Vacuum 203 (2022) 111237

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Towards the first direct measurement of the dynamic viscosity of gaseous tritium at cryogenic temperatures

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ARTICLE INFO

Keywords:
Viscosity
Spinning rotor gauge
Tritium

ABSTRACT

Accurate values for the viscosity of the radioactive hydrogen isotope tritium (T_2) at cryogenic temperatures are unavailable. Values for tritium found in literature are based on extrapolation by mass ratios as well as an empirical factor derived from hydrogen (H_2) and deuterium (D_2) viscosity measurements, or classical kinetic theory which does not handle quantum effects. Accurate data of the tritium viscosity will help to improve the modelling of the viscosity of diatomic molecules and can be used as a test of their interaction potentials.

With this contribution we report a major step towards a fully tritium and cryogenic temperature compatible setup for the accurate measurement of the viscosity of gases, using a spinning rotor gauge (SRG) at the Tritium Laboratory Karlsruhe. After calibration with helium, measurements with hydrogen and deuterium conducted at room temperature agree with literature values within 2%. The performance at liquid nitrogen (LN_2) temperature has been successfully demonstrated with a second setup in a liquid nitrogen bath. Again after calibration with helium at LN_2 temperature, the viscosities of H_2 and D_2 were determined and are in agreement with literature to about 2%.

1. Introduction

The radioactive hydrogen isotope tritium in significant amounts is of interest in astroparticle physics and as a fuel for fusion power plants. In particular, tritium with its low energy beta spectrum is one of the most promising candidates in experiments aiming at the direct kinematic measurement of the electron anti neutrino mass [1–3].

The Karlsruhe Tritium Neutrino Experiment (KATRIN) collaboration has recently published the world leading experimental upper limit of 1.1 eV on the electron anti-neutrino mass from high precision β-spectroscopy of tritium [4,5]. In the KATRIN experiment, a windowless gaseous tritium source (WGTS) is used. In this source, tritium gas is injected into the centre of a beam tube, which is kept at a chooseable but ultra stable temperature between 30 K and 100 K, and then pumped out at its ends [6].

The density profile of the tritium gas, which determines the total luminosity, energy loss and other systematic effects of the β-decay electrons is therefore an important systematic for the neutrino mass analysis. From direct measurements, like electron transmission and energy loss measurements with an electron-gun and monoenergetic sources like krypton [7], only the longitudinal path integral of the gas density profile can be measured directly. Therefore, accurate cross-calibration with simulations of the gas density profile is crucial to understand its systematic influence to the neutrino mass analysis. One important input parameter for these simulations is the dynamic viscosity of gaseous tritium. To the best knowledge of the authors, experimental and theoretical literature values for this transport property of tritium in the temperature region of interest for KATRIN are unavailable. Theoretical values for the viscosity of tritium above 300 K calculated using a classical approach have been published recently [8], however, as this calculation is neglecting quantum mechanical effects, no results were given for low temperatures. The values currently used in KATRIN for the viscosity of tritium at cryogenic temperatures are extrapolations from the viscosities of hydrogen (H_2) and deuterium (D_2) based on their atomic mass ratios and an empirical factor [6,9]. As a result, the relative uncertainty on the tritium viscosity at cryogenic temperatures has been assumed to be ≈10% [6,10]. In order to improve this, an experimental setup has been built to measure the tritium viscosity at cryogenic temperatures with an accuracy <2%.

The gas viscosity can be measured using several different methods such as the capillary flow, the oscillating body, the vibrating wire or

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https://doi.org/10.1016/j.vacuum.2022.111237
Received 5 January 2022; Received in revised form 27 April 2022; Accepted 7 June 2022
Available online 10 June 2022

the torsional crystal method [11]. Our selection was based on cryogenic and tritium compatibility, which means fully metal vacuum systems including metallic seals [12]. For these reasons, a variation of the oscillating body method using a spinning rotor gauge (SRG) [13] was chosen. A spinning rotor gauge consists of a spherical, metallic rotor which is magnetically levitated inside of a vacuum system. This rotor is accelerated magnetically to rotate at frequencies of several hundred Hz. Interaction with gas atoms or molecules around the rotor cause a drag force, slowing down the rotor, which can be measured as a decrease in rotation frequency. In the slip regime, this drag force depends on the viscosity of the surrounding medium, allowing for a measurement of the viscosity.

In this article, we report on the construction of a demonstrator setup for the tritium-compatible measurement of gas viscosities at cryogenic temperatures and demonstrate the operational readiness with calibration using helium and measurements of the viscosities of H_2 and D_2 at room and liquid nitrogen temperature.

2. Measurement principle

The measurement of gas viscosity with a spinning rotor gauge is based on the decrease of the angular rotor speed Ω due to interaction with the surrounding gas, which is measured in the form of the (normalized) angular deceleration rate of the rotor:

$$\frac{\mathrm{d}\Omega/\mathrm{d}t}{\Omega}. \tag{1}$$

Using the moment of inertia of a spherical rotor with diameter a_1 and density ρ:

$$I = \frac{8\pi}{15} a_1^5 \rho, \tag{2}$$

the torque D exerted onto the SRG rotor by gas molecules can be obtained from the angular deceleration rate:

$$\frac{D}{\Omega} = I \frac{\mathrm{d}\Omega/\mathrm{d}t}{\Omega}. \tag{3}$$

For the slip regime, an equation relating the left-hand side of Eq. (3) to the gas viscosity μ has been derived in [13–15] (which used the symbol T for the torque D as well as for the temperature T):

$$\frac{1}{D/\Omega} = \frac{1}{8\pi a_1^3 C_0 \mu} + \frac{1}{p} \cdot \sqrt{\frac{2k_B T}{m}} \left(\frac{c_m}{8\pi a_1^3 C_0} \left(\frac{3}{a_1} + \frac{1}{a_2} \right) \right), \tag{4}$$

where a_2 is the radius of a cylinder surrounding the SRG rotor, k_B the Boltzmann constant, T the temperature of the gas, m the molecular mass of the gas, c_m the velocity slip coefficient, p the pressure surrounding the SRG rotor, and C_0 a factor describing the flow around a sphere rotating around an axis perpendicular to the axis of a surrounding cylinder. No analytic expression for the parameter C_0 has been derived at present, so it needs to be determined experimentally [14]. The pressure p needs to be determined with an separate pressure sensor, which needs to be gas species independent.

Using the linear relation of Eq. (4) between the inverse quantities $(D/\Omega)^{-1}$ and p^{-1}, which are both experimentally accessible, the viscosity μ and the velocity slip coefficient c_m can be obtained by fitting a linear function. In particular, the viscosity is determined by the offset y_0 of the linear fit:

$$\mu = \frac{1}{8\pi a_1^3 C_0 y_0}. \tag{5}$$

The unknown parameter C_0 in this equation can be determined via a calibration measurement using helium as the viscosity of helium μ_{He} is known very accurately (uncertainty ≈0.1% from ab initio calculations [16]). Taking μ_{He} as given, Eq. (5) can be rearranged to derive C_0. Care has to be taken that both, this calibration and later measurements, are taken at the same temperature T to exclude any implicit temperature dependency of C_0.

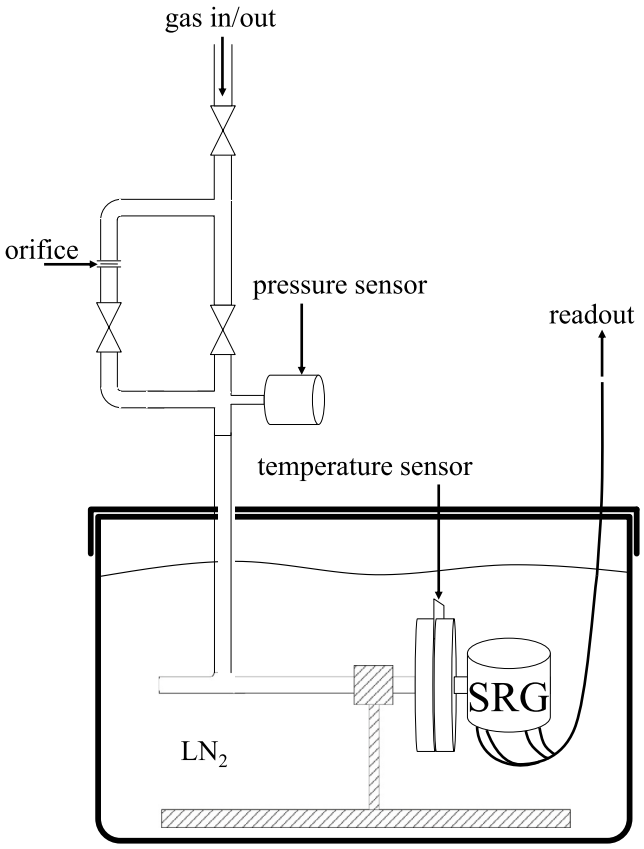

Fig. 1. Experimental setup for first cryogenic measurements. The SRG is placed inside a liquid nitrogen bath.

3. Experimental setup

A schematic drawing of the experimental setup is shown in Fig. 1. The entire experiment is built using stainless steel components and metal sealings (VCR® and ConFlat®-compatible) to ensure compatibility with tritium operation in the future. The sample gas is transferred to the measurement volume (or cavity) of the spinning rotor gauge (SRG) via an orifice, giving the possibility of adjusting the pressure of the gas on the 1 Pa level. As a gas species independent pressure sensor, a capacitive diaphragm manometer (MKS 627) is used. The heart of the setup is the spinning rotor gauge (MKS SRG-3), with an inner cylinder diameter of 0.7 cm and a rotor diameter of 0.45 cm. Two such setups, ViMA1 and ViMA2 (Viscosity Measurement Apparatus), have been built. For the measurements reported in this paper, the temperature sensor was mounted to the outside surface of the vacuum system, and therefore does not directly measure the temperature of the gas. For future measurements, a Pt100 temperature sensor has been installed inside the gas volume close to the SRG. In the ViMA2 setup, the SRG is placed inside of a cold box which can be filled with liquid nitrogen. The capacitive pressure sensor is connected via flexible metal tubing from outside of the cold box, allowing for the pressure sensor to stay at room temperature. Directly after installation, the whole system is evacuated over the weekend with a turbo molecular pump. The gas bottles we used were specified by the manufacturer with 99.8% atomic purity for deuterium and 99.9999% purity for hydrogen, where the major impurities in the deuterium is hydrogen and other species are 1 ppm.

4. Data taking and analysis

Measurement procedure. The first step of each measurement is the purging of the setup with sample gas to minimize impurities. For

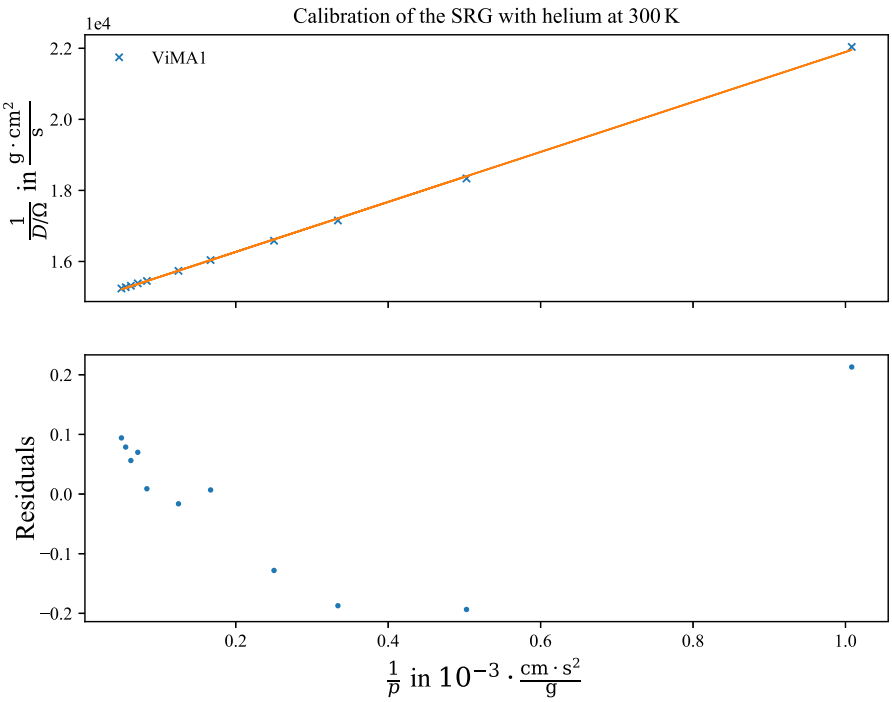

Fig. 2. Sample linear regression for the calculation of the calibration factor C_0 by the measurement with helium.

cryogenic measurements, liquid nitrogen is filled into the cold box, otherwise this setup can be operated at room temperature. Afterwards, the sample gas is filled into the SRG cavity up to a chosen pressure. The measurement starts, when the rotational speed of the rotor inside the SRG has reached its upper limit, 440 Hz in our case. The deceleration of the rotor is measured together with the pressure of the gas, the temperature and the rotational speed of the rotor. When the rotor speed reaches its lower limit, (420 Hz in our case), the data taking is paused, until the rotor is re-accelerated to its upper speed limit. This procedure is repeated until the measured deceleration reaches a plateau, meaning it has equilibrated, which happens after a few minutes. This has to be done, since the rotor is heated up during the initial acceleration from 0 Hz (compare to [13]). When the system is in equilibrium, data is taken for several minutes before the measurement is stopped. This procedure is repeated for different pressure values, to accumulate enough data for the linear fit described in Section 2. The pressure values are stepped through upwards and downwards to check for hysteresis effects. These two options produce slightly different values for the viscosity with a deviation of about 0.2%, which is likely caused by heating effects of the rotor. A detailed investigation of this effect is currently being carried out to further increase the accuracy of the measurement. Since the used method requires slip regime conditions, the pressures for which data is taken are 100 Pa to 2000 Pa at room temperature and 20 Pa to 500 Pa at liquid nitrogen temperature.

Uncertainty estimation. By operating the viscosity measurement setup at the same temperature T_0 for the measurement of μ and a calibration measurement of the viscosity of helium μ_{He}, the uncertainty of the measured viscosity is determined by the uncertainty of the y-intercept of the linear fit. The statistical uncertainty on the y-intercept σ_{y_0} we find in our measurements is on the order of 0.1%. The uncertainty of the viscosity of helium from ab initio calculations is <0.1% in the temperature range from 10 K to 10^4 K [16], and can be considered to be negligible in our case. A comparatively large measurement uncertainty σ_{T_0} on the temperature T_0 of ≈0.5 K in our current setup leads to a much larger impact on the value of $\mu_{He}(T_0)$ used to calculate C_0. This temperature uncertainty will be improved with a better temperature measurement instrumentation, planned for the final setup. Other thermal effects are currently under investigation and not implemented yet in the actual data analysis, so the uncertainties given for the results are all only statistical uncertainties. Since temperature stabilization for the measurements at room temperature is not given with the current setup, the temperature between two viscosity measurements can vary by up to 5 K, which is one of the reasons for a deviation between two measurements of up to 2%. This will be improved on the way to a fully tritium compatible setup.

5. Measurements and results

An example of a C_0 calibration as described in Section 2 is shown in Fig. 2. Several calibration factor measurements were done at both room and liquid nitrogen temperature. Their mean values and 1 σ standard deviations are listed in Table 1. We have found good agreement between both of our systems ViMA1 and ViMA2 with each other and with the C_0 values measured in [13] using an MKS SRG2 system, the predecessor model to the SRG used in this work with similar cylinder and rotor geometry. We have also found a slight decrease of C_0 at liquid nitrogen temperature compared to room temperature. Using the calibration factors for our systems, we have measured the viscosity of H_2 and D_2 at both room and liquid nitrogen temperature. The data of several such measurements for H_2 at room temperature is shown in Fig. 3. The mean values and standard deviations of these viscosities are listed in Table 2.

All values agree with literature values within about 2% [17,18]. The values measured in the liquid nitrogen bath tend to be systematically above the literature values. A possible cause might be a slight heating of the gas around the SRG rotor compared to the temperature of the surrounding vacuum vessel walls. Therefore, great care must be put in temperature calibration and understanding of temperature gradients in the successor setup. At room temperature our values differ from the literature values in both directions due to the influence of varying laboratory temperature in our first setup. In general, we conclude that our measured values for the viscosity of hydrogen and deuterium are in agreement with the literature values within <2%. By this, we are confident to obtain experimental values which improve on the accuracy of the tritium viscosity compared to presently available estimations. In addition, temperature stabilization in an improved setup and in-depth study of the systematic effects will help to further improve the accuracy of the measurement.

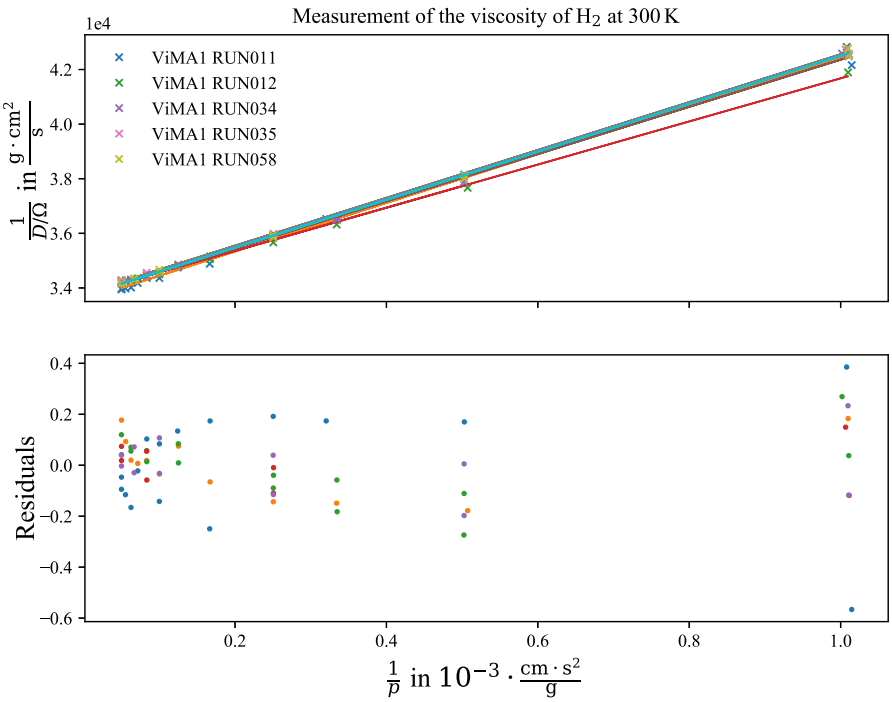

Fig. 3. Linear regressions for the calculation of the viscosity of hydrogen at room temperature. The temperature between these measurements varies between (299.0 ± 2.5) K.

Table 1
Average calibration constant C_0 and its standard deviation σ_{C_0} for ViMA1 and ViMA2 at room temperature and liquid nitrogen temperature, compared to [13]. For liquid nitrogen temperature no literature value is available. A slight temperature dependence of C_0 can be seen.

Setup	Temperature in K	C_0	σ_{C_0}	Literature
ViMA1	299.0 ± 2.5	1.184	0.02	1.1783
ViMA2	299.0 ± 2.5	1.199	0.02	
	LN_2	1.134	0.02	

Table 2
Average viscosity and its standard deviation for different gases and temperature, compared to earlier measured values. For H_2 and D_2 at 299 K, only two measurements each are available for ViMA2. Therefore, the standard deviation is omitted due to lack of statistics. The literature values given by a range vary between 298 K and 301 K.

Setup	Gas	Temperature in K	μ in µPa s	σ_μ in µPa s	Literature
ViMA1	H2	299.0 ± 2.5	8.74	0.15	8.91–8.97 [17]
	D2	299.0 ± 2.5	12.93	0.1	12.67–12.77 [17]
ViMA2	H2	299.0 ± 2.5	8.891		
		LN_2	3.51	0.02	3.454 [18]
	D2	299.0 ± 2.5	12.581		
		LN_2	4.83	0.02	4.79 [18]

6. Summary & outlook

A demonstrator setup for the tritium compatible measurement of gas viscosities at liquid nitrogen temperature and room temperature based on a spinning rotor gauge has been reported in this paper. By extending a setup presented in literature with a cold box, we managed to measure the viscosity of hydrogen and deuterium at liquid nitrogen temperature and 300 K with an accuracy of <2%, and achieved values which are in good agreement with literature values. The dominant source of uncertainty in the current setup is the uncertainty on the gas temperature inside the system and the inability to stabilize it against fluctuations of the laboratory temperature of up to several K.

To improve on our results in the short term, a dedicated temperature sensor inside the vacuum system has been installed, which measures the temperature of the gas close to the SRG rotor instead of the temperature of the walls of the vacuum system. To improve the uncertainty in the long term, and to measure the viscosity of tritium over a wide temperature range, a tritium compatible cryostat is currently under construction, which will allow temperature stabilized ($\Delta T < 0.1$ K) measurements over the entire temperature range from liquid nitrogen temperature to 330 K.

CRediT authorship contribution statement

Johanna Wydra: Writing – original draft, Visualization, Supervision, Investigation. **Alexander Marsteller:** Writing – original draft, Supervision, Software, Conceptualization. **Robin Größle:** Writing – review & editing, Writing – original draft, Supervision. **Linus Schlee:** Investigation. **Florian Priester:** Writing – review & editing, Conceptualization. **Michael Sturm:** Writing – review & editing, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

We acknowledge the support of the Helmholtz Association (HGF), Germany, the German Ministry for Education and Research BMBF (05A20VK3) and the DFG graduate school KSETA, Germany (GSC 1085).

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