Fusion Science and Technology

ISSN: 1536-1055 (Print) 1943-7641 (Online) Journal homepage: http://www.tandfonline.com/loi/ufst20

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To cite this article: Simon Niemes, Michael Sturm, Robert Michling & Beate Bornschein (2015) High Level Tritiated Water Monitoring by Bremsstrahlung Counting Using a Silicon Drift Detector, Fusion Science and Technology, 67:3, 507-510, DOI: 10.13182/FST14-T66

To link to this article: https://doi.org/10.13182/FST14-T66

Published online: 23 Mar 2017.

HIGH LEVEL TRITIATED WATER MONITORING BY BREMSSTRAHLUNG COUNTING USING A SILICON DRIFT DETECTOR

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The β-ray induced X-ray spectrometry (BIXS) is a promising technique to monitor the tritium concentration in a fuel cycle of a fusion reactor. For in-situ measurements of high level tritiated water by bremsstrahlung counting, the characteristics of a low-noise silicon drift detector (SDD) have been examined at the Tritium Laboratory Karlsruhe (TLK). In static measurements with constant sample volume and tritium concentration, the bremsstrahlung spectra of tritiated water samples in a concentration range of 0.02 to 15 MBq/ml have been observed. The volume has been kept constant at 5 cm^3. The observed spectra are well above the noise threshold. In addition to X-rays induced by β-rays, the spectra feature X-ray fluorescence peaks of the surrounding materials. No indications of memory effects have been observed. A linear relation between the X-ray intensity and the tritium concentration was obtained and the lower detection limit of the setup has been determined to 1 MBq ml^{-1}, assessed by the Currie criterion. In addition, the spectra obtained experimentally could be reproduced with high agreement by Monte-Carlo simulations using the Geant4-toolkit. It was found that the present detection system is applicable to non-invasive measurements of high-level tritiated water and the SDD is a convenient tool to detect the low energy bremsstrahlung X-rays.

I. INTRODUCTION

A fundamental task in tritium processing in the ITER tritium plant is, among others, the monitoring of high level tritiated water (HTO) regarding process control and accountancy.

The established method of Liquid Scintillation Counting (LSC) is suitable to measure any tritium concentration in liquids, but is inapplicable for an in-situ measurement of the expected concentration levels of around 20 MBq/ml in the tritium plant[1]. This is due to the necessary dilution of the samples and the offline nature of the measurement process. Furthermore, tritiated water is removed from the intended closed fuel cycle and each measurement produces radioactive organic waste containing tritium.

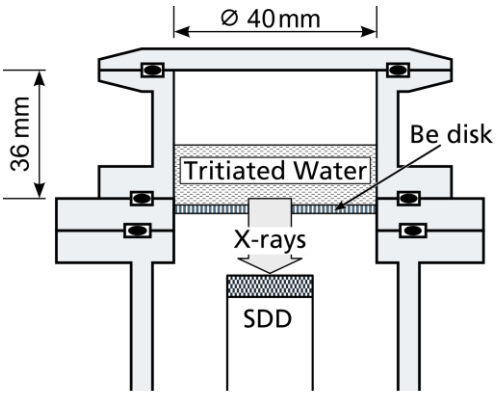


Fig. 1. Cross-section view of the experimental setup and BIXS principle.

A promising in-situ measuring technique is β-induced X-ray spectrometry (BIXS). This technique is based on the observation of X-ray bremsstrahlung resulting from the interaction of the β-rays and the nuclei of all atoms in tritiated water, mainly oxygen due to the higher Z number. By efficiently detecting those X-rays and calibrating the observed spectrum to known concentrations, the tritium concentration in water can be measured (Fig. 1).

Such a detection device could be used in the Water Detritiation System (WDS), a scaled prototype facility as part of the ITER fuel cycle development at the Tritium Laboratory Karlsruhe (TLK).

To detect the low energy and low intensity spectrum, different detector types and X-ray window combinations have been examined. Nishizawa et al.[2] performed measurements using a NaI(Tl) scintillation counter with a beryllium window and custom electronics, achieving a lower detection limit of 0.037 MBq ml^{-1}. Using a similar setup but with commercial electronics, shielding and a vial for loading the HTO sample, Matsuyama et al.[3] achieved a sensitivity of 1.1×10^{-1} counts s^{-1} Bq^{-1}, three times higher than the reported sensitivity by Nishizawa. Matsuyama et al. also used a Germanium-detector to

examine the shape of the bremsstrahlung spectrum and developed a computational simulation to investigate the influence of the sample volume.

To evaluate additional detector choices and further improve HTO monitoring, the TLK has set up different experimental setups. A key component of this research is the possible integration into the WDS.

The presented experiment has been set up to investigate the performance of a silicon drift detector (SDD) to detect the bremsstrahlung spectrum. The setup is able to perform measurements on HTO in a metallic sample chamber. The SDD enables the user to discriminate better between signal and background compared to scintillation counters and simplifies the whole setup (e.g. no high voltage). The excellent usability of a SDD detecting bremsstrahlung originating from a gaseous tritium source has already been reported.[4] In a similar approach, the usability of a CaF_2 scintillation counter has also been examined.

In this paper, a review of the applicability of a SDD for BIXS measurements is presented. The performance of the SDD is compared to the aforementioned CaF2 scintillation counter. In addition, the ability of the SDD to observe the shape of the bremsstrahlung spectrum as well as the achievable sensitivity are evaluated. Furthermore, a Monte-Carlo-simulation (MC) using the Geant4-toolkit[5] is presented to further analyze the observed spectra.

II. EXPERIMENTAL SETUP

The experimental setup for BIXS measurements consists of a stainless steel sample chamber, X-ray window, SDD chip in a vacuum vessel and detector electronics, as shown in figures 1 and 2.

The detector itself is a conventional SDD of 109 mm^2 collimated to 80 mm^2 sensor surface with a zirconium collimator (KETEK Vitus H80 Premium with AXAS-M housing and signal processing).

The detector chip is cooled by a peltier element and has an integrated JFET preamp to improve the signal-to-noise-ratio. The sensor chip is used without the standard protective cap since an external X-ray window is used. To avoid condensation damage on the cold sensor surface, the chip is operated in a vacuum of 5 Pa.

The external X-ray window separating sample chamber and vacuum vessel consists of a beryllium disk of 0.25 mm thickness and a diameter of 40 mm with a 200 nm protective gold coating. The disk is brazed in a stainless steel flange.

The sample chamber is constructed from a 316L stainless steel tube with an internal volume of approx. 15 cm^3 that is connected to the X-ray window flange. The volume can be sealed off by a blind flange to prevent evaporation into the fume.

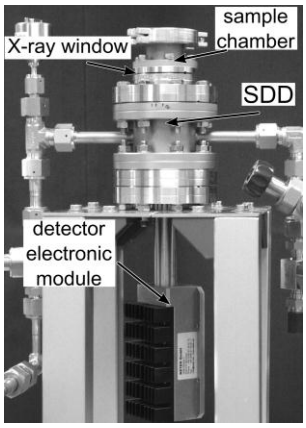


Fig.2. Overview of the experimental setup.

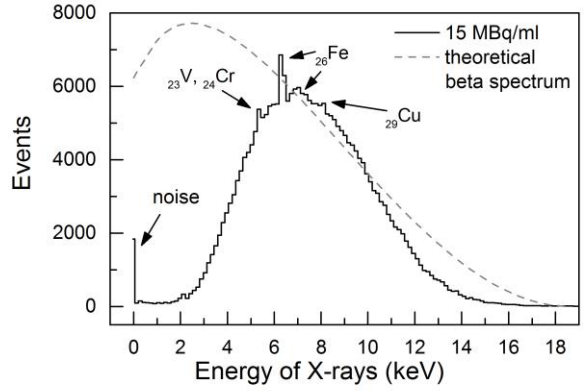
The detector electronic module was controlled by a computer and X-ray intensity data and measuring time were automatically recorded for every measurement.

The detector setup has been calibrated using an Americium-241 calibration source from Amersham-Buchler. The source includes metallic fluorescence foils to produce characteristic X-rays. To cover the range from 5 to 22 keV, the elements copper, rubidium and molybdenum have been used. In addition to the calibration source, characteristic fluorescence lines excited in the surrounding setup material have also been used for the energy calibration. The measured energy resolution is about 140 eV FWHM at 5.9 keV. This equals to a channel width of about 2.55 eV. In 14 000 measurements with constant detector parameters but various measurement times up to 10 000 s, no detectable drift in neither energy calibration nor count rate efficiency has been observed.

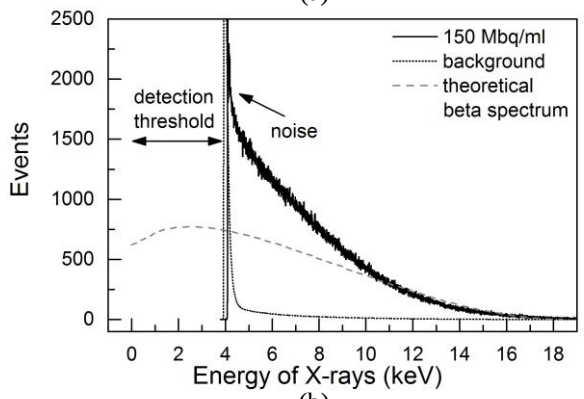
III. EXPERIMENTAL PROCEDURES

For experiments with tritiated water, the sample chamber was filled with 5 ml of deionized water for background measurements. In the first step, 0.5 ml of the water sample were removed and replaced by the same volume of tritiated water with a tritium concentration of 20 MBq/ml. The concentration of the sample was determined by 3 separate samples of the remaining sample volume of 3 µl each using LSC (Perkin Elmer TriCarb 2800 TR). In each following measurement, a certain volume of the tritiated water sample was removed and replaced by the same volume. The volume removed for LSC measurements was also replenished. This way the sample volume was kept constant at all times and the tritium concentration varied in the range from 0.02 to 15 MBq/ml.

To investigate a possible memory effect, the concentration in the sample chamber was increased and decreased several times using deionized water and highly tritiated water as diluent.



(a)



(b)

Fig. 3. Comparison of the observed bremsstrahlung spectrum measured with (a) SDD and (b) CaF_2 scintillation counter. Also shown is the calculated beta emission spectrum of tritium

IV. RESULTS & DISCUSSION

The requirements for a suitable detector are the ability to observe the shape of the bremsstrahlung spectrum without significant interference of noise and sufficient energy resolution. Figure 3 shows a typical spectrum measured with the SDD and the CaF_2 detector.

The bremsstrahlung spectrum obtained by the SDD is well above noise threshold and superimposed by characteristic X-ray fluorescence from the surrounding materials. These peaks mainly originate from stainless steel constituents and can be used as an online method for energy calibration. To make spectral features more prominent, a channel binning with a size of 50 channels has been used. With suitable signal processing settings, background was determined to (0.026 ± 0.0013) cps.

Compared to the spectrum obtained with the previous CaF_2 setup, the SDD clearly demonstrates both higher resolution and signal-to-noise-ratio. Using the CaF_2 detector, the low energy portion of the spectrum is cut off because of the high detection threshold. Due to the low energy resolution, no prominent features can be identified.

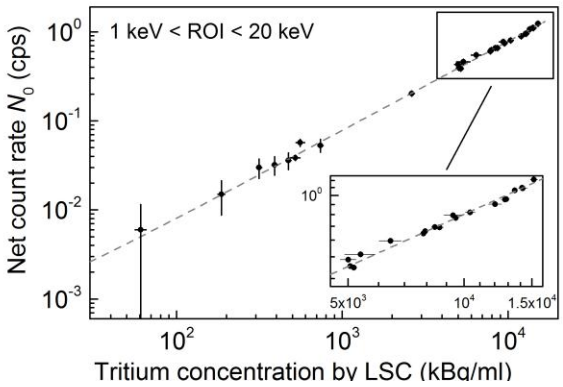


Fig. 4. Linear calibration curve for tritium concentration, as measured by LSC.

In figure 3, the calculated beta emission spectrum of tritium is also shown with arbitrary intensity for comparison. The mean energy of the bremsstrahlung spectrum is shifted to a higher energy relative to the beta emission spectrum. The lower energy part of the bremsstrahlung is suppressed due to the strong energy dependence of both the bremsstrahlung interaction cross-section and X-ray.

To examine the relationship between tritium concentration and detection count rate, each spectrum was integrated from 1 to 20 keV. As seen in figure 4, a linear relationship over a wide concentration range has been observed. The linear regression was performed utilizing York's algorithm[6] to include uncertainties in both concentration and count rate. The correlation between x- and y-uncertainties has been set to zero, because the values derived from different measurements. The relation is described by the following equation:

$$N(C_{HTO}) = (77.73\pm0.52) \times C_{HTO} + (23.9\pm1.9)\times10^{-3} \text{ cps},$$

where C_{HTO} represents the concentration of tritium in kBq/ml.

During all measurements, no indications of a memory effect have been observed. Although the background count rate increased by a factor of 2 after completely emptying the sample chamber, the count rate reverted to the initial rate after approximately 1 hour. This is due to evaporation of water residues inside the sample chamber and it did not influence concentration measurements.

To determine the minimum detectable quantity (MDQ), the Currie equation[7] for paired observations has been used. The criterion defines the MDQ by limiting the possibility of both a false-positive and false-negative result to 5 %. With a confidence level of 99 % (k=3), a sensitivity of 1 MBq/ml in 1000 s can be achieved. The mean detection efficiency of the setup was evaluated as 2.2×10^{-8} counts s^{-1} Bq^{-1}.

For practical use in the WDS system, the sensitivity of the system has to be further improved, although a

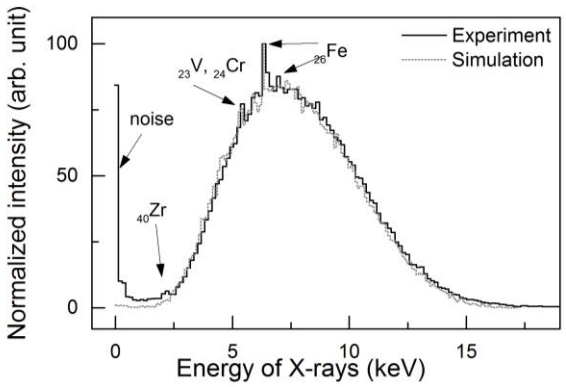


Fig. 5. Comparison of the measured and simulated spectra.

similar setup can be used to monitor storage tanks with highly tritiated water.

Although the sensitivity is lower than the values reported by Matsuyama et al.[4], the effective surface of the SDD sensor and therefore the covered solid angle is about 40 times smaller compared to the scintillation probe. Furthermore, the advantageous energy resolution and noise characteristic of the SDD allow for a better understanding of the shape of the bremsstrahlung spectrum. Considered ways to improve the sensitivity include lead shielding, a detector chip with larger surface area and a multi-detector type setup similar to the one mentioned in Ref. 8.

To investigate the bremsstrahlung spectrum and the influence of different parts of the setup, a MC-simulation using the Geant4-toolkit[6] and the Livermore physic models has been developed. The Livermore package gives a tested, standardized list of interaction models, suitable for the observed low keV energy range. In figure 5, the experimental data is compared to the simulated spectra. Both spectra have been normalized with reference to the K_α-line of $_{26}$Fe. The simulated bremsstrahlung spectrum agrees well with the experimental spectrum. Both spectra feature the same characteristic X-ray lines and relative peak intensities. Because of the simplified detector structure within the simulation, some features like the $_{40}$Zr peak due to the collimator of the SDD chip are missing.

In consequence of the low bremsstrahlung interaction cross-section, variance reduction techniques, like increased interaction probability, have been used to reduce computing time.

Based on the good agreement, the simulation can be used to further study and optimize the geometry of the experimental setup.

V. CONCLUSIONS

To establish the silicon drift detector (SDD) as a suitable detector for tritium activity measurement in highly tritiated water using BIXS, an experimental test setup has been built at the TLK.

To evaluate the SDD performance, the bremsstrahlung spectra of tritiated water in a concentration range from 0.02 to 15 MBq/ml have been obtained. The observed spectra are well above the noise threshold and include characteristic X-ray fluorescence of the materials used in the setup.

The used SDD clearly outperforms the CaF_2-detector from the previous setup. The detection limit of the new setup has been determined to 1 MBq/ml within 1 000 s measurement time using the Currie criteria.

A Monte-Carlo simulation using Geant4[6] and a reference physics list has been developed to validate the obtained spectra. The simulated data agrees well with the experimental data.

Based on the performance of the SDD, an optimized experimental setup including dynamic measurements of tritium concentration in flowing water is proposed. This setup needs to investigate performance improvements in detection limit and acquisition time, both necessary for a possible integration into the WDS.

ACKNOWLEDGMENTS

The authors would like to thank all their colleagues from TLK for the support during this work.

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