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Measurement system for in-pile tritium monitoring from Li_2TiO_3 ceramics at WWRK reactor

Y. Chikhray [a,*], V. Shestakov [a], T. Kulsartov [a], I. Tazhibayeva [b], H. Kawamura [c], A. Kuykabaeva [a]

[a] Kazakhstan State University, Tole-Bi-Street 96a, Almaty, Kazakhstan
[b] National Nuclear Center, Lenin-Street 6, Kurchatov, Kazakhstan
[c] Japan Atomic Energy Research Institute, Tokyo, Japan

Abstract

A 220-day irradiation of Li_2TiO_3 ceramics with 96% enrichment of isotope ^6Li was carried out at WWRK reactor. One of the study goals was to examine tritium release behavior during Li burn-up. To achieve this goal three types of ceramics samples were examined simultaneously using a system for in-pile tritium monitoring: one (pebbles) – under constant temperature of 650 °C, and two (pebbles and pellets) – within temperature change ranges from 500 to 900 °C. Flows of tritium release from ceramics during the various reactor campaigns, as well as tritium generation rates for each ampoule are presented in the paper. The main result is justification of burn-up in ^6Li reaching up to 20% of Li_2TiO_3 ceramics.

1. Introduction

Design of a tritium measurement system (named 'Sakura') was part of the three-year project K-578 of the International Scientific Technical Center (ISTC) directed to investigations of Li_2TiO_3 ceramics as a candidate material for DEMO reactor's blanket. Li_2TiO_3 ceramics enriched with the ^6Li isotope are considered as an appropriate material for such a blanket taking into account both the radiation hardness and ability of tritium generation [1–3]. The mission of the K-578 project was long-term neutron irradiation of lithium ceramics in the Water-Water Reactor-Kazakhstan (WWRK) until ~20% burn-up of ^6Li isotope occurred, with 'in situ' measurements of tritium generated in ceramics at various temperatures. The total quantity of the lithium ceramics under study was 16.2 g. The procedure of Li_2TiO_3 pebbles fabrication includes a fabrication process of gel-spheres, and subsequent dropping, drying, calcinations and sintering processes; main steps of Li_2TiO_3 pellet fabrication include mixing of the Li_2TiO_3 and TiO_2 powders, cold pressing and sintering [4]. In accordance with the adopted experimental concept the ceramics were heated by radiative heating of the ceramic itself (ceramic was placed in vacuum capsule) and of the capsule material (stainless-steel). Ceramic temperature was controlled within the 500–900 °C range by changing the heat-exchange between capsule

* Corresponding author.
E-mail address: istcvova@kazmail.asdc.kz (Y. Chikhray).

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and outer case (ampoule) by letting in high purity helium. Tritium generation rate was monitored from three capsules containing the 1 mm pebbles of Li_2TiO_3 at different temperatures: In capsules A1 and A2 the temperature was changed from 500 to 900 °C every 48 h with 100 °C step; and in A3 capsule the temperature was kept at 650 °C during the entire irradiation campaign.

2. Experiment outline

The tritium measurement system 'Sakura' was installed in a temporarily unused box of vacuum pumps (so called 'TMN box'), located nearby of reactor's top cover. The length of inlet path from capsules to the 'Sakura' was ~9 m. Fig. 1 shows the vertical section of WWRK reactor with location of the TMN box. The neutron energy and flux density were as follows: for neutron energy less than 0.4 eV the neutron flux density was about $(5.7 \pm 0.4) \times 10^8$ cm^{-2} c^{-1}; for the neutrons with energy more than 1.15 eV the neutron flux density was about $(5.5 \pm 0.8) \times 10^8$ cm^{-2} c^{-1}; and for energies more than 2.35 eV – $(2.9 \pm 0.4) \times 10^8$ cm^{-2} c^{-1}. Three experimental ampoules and the three-channel tritium measurement system have been merged with stationary WWRK's multi-purpose loop facility (MLF). Detailed description of reactor experiment techniques is given in [5].

Tritium and HT concentrations should be measured in a mixture of gases inside the capsule with ceramics (biased with a significant amount of helium); therefore, 'hydrogen windows' of a palladium–silver alloy were used to prevent helium penetration into the analytical systems. It is well known that at operating temperatures about 450 °C such a filter passes only hydrogen and its isotopes.

Before empirical measurements of tritium yield from lithium ceramics, the system was calibrated in a test experiment under laboratory conditions. Calibration was intended to determine the constant of proportionality between hydrogen pressures at filter inlet side with the indications of omegatron-type mass-analyzer (RMO-13 tube). Filter temperature was 400 °C.

The inlet side of hydrogen filter was exposed to hydrogen and the process of its penetration into the analytical system was registered with the mass-analyzer (see Fig. 2). The area under the registered curve of hydrogen flux decay gives the total amount of hydrogen Q (moles) passed through the filter at moment t_0 (seconds)

$$Q = k \int_0^{t_0} U(t)\mathrm{d}t, \tag{1}$$

where $U(t)$ is the indication of mass-analyzer (mV), k – required constant of proportionality (mol/(s mV)).

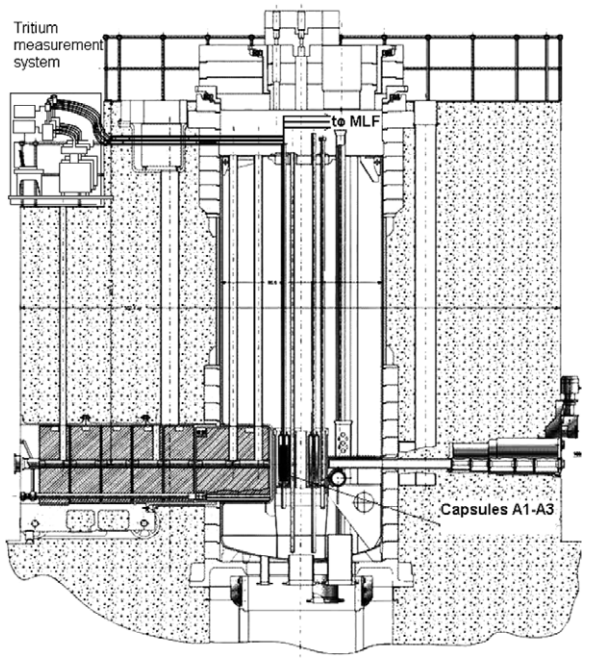

Fig. 1. Location of capsules with ceramics and the tritium measurement system at WWRK reactor.

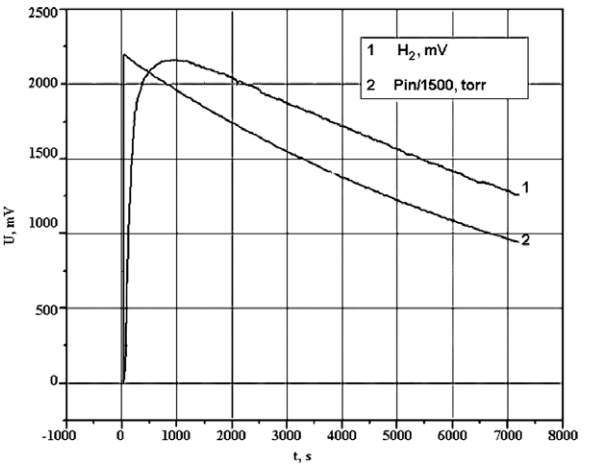

Fig. 2. Changes of inlet hydrogen pressure and process of hydrogen permeation via the filter.

On the other hand the amount of passed hydrogen is determined via the hydrogen pressure drop on the inlet side of filter at the same time

$$Q = \frac{V \cdot (P_0 - P_1)}{R \cdot T}, \quad (2)$$

where $V = 2.6 \times 10^{-5}\,\mathrm{m}^3$ is the inlet volume, $P_0 - P_1 = 113\,\mathrm{Pa}$ is the pressure drop, $R = 8.31$ J/(mol K), $T = 293$ K is the temperature of hydrogen in the inlet volume.

Therefore, the constant of proportionality between the hydrogen pressure on the filter inlet side with the indications of mass-analyzer was derived from (1) and (2) and is $k = (8 \pm 0.8) \times 10^{-12}$ mol/(s mV). Such curves were registered for the entire range of possible inlet pressures of hydrogen and gave perfect coincidence in all cases.

Similar experiments were carried out to determine the $U(P)$ transfer function for the tritium measurement system with Pd–Ag filter. The goal was to determine the link coefficient between the hydrogen pressure P on the inlet side with hydrogen flow into the mass-analyzer U. Measurements were carried out in the pressure range 10^{-2}–300 Pa. The coefficient between hydrogen flow into the measurement system and its inlet pressure was derived equal $K = 1.0 \times 10^{-10}$ mol/Pa.

The response limit of the tritium measurement system (minimal pressure at filter inlet which could be registered) was determined as 0.1 Pa and met the requirements for measurements.

3. Tritium measurement system

The tritium measurement system 'Sakura' (Table 1, Fig. 3) consisted of three analytical systems and an automated measurement system (AMS). Each analytical system contained a hydrogen filter with its temperature control, vacuum system and omegatron-type mass-analyzer.

Automated measurement system provided for control of three mass-analyzers and registration of their signals (mass-spectra). The AMS comprised a programmable oscillator of operating frequency for mass-analyzers, an analog switch with three signals for mass-analyzers to one output, and a digital voltmeter linked with the control computer via a RS–232 serial interface.

The Ms3T program on the control computer (server) controls the oscillator and relay switch via the parallel port LPT and reads a measured value from the voltmeter via serial port COM. There is a second computer assigned for remote control of measurement system during the reactor operation. This remote computer (host) is equipped with an Ethernet card linked by UTP cable with the server and runs special software for remotely administering the server using TCP/IP protocol. From the safe distance of about 70 m, the host computer provides full access to server running Ms3T program: e.g. run and stop

Table 1
Technical specifications of tritium measurement system 'Sakura'

Number of measurement capsules	3
Mass-analyzer (type)	Omegatron, RMO–13 tube
Examined masses (Big Scan)	2–6 (H_2, HD, HT, DT, T_2)
Examined masses (Single Scan)	2–43
Maximum pressure of H_2/T_2 in capsule	$<5 \times 10^4$ Pa
Minimum pressure of H_2/T_2 in capsule	10^{-1} Pa
Maximum residual pressure in analyzer	$<10^{-8}$ Pa
Frequency range of mass-analyzers	30–2300 kHz, step 100 Hz
Induction in analyzer's magnet gap	3900 G (0.39 T)
Input resistance of electrometers	100 GΩ
Output signal	Analogous, 10^{-4}–10 V
Readiness time after system turn-on	<30 min

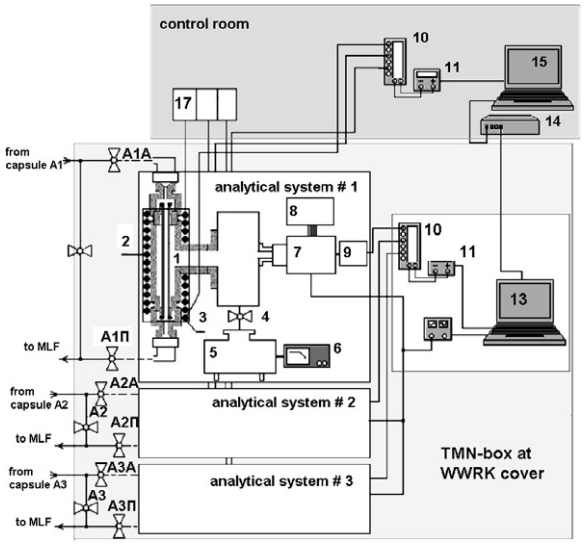

Fig. 3. Block-scheme of tritium measurement system 'Sakura': 1 – 'hydrogen window' (PdAg-filter); 2 – heater; 3 – thermocouple; 4 – vacuum gate; 5 – vacuum pump (NORD); 6 – power supply for pump; 7 – RMO-13 (omegatron); 8 – power supply for omegatron; 9 – electrometer; 10 – switch-box; 11 – voltmeter; 12 – programmable oscillator; 13 – server; 14 – hub, 15 – remote terminal (host), 17 – heater control.

self-diagnostic or measuring routines, set-up measurements, select needed channel (capsule) etc.

Ms3T software provides two operating modes: so-called 'Big Scan' – regime of alternate measurement of concentrations of three selected gases (tritium, HT and hydrogen) in each mass-analyzer (i.e. in capsules A1–A3) and 'Single Scan' – regime of ordinary plain mass-scan in selected capsule within specified interval of masses.

'Big Scan' is the main working mode used for time-domain monitoring of tritium, HT and hydrogen concentrations penetrated via PdAg-filters. The peak of every corresponding gas is scanned from the beginning of its rise to the end of decay to avoid the possible error in evaluation of peak height related with d.c. zero drift. The duration of such scan takes about five minutes per capsule. 'Single Scan' mode is used as optional for controlling the entire gas composition in the case of need (usually before the start and at the end of irradiation campaign). The operator can specify the scanned range of gasses.

Regardless of system's operation mode, all measured data are represented graphically on the computer display, recorded to log-file and stored in data-base.

4. System installation and carrying-out of experiment

Before the system 'Sakura' was installed in the working site, all three analytic systems were pumped-out to the pressure of 10^{-8} Pa and sealed. After its installation in the 'TMN-box' and porting to examined capsules and MLF, the system vacuum was checked again. Such a check-up was conducted at the beginning of every irradiation campaign. After that, the 'Sakura' system was turned-on and linked to the remote computer. From this point the system was operated under remote control from the control room located in the same place as MLF panel.

Usually, at the beginning of each irradiation campaign (before and after the hydrogen filter's heaters are turned-on), the spectrum of residual gases in the analytical system was registered to look for peaks of hydrogen, water and oxygen to detect possible leakage in the vacuum system.

After the system is checked and MLF is ready for irradiation, the heaters of PdAg-filters are turned-on and heated-up to operating temperature 450 °C. Heater's temperature control was implement from MLF panel using established communication means in TMN-box. Filter's temperatures were controlled with the second pair of switch-voltmeter and special software PdAg-filter, running on the host computer. PdAg-filter software allowed logging the filter's temperature history for an entire irradiation campaign or up to 30 days. After all filters are heated-up to operating temperatures the 'Big Scan' is started and system 'Sakura' gives the readiness signal for reactor power increase to 6 MWt in a new irradiation campaign.

5. Results

The tritium measurement system 'Sakura' was designed and implemented to provide the measurements of tritium generation rate in Li_2TiO_3 ceramics during its long-term (220 days) irradiation in the WWRK reactor. 'Sakura' system provided reliable and continuous registration of dynamics of tritium yield from examined ceramic samples in three experimental capsules during the entire irradiation period (more them 5000 h). Fig. 4 shows the example of data registered with the 'Sakura' system: the dynamics of tritium release (curve 2) from capsule A1 during irradiation campaign 13 (two weeks). Characteristic bursts of tritium yield are caused by the changes of ceramic temperature (curve 1).

After completing the experiment, the hydrogen filters and pipes of each analytical system were annealed during pump-out by their ion pumps NORD-250 below the acceptable level of tritium contents and after that the pumps were disconnected, sealed and stored as radioactive sources in special storage. According to our evaluation, the maximum amount of tritium contained in each pump (as titanium tritide) does not exceed 50 Ci. Main results of the tritium generation rate for each ampoule are represented in Table 2. The tritium

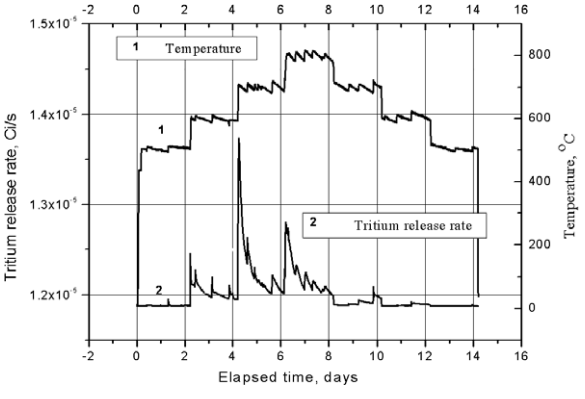

Fig. 4. Dynamics of tritium yield from A1 capsule during irradiation in Campaign 14. Ceramic temperature is curve 1.

Table 2
Tritium generation rate for each ampoule (evaluation and experimental data)

Capsule no.	A1 T release rate Ci/s		A2 T release rate Ci/s		A3 T release rate Ci/s	
Test no.	Evaluation	Experiment	Evaluation	Experiment	Evaluation	Experiment
Campaign 1	1.57E−05		1.57E−05	1.67E−05	1.57E−05	1.80E−05
Campaign 2	1.61E−05	1.66E−05	1.61E−05	2.37E−05	1.61E−05	1.59E−05
Campaign 3	1.64E−05	1.67E−05	1.64E−05	2.58E−05	1.64E−05	1.56E−05
Campaign 4	1.59E−05	1.83E−05	1.59E−05	2.32E−05	1.59E−05	1.64E−05
Campaign 5	1.50E−05	1.44E−05	1.50E−05	2.07E−05	1.50E−05	1.55E−05
Campaign 6	1.34E−05	1.27E−05	1.34E−05	2.05E−05	1.34E−05	1.26E−05
Campaign 7	1.28E−05	1.42E−05	1.28E−05		1.28E−05	1.36E−05
Campaign 8	1.23E−05	1.27E−05	1.23E−05		1.23E−05	1.35E−05
Campaign 9	1.22E−05	1.34E−05	1.22E−05		1.22E−05	1.39E−05
Campaign 10	1.17E−05	1.25E−05	1.17E−05		1.17E−05	1.15E−05
Campaign 11	1.09E−05	1.04E−05	1.09E−05		1.09E−05	1.01E−05
Campaign 12	1.11E−05	1.13E−05	1.11E−05		1.11E−05	1.06E−05
Campaign 13	1.12E−05	1.20E−05	1.12E−05		1.12E−05	1.17E−05
Campaign 14	1.16E−05	1.24E−05	1.16E−05		1.16E−05	1.11E−05
Campaign 15	1.19E−05	1.16E−05	1.19E−05		1.19E−05	1.23E−05

generation rate (lithium burn-up) was evaluated by using the MCNP codes in accordance with formula

$$R = N \cdot \int_0^{20\ \mathrm{MeV}} \varphi(E) \cdot \sigma_{\mathrm{E}} \cdot \mathrm{d}E, \tag{3}$$

where N is the density of lithium-6 atoms, $\varphi(E)$ is the flux of the neutrons; and σ_{E} is the section of the reaction $(\mathrm{Li}^6, \mathrm{n}) \rightarrow (\mathrm{T}, \mathrm{He})$.

6. Conclusions

The developed technique of tritium measurement from the gas mixture was used in a real time intra-channel experiment to study lithium burn-up from lithium ceramic Li_2TiO_3 enriched up to 96% in isotope ^{6}Li. Lithium burn-up reached 23% for the active ampoule pebbles, 20% for the passive ampoule pebbles, and 18% for the pellets. The tritium measurement system permitted the tritium yield rate to be determined under long-term irradiation of lithium ceramic Li_2TiO_3.

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References

[1] J.G. Van der Laan, R. Conrad, K. Bakker, N. Roux, M.P. Stijkel, in: Proceedings of the 20th Symposium on Fusion Technology, France, 1998, p. 1239.
[2] K. Tsuchiya, N. Nakamichi, Y. Nagao, M. Enoeda, T. Osaki, S. Tanak, H. Kawamura, J. Nucl. Sci. Technol. 38 (11) (2001) 996.
[3] J.G. Van der Laan, L.V. Boccaccini, R. Conrad, J.H. Fokkens, M. Jong, A.J. Magielsen, B.J. Pijlgroms, J. Reimann, M.P. Stijkel, S. Malang, Fusion Eng. Des. 61&62 (2002) 383.
[4] K. Tsuchiya, H. Kawamura, Fabrication and Characterizationn of ^{6}Li-enriched Li_2TiO_3 Pebbles for a High Li-burnup Irradiation Tests, Japan Atomic Energy Agency, October, 2006.
[5] V. Shestakov, I. Tazhibayeva, H. Kawamura, Y. Kenzhin, T. Kulsartov, Y. Chikhray, A. Kolbaenkov, F. Arinkin, Sh. Gizatulin, P. Chakrov, Fusion Sci. Technol. 47 (2005) 1084.