properties of H₂ mixed with other gases found within the storage system is therefore essential for the efficient design for the processes involved in this system chain. In this study, we used the established and regarded GERG-2008 Equation of State (EoS) and SuperTRAPP model to predict the thermo-physical properties of H₂ mixed with CH₄, N₂, CO₂, and a typical natural gas from the North-Sea. The data covers a wide range of mole fraction of H₂ (10–90 Mole%), pressures (0.01–100 MPa), and temperatures (200–500 K) with high accuracy and precision. Moreover, to increase ease of access to the data, a user-friendly software (H2Themobank) is developed and made publicly available.

Background & Summary

To meet the Paris Agreement climate targets, global carbon emissions need to reach net-zero by 2050¹. To achieve this the emissions from fossil fuels must be reduced and the energy mix transition to low carbon energy sources must be accelerated. Hydrogen can support this transition by replacing natural gas for domestic and industrial uses; replacing coal and natural gas for power generation; replacing fuel oil and gasoline to decarbonise transport and facilitating increased renewable energy by acting as an energy carrier to balance supply and demand. To enable hydrogen as a low carbon energy pathway, gigawatt-scale storage will be required^{2–5}. Geological gas storage in underground salt caverns, depleted oil and gas fields and deep aquifers are proven technologies that could provide the necessary scales for hydrogen storage^{6–8}. Hydrogen-rich town gas mixtures have been stored in geological formations since the 1970's⁹ and currently, over 1,000,000 m³ of hydrogen is stored in underground salt caverns^{10,11}. Furthermore, several types of gas have been successfully stored in geological formations, such as natural gas, compressed air and CO₂. Recent work has shown that leakage of injected and stored gas is unlikely, if rigorous standards are in place¹².

The storage of gas in the subsurface as chemical energy storage, whether as natural gas or hydrogen (the working gas), requires a cushion gas (30–70% of the total gas storage volume¹³) to prevent brine from entering the production stream and to maintain the required reservoir pressure ensuring deliverability. As depleted gas fields are being considered as storage sites for subsurface hydrogen storage, the *in situ* gas could be used as cushion gas and hence the working and cushion gasses will be of different compositions⁷. For gas storage in saline aquifers, where there is very little *in situ* gas present, there is a requirement to use a cushion gas that is significantly cheaper than the working gas. Considered options for aquifer storage cushion gasses are nitrogen, due to its low price, and CO₂ due to its high compressibility and potential for secure storage of this greenhouse gas^{14–16}. During the injection/production cycles, mixing of the gas components is inevitable and is determined by parameters such as mobility ratios, density differences, molecular diffusion and mechanical dispersion¹⁷. The numerical simulation of any storage scenario must confirm that the working gas can be produced with minimal cushion gas

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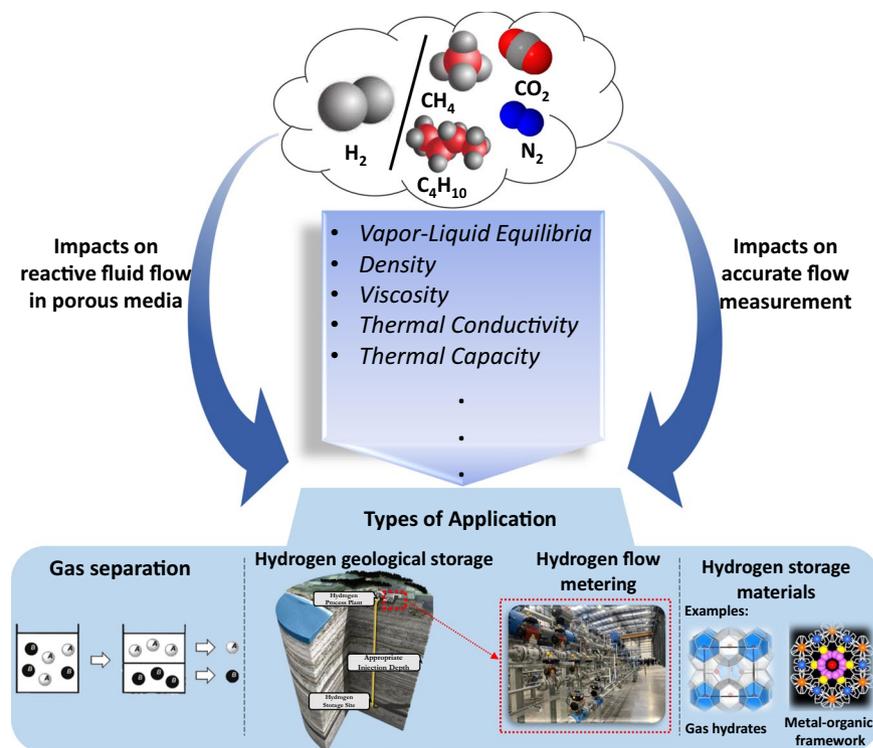


Fig. 1 Potential applications of thermodynamic properties of hydrogen-containing streams for geological hydrogen storage, flow metering, gas separation⁶² and other types of hydrogen storage purposes such as gas hydrates^{63,64} or metal-organic framework⁶⁵.

contamination. Therefore, if the cushion gas and working gas are of different compositions, the accurate quantification of the cushion gas/working gas mixing zone is of paramount importance. Once mixing takes place, the different gaseous components will alter the properties of the gas and introduce significant uncertainty into the expected behaviour of the injected, stored and produced gas, as shown for different gas storage applications^{18,19}.

For gas storage modelling, accurate thermodynamic reference data for relevant fluid mixtures, which can either be directly imported into fluid flow modelling software or can be used to confirm existing reservoir engineering software outputs, is an important tool to enhance the compliance for scenario modelling results. Furthermore, the thermodynamic data for hydrogen-containing systems can enable scientists to have a deeper understanding of reactive flow through porous media during the hydrogen storage process. Another target in a hydrogen-based economy is to establish a fundamental understanding of metering technologies and the flow measurement principles behind them. In this regard, the thermo-physical properties of hydrogen mixed gases are crucial to understand and model hydrogen transportation and flow measurement processes. Thermo-physical properties of hydrogen-containing gas mixtures over a wide range of pressures and temperatures are pivotal to the design and optimisation of hydrogen production units, transportation, and storage processes (see Fig. 1).

Significant effort has been made to investigate the thermodynamic properties of hydrogen-containing mixtures systematically^{20–35}. In addition, the phase equilibria and solubility of Hydrogen-natural gas components contained blends have been studied by researchers (see Table 1).

While the thermodynamic properties of pure hydrogen are well established^{36,37}, published properties of gas mixtures in relation to geological hydrogen storage^{17,38–45} do not cover the full range of additional gasses and often do not encompass the pressures and temperatures encountered within the hydrogen storage system (see Fig. 2). This data study will quantify the impacts of these additional gas components, to enable the accurate simulation of hydrogen mixed with various gases, all of which are essential for the modelling of the transportation, injection, geological storage, and production of hydrogen over multiple injection/production cycles.

Methods

Density and other derived thermodynamic properties using GERG-2008 Equation of State (EoS).

To efficiently design and operate the technical processes involved in gas-based energy industries, precise representation of the thermodynamic properties using an accurate EoS is essential. Here, the established and well regarded GERG-2008 EoS⁴⁶ was used to predict phase behaviour and density of gas mixtures relevant to hydrogen storage, covering the thermodynamic properties of gas phase, liquid phase and supercritical regions. This equation is valid over a wide range of pressures and temperatures for 21 gas components including 1-methane, 2-nitrogen, 3-carbon dioxide, 4-ethane, 5-propane, 6-n-butane, 7-iso-butane, 8-n-pentane, 9-isopentane, 10-n-hexane, 11-n-heptane, 12-n-octane, 13-hydrogen, 14-oxygen, 15-carbon monoxide, 16-water, 17-helium, 18-argon, 19-n-nonane, 20-n-decane, and 21-hydrogen sulphide. The thermodynamic properties of the fluids

| No. | System: H ₂ (1 st Component) + X (2 nd Component and beyond) | Property Type | Pressure Range (MPa) | Temperature Range (K) | x ₁ range (1 st component liquid mole fraction) | y ₁ range (1 st component gas mole fraction) | Reference |
|-----|---|---|----------------------|-----------------------|---|--|--------------------|
| 1 | CH ₄ | VLE/Solubility/Density/Viscosity/Compressibility/Thermal Conductivity | 0.22–141.40 | 66.89–350.00 | 0.002–0.859 | 0.034–1.000 | 20–35 |
| 2 | C ₂ H ₆ | VLE/Compressibility | 0.27–562.50 | 83.00–283.15 | 0.002–0.800 | 0.085–1.000 | 26,32,71–75 |
| 3 | C ₃ H ₈ | VLE/Compressibility | 0.69–55.16 | 93.15–366.40 | 0.001–0.669 | 0.110–0.999 | 26,71,76–79 |
| 4 | C ₄ H ₁₀ | VLE/Solubility | 2.07–53.43 | 144.26–394.25 | 0.008–0.341 | 0.213–0.999 | 20,80–82 |
| 5 | C ₅ H ₁₂ | VLE/Solubility | 0.69–27.59 | 273.15–463.15 | 0.004–0.259 | 0.373–0.997 | 83,84 |
| 6 | C ₆ H ₁₄ | VLE/Solubility | 1.24–68.95 | 277.59–506.48 | 0.011–0.700 | 0.100–0.998 | 85–87 |
| 7 | Cyclo-C ₆ H ₁₄ | VLE/Solubility | 0.10–69.04 | 293.15–523.15 | 0.000–0.367 | 0.549–0.997 | 83,88–96 |
| 8 | N ₂ | VLE/Solubility/Heat Capacity/Compressibility | 0.13–101.33 | 20.10–122.04 | 0.012–0.620 | 0.082–1.00 | 20,28,31,34,97–113 |
| 9 | CO ₂ | VLE/Viscosity/Density/Thermal Conductivity | 0.93–191.80 | 219.90–298.15 | 0.001–0.744 | 0.043–0.934 | 108,114–118 |
| 10 | H ₂ S | Solubility | 1.01–5.07 | 243.15–273.15 | 0.002–0.020 | 0.322–0.910 | 119,120 |
| 11 | CO | VLE/Viscosity/Thermal Conductivity/Density | 0.13–5.07 | 20.10–122.04 | 0.012–0.731 | 0.082–1.00 | 27,110,121,122 |
| 12 | CH ₄ + C ₂ H ₆ | VLE | 0.27–562.50 | 83.00–283.15 | 0.002–0.800 | 0.085–1.000 | 72 |
| 13 | C ₃ H ₈ + CO | VLE | 0.69–20.68 | 88.15–348.15 | 0.005–0.107 | 0.034–0.847 | 79 |
| 14 | CH ₄ + CO ₂ | VLE | 6.90–27.60 | 227.35–258.15 | 0.004–0.259 | 0.373–0.997 | 84 |
| 15 | CH ₄ + CO | VLE | 2.90–5.00 | 120.00–140.00 | 0.000–0.110 | 0.000–0.926 | 31 |
| 16 | C ₅ H ₁₂ + CO ₂ | VLE | 6.90–27.60 | 273.15–323.15 | 0.004–0.259 | 0.373–0.997 | 84 |
| 17 | N ₂ + CO | VLE | 0.003–22.80 | 58.15–122.04 | 0.012–0.930 | 0.082–1.00 | 100,109–111 |
| 18 | CH ₄ + N ₂ | VLE | 3.40–10.00 | 80.00–144.00 | 0.009–0.720 | 0.060–1.00 | 98 |

Table 1. Database of available experimental thermodynamic properties data in the literature for hydrogen-containing systems along with the temperature, pressure and composition range with respect to hydrogen for each binary/ternary system.

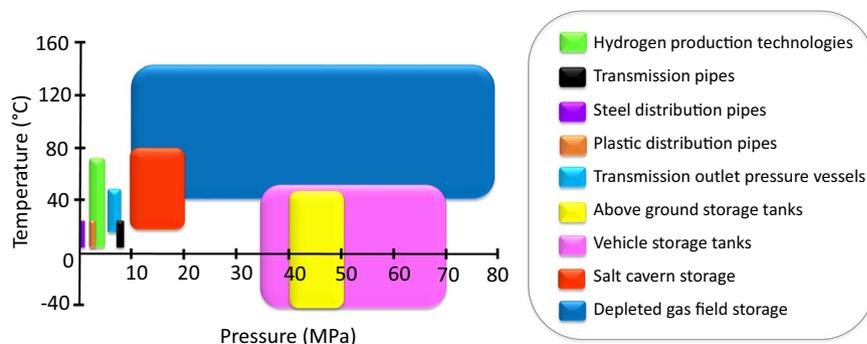


Fig. 2 Pressure and temperature ranges for various hydrogen-based economy systems. Note that the pressure range for depleted gas fields is based on the data available for the UK only^{66–69}.

that are predicted here at certain temperatures (T) are based on a multi-fluid approximation using the dimensionless Helmholtz energy obtained from:

$$\alpha(\delta, \tau, \mathbf{x}) = \alpha^0(\rho, T, \mathbf{x}) + \alpha^r(\delta, \tau, \mathbf{x}) \quad (1)$$

where ρ is the mixture density and \mathbf{x} is the molar composition vector. The term $\tau = T/T_r$ is the inverse reduced temperature, and the term δ is the reduced density, both of which are composition-dependent i.e. they depend on the molar composition vector. The ideal-gas contribution (α^0) is related to the number of mixture components (N), the mole fraction of each component i (x_i), and the dimensionless Helmholtz energy of component i in the ideal-gas phase (α_{oi}^0) by:

$$\alpha^0(\rho, T, \mathbf{x}) = \sum_{i=1}^N x_i [\alpha_{oi}^0(\rho, T) + \ln x_i] \quad (2)$$

The residual part of the dimensionless Helmholtz energy (α^r) is composed of two different parts; the linear summation of the residual part of the reduced Helmholtz free energy of each component i (α_{oi}^r) and the so-called departure function ($\Delta\alpha^r$) which is also a function of the mixture composition, the inverse reduced mixture temperature, and the reduced mixture density. The residual part of the dimensionless Helmholtz energy is given by:

$$\alpha^r(\delta, \tau, \mathbf{x}) = \sum_{i=1}^N x_i \alpha_{oi}^r(\delta, \tau) + \Delta \alpha^r(\delta, \tau, \mathbf{x}) \quad (3)$$

The advantage of using Helmholtz energy in the given form is that all the other thermodynamic properties can be derived analytically from terms α^o and α^r and their derivatives. One example is isobaric heat capacity which is given by:

$$c_p(\delta, \tau, \mathbf{x}) = R \left[-\tau^2 (\alpha_{\tau\tau}^o + \alpha_{\tau\tau}^r) + \frac{(1 + \delta \alpha_{\delta}^r + \delta \tau \alpha_{\delta\tau}^r)^2}{1 + 2 \delta \alpha_{\delta}^r + \delta^2 \alpha_{\delta\delta}^r} \right] \quad (4)$$

where R is the gas constant. The subscriptions of α^o and α^r denote the order of their derivatives with respect to τ and δ . For example $\alpha_{\tau\tau}^r$ denotes the second-order derivatives of α^r with respect to τ . Similarly, enthalpy (h), entropy (s), Gibbs free energy (g), pressure (P) can be obtained from:

$$P(\delta, \tau, \mathbf{x}) = RT\rho[1 + \delta \alpha_{\delta}^r] \quad (5)$$

$$h(\delta, \tau, \mathbf{x}) = RT[1 + \tau (\alpha_{\tau}^o + \alpha_{\tau}^r) + \delta \alpha_{\delta}^r] \quad (6)$$

$$s(\delta, \tau, \mathbf{x}) = R[\tau (\alpha_{\tau}^o + \alpha_{\tau}^r) - \alpha^o - \alpha^r] \quad (7)$$

$$g(\delta, \tau, \mathbf{x}) = RT[1 + \alpha_{\tau}^o + \alpha_{\tau}^r + \delta \alpha_{\delta}^r] \quad (8)$$

Other thermodynamic properties such as compression factor, internal energy, speed of sound, Joule-Thomson coefficient, etc. can be defined similarly. Kunz. *et al.*⁴⁶ provides comprehensive coverage of these derivatives and thermodynamic properties. In GERG-2008 EoS, terms ρ_r and T_r are calculated using quadratic mixing rules proposed by Klimeck⁴⁷:

$$\frac{1}{\rho_r(\mathbf{x})} = \sum_{i=1}^N x_i^2 \frac{1}{\rho_{c,i}} + \sum_{i=1}^{N-1} \sum_{j=i+1}^N \frac{2x_i x_j}{\rho_{c,ij}} \quad (9)$$

$$T_r(\mathbf{x}) = \sum_{i=1}^N x_i^2 T_{c,i} + \sum_{i=1}^{N-1} \sum_{j=i+1}^N 2x_i x_j T_{c,ij} \quad (10)$$

where $\rho_{c,i}$ is the critical density of component i, .. is the critical temperature of component i. The parameters for the components studied in this study are provided in figshare entry⁴⁸. $T_{c,ij}$ and $\rho_{c,ij}$ are obtained from:

$$\frac{1}{\rho_{c,ij}} = \beta_{\nu,ij} \gamma_{\nu,ij} \frac{x_i + x_j}{\beta_{\nu,ij}^2 x_i + x_j} \cdot \frac{1}{8} \left(\frac{1}{\rho_{c,i}^{1/3}} + \frac{1}{\rho_{c,j}^{1/3}} \right)^3 \quad (11)$$

$$T_{c,ij} = \beta_{T,ij} \gamma_{T,ij} \cdot \frac{x_i + x_j}{\beta_{T,ij}^2 x_i + x_j} (T_{c,i} T_{c,j})^{1/2} \quad (12)$$

where $\beta_{\nu,ij}$, $\gamma_{\nu,ij}$, $\beta_{T,ij}$, and $\gamma_{T,ij}$ are the four adjustable binary interaction parameters. The binary interaction parameters used for the components in this study are provided in figshare entry⁴⁸.

An example of the calculated densities and isobaric heat capacity for H₂ + CH₄ mixtures over a range of pressure and temperature for various H₂ mole fractions is provided in Figs. 3 and 4, respectively. Plots of the other derived thermodynamic properties of H₂ with CH₄ and the thermodynamic properties of H₂ with CO₂, N₂, and the typical natural gas are presented in figshare entry⁴⁸.

Viscosity and thermal conductivity using SuperTRAPP model. For calculating viscosity of the system we used SuperTRAPP model⁴⁹ that is based on the corresponding-states model. SuperTRAPP viscosity model is composed of a dilute-gas and residual contribution part, where only the latter is treated with corresponding states. The viscosity (μ) is a function of density and pressure and is obtained from:

$$\mu(T, \rho) = \mu^*(T) + \Delta \mu_0(T_0, \rho_0) F_{\mu}(T, \rho) \quad (13)$$

where * refers to dilute gas and 0 refers to a reference fluid. The dilute gas viscosity is calculated using Chung *et al.*⁵⁰ theory which is a modification of the original model by Chapman-Enskog⁵¹. The function F_{μ} can be obtained from:

$$F_{\mu}(T, \rho) = \sqrt{f} h^{-2/3} \sqrt{\frac{m}{m_0}} \quad (14)$$

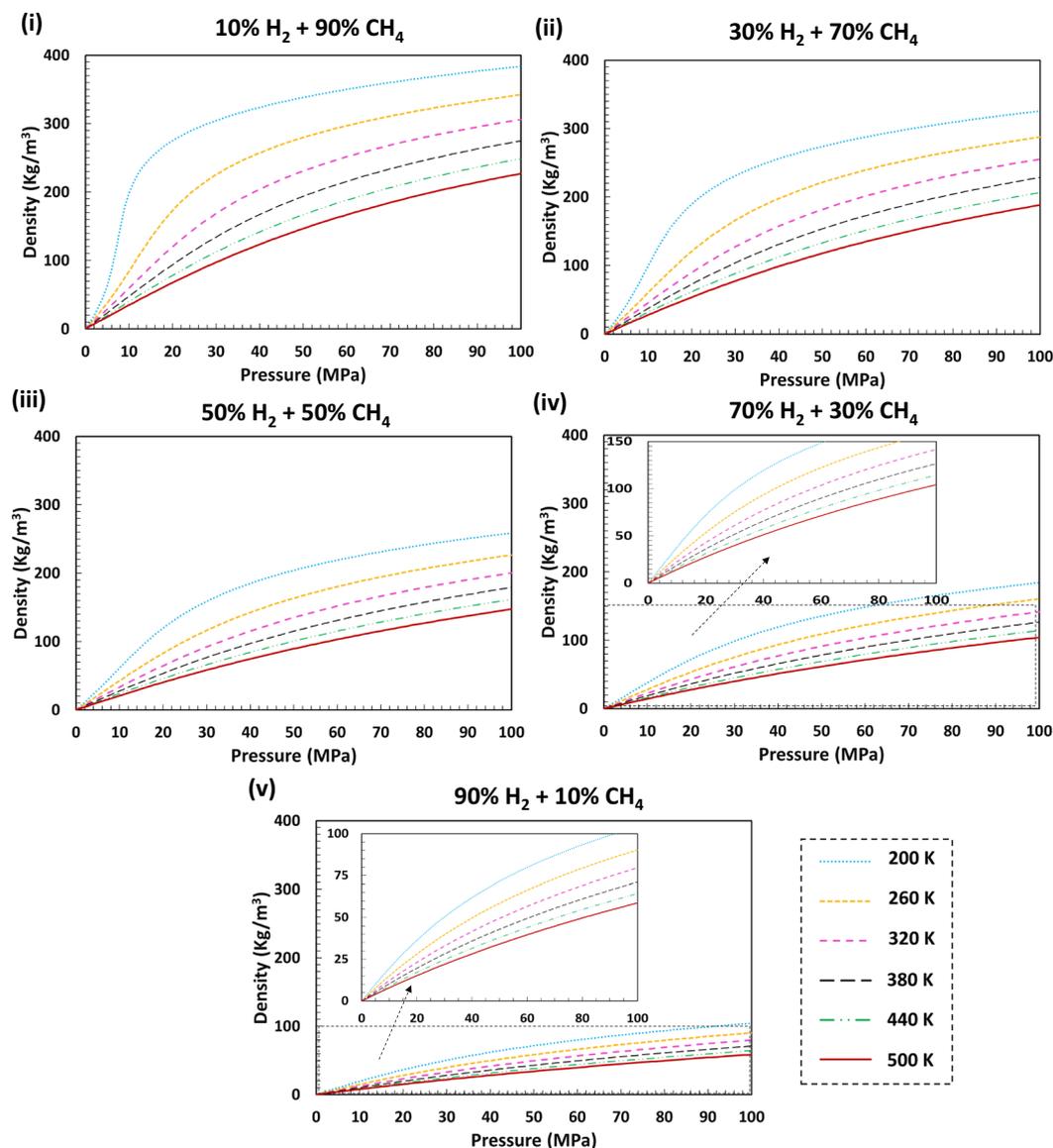


Fig. 3 Predicted densities for different H₂ + CH₄ mixtures for various mole H₂ fractions over a wide range of pressures and temperatures using GERG-2008 EoS. Density values are greater in the presence of higher mole fractions of CH₄ in the studied systems as the density of CH₄ is considerably higher than that of H₂. The densities increase with increasing pressure (Boyle's Law) for all isotherms and reduce with increasing temperature (Charles's Law).

where m , and m_0 are the molar mass of the main fluid and the reference fluid, respectively. The terms f and h are so-called equivalent substance reducing ratios, relating the reference fluid to the studying fluid using critical parameter ratios. For a more detailed examination of the formulations used for calculating viscosity, the reader is referred to reference⁴⁹. An example of calculated viscosities for H₂ + CH₄ mixtures over a range of pressures and temperatures for various H₂ mole fractions is provided in Fig. 5. Plots of the viscosity of H₂ with CO₂, N₂, and typical natural gas are presented in figshare entry⁴⁸. To calculate the thermal conductivity of the fluids (γ) we also used SuperTRAPP model⁴⁹. The thermal conductivity is obtained from:

$$\gamma(T, \rho) = \gamma^{int}(T) + \overbrace{\gamma^*(T) + \gamma^r(T, \rho) + \gamma^{crit}(T, \rho)}^{trans} \quad (15)$$

This method is based on the Ely and Hanley procedure⁵² for calculating the thermal conductivity, where the model considers the effect of collisions between molecules (translational energy transfer) (γ^{trans}), and the internal motions of the molecules (γ^{int} , calculated using modified Eucken correlation). The former term can be further divided into three contributions i.e. dilute gas (γ^*), residual (γ^r) and critical enhancement (γ^{crit}). We refer the reader to the article by Huber⁴⁹ for detailed formulation and parameters of the thermal conductivity. An example of the calculated thermal conductivities for H₂ + CH₄ mixtures over a range of pressures and temperatures for

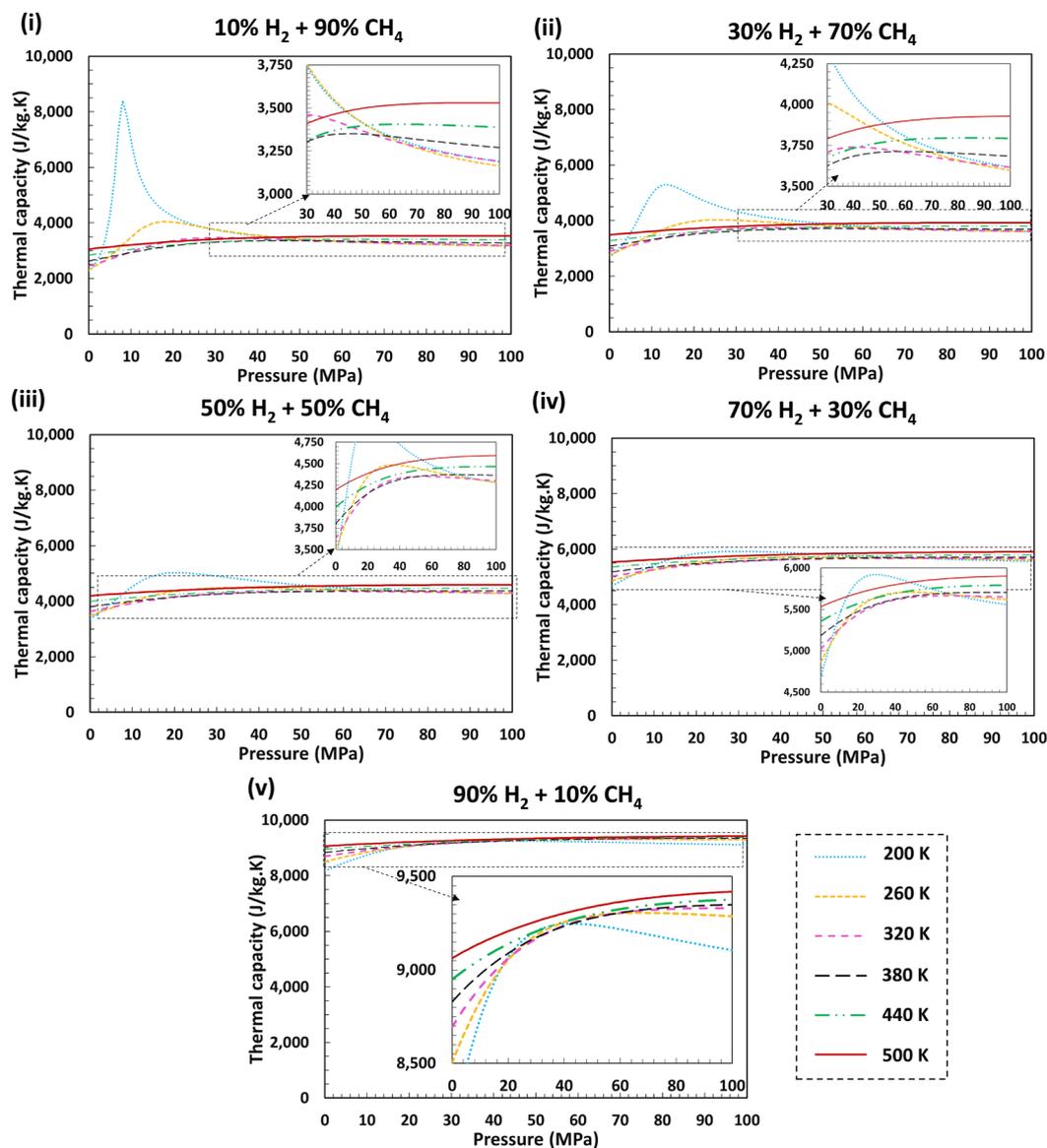


Fig. 4 Predicted isobaric heat capacities for different $\text{H}_2 + \text{CH}_4$ mixtures for various H_2 mole fractions over a wide range of pressures and temperatures using GERG-2008 EoS. Thermal capacities have higher values for higher H_2 mole fractions as the heat capacity of pure H_2 is significantly higher than that of pure CH_4 at temperatures and pressures above the critical point of CH_4 . Generally, it can be noted that with increasing pressure, the thermal capacities increase for all temperature conditions due to increased intermolecular forces. The peaks in the graphs can be attributed to the fact that near the critical points of the components the heat capacities undergo sudden changes because of the changes in their phase. In these examples, as the temperatures and pressures are close to the critical conditions of CH_4 , peaks have emerged. Reducing the mole fraction of CH_4 in the system composition moves the system away from the critical point and as such the peaks reduce or do not appear in the graphs (iv and v).

various H_2 mole fractions is provided in Fig. 6. The plots of thermal conductivity of H_2 with CO_2 , N_2 , and the typical natural gas are presented in figshare entry⁴⁸.

Vapour liquid Equilibria and isothermal flash. For calculating phase equilibrium of the studied mixtures, we used a method established by Michelsen⁵³. The vapour–liquid phase envelopes calculated for the system studied are provided in Fig. 7. As can be seen, some parts of two-phase envelope of the $\text{H}_2 + \text{CO}_2$ and the $\text{H}_2 + \text{Natural gas}$ systems occur within the pressure and temperature ranges of geological storage and as such are used in this study (temperature 200–500 K and pressure 0.01–100 MPa). For these points, we used isothermal multi-phase flash to calculate fraction and composition of gas and liquid phase. Here, we followed stability analysis by the successive substitution method which was introduced by Michelsen⁵⁴ to minimise the Gibbs energy of the system. This was followed by the calculation of thermodynamic properties for each phase.

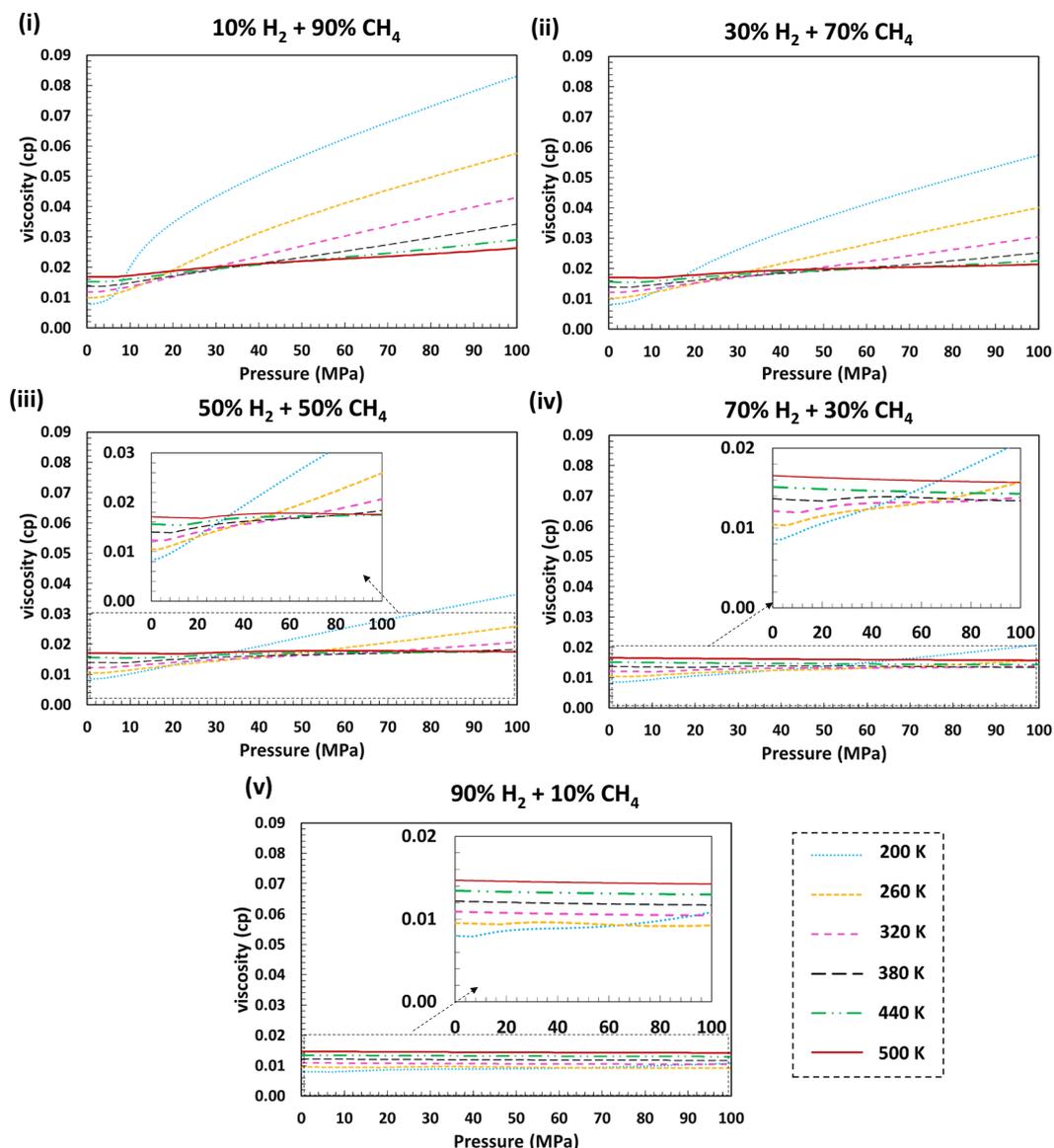


Fig. 5 Modelled viscosity values for various $\text{H}_2 + \text{CH}_4$ blends for different H_2 mole fractions over a wide range of pressures and temperatures using GERG-2008 EoS and SuperTRAPP model. The viscosities of the mixtures are suppressed with increasing H_2 mole fractions in the system as H_2 has a significantly lower viscosity than CH_4 due to its smaller molecule size. The viscosities of the blends increase with increasing pressure and temperature. This can be attributed to the fact that an increase in pressure or temperature increases the velocities of the random motion of molecules and as such collisions of gas molecules increase, which resists the flow of gas and increases the viscosity. The unusual behaviour of CH_4 -rich blends at lower temperatures is because of their proximity to the CH_4 critical point.

Data Records

Viscosity, thermal conductivity, density, and other derived thermodynamic properties of H_2 mixed with N_2 , CO_2 , CH_4 , and a typical natural gas from the UK North Sea (see Table 2) is provided for temperatures between 200–500 K, pressures between 0.01–100 MPa and mole fractions of hydrogen and additional gases between 10–90% using the described method. Note that the presence of water vapour, other impurities within the natural gas composition or selecting a different natural gas composition will affect the accuracy of the properties calculated.

The calculated data is publicly available and can be obtained using the following sources:

H2ThermoBank. An open-source user-friendly software developed using C# code in visual studio to ease the access of data for any user.

Excel format. The data is uploaded to (figshare) and is publicly accessible⁴⁸.

Figure format. Some selected data points with large steps are plotted and provided in figshare entry⁴⁸.

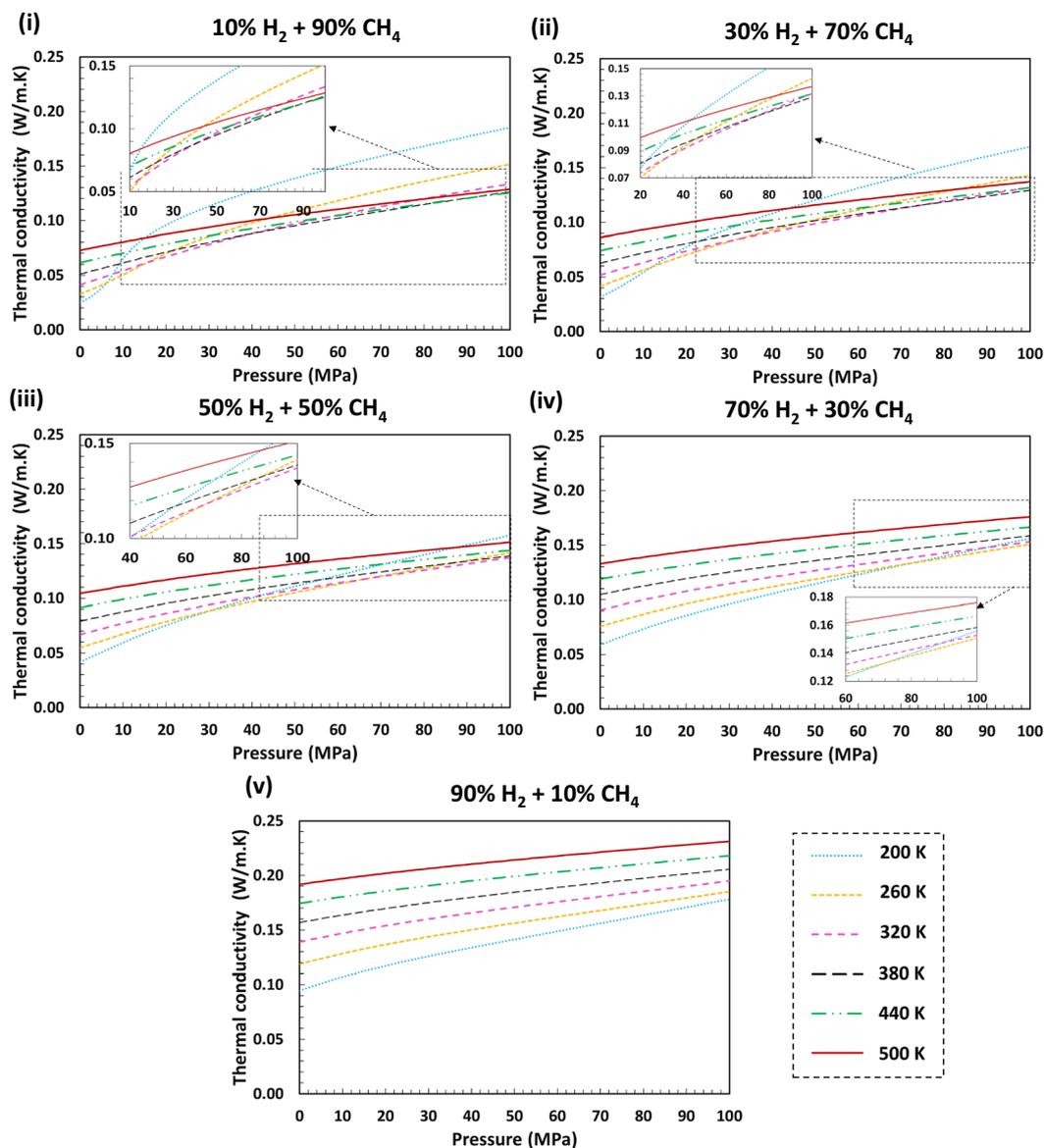


Fig. 6 Estimated thermal conductivities for different $\text{H}_2 + \text{CH}_4$ blends with a range of H_2 mole fractions over a wide range of pressures and temperatures using GERG-2008 EoS and SuperTRAPP model. It can be inferred that for all hydrogen/methane mixtures, the thermal conductivity values increase with increasing pressure for all isotherms. Thermal conductivities also increase with temperature for high H_2 mole fraction systems (above 50%). These behaviours can be attributed to the fact that increasing pressure or temperature increases the molecular motion and as such improves the conduction of heat within gas molecules. The unusual behaviour of CH_4 -rich streams at lower temperatures is because of the proximity of these points to the CH_4 critical point. Generally, thermal conductivity values increase with increasing hydrogen mole fractions as pure H_2 has a considerably higher thermal conductivity than CH_4 .

Technical Validation

The validity of the code developed for this study is checked by comparing the calculated results for a sample natural gas with existing data in the literature. Numerous pressure and temperature points were randomly selected for this comparison. The manual validation revealed no error in the written script for this study.

The GERG-2008 EoS is valid over a wide range of temperatures, pressures, and gas compositions achieving high accuracy in the prediction of thermodynamic properties of the 21 components listed in the methods section. Although the GERG-2008 EoS has been fitted to a wide range of experimental data, for some binary mixtures only the reducing functions were used. This is because predicting a general rule for accuracy of the GERG-2008 EoS for such binary mixtures is a very challenging task as there is no experimental data available for some ranges, therefore the absolute value of error for such ranges is considered to be unknown. However, the GERG-2008 EoS has been extensively compared with available data and its validity range can be divided into three pressure/temperature ranges:

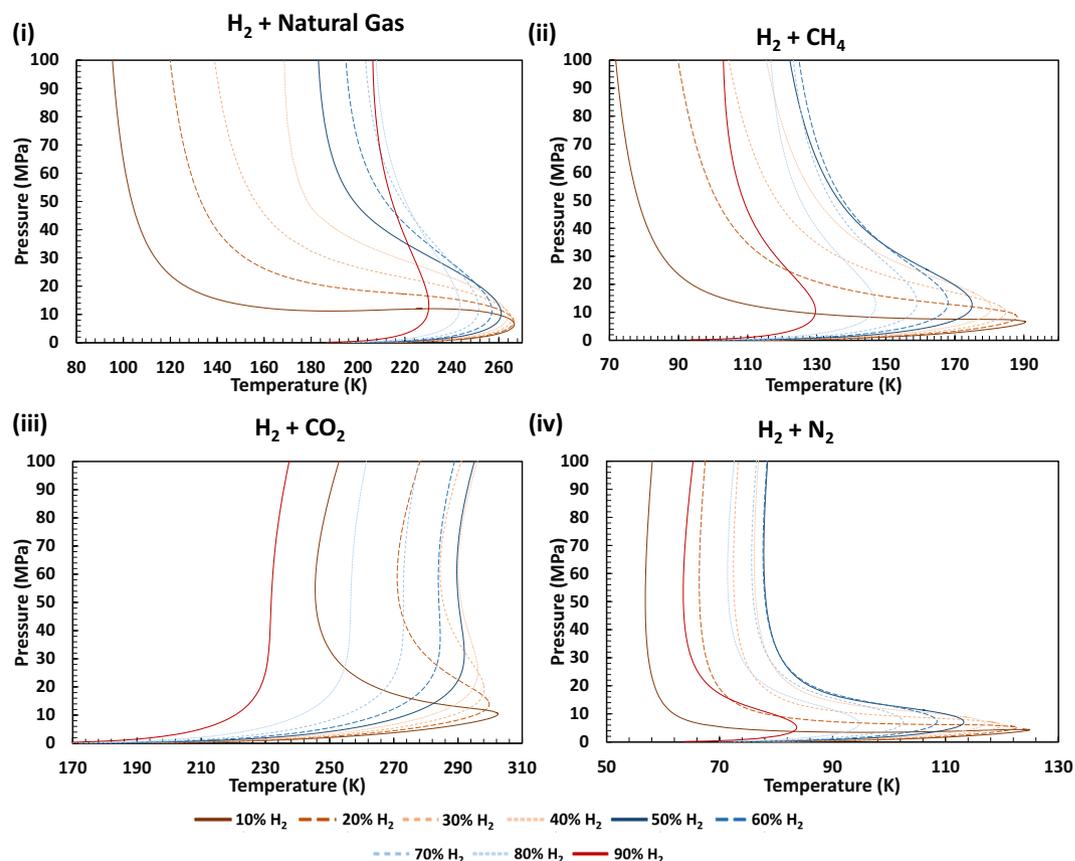


Fig. 7 Modelled Vapour liquid Equilibria (VLE) diagrams using the developed tool in this study for various H₂ containing mixtures with different H₂ mole fractions over a wide range of pressures and temperatures. Comparing Fig. 2 with the above figure highlights that the possibility of entering into a two-phase region only exists in systems with higher CO₂ concentrations. We refer readers to an excellent book on thermodynamics and phase behaviour of fluids to read more details about the behaviour of mixed fluids under various pressure and temperature conditions⁷⁰.

| Component | Mole% |
|-------------------------------|-------|
| CH ₄ | 83.60 |
| C ₂ H ₄ | 7.48 |
| C ₃ H ₈ | 3.92 |
| n-C ₄ | 0.81 |
| i-C ₄ | 0.81 |
| n-C ₅ | 0.15 |
| i-C ₅ | 0.14 |
| N ₂ | 1.95 |
| CO ₂ | 1.14 |

Table 2. Natural gas molar composition used in this study. The composition is based on data obtained from a typical UK North Sea natural gas¹²³.

- Normal range: which covers temperatures between 90 K and 450 K and pressures up to 35 MPa (The uncertainty of calculated density is less than 0.1% over the major part of this range)
- Extended range: which covers temperatures between 60 K and 700 K and pressures up to 70 MPa (The uncertainty of calculated density is less than 0.5% over the major part of this range)
- Extrapolated range: which covers temperatures and pressures beyond the previous range. (The uncertainty of calculated density is less than 1% over the major part of this range up to 100 MPa)

A number of studies using the same thermodynamic models have demonstrated the validity and accuracy of GERG-2008 EoS for different mixtures such as CH₄ mixtures^{55,56}, CO₂ mixtures^{57,58}, natural gas^{59,60} and compressed air⁶¹.

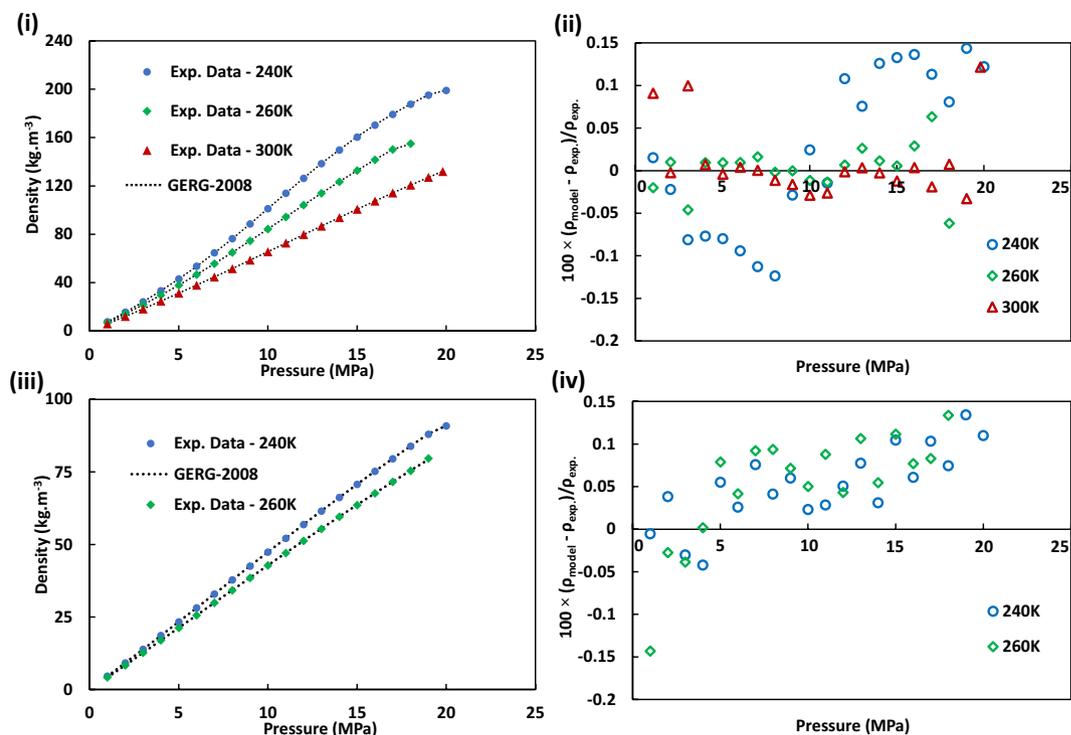


Fig. 8 Thermodynamic modelling and experimental results of density of hydrogen/methane mixtures at a range of temperatures and pressures: i & ii are the results for a 10% H_2 + 90% CH_4 mixture and iii & iv are the results for a 50% H_2 + 50% CH_4 mixture at different pressure and temperatures. ii and iv show the relative deviations in density values predicted by GERG-2008 equation of state, ρ_{model} , from the density from the experimental (ρ_{exp}) data²³ versus pressure at different temperatures. The relative expanded uncertainties in experimental density ($k=2$) $U(\rho_{\text{exp}})$ for the 10% H_2 + 90% CH_4 mixture and the 50% H_2 + 50% CH_4 mixture are $0.024 \leq U(\rho_{\text{exp}}) \leq 0.046$ and $0.024 \leq U(\rho_{\text{exp}}) \leq 0.034$, respectively.

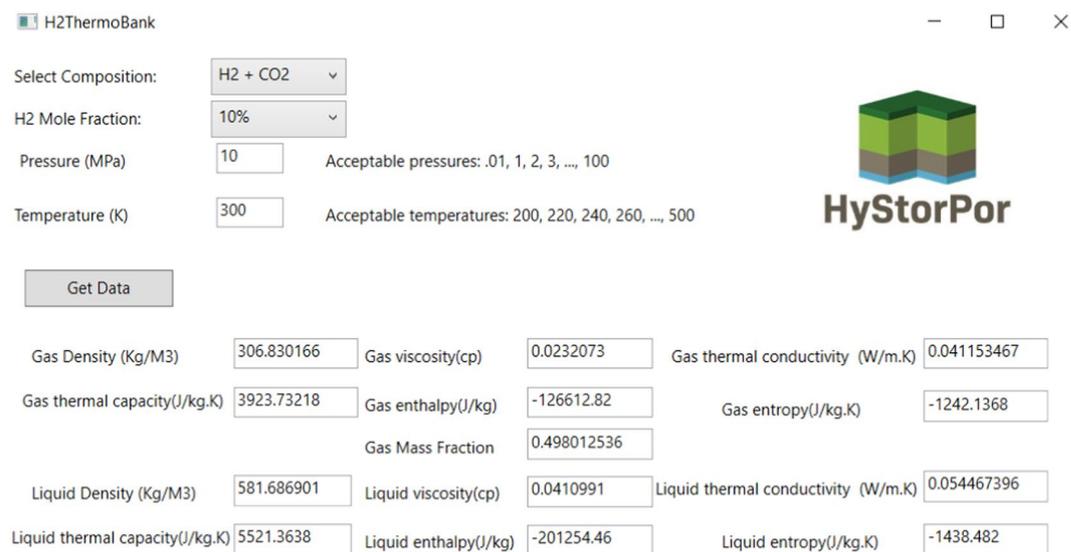


Fig. 9 Example of graphical user interface of H2ThermoBank. Here, we chose a mixture of CO_2 and H_2 with 10% H_2 and 90% CO_2 , at 10 MPa and 300 K and clicked on the “Get Data” button. The image presents the calculated thermodynamic and transport properties of the liquid phase and the gas phase. In addition, the gas mass fraction for this mixture at the entered condition is obtained.

We have utilised published density data of hydrogen/methane blends at a range of pressure and temperature conditions to statistically analyse and assess the reliability and accuracy of the attained modelling data from the GERG-2008 EoS. Figure 8 presents the relative deviations of the predicted densities of GERG-2008 EoS from the

published hydrogen/methane blend experimental data²³. The average absolute deviations (AADs) of the GERG-2008 EoS calculated data used in this study from the experimental data for the various pressures and temperatures are 0.044 for the 10% H_2 + 90% CH_4 mixture and 0.006 for the 50% H_2 + 50% CH_4 mixture. The low AAD values confirm the high accuracy of GERG-2008 EoS predictions with relatively low errors as discussed above.

The errors in viscosity and thermal conductivity estimates at various pressures, temperatures and gas compositions are uncertain due to the lack of experimental data. It is extremely time consuming and almost impossible to measure all the data required using existing laboratory methods. However, the thermal conductivity and viscosity of various compositions calculated from SuperTRAPP model have been compared with experimental data, with an error range of 0–15% reported in the literature⁴⁹.

Usage Notes

H2ThermoBank. A screenshot of the data bank software is provided in Fig. 9. To calculate the required data, the user initially needs to select the gas composition of the closed system. There are 4 options (H_2 + CH_4 , H_2 + N_2 , H_2 + CO_2 , and H_2 + Natural gas). Following this, the H_2 mole fraction in the closed system should be selected. After entering the desired pressure and temperature the “Get Data” button should be clicked to collect the data. For systems of H_2 + CH_4 and H_2 + N_2 there is only one phase exist in the covered range. It is important to note that for H_2 + CO_2 and H_2 + Natural gas system some of the two-phase region is covered in this study (see Fig. 7). For these systems, the user will be able to get both liquid and gas properties together with the mass fraction of the gas phase.

In this study, we have utilised a newly developed tool based on the GERG-2008 EoS and SuperTRAPP model to predict different thermo-physical properties of hydrogen (e.g. density, viscosity, thermal conductivity, etc.) when mixed with other gaseous species including methane, nitrogen, carbon dioxide and a typical North Sea natural gas. The model has been applied to a wide range of pressures, temperatures, and gas mixture compositions which cover the temperature and pressure conditions experienced within the whole hydrogen-based energy system from production to storage in geological formations. The obtained results could be employed by a range of different stakeholders to effectually design and develop innovative infrastructure for the hydrogen economy.

Excel format. To enable easy access to the data over a wide range of temperature, pressure and concentration conditions without requiring running the abovementioned application for each point, four excel files for each of gas mixture systems are provided. Each worksheet in the excel files is allocated to a different mole fraction of hydrogen. Data provided here could be sorted and selected for a required range.

Code availability

The code for H2ThermoBank has been made available on the H2ThermoBank GitHub page (<https://github.com/aliakbarhssnpr/H2ThermoBank>).

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Author contributions

Aliakbar Hassanpouryouzband conceived the idea presented in the manuscript, undertook the modelling study and wrote the manuscript. Edris Joonaki conducted the literature review, undertook the uncertainty/error analysis, evaluated GERG-2008 EoS respect to the experimental data, and contributed to the writing of the manuscript. Katriona Edlmann contributed to the writing of the manuscript and supervised the research. Niklas Heinemann contributed to the writing of the manuscript and edited the final manuscript. Jinhai edited the final manuscript and supervised the modelling study. All the authors discussed the results and contributed to the final manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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