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In-situ measurement of tritium accumulation and decontamination of tungsten

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ABSTRACT

Tritium accumulation on surfaces, also known as the memory effect, has a wide range of implications for operating and maintaining fusion reactors, astroparticle physics experiments using tritium, and tritium analytics. At Tritium Laboratory Karlsruhe (TLK), the Tritium Activity Chamber Experiment (TRACE) was designed and built to investigate the tritium memory effect of a variety of materials. With TRACE, it is possible to expose samples to high-purity tritium at ambient temperature and measure their retained near-surface activity in-situ via beta-induced X-ray spectrometry (BIXS). Within this work, the relative memory effect of two tungsten samples — one pre-contaminated — after a cumulative exposure to tritium of 2100 mbar·h was determined. The time evolutions of their activity prove the repeatability of the experiment. Attempts were made to decontaminate one of the samples in-situ. During continuous evacuation, merely 20% of its near-surface activity could be desorbed at a bake-out temperature of 200 °C.

1. Introduction

When handling gaseous tritium, the wetted surfaces are inevitably contaminated due to adsorption, also known as the tritium memory effect. This tritium is unavailable for processing, which poses a challenge in a variety of situations, including the fuel cycle of fusion reactors and their necessary tritium accountancy. Adsorbed tritium also leads to systematic effects in the Karlsruhe Tritium Neutrino (KATRIN) experiment [1], which developed a suitable decontamination strategy [2]. Additionally, the tritium memory effect in analytical systems negatively impacts their performance in the form of a history-dependent background signal [3]. Hence, the knowledge of material-specific tritium accumulation on surfaces is crucial for various areas in fusion, e.g. fuel cycle modelling, fuel balance, and maintenance, as well as experimental astroparticle physics.

This work focuses on the Tritium Activity Chamber Experiment (TRACE), which enables tritium loading of samples, accompanied by in-situ near-surface activity measurements. Within the scope of this study, tritium adsorption on two tungsten samples is investigated and checked for repeatability. In addition, the decontamination efficiency of continuous evacuation at ambient and elevated temperatures is quantified.

2. The tritium activity chamber experiment (TRACE)

2.1. Experimental setup

The centrepiece of the experimental setup [4] (Fig. 1) contains the solid sample and a silicon drift detector (SDD). A beryllium window separates the SDD from the gas volume in order to prevent contamination of the detector. The activity is measured via beta-induced X-ray spectrometry (BIXS) by counting the number of X-rays generated by the β-electron originating from tritium decay [5–7]. The beryllium, stainless steel and tungsten surface areas exposed to tritium are comparable in size. However, the tungsten sample is the only object in the detector's direct field of view. This is ensured by the specifically designed sample holder, which creates a distance of roughly 3 mm between the tungsten surface and the beryllium window. A heating sleeve was fitted around the recipient to homogeneously bake out the entire sample cell. Fig. 2 displays how the sample cell, labelled "BIXS", is connected to the tritium gas supply and the pumping section. This setup allows exposing the sample to high-purity (> 95%) tritium in a pressure range of $(10^{-4} – 1000)$ mbar, as well as creating UHV conditions within the cell, while continuously monitoring the activity. After exposure measurements, the gas mixture is then transferred batch-wise to the CAPER facility [8] for exhaust purification.

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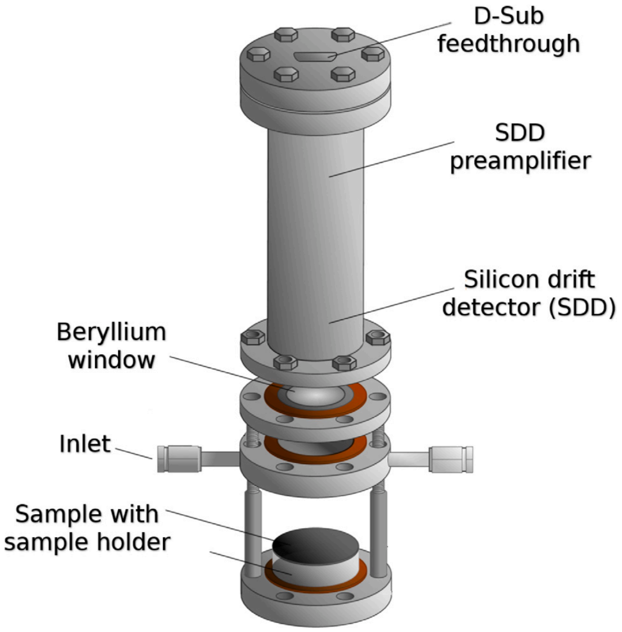

Fig. 1. Illustration of the BIXS measurement cell.

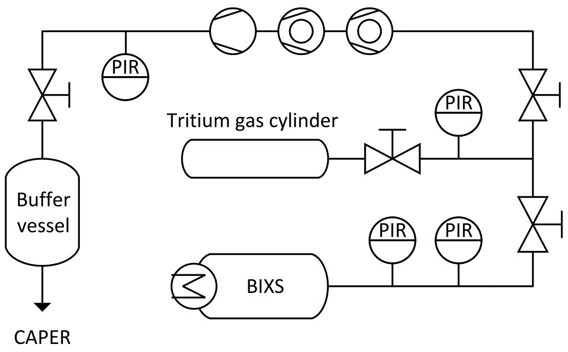

Fig. 2. Schematic of TRACE and its connection to the pumping section and TLK infrastructure.

2.2. BIXS activity calibration

The activity calibration of the BIXS system must be performed for each sample material due to differences in X-ray yield. Gaseous tritium with known isotopic composition, determined by e.g. Raman spectroscopy [9,10], is filled into the cell and the resulting count rate is recorded. By measuring pressure, temperature and volume, the total activity within the chamber can be calculated and compared to the count rate. One count per second corresponds to an activity of $(9.1 \pm 1.8)\cdot 10^5$ Bq and $(6.9 \pm 1.4)\cdot 10^5$ Bq for sample #1 and #2, respectively, with the volume being the dominant contribution to the uncertainties.

3. Tritium sorption behaviour of tungsten

3.1. Sample properties

The samples were cut from rolled tungsten sheets to circular shape with a diameter of 35 mm and a thickness of 1 mm. After the manufacturing processes, X-ray fluorescence analysis revealed an elemental purity of more than 99%.

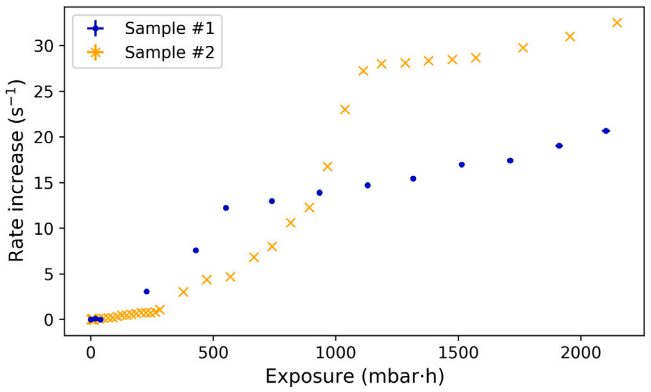

Fig. 3. Measured activity increase of two tungsten samples after repeated exposure to tritium. The 1 σ error bars are smaller than the data points.

3.2. Adsorption

Prior to the first tritium exposure of the samples, the background count rate of the system was measured for a prolonged time. For samples #1 and #2, the background rate was 17.2 and 33.4 s^{-1}, respectively. The background increase can be explained by the fact that other samples were measured in between. A measurement campaign consists of multiple cycles of tritium exposure at ambient temperature with a subsequent evacuation period, during which the retained near-surface activity by the sample is quantified via BIXS. A single cycle was divided into an exposure time of 20 h and an evacuation time of four hours. After acquiring the spectra during evacuation, the low-energy noise is removed and the average count rate is calculated. The count rate follows Poisson statistics. With an integration time of four hours, the relative uncertainties of the reported rates are below 0.2%. For the pressure sensors, an uncertainty of 0.12% of the reading was specified, which is propagated across the exposure cycles. Fig. 3 shows the rate increase of the two tungsten samples as a function of total exposure to tritium. For sample #1, the first two loadings were performed at a tritium pressure of 1 mbar, which showed no significant rate change. Afterwards, the loading pressure was increased to 10 mbar, which resulted in a steep rate increase of 12.2 s^{-1} until a total exposure of about 550 mbar h was reached. Beyond this point, the rate increased more slowly, with an additional 8.4 s^{-1} over the remaining 1550 mbar h.

To validate this change in adsorption behaviour, the procedure was repeated with more finely resolved exposures for a second tungsten sample. Similar to sample #1, the first seven exposures at a lowest pressure of in this case 0.1 mbar led to no significant rate increase. Consequently, the loading pressure was set to 1 mbar, which resulted in a slight linear rate increase of roughly 1.1 s^{-1} over an exposure of 280 mbar h. Further increasing the pressure to 5 mbar induced a transition of the adsorption regime resembling exponential growth to 27.2 s^{-1} at about 1100 mbar h, followed by a slower rate increase. As such, sample #2 confirms the previously observed transitions in tritium adsorption behaviour.

3.3. Desorption

Two decontamination procedures were applied to sample #1 to conclude its measurement campaign. Firstly, the system including the sample was baked out at 200 °C during evacuation to prevent the recontamination of nearby surfaces. While baking out, the detector needed to remain switched off to prevent damaging its Peltier element. To measure the activity with BIXS, the bake-out was interrupted multiple times as depicted in Fig. 4. Approximately five hours at elevated temperature led to an initial relative activity reduction of 12.5%.

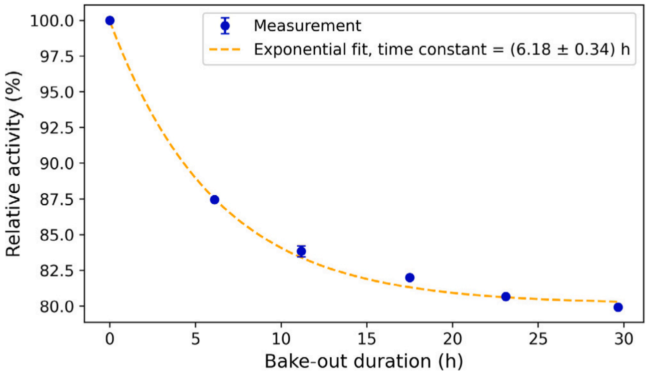

Fig. 4. Measured relative activity of the tungsten sample during bake-out at 200 °C. The 1 σ error bars are not visible for most of the data points.

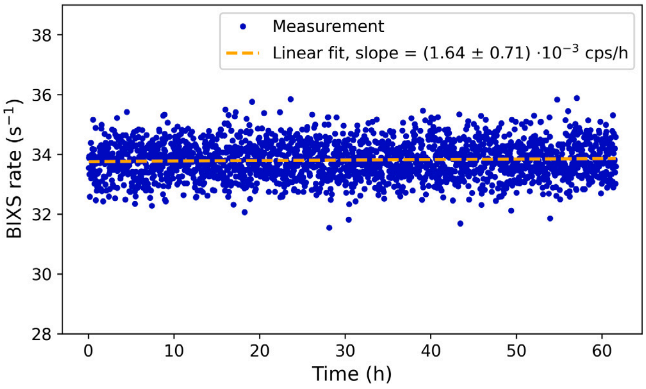

Fig. 5. Measured activity of the tungsten sample during continuous evacuation at ambient temperature after a bake-out of 5 h.

To verify the BIXS rate stability in absence of increased temperature, the second data point was measured for a prolonged time. Fig. 5 shows its time evolution during continuous evacuation at ambient temperature. Over the course of 60 h, no reduction of surface activity could be detected.

Hence, four more cycles of bake-out with activity measurement were applied and after 30 h overall, no further activity reduction was expected. In total, the activity decreased exponentially by 20% with a time constant of 6.2 h.

4. Discussion

Overall, the two tungsten samples show consistent behaviour regarding tritium accumulation. Furthermore, the tritium accumulation showed a kink-like transition towards reduced adsorption rates for both samples. This could be interpreted as the onset from monolayer to multilayer tritium adsorption. Sample #1 reaches this transition sooner, since it was already contaminated from a previous iteration of the experiment, while sample #2 was uncontaminated. Hence, the onsets cannot be compared directly.

To be able to compare tungsten regarding tritium uptake to other materials studied in [11], the relative tritium memory effect will be defined as the ratio of the count rate during evacuation after a total exposure of 2100 mbar h, and the rate during the preceding tritium loading. In the previous study [11] using a similar setup, stainless steel coated by various materials, e.g. gold and tungsten, was investigated. As a reference, the memory effect of uncoated type 316LN stainless was determined to be 1.28% and a tungsten coating approached 12%. Of all the materials studied, gold proved to have the lowest memory effect with 0.82%.

For the two tungsten samples used in this study, the initially measured background is included in both rates. This yields a value of (2.04 ± 0.02) % for sample #1 and (2.95 ± 0.03) % for sample #2. When subtracting the background rate prior to the calculation of the memory effect, this changes to (1.12 ± 0.03) % and (1.45 ± 0.03) % for sample #1 and #2, respectively. Only the statistical uncertainties are considered, however, neglecting the fact that sample #1 was already contaminated at the beginning of this study leads to an underestimation of its relative memory effect. In both cases, the elemental tungsten sample showed a significantly lower memory effect than the tungsten coating studied in [11].

In terms of total attained surface activities, (1.95 ± 0.39) MBq cm^{-2} and (2.31 ± 0.47) MBq cm^{-2} on the samples were determined.

It is currently not possible to entirely disentangle tritium accumulation on the sample from adsorption on nearby stainless steel and beryllium surfaces. In order to do so, a more sophisticated background model is required.

During 30 h of bake-out at 200 °C, the visible activity of the system including tungsten sample #1 could be reduced by roughly 20%, or (7.0 ± 1.4) MBq. It has to be emphasized that with BIXS, tritium desorption and temperature-induced migration deeper into the bulk cannot be distinguished, since it is only sensitive to near-surface activity. One limited possibility to distinguish diffusion into the bulk from desorption is to bake out the sample in a vacuum oven and trap the released tritium by converting it to water via oxidation. Liquid scintillation counting of the generated tritiated water could offer insights on the sample's bulk activity. Another possibility would be to analyse the sample's elemental depth profile using time-of-flight secondary-ion mass spectrometry (TOF-SIMS). Contaminating two samples but baking out only one prior to the analysis and comparing the results could reveal information on the respective surface and bulk activities. Both methods can only be applied ex-situ and require removing the sample from the recipient, which potentially influences the state of the sample.

During bake-out, the activity might have been released in the form of Q_2O (Q = H, D, T) or tritiated volatile hydrocarbons, in view of the need for elevated temperatures. With higher temperatures, the activity might have been reduced even further. Within TLK's safety framework, temperatures above 200 °C require additional safety measures [12] due to increasing tritium permeation. Hence, a decontamination method that is also effective at ambient temperatures is advantageous. One of these methods is UV/ozone cleaning [2,13–15], which appears to be a promising alternative for in-situ decontamination that needs to be investigated further.

5. Conclusion and outlook

The TRACE setup allows for in-situ measurements of tritium adsorption on and desorption from solid samples. Within this study, the tritium memory effect of two tungsten samples was investigated by repeated expositions to high-purity tritium and continuous activity monitoring via BIXS. Both samples showed consistent behaviour of accumulation and after an exposure of 2100 mbar h, their near-surface activities amounted to (1.95 ± 0.39) MBq cm^{-2} and (2.31 ± 0.47) MBq cm^{-2}. A reduction of near-surface activity during continuous evacuation was only observed at elevated temperatures.

A redesign of the experimental setup is in progress. This includes an extension of the temperature range during tritium loading to > 1000 K to more realistically simulate conditions within the fusion fuel cycle, as well as an integrated UV source to investigate the efficiency of UV/ozone decontamination. Furthermore, simulations of spatial activity distributions are being carried out to facilitate the decoupling of sample activity from the activity of the surrounding vessel surfaces.

CRediT authorship contribution statement

Dominic Batzler: Writing – original draft, Visualization, Supervision, Investigation, Formal analysis, Conceptualization, Writing – review & editing. **Max Aker:** Writing – review & editing, Conceptualization. **James Robert Braun:** Software, Investigation, Formal analysis, Conceptualization. **Robin Größle:** Writing – review & editing, Supervision, Conceptualization. **Philipp Haag:** Software, Investigation, Formal analysis, Conceptualization. **Marco Röllig:** Writing – review & editing, Supervision, Resources, Conceptualization. **Dylan Ray Roodt:** Software, Investigation, Formal analysis, Conceptualization. **Marie-Christine Schäfer:** Investigation, Conceptualization. **Marius Schaufelberger:** Visualization, Investigation, Formal analysis, 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.

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Data availability

Data will be made available on request.

References

[1] M. Aker, D. Batzler, A. Beglarian, J. Behrens, J. Beisenkötter, M. Biassoni, B. Bieringer, Y. Biondi, F. Block, S. Bobien, M. Böttcher, B. Bornschein, L. Bornschein, T.S. Caldwell, M. Carminati, A. Chatrabhuti, S. Chilingaryan, B.A. Daniel, K. Debowski, M. Descher, D.D. Barrero, P.J. Doe, O. Dragoun, G. Drexlin, F. Edzards, K. Eitel, E. Ellinger, R. Engel, S. Enomoto, A. Felden, C. Fengler, C. Fiorini, J.A. Formaggio, C. Forstner, F.M. Fränkle, K. Gauda, A.S. Gavin, W. Gil, F. Glück, S. Grohmann, R. Grössle, R. Gumbsheimer, N. Gutknecht, V. Hannen, L. Hasselmann, N. Haußmann, K. Helbing, H. Henke, S. Heyns, S. Hickford, R. Hiller, D. Hillesheimer, D. Hinz, T. Höhn, A. Huber, A. Jansen, C. Karl, J. Kellerer, K. Khosonthongkee, M. Kleifges, M. Klein, J. Kohpeiß, C. Köhler, L. Köllenberger, A. Kopmann, N. Kovač, A. Kovalík, H. Krause, L.L. Cascio, T. Lasserre, J. Lauer, T. Le, O. Lebeda, B. Lehnert, G. Li, A. Lokhov, M. Machatschek, M. Mark, A. Marsteller, E.L. Martin, C. Melzer, S. Mertens, S. Mohanty, J. Mostafa, K. Müller, A. Nava, H. Neumann, S.N. andA. Onillon, D.S. Parno, M. Pavan, U. Pinsook, A.W.P. Poon, J.M.L. Poyato, S. Pozzi, F. Priester, J. Ráliš, S. Ramachandran, R.G.H. Robertson, C. Rodenbeck, M. Röllig, C. Röttele, M. Ryšavý, R. Sack, A. Saenz, R. Salomon, P. Schäfer, M. Schlösser, K. Schlösser, L. Schlüter, S. Schneidewind, U. Schnurr, M. Schrank, J. Schürmann, A. Schütz, A. Schwemmer, A. Schwenck, M. Šefčík, D. Siegmann, F. Simon, F. Spanier, D. Spreng, W. Sreethawong, M. Steidl, J. Štorek, X. Stribl, M. Sturm, N. Suwonjandee, N.T. Jerome, H.H. Telle, L.A. Thorne, T. Thümmler, S. Tirolf, N. Titov, I. Tkachev, K. Urban, K. Valerius, D. Vénos, C. Weinheimer, S. Welte, J. Wendel, C. Wiesinger, J.F. Wilkerson, J. Wolf, S. Wüstling, J. Wydra, W. Xu, S. Zadorozhny, G. Zeller, Direct neutrino-mass measurement based on 259 days of KATRIN data, 2024, http://dx.doi.org/10.48550/arXiv.2406.13516, arXiv:2406.13516.

[2] M. Aker, M. Sturm, F. Priester, S. Tirolf, D. Batzler, R. Größle, A. Marsteller, M. Röllig, M. Schlösser, In situ tritium decontamination of the KATRIN rear wall using an ultraviolet/ozone treatment, Fusion Sci. Technol. 80 (3–4) (2024) 303–310, http://dx.doi.org/10.1080/15361055.2023.2214695.

[3] M. Nishikawa, T. Takeishi, Y. Matsumoto, I. Kumabe, Ionization chamber system to eliminate the memory effect of tritium, Nucl. Instrum. Methods Phys. Res. A 278 (2) (1989) 525–531, http://dx.doi.org/10.1016/0168-9002(89)90875-9.

[4] M. Röllig, S. Ebenhöch, S. Niemes, F. Priester, M. Sturm, Development of a compact tritium activity monitor and first tritium measurements, Fusion Eng. Des. 100 (2015) 177–180, http://dx.doi.org/10.1016/j.fusengdes.2015.05.056.

[5] T. Westermark, L. Devell, N. Ghanem, On the use of bremsstrahlung for the determination of tritium in aqueous and organic systems, Nucl. Instrum. Methods 9 (2) (1960) 141–144, http://dx.doi.org/10.1016/0029-554X(60)90093-8.

[6] M. Matsuyama, K. Watanabe, K. Hasegawa, Tritium assay in materials by the bremsstrahlung counting method, Fusion Eng. Des. 39–40 (1998) 929–936, http://dx.doi.org/10.1016/S0920-3796(98)00232-4.

[7] M. Röllig, Tritium Analytics by Beta Induced X-ray Spectrometry (Ph.D. thesis), Karlsruhe, Karlsruher Institut für Technologie (KIT), 2015, http://dx.doi.org/10.5445/IR/1000054050.

[8] D. Demange, E. Fanghänel, S. Fischer, T.L. Le, F. Priester, M. Röllig, M. Schlösser, K.H. Simon, CAPER as central and crucial facility to support R&D with tritium at TLK, Fusion Sci. Technol. 67 (2) (2015) 308–311, http://dx.doi.org/10.13182/FST14-T17.

[9] S. Niemes, H.H. Telle, B. Bornschein, L. Fasselt, R. Größle, F. Priester, M. Schlösser, M. Sturm, S. Welte, G. Zeller, Accurate reference gas mixtures containing tritiated molecules: Their production and Raman-based analysis, Sensors 21 (18) (2021) http://dx.doi.org/10.3390/s21186170.

[10] F. Priester, A. Marsteller, S. Niemes, N. Tuchscherer, S. Welte, μRA—A new compact easy-to-use Raman system for all hydrogen isotopologues, Sensors 22 (10) (2022) http://dx.doi.org/10.3390/s22103952.

[11] M. Aker, M. Röllig, Material studies to reduce the tritium memory effect in BIXS analytic systems, Fusion Sci. Technol. 76 (3) (2020) 373–378, http://dx.doi.org/10.1080/15361055.2020.1712989.

[12] S. Welte, U. Besserer, D. Osenberg, J. Wendel, Tritium laboratory karlsruhe: Administrative and technical framework for isotope laboratory operation, Fusion Sci. Technol. 67 (3) (2015) 635–638, http://dx.doi.org/10.13182/FST14-T98.

[13] J.R. Vig, Ultraviolet-ozone cleaning of semiconductor surfaces, 1992, Research and Development Technical Report SLCET-TR-91-33.

[14] J.P. Krasznai, R. Mowat, UV/Ozone treatment to decontaminate tritium contaminated surfaces, Fusion Technol. 28 (3P2) (1995) 1336–1341, http://dx.doi.org/10.13182/FST95-A30597.

[15] D. Batzler, M. Aker, R. Größle, D. Kurz, A. Marsteller, F. Priester, M. Sturm, P. Winney, Monitoring of ozone production and depletion rates in a tritium-compatible system, Fusion Eng. Des. 203 (2024) 114425, http://dx.doi.org/10.1016/j.fusengdes.2024.114425.