

Computer-aided and experimental determination of thermophysical properties of gas mixtures containing hydrogen

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Abstract—In the project H2MIXPROP, highly accurate data for several thermophysical properties of gaseous mixtures containing molecular hydrogen will be obtained by state-of-the-art theoretical approaches and experimental methods. Such data are required for many technical applications in the transition of the energy supply system to renewable energy sources, in which hydrogen is expected to play a prominent role. This contribution describes the used theoretical and experimental methods, safety measures that have to be applied when handling hydrogen in the laboratories, concepts for adapting existing experimental setups for measurements of mixtures containing hydrogen, the design of two new experimental setups for measurements of second virial coefficients of the mixtures, and the current status of the different tasks in the project.

Index Terms—hydrogen, mixture, computer simulation, Burnett method, virial coefficient, viscosity

I. INTRODUCTION

Thermophysical properties of mixtures containing hydrogen (H_2) are required in the transition of the energy supply system to renewable energy sources in a multitude of technical applications, e.g., in the injection of hydrogen into the natural gas grid or the storage of hydrogen in subsurface caverns. In our project, we predict virial coefficients, viscosity, thermal conductivity, and diffusion coefficients of hydrogen-rich gaseous mixtures on the basis of quantum-chemical *ab initio* calculations of the intermolecular interaction energies. We investigate mixtures of hydrogen with methane (CH_4), nitrogen (N_2), oxygen (O_2), water (H_2O), carbon dioxide (CO_2), hydrogen sulfide (H_2S), ethane (C_2H_6), and propane (C_3H_8). In order to validate the theoretically determined data, experiments for selected mixtures will be carried out with newly developed Burnett instruments at Helmut Schmidt University (HSU) in Hamburg and Physikalisch-Technische Bundesanstalt (PTB) in Berlin, which complement each other with their operating

conditions. Moreover, viscosity measurements will be carried out at HSU with two different viscometers. Eurotechnica consults PTB and HSU with regard to laboratory safety for handling of hydrogen and contributes measurements of density, viscosity, phase equilibria, and diffusion coefficients for selected mixtures. For the use in practical applications, the data determined in the project will be represented by correlating equations. Beyond the data for practical applications, the project will yield valuable insights about the possibilities to replace time-consuming, expensive, and possibly even dangerous experiments (e.g., with hydrogen/oxygen mixtures, so-called oxyhydrogen gas) increasingly by computer simulations in the future. Especially at the high temperatures relevant for combustion processes, experiments are virtually impossible, and computer simulations are the only source for the thermophysical properties at these conditions.

II. FIRST-PRINCIPLES DETERMINATION OF THERMOPHYSICAL PROPERTIES

Thermophysical properties of fluids can today be determined very accurately from first principles (i.e., purely from theory). A prerequisite for such calculations are pair potentials, which describe the interaction energy between two molecules as a function of their separation and mutual orientation. For calculations on dense gases and liquids, so-called nonadditive three-body potentials are additionally required. In the group at HSU, interaction energies are computed by means of the supermolecular approach using standard quantum-chemical program packages. Such calculations have to be carried out for a large number of interparticle separations and mutual orientations in order to obtain a complete description of the intermolecular interaction.

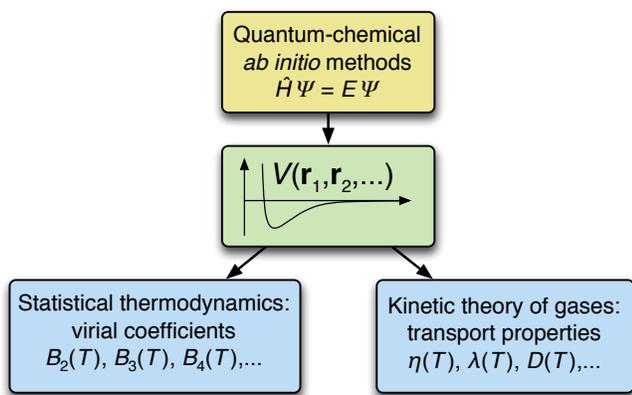


FIGURE 1. ILLUSTRATION OF THE FIRST-PRINCIPLES APPROACH USED AT HSU TO DETERMINE VARIOUS THERMOPHYSICAL PROPERTIES OF GASES AND GAS MIXTURES. THE QUANTITY V IS THE INTERACTION ENERGY BETWEEN TWO OR MORE MOLECULES.

In the next step, suitable mathematical functions are fitted to the calculated interaction energies. Using these potential functions, we can calculate second and higher virial coefficients employing statistical thermodynamics and transport properties in low-density gases and gas mixtures using the kinetic theory of gases. These calculations are performed using unique in-house computer codes.

Whenever possible, the computed property values are compared with experimental data in order to enable a mutual validation of theory and experiment. A great advantage of first-principles approaches over experiments is that a high accuracy can also be achieved at extreme temperatures and for toxic, corrosive, and explosive fluids and fluid mixtures. The experimental effort is often unjustifiably high in these cases.

The described theoretical approach, illustrated as a flow chart in FIGURE 1, is used in this project to determine second virial coefficients, shear viscosities, thermal conductivities, and mutual diffusion coefficients of eight binary systems containing H_2 . In addition to mixtures of H_2 with CH_4 , C_2H_6 , C_3H_8 , N_2 , and CO_2 , for which also extensive experimental work is conducted in this project (see below), mixtures of H_2 with H_2O , H_2S , and O_2 are investigated theoretically. The mixtures with H_2O and H_2S are difficult to handle in experiments due to the corrosive impact of these two gases on the equipment and due to the high toxicity of H_2S . In the case of mixtures of H_2 and O_2 (oxyhydrogen gas), experimental investigations are impossible because this mixture is highly explosive.

Most of the nine pure gases from which the eight selected binary systems are formed were already studied previously, see, e.g., the work of Hellmann on C_2H_6 [1], C_3H_8 [2], and CO_2 [3] or the work of Patkowski et al. [4] and Mehl et al. [5] on pure H_2 . A detailed theoretical investigation of the thermophysical properties of pure O_2 is part of the present project.

For six of the eight binary systems and for pure O_2 , the quantum-chemical calculations are completed by now. The next work step for these systems, the fit of mathematical functions to the interaction energies, is well underway, so that the calculation of the thermophysical properties will soon begin.

III. LABORATORY SAFETY FOR HANDLING HYDROGEN

Some components of the investigated mixtures, e.g., hydrogen, methane, ethane, or propane, form explosive mixtures when mixed with ambient air. Therefore, special safety measures must be applied when experiments with these mixtures are carried out. Our safety concept for the laboratory is based on the principle of “permanently technically tight systems” according to the Technical Rule for Hazardous Substances TRGS 722 for all experimental setups because this way the laboratory room does not need to be declared as an explosive zone. Permanently technically tight systems form also a basic requirement for conducting highly accurate measurements with gas mixtures, especially for the Burnett method. In order to realize permanently technically tight experimental setups, the gas mixtures must be prevented from leaking from the high pressure zones of the experimental setups into the laboratory room. In particular, hydrogen imposes high requirements on the tightness because of the small size of its molecules. Therefore, the tightness of all connections in the high-pressure valve systems and at the pressure vessels is checked by carrying out leak tests with helium before commissioning. These leak tests are always repeated before an apparatus is filled with a new gas mixture. When an experimental setup is filled with hydrogen or a mixture containing hydrogen, rather hydrogen instead of helium is used as test gas. With the leak tests, it is ensured that leaks are detected and cleared before a new measurement campaign is started.

The pressure vessels for the experimental setups are developed by Eurotechnica based on the European pressure directive and will be inspected and certified by TÜV (notified body as per European pressure directive). Hydrogen causes hydrogen embrittlement in some steels, resulting in a drastic reduction of the elongation at fracture. All parts that are in contact with hydrogen and whose fatigue poses a safety risk must be made of materials compatible with hydrogen. To counteract hydrogen embrittlement, stainless steels with a nickel content above 12 % and a low molybdenum content of 2 % to 3 % must be employed [6].

To maximize safety in the laboratory, several worst-case scenarios were considered and evaluated. Due to the small internal volumes of the experimental setups between 100 ml and 1000 ml and the comparatively high volume of the laboratory room (e.g., 152 m³ at HSU), even an unnoticed leakage of the entire filling of a setup could be classified as non-critical because the limit of flammability for all pure gases and gas mixtures will not be exceeded. In addition to the safety measures taken, sensors for the detection of hydrogen,

methane, ethane, and propane are installed in the room to indicate when concentrations of explosive gases exceed the flammability limits in the laboratory room. When the sensors indicate increased concentrations of the explosive gases, an emergency plan, which describes the required measures, applies.

Other scenarios examined are the failure of the cooling system of the thermostat of the torsional crystal viscometer, the temperature increase of hydrogen caused by the Joule–Thomson effect, when a pure explosive gas or a gas mixture is released to the environment, and the formation of an explosive mixture with ambient air upon discharge of the entire filling of an experimental setup to the environment. When the cooling system of the thermostat of the torsional crystal viscometer during operation at temperatures below ambient fails, the temperature and pressure of the gas mixture in the pressure vessel increase. When this failure occurs at low temperatures between 200 K and 240 K at the highest measured pressures between 80 MPa and 100 MPa, the upper pressure limit of the pressure vessel can be exceeded when the thermostat heats up to ambient temperature. Thus, in this range of temperature and pressure, the measurements must not be carried out by automatic operation, but under surveillance of the operator. The release of hydrogen from any of the experimental setups to the environment results only in a temperature increase of 20 K at maximum. Even at the highest operating temperature of the vibrating-wire viscometer, the spontaneous ignition temperature of hydrogen of 858 K will never be reached. The outlet of all high-pressure systems at HSU is above the roof of the laboratory building. When an entire filling of an experimental setup is discharged to the environment, an explosive mixture within a spherical volume of 2.7 m radius around the outlet can form in the worst case. Therefore, warning signs “No Smoking or Open Fires” will be mounted near the outlet on the roof and at the side wall of the building. Overall, the chosen concept ensures safe operation of the experimental setups with hydrogen and mixtures containing hydrogen in the laboratory room.

IV. VIRIAL COEFFICIENTS FROM THE BURNETT METHOD

A. Working Principle of the Burnett Method

The equation of state (EOS) of a gas plays an important role in both academic and industrial applications. The Burnett technique provides a simple experimental method, in which neither the mass nor the volume of gas has to be measured.

The principle of a Burnett apparatus is sketched in FIGURE 2. Two vessels with volumes V_A and V_B are placed in an isothermal bath. The gas to be measured is filled into volume V_A via the supply valve v_1 at a high pressure p_0 . After thermal equilibrium has been established, the EOS of the gas is expressed by

$$p_0 V_A = Z_0 m R T, \quad (1)$$

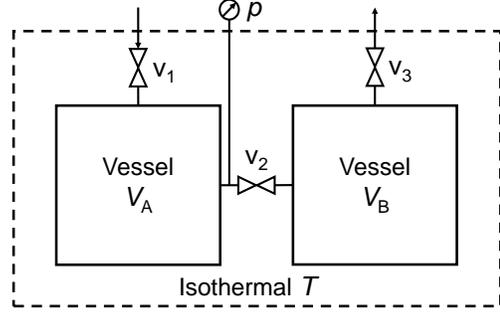


FIGURE 2. WORKING PRINCIPLE OF A BURNETT APPARATUS.

where Z_0 is the compressibility factor at pressure p_0 , m is the mass of the gas, and R is the specific gas constant.

When the expansion valve v_2 is opened, the gas expands into the second vessel. Once thermal equilibrium is established again, the pressure in the system is p_1 , and the EOS is expressed by

$$p_1 (V_A + V_B) = Z_1 m R T, \quad (2)$$

where Z_1 is the compressibility factor at pressure p_1 . Dividing (2) by (1) yields

$$\frac{p_1 (V_A + V_B)}{p_0 V_A} = \frac{Z_1}{Z_0}, \quad (3)$$

in which $(V_A + V_B)/V_A$ defines the volume ratio N of the apparatus.

After closing the expansion valve v_2 and evacuating volume V_B , the remaining gas in volume V_A is expanded again. This process is repeated until the absolute pressure reaches the lowest value that can be accurately measured. For the i th expansion, (3) is written as

$$\frac{p_i N}{p_{i-1}} = \frac{Z_i}{Z_{i-1}}. \quad (4)$$

If the expansion is repeated infinitely often, the gas will be diluted to the state of an ideal gas, which corresponds to the compressibility factor $Z_\infty = Z_{\infty-1} = 1$ and $N = p_{\infty-1}/p_\infty$. The relation of the pressure ratio p_{i-1}/p_i to p_i can be represented by a polynomial:

$$\frac{p_{i-1}}{p_i} = a + b p_i + c p_i^2 + \dots \quad (5)$$

Thus, the volume ratio N can be determined by the intercept of this polynomial when p_i is equal to zero after infinitely many expansions.

From (4), we obtain

$$\prod_{i=1}^n \frac{p_i}{p_{i-1}} N^n = \prod_{i=1}^n \frac{Z_i}{Z_{i-1}}, \quad (6)$$

which can be simplified to

$$\frac{p_n N^n}{p_0} = \frac{Z_n}{Z_0}. \quad (7)$$

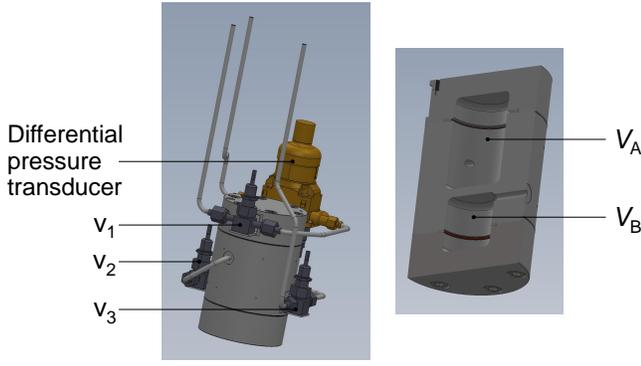


FIGURE 3. DESIGN OF A NEW BURNETT APPARATUS AT HSU. THE LEFT SUBFIGURE SHOWS THE PRESSURE VESSEL WITH THE VALVE SYSTEM AND DIFFERENTIAL PRESSURE TRANSDUCER, AND THE RIGHT SUBFIGURE IS A CROSS SECTION OF THE PRESSURE VESSEL.

Considering $p_n N^n$ as a polynomial in p_n and assuming that n is infinite, so that $Z_\infty = 1$, the intercept of the polynomial at $p_\infty = 0$ is $p_\infty N^\infty$, which by (7) is equal to p_0/Z_0 .

Combining this result with (7), the compressibility factor Z_i for each expansion step is obtained as

$$\frac{p_i N^i}{p_\infty N^\infty} = \frac{Z_0 p_i N^i}{p_0} = Z_i. \quad (8)$$

The virial coefficients are obtained by a least-squares fit of the virial expansion

$$Z_i = 1 + b_2(T)p_i^2 + b_3(T)p_i^3 + \dots, \quad (9)$$

where b_i denotes the i -th pressure virial coefficient, to the compressibility factors of the expansion series as a function of pressure. Finally, the pressure virial coefficients b_i are converted to the density virial coefficients B_i of the density expansion

$$Z = 1 + B_2(T)\rho^2 + B_3(T)\rho^3 + \dots \quad (10)$$

by well-known relations.

B. Design of a New Burnett Apparatus at HSU

In the group at HSU, a new improved Burnett apparatus has been designed and is being set up, as shown in FIGURE 3. It is designed for the temperature range from 233 K to 343 K with pressures up to 7 MPa. The two chambers V_A and V_B have a volume ratio of approximately 1.45 and are dug out from both ends of a pressure vessel, which is made of stainless steel 1.4980. This material is an austenitic precipitation-hardened stainless steel, has a high creep resistance, and is compatible with hydrogen in the working range of the apparatus. The three main valves for supply (v_1), expansion (v_2) and discharge (v_3) of the gas are retrofitted with motors, which are controlled and operated automatically by the data acquisition computer. The hydrogen mixture is separated from the absolute pressure transducer by a differential pressure transducer, in which a membrane with gold plating on the hydrogen side acts as a separator. The branch of the valve system with the absolute

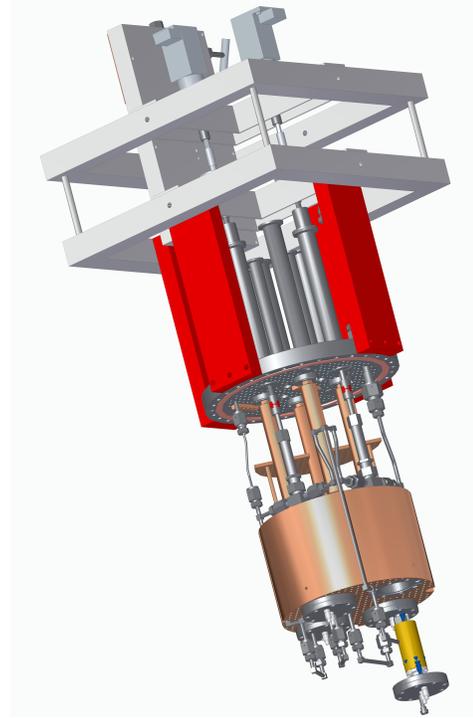


FIGURE 4. DESIGN OF THE NEW COMBINED EXPANSION APPARATUS. CAPACITORS ARE SITUATED IN THE COPPER BLOCK WITH THE EXPANSION VALVES MOUNTED ON TOP. THESE PARTS ARE SURROUNDED BY A VACUUM CHAMBER IMMERSED IN A CONTROLLED WATER BATH. THE PRESSURE SENSORS AS WELL AS THE MOTORS AND THE GEARS FOR THE EXPANSION VALVES ARE OUTSIDE THE WATER BATH.

pressure transducer is filled with nitrogen. The pressure vessel, the three valves v_1 , v_2 , and v_3 and the differential pressure transducer are thermostated in a circulating liquid bath thermostated with silicone oil as thermostating liquid. The thermostat ensures a temperature stability better than 0.5 mK. Since the uncertainty of the measured virial coefficients is mostly influenced by the uncertainty of the pressure measurement, the absolute pressure transducer is calibrated before each expansion series with a highly accurate gas piston gauge. In order to avoid zero point shifts of the differential pressure transducer during a measurement campaign, the zero point is also calibrated before each expansion series. The temperature is measured by a long-stem standard platinum resistance thermometer calibrated on the ITS-90 in the wall of the pressure vessel with an uncertainty of a few mK by a resistance bridge system.

The design of the pressure vessel, valve system, and pressure measurement system is completed. Technical drawings for the pressure vessel and all mechanical parts were issued, and the manufacturing has been started. Moreover, a concept for automation of the calibration of the pressure sensors and measurement of an expansion series was developed, and some components of the data acquisition software have been coded in the graphic programming environment LabVIEW.

C. Implementation of a Burnett Apparatus at PTB

The apparatus designed at PTB is a combination of three different methods. The first and extensively explored method is the so-called Dielectric-Constant Gas Thermometry (DCGT) based on replacing the density in the equation of state of a gas by the dielectric constant. The dielectric constant is determined via the change of the capacitance of a capacitor measured with and without the gas. This method of primary thermometry has already been successfully used to determine combinations of density and dielectric virial coefficients [7], [8]. The drawback of this approach is the impossibility to differentiate between both contributions since capacitance and pressure data is utilized in combination. That is why these experiments are coupled with Burnett and dielectric expansion experiments where pressure and capacitance ratios are defined by repeated expansion of gas from one into another volume as mentioned in Section IV-A. From these ratios, the independent density and dielectric virial coefficients can be determined without the need to assess the absolute particle density. Both methods typically require large experimental setups whose manual operation is time consuming. In the last years at PTB, a compact apparatus was developed that combines both experimental approaches, in which the expansions are fully automated [9], [10]. This apparatus was used for experimental studies on noble gases. Within the framework of the project, a new improved apparatus especially dedicated to measurements of hydrogen/methane mixtures will be built (see FIGURE 4). In contrast to the forerunner, the expansion valves will be situated inside the vacuum chamber, reducing the amount of gas not being thermostated precisely at the designated temperature. Since, for thermal reasons, the motors for the valves are outside the vacuum chamber, the use of appropriate vacuum feedthroughs, torque regulators, and so forth must be included in the design. Furthermore, the new apparatus will have an improved thermal environment of the expansion cells. The materials for the parts in contact with the gas have been proven to be compatible with hydrogen. Beside these large changes, there are other lessons learned from the first apparatus, so that the data evaluation should be more straightforward for the new setup.

V. VIBRATING-WIRE AND TORSIONAL CRYSTAL VISCOMETERS

A. Vibrating-Wire Viscometer

The vibrating-wire viscometer at HSU was originally developed by Seibt [11] at the University of Rostock in the group of Prof. Eckhard Vogel and was applied there to measure the viscosity of gaseous nitrogen, methane, ethane, propane, *n*-butane, and isobutane [11], [12]. After the retirement of Prof. Vogel, it was transferred to HSU, rebuilt in our laboratory, and used to measure the viscosity of the noble gas neon by Kochan-Eilers [13]. The experimental setup (FIGURE 5) consists of a single-sinker densimeter and three vibrating-wire

sensors (FIGURE 6) in a single pressure vessel and enables simultaneous measurements of the density and viscosity of a gas. It covers the temperature range between 293 K to 498 K at pressures up to 30 MPa. The density measurements are characterized by a relative expanded uncertainty (coverage factor $k = 2$) of 0.1 %, except in the low density range, where it is larger, while the relative expanded uncertainty ($k = 2$) in the viscosity is estimated to be 0.25 % to 0.3 %.

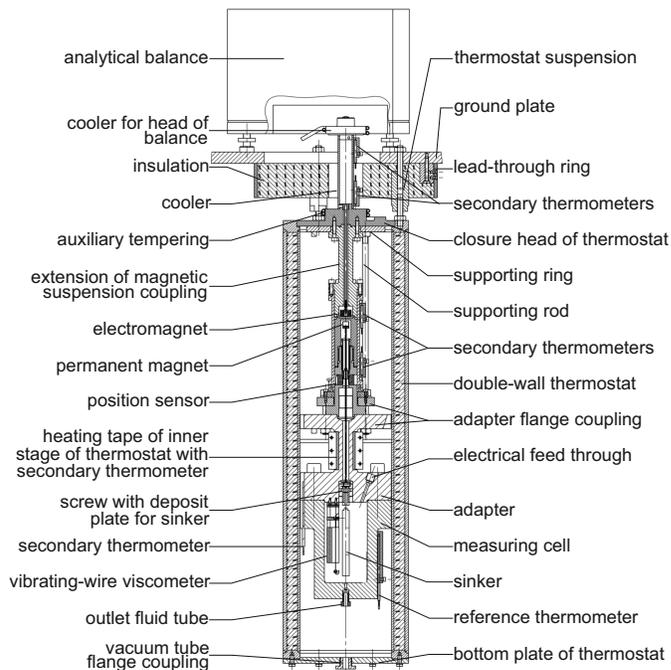


FIGURE 5. CROSS-SECTION OF THE VIBRATING-WIRE VISCOMETER USED AT HSU.



FIGURE 6. PHOTO OF A VIBRATING-WIRE SENSOR (WIRE NOT TENSIONED).

A vibrating-wire sensor (FIGURE 6) consists of a Chromel[®] wire with a diameter of 25 μm , which is clamped at both ends and tensioned between two permanent samarium-cobalt magnets by a lever arm with a weight at its lower end. By applying a sinusoidal alternating voltage along the wire, it is excited to vibrate. After switching off the excitation voltage, the magnetic field between the two magnets induces an alternating current in the freely vibrating wire. By measuring the voltage along the wire as a function of time, the damping of the vibration can be recorded and evaluated. The viscosity η of the gas is determined by the working equation

$$\eta = \frac{2\pi f_F \rho R^2}{\Omega}, \quad (11)$$

where R is the radius of the wire, ρ is the density of the fluid, f_F is the resonant frequency of the wire in the gas, Ω is a function of the mechanical properties of the wire, and the resonant frequency and logarithmic decrement of the vibration of the wire in vacuum and in the fluid. The radius of the wire is determined from calibration measurements with helium, while the density of the gas can either be measured with the single-sinker densimeter or calculated with an equation of state if an accurate one is available for the gas. The resonant frequency and logarithmic decrement of the vibration in vacuum and in the fluid are determined from the measured voltage data of the decay of the free vibration.

For measurements of hydrogen and mixtures containing hydrogen, some components of the valve system, a differential pressure transducer, which couples the measured gas with the nitrogen in the pressure measurement system, and the pressure vessel must be replaced by components, in which the parts in contact with hydrogen are made of materials compatible with hydrogen. The differential pressure transducer (Emerson, type 3051) is replaced by a new transducer of the same type, in which the stainless steel membrane, which separates the measured gas from the nitrogen, is gold plated on the side in contact with hydrogen. The gold plating prevents hydrogen from diffusing into and through the membrane. A new pressure vessel made of a copper alloy with high mechanical strength and high thermal conductivity is designed by Eurotechnica. The certification of the pressure vessel by the TÜV requires more thorough proofs than for pressure vessels of conventional materials because copper alloys have not often been applied yet for pressure vessels operated with hydrogen in the temperature and pressure range of the vibrating-wire viscometer.

Additionally, further changes will be introduced to improve the performance of the viscometer. Beside the Chromel wire, a gold-plated tungsten wire with a smooth surface will be tested. Tungsten has a higher density, a higher elastic modulus, and a lower coefficient of thermal expansion than Chromel, and the application of tungsten wires is expected to improve the reproducibility of the measurements. In previous realizations of vibrating-wire viscometers, mostly pure tungsten wires were used, which had a very rough surface [14]. With the gold

plating, the surface roughness of tungsten wires is reduced considerably.

With the current setup, the temperature control of the thermostat of the viscometer was sometimes unstable because of heat losses at the top cover of the thermostat. A new temperature-controlled heating coil will be applied near the top cover to decouple the pressure vessel in the thermostat thermally from the laboratory environment. In addition, a stand-alone setup with a single vibrating-wire sensor in a new pressure vessel will be built. The pressure vessel will be thermostatted in a circulating liquid oil bath of a calibration thermostat with a temperature stability of 5 mK (Fluke Calibration 7341). The pressure vessel is also designed by Eurotechnica and made of the stainless steel 1.4980. It is expected that thermal equilibrium in the pressure vessel and gas is attained much faster than in the rather complex combined densimeter viscometer due to the convective heat transfer in the liquid bath thermostat.

Moreover, the operation of the viscometer will be automated. A pressure controller (WIKA, type CPC8000) will be used to control the pressure of the nitrogen in the pressure measurement system branch of the valve system. The manual valves for discharging gas from the high pressure system will be replaced by pneumatic valves, which will be controlled by the data acquisition computer. By using a computer-controlled relay switch system, the data acquisition computer can automatically switch between the three different vibrating-wire sensors.

B. Torsionally Vibrating Crystal Viscometer

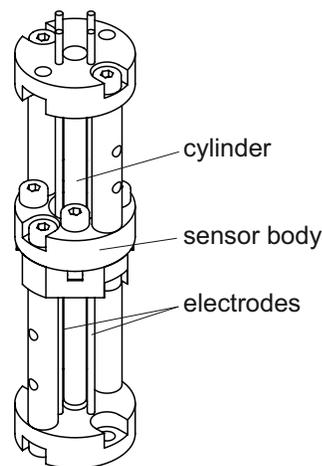


FIGURE 7. THREE-DIMENSIONAL VIEW OF THE SENSOR OF THE TORSIONALLY VIBRATING CRYSTAL VISCOMETER.

The use of piezoelectric cylinders, which vibrate in the fundamental torsional mode, as viscosity sensors was first suggested by Mason [15]. Almost exclusively piezoelectric quartz cylinders with diameters of a few mm and lengths of a few cm have been applied, whose cylinder axis is

aligned in parallel with the X axis of the crystallographic lattice coordinate system of quartz. Four separate electrodes with their centers at angles of $\pi/4$ between the Y and Z axes surround the crystal as separate parts in a quadrupole arrangement. When an alternating electric field is applied to the electrodes, the cylinder is excited to torsional vibrations, which are damped by the surrounding fluid. The working equation of the torsionally vibrating crystal viscometer is given by

$$\eta\rho = \left(\frac{m}{S}\right)^2 \pi f_F \left(\frac{\Delta f_F}{f_F} - \frac{\Delta f_0}{f_0}\right)^2, \quad (12)$$

where m is the mass of the cylinder and S denotes its surface area. The resonant frequency f_F and the bandwidth Δf_F in the fluid and the respective quantities in vacuum f_0 and Δf_0 are determined from measured curves of the conductance and susceptance of the viscosity sensor in the vicinity of the resonant frequency in the frequency domain. The mass of the cylinder is accurately determined by weighing with a mass comparator, while the surface area can in principle be determined from dimensional measurements of the length and of the curved surface of the cylinder. In practise, an effective radius of the cylinder is determined by calibration measurements with a liquid viscosity standard. The density must either be measured separately or calculated with an equation of state.

Junker developed and operated a torsionally vibrating crystal viscometer for measurements of liquids in the temperature range between 240 K and 420 K with pressures up to 100 MPa at HSU [16]. As part of that work, the electric field in the viscosity sensor was analyzed [17], and the deformation of the anisotropic quartz cylinder in the fundamental torsional mode was investigated in detail [18]. Here, the viscometer will be used to measure the viscosity of hydrogen mixtures, especially at temperatures below ambient, where the vibrating-wire viscometer cannot be applied. The pressure vessel will be replaced by a new one made of stainless steel 1.4980 developed by Eurotechnica, while all parts of the valve system (SITEC, 1000 Micro) are made of stainless steel 1.4571 and are compatible with hydrogen. The computer-controlled syringe pump used to set the pressure when measuring liquids will be replaced by an electric pump with a larger swept volume for measurements of gases (SITEC 750.6101-1-sp with hydrogen-compatible materials).

The data acquisition system of the viscometer will be extended by a fast digital oscilloscope card (Spectrum Instruments, type M4i4420-x8) for measurements of the free vibration in the time domain, which reduces the required time for a measurement considerably compared to measurements of complete resonance curves in the frequency domain. This technique was first realized with quantitative results by Hafer and Laesecke [19]. Furthermore, it will be examined if the dimensions of the cylinder can be determined by measurements with an optical technique with sufficient accuracy to realize absolute measurements of the viscosity without the need for

calibration. Additionally, the surface roughness of the cylinder will be characterized, and its influence on the results for the viscosity will be examined.

VI. OUTLOOK

In the first phase of the project, the potential energy surfaces for the eight investigated binary mixtures with hydrogen and pure oxygen are developed in the theoretical part of the project. On the experimental side, the Burnett apparatuses at HSU and PTB were drafted, technical drawings for the main subassemblies were prepared, wrought materials, components, and measurement devices were selected and ordered, and manufacturing of the mechanical parts of the setups was started. Moreover, a concept for automation of the Burnett apparatus at HSU was developed and some components of the data acquisition software were written. For the vibrating-wire and torsionally vibrating crystal viscometers at HSU, concepts for their adaption for measurements of hydrogen mixtures and their automation were developed.

In the next phase, the potential energy surfaces will be finalized and applied to calculate the thermophysical properties of the mixtures and pure oxygen. Moreover, the experimental setups at HSU and PTB will be assembled in the laboratories and commissioned. Before the measurements of the hydrogen mixtures, calibration measurements of the new Burnett apparatuses and the vibrating-wire viscometer with helium will be carried out. Subsequently, the Burnett apparatuses will be validated by measurements with argon and nitrogen, for which the second virial coefficients are very accurately known.

ACKNOWLEDGMENT

This work was funded by dtec.bw – Digitalization and Technology Research Center of the Bundeswehr [Energy and Digitalization].

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