DEVELOPMENT OF METHODOLOGIES FOR ISOTOPE ANALYSIS OF HEAVY WATER COOLANT AT THE PIK REACTOR

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PIK is a research heavy water reactor facility. According to the project, the PIK reactor in corporates two heavy-water circuits: a heavy water reflector (HWR) circuit and a liquid control loop (LCL). This requires the organization of control of the isotopic composition of heavy water (the content of deuterium and tritium), since:

1. Heavy water is hygroscopic, the rate of its dilution in the reactors is estimated as (0.1-0.3) atomic % per year. Calculations suggest that dilution (decrease of deuterium concentration) of heavy water in the reflector of the PIK reactor by 1% would result in a decrease in the thermal neutron flux by 20% [1].
2. A significant part (up to 40%) of the occupational dose-at the heavy water reactor facility is due to tritium dose. At the PIK reactor, the maximum volumetric activity of heavy water by tritium should not exceed 2 Ci / dm^3 [2].

Special methodologies are developed at the PIK reactor complex to determine the content of deuterium and tritium in heavy water.

To analyze the content of tritium in heavy water, the most sensitive and accurate radiometric method was chosen. In the laboratory of industrial chemical control, such an analysis is performed on a liquid-scintillation radiometer "Thriathler LSC". For several years, the technique "Measurement of the volume activity of tritium in light and heavy water by the method of liquid-scintillation radiometry" has been developed.

The work was performed using SSS (Standard State Sample) of tritium water, calibration standards (CS) were prepared from the SSS by the weight method. Based on the CS obtained, calibration dependences of the volumetric tritium activity on the radiometer count were constructed, the errors and the measurement range of the method were calculated. In addition, the influence of the measurement time on the result of the analysis was determined. In order to extend the measurement range of the method, different ratios of V_{sample}/V_{SL}, where SL is a Scintillation Liquid, were used. Approved in 2016, the technique allows determining the volumetric activity of tritium water from $3.7 \cdot 10^{-7}$Ci/dm^3 to 6 Ci/dm^3 [2]. The equations of calibration dependences are presented in Table 1.

Table 1. Equations of calibration dependences for various ratios V_{sample}/V_{SL} and their characteristics.

V_{sample}/V_{SL}	Dependence equation C_T (Ci/dm^3) of I (imp/sec)	C_{min}, Ci/dm^3	C_{max}, Ci/dm^3
0.1/10	$4.25 \cdot 10^{-8} \cdot (I - I_0)$	$8.89 \cdot 10^{-6}$	$8.22 \cdot 10^{-2}$
1/10	$5.68 \cdot 10^{-9} \cdot (I - I_0)$	$1.15 \cdot 10^{-6}$	$1.11 \cdot 10^{-2}$
8/10	$1.83 \cdot 10^{-9} \cdot (I - I_0)$	$4.77 \cdot 10^{-7}$	$3.95 \cdot 10^{-3}$

The work is planned to be continued: it is necessary to study the influence of impurities in the coolant on the analysis result.

The next parameter that is important to control in the heavy water coolant is the deuterium content. Due to the high hygroscopicity of heavy water, working with it requires certain skills [and conditions]: tightness of the sample and storage tanks, low air humidity in the laboratory, etc. At the PIK facility, an IR spectrometry method was chosen for this analysis; this method is fast and sensitive enough, does not require additional preparation or a large amount of an analyzed sample. In the laboratory of industrial chemical control, measurements are performed on an IR Fourier spectrometer Tensor-37.

Several years ago, 14 calibration solutions of heavy water were prepared and measured for the development of a technique for measuring the deuterium concentration by the weight method. Two calibration curves were constructed from the spectrum obtained: for concentrated heavy water (99.0 – 99.9) atomic % and for the entire concentration range (0-100) atomic %. The error of curves was 0.005 atomic % and 0.4 atomic % respectively. These dependencies were used to analyze the heavy water of the PIK reactor HWR, but the analysis error over a wide range of heavy water concentrations turned out to be high. Therefore, the work was continued. For this purpose, 46 calibration solutions of heavy water were prepared, their spectra were measured and several calibration dependences were constructed. Moreover, the range of measured concentrations has been experimentally divided into several parts. The data are presented in Table 2. As a result, it was possible to reduce the measurement error by an order of magnitude. Today, these calibration dependencies are used for analysis.

Table 2. Calibration dependence equations and their error for different concentration ranges.

Concentration range, atomic %	Calibration dependence equation	Analysis error, Δ, %
99.50 – 99.90	$a_D = 99.689 - 0.00166 \cdot S$	0.005
99.00 – 99.90	$a_D = 99.687 - 0.00168 \cdot S$	0.01
90.00 – 99.00	$a_D = 152.6 - 0.1586 \cdot S + 0.00095 \cdot S^2$	0.02
50.00 – 90.00	$a_D = 131.94 - 0.3712 \cdot S + 0.00023 \cdot S^2$	0.03
10.00 – 50.00	$a_D = 123.16 - 0.29924 \cdot S + 0.00008 \cdot S^2$	0.06
1.00 – 10.00	$a_D = 56.968 - 0.20462 \cdot S + 0.00012 \cdot S^2$	0.03
0.015 – 1.00	$a_D = 0.49215 + 0.00463 \cdot S$	0.003
0.015 – 99.90	$a_D = 0.49215 + 0.00463 \cdot S$	0.4

Currently, the measurement technique of deuterium in heavy water is under design.

REFERENCES

1 Petrov Yu.V. Choice of reactor parameters for physical researches // Preprint LNPI – 802. L., 1982. P. 62.

2. Voronina T.V., Romanova D.E., Tugusheva D.Yu. "Determination of the volumetric activity of tritium in the PIK reactor coolants by the liquid-scintillation radiometry method". Main results of scientific work 2015. Gatchina, 2016, P.106.