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First Experimental Results on the Tritium Viscosity in the Zero-Density Limit from 110 k to 300 k

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Abstract — *The viscosity of tritium is an important property for fusion science and technology as well as fundamental research concerning tritium-based astroparticle physics experiments such as the KATRIN experiment. As of now, only theoretically calculated values with uncertainties of up to 7% are publicly available. No experimental results are found in literature, as there are not many laboratories with the license to handle the macroscopic amounts of tritium required to conduct experiments measuring material properties such as viscosity. The Tritium Laboratory Karlsruhe is unique in Europe, with a license to handle up to 40 g of tritium and around 30 g of tritium on site. With this contribution, we first present experimental values for the viscosity of tritium in the zero-density limit in a temperature range from 110 K to 300 K with a refined analysis. The measurement setup Cryogenic Viscosity Measurement Apparatus (Cryo-ViMA) is based on a spinning rotor gauge and was previously tested with helium, hydrogen, and deuterium in a temperature range from 200 K to 300 K. These results were reproduced with the final setup inside a glovebox, with a wider temperature range of 100 K to 300 K, before the measurement was conducted with tritium. The cooling is realized with a cold gas system utilizing liquid nitrogen. With this, temperatures down to 100 K can be reached, and there is a plan to upgrade the system for viscosity measurements at lower temperatures with the aim of reaching an accuracy of 2%. Through thermal cycling of the system, high resolution of the temperature dependency of the viscosity of tritium can be reached to test previous calculations.*

Keywords — *Tritium, viscosity, spinning rotor gauge, cryogenic temperatures.*

I. INTRODUCTION

Over recent decades, the importance of tritium has increased dramatically, mainly caused by fusion technology [1–4], which uses tritium as as fuel for clean, base load power generation. As the development of the fusion reactor components progresses, the knowledge of tritium properties is gaining importance as well. One important material property of interest is the viscosity of tritium. Ab initio calculations of the viscosity μ have been performed based on kinetic theory [2]:

$$\mu = \frac{5 \cdot \sqrt{\pi \cdot m \cdot k_B \cdot T}}{16 \cdot \pi \cdot \sigma^2 \cdot \Omega^{(2,2)^*}} \cdot f_\mu \tag{1}$$

where m is the mass of the molecule, k_B is the Bolzmann constant, T is the temperature of the gas, σ is the interaction cross-section, and $\Omega^{(2,2)^*}$ the collision integral of the molecule. The correction factor f_μ accounts for different

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types of interactions between the molecules [2]. As the viscosity extrapolated from measurements with the spinning rotor gauge (SRG) is the viscosity in the zero-density limit where interactions between more than two molecules are rare, f_μ must be close to 1. Another way to calculate the viscosity of tritium also utilizes Eq. (1), but uses it to extrapolate the viscosity of hydrogen to that of tritium by scaling with the mass ratio [5]. The validity of this scaling assumption has not previously been experimentally verified. Recently, initial experimental results on the viscosity of tritium were published [6]. Within this paper, we will show why these values must be reevaluated with a new calibration.

II. MEASUREMENT PROCEDURE AND ANALYSIS WITH A SPINNING ROTOR GAUGE

The measurement setup is described in detail in prior publications [6,7]. The measurement setup is based on an SRG, which consists of a stainless steel cylinder, in which a rotating sphere, the rotor, is held in suspension by a magnet system. The magnet system accelerates the rotor to a frequency of 440 Hz. Once acceleration ceases, the subsequent deceleration of the rotor is measured in parallel with the pressure and temperature of the sample gas inside the cylinder. When the frequency decreases to 420 Hz, the rotor is re-accelerated. The SRG is mounted inside a cryostat that can be cooled to 100 K using a cold-gas system. For viscosity measurements, tritium is filled into the cylinder at a defined pressure, after which the cylinder volume is closed, leading to an isochoric setup with a fixed number density. During the measurement at this given filling pressure setpoint, the cryostat is first cooled to approximately 100 K and then gradually warmed up again. Only the warmup phase is used for viscosity analysis, since the cooling process is too rapid for the system to reach sufficiently stable conditions for a measurement with low systematic error. After completing one such thermal cycle, the cylinder pressure is adjusted and the measurement sequence is repeated.

The analysis of the measurement consists of linearly fitting the inverted normalized deceleration rate (DCR) of the SRG rotor to the inverted pressure p in the form of

$$\frac{1}{\frac{\partial\Omega}{\partial t}/\Omega} = \frac{I}{8\pi a_1^3 C_0 \mu} + \frac{I}{p}\cdot\sqrt{\frac{2k_B T}{m}}\left(\frac{c_m}{8\pi a_1^3 C_0}\left(\frac{3}{a_1}+\frac{1}{a_2}\right)\right) \quad (2)$$

as first shown in Ref. [8]. In this equation, $\frac{\partial\Omega}{\partial t}/\Omega$ is the *DCR*, I is the moment of inertia of the rotor, a_1 and a_2 correspond to the radii of the rotor and the cylinder, respectively, C_0 is a calibration constant, and c_m the velocity slip coefficient. The calibration constant cannot be determined analytically for the present setup and and is derived experimentally by calibrating against helium. The calibration procedure follows the same procedure as the viscosity measurements. The potential influence of tritium on this calibration is discussed in the following section.

III. CALIBRATION RESULTS OF MEASUREMENTS BEFORE AND AFTER CONTACT WITH TRITIUM

Viscosity measurements with an SRG in our setup are relative, since the calibration factor C_0 can only be determined analytically when the axis of rotation is aligned parallel to the cylinder axis. In all commercially available SRGs, however, the axis of rotation is oriented perpendicular to the cylinder axis, so no analytical solution for C_0 exists. The system is therefore calibrated using helium, the viscosity of which is known with high precision [9]. From this measurement, the calibration factor as a function of temperature is derived. Whenever the system geometry or surface properties are modified, the calibration must be repeated. In the case of tritium, radiochemical reactions may occur that generate tritiated hydrocarbons from carbon impurities and water from oxygen at the stainless-steel rotor surface [10,11]. Such processes alter the rotor surface, raising the question of whether the viscosity measurement is sensitive to these subtle modifications. To investigate this effect, the system was calibrated with helium both before exposure to tritium and again after the burn-in phase and subsequent viscosity measurements with tritium. The results are shown in Fig. 1: blue dots represent the initial calibration, while orange crosses indicate the calibration after the T_2 measurements. The discrepancy between the two calibrations reaches up to 15%.

IV. REFINED FIT OF THE TEMPERATURE DEPENDENCE OF THE VISCOSITY OF TRITIUM

Before the final viscosity measurements of T_2, the system had been exposed to tritium with a purity of 95% at a pressure of 2500 Pa for about four weeks. This was followed by an additional four weeks of viscosity measurements using the same T_2 gas, as well as a second batch of

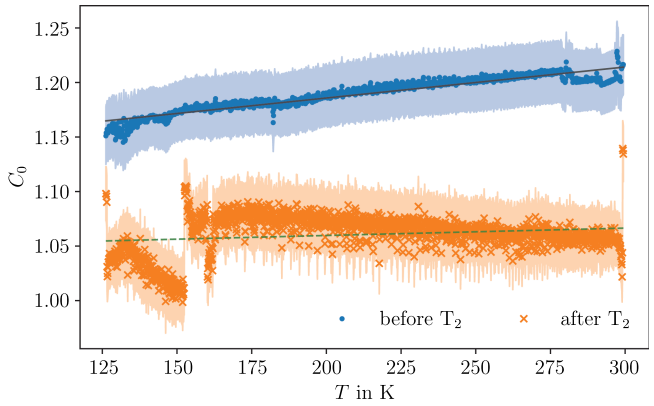

Fig. 1. Calibration factor dependent on the temperature calculated from the measurement before the system had contact with tritium is shown in blue, and after the burn-in phase in orange. The discrepancy between both measurements ranges up to 11%, while the uncertainty of the measurements is on the order of 4%.

tritium with 99% purity. For the evaluation, only the last two weeks of data with high-purity tritium at pressure setpoints between 300 Pa and 10 Pa were considered. Since the calibration performed directly after these tritium measurements deviated significantly from the initial calibration, the T_2 data had to be reevaluated using the new calibration curve. The results are shown as orange dots in Fig. 2. The green dotted line represents the viscosity of tritium extrapolated from H_2 by mass scaling, while the red dashed line shows the same values scaled by an additional 5%, as applied in the extrapolation from H_2 to D_2. The blue points correspond to the earlier evaluation with the initial calibration, obtained before the system was exposed to T_2, as reported in Ref. [6]. The discrepancy between the two evaluations of the same data reaches up to 20%. Fig. 3 compares different fit models that reproduce the viscosity of tritium with an accuracy better than 0.5% across a wide temperature range. The models are as follows:

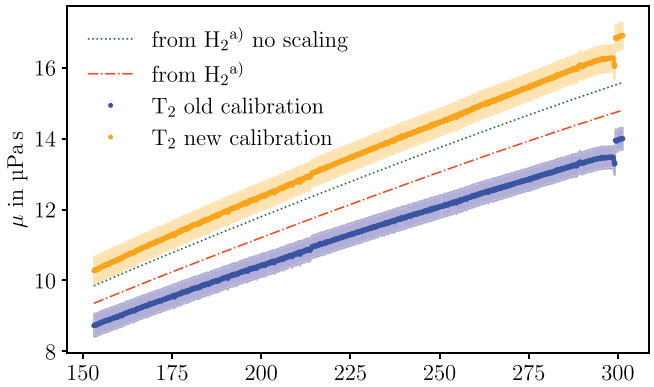

Fig. 2. The viscosity is shown dependent on the temperature.

1. The square-root model (sqrt model)

$$\mu(T) = a \cdot \sqrt{T} + b \ . \tag{3}$$

2. The power model

$$\mu(T) = a \cdot T^b \ . \tag{4}$$

3. The Sharipov model

$$\begin{aligned} \mu_{D_2}(T) &= \mu_{H_2} \cdot \sqrt{m_{D_2}/m_{H_2}} \cdot (1 - \exp\left(-a \cdot (T/b)^c\right) \\ \mu_{T_2}(T) &= d \cdot \sqrt{m_{T_2}/m_{D_2}} \cdot \mu_{D_2}(T) \ . \end{aligned} \tag{5}$$

The square-root model relies on two assumptions: first, that Eq. (1) remains approximately valid for gases containing small amounts of impurities and, second, that the viscosity does not vanish at 0 K. This can be understood from the definition of viscosity, where Eq. (1) represents only the first term of an infinite series. In principle, the parameter b would be expected to be small and positive. However, the fit yields a comparatively large negative value, which is physically meaningless. Moreover, as shown in Fig. 3, the square-root model does not reproduce the data: It underestimates the viscosity between 175 K and 250 K, while at higher temperatures it increasingly overestimates it compared to the other two models.

For the power model it is assumed that the viscosity may not follow a square-root dependence on temperature due to the radioactivity of tritium. This approach agrees well with the measurements above 250 K but fails at lower temperatures.

The Sharipov model, in contrast, is based on extrapolating the viscosity from H_2 through D_2 to T_2. The parameters a, b, and c are normally gained from calculations of the hydrogen molecule. However, for the fit they are not fixed to the H_2 values but treated as free fit parameters. This model reproduces the data across the entire range from 175 K to 300 K, and even below this interval it provides the best agreement, as the measurements show larger fluctuations there. The corresponding fit parameters for each model are summarized in Table 1.

V. DISCUSSION

The deviation between the two calculations of tritium viscosity is not surprising in view of the deviation observed between the C_0 before and after the system had been in contact with tritium. The key question is whether this calibration shift is solely caused by changes

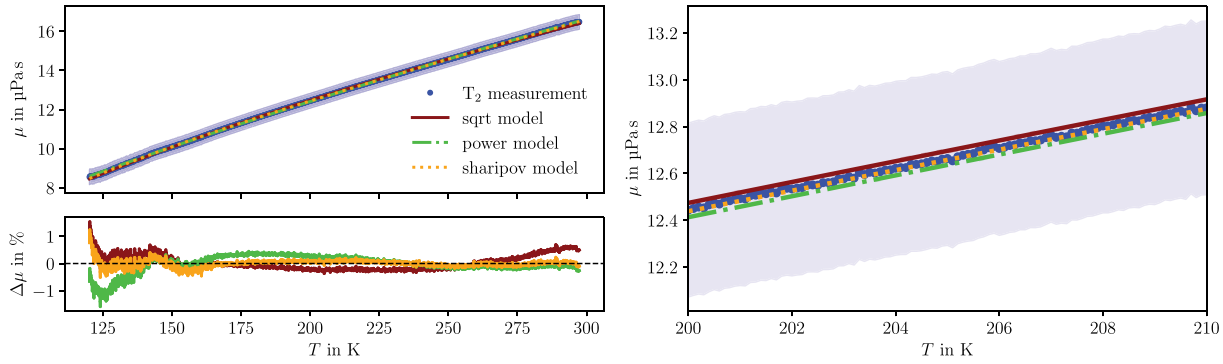

Fig. 3. (a) Results of the different fit models for the viscosity of T_2. (b) A zoom in at 200 K for better visualization of the best fit.

TABLE 1

Results for the Parameters of the Different Fit Models. For the Sharipov Model the Values for a and b are Correlated, Leading to Very High Uncertainties. Nevertheless, the Sharipov Model Reproduces the Measured Results Best.

Model	sqrt	power	Sharipov
a	1.2659(5)	0.2715(4)	3.3
b	−5.428(7)	0.7214(3)	120.9
c			0.973(7)
d			1.0701(1)

to the surfaces of the rotor and cylinder of the SRG. To address this, additional measurements with D_2 are currently being performed. From a measurement with the pressure-cycling mode with D_2 in Cryo-ViMA, it is clear that the new calibration does not influence the accuracy of the new D_2 measurement results. In this mode, the viscosity is measured only at room temperature, but the result of 12.8 μPa s fits within the uncertainty of the measurement to literature values by [12]. This means that the D_2 data analyzed using the post-tritium calibration agree with the initial D_2 data analyzed using the pre-tritium calibration, which provides strong evidence that exposure to tritium has indeed altered the surfaces of both rotor and cylinder. Further confirmation will come from analyzing the measured data with a focus on the surface-related parameters velocity slip coefficient (VSC) and tangential momentum accommodation coefficient (TMAC). These coefficients can be derived from the slope of Eq. (2). Significant surface modifications should manifest as changes in the VSC values and consequently in the TMAC values as well. If both lines of evidence are consistent, the new calibration can be considered reliable, and the viscosity of tritium is indeed substantially higher than previously reported in Ref. [6].

VI. CONCLUSION

In this work, reanalyzed viscosity values for tritium obtained with Cryo-ViMA were presented. The results demonstrate that the burn-in of tritium, likely resulting in a modification of the stainless-steel surfaces, significantly alters the calibration, causing a systematic deviation of the measured viscosity by approximately 20% compared to the values reported in Ref. [6]. Furthermore, the theoretically extrapolated values in Ref. [6] were corrected in the wrong direction to lower values because the burn-in effect was not taken into account. Ongoing experiments will include measurements with hydrogen, deuterium, tritium, and mixtures of all three hydrogen isotopologues. From these data, the viscosities as well as surface-related parameters—such as the velocity-slip coefficient and the tangential-momentum accommodation coefficient—will be determined, following the approach of Ref. [13]. Whether the magnitude of the burn-in–induced effects exceeds the intrinsic measurement uncertainty remains a central question. Answering this will clarify the reliability of viscosity data for tritium-containing systems and guide the development of more accurate predictive models.

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Author Contributions

CRediT: **Johanna Wydra:** Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Project

administration, Supervision, Validation, Visualization, Writing – original draft; **Robin Größle:** Conceptualization, Supervision, Writing – review & editing; **Alexander Marsteller:** Conceptualization, Formal analysis, Methodology, Software, Writing – review & editing; **Simon Niemes:** Supervision, Writing – review & editing; **Tim Poppe:** Investigation, Writing – review & editing; **Florian Priester:** Conceptualization, Supervision, Writing – review & editing; **Vincent Skrobocz:** Investigation; **Michael Sturm:** Conceptualization, Supervision, Writing – review & editing.

Disclosure Statement

No potential conflict of interest was reported by the author(s).

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