ANALYSIS OF GAMMA ACTIVITY OF HEAVY WATER AT RB REACTOR

M. Pešić, V. Ljubenov, T. Milovanović, S. Avdić, S. Milovanović
The Institute of Nuclear Sciences "Vinča",
Nuclear Engineering Laboratory,
P.O.Box 522, 11001 Belgrade, Yugoslavia

Abstract

The RB experimental nuclear reactor still works with heavy water obtained in 1959 from the former USSR. Gamma activity of heavy water was periodically controlled during the past time. In this experiment measurements were carried out with two samples: D_2O taken from RB reactor and D_2O that has never been used in reactor. Two germanium spectrometers were used as detectors. Gamma spectra's data were evaluated manually and using several computer codes. Results of the experiment show that gamma activity D_2O of RB reactor is at the level of background in "Vinča" Institute, without contamination with fission products.

Key words: Reactor RB, heavy water, gamma activity

1. Introduction

The RB zero-power, heavy water reactor started operation in 1958 with heavy water obtained in former USSR. That water was removed to RA reactor at the end of 1959, when the RA reactor was put in operation. New heavy water was obtained from USSR and it is still used in RB reactor, after 37 years of continual operation. This heavy water is controlled each year, and even more frequently if it is necessary. Control is related on isotope composition of heavy water, it's pH factor and it's possible contamination with fission products.

2. Experimental procedure

Gamma activity analysis of RB reactor heavy water was performed in 1995, for the first time on low-background (Pb shielded) germanium spectrometer. Using that measurement system, analyses of gamma background were carried out up to energies of 2.85 MeV with average integral count rate of 0.5 cps. Gamma background spectrum was also, as usual, measured using coaxial Ge detector with corresponding nuclear electronic modules, placed in experimental room of the RB reactor. These measurements were carried out in energy range between 50 keV and 1.85 MeV, with average integral count rate 16.167 ± 0.009 cps, because it has no special shielding from the background radiation.

For the first time, in order to verify results of measurements, compared gamma activity analyses were carried out with samples of D_2O taken from the RA reactor. That heavy water was bought in former USSR in late 1980's and never used in reactor.

Calibration gamma radioactive sources were used in energy range from 30 keV and 2.7 MeV for energy calibration and determination of the efficiency of the low-background Ge gamma spectrometer. These sources were specially made solutions of naturally gamma radioactive materials in water, placed in plastic bottles of 300 ml volume, "Cedevita" type. One of these calibration sources

contains 12.633 g of KCl solved in 300 ml distillate water, and it provides gamma line with energy 1460.8 keV from ^{40}K nuclide. Second calibration gamma source was made as a solution of 15.063 g $La(NO_3)_3$ x $6H_2O$ in 300 ml distillate water, and it provides gamma lines with energies 788 keV and 1436 keV from ^{138}La, as well as gamma lines with energies 154 keV, 270 keV, 351 keV, 403 keV and 832 keV from radioactive successors in ^{227}Th natural radioactivity chain.

Energy calibration and absolute efficiency curve were determined by using gamma lines and their absolute activities determined from pulse amplitude spectrum, collected in Canberra series-35 MCA with 100 MHz ADC and 8192 channel memory:

$$E_\gamma[keV] = 0.34999 \cdot ch + 8.711, \quad ch \in [1,8192] \tag{1}$$

The absolute activities of calibration sources were determined using known mass of radioactive nuclides in the solution. The absolute efficiency of the spectrometer in whole energy range was obtained by least square fitting method, through efficiency-energy points, as a hyperbolical function:

$$\varepsilon = \frac{4.65699}{E_\gamma(keV)^{1.04466}} \tag{2}$$

Samples of 300 ml of heavy water from RB and RA reactors were prepared on the same way as the calibration sources (in plastic bottles of 300 ml volume, "Cedevita" type) and analyzed by low-background Ge gamma spectrometer. Measurements of the samples of heavy water from RB reactor were carried out for 240000 s, with total integral of 135093 counts, that provides average integral count rate of 0.563 cps (i.e., at the gamma background level of the spectrometer). Measurements of the samples of heavy water from RA reactor were carried out for 96 000 s, with total integral of 55751 counts, which provides average integral count rate of 0.581 cps (i.e., again at the gamma background level of the spectrometer).

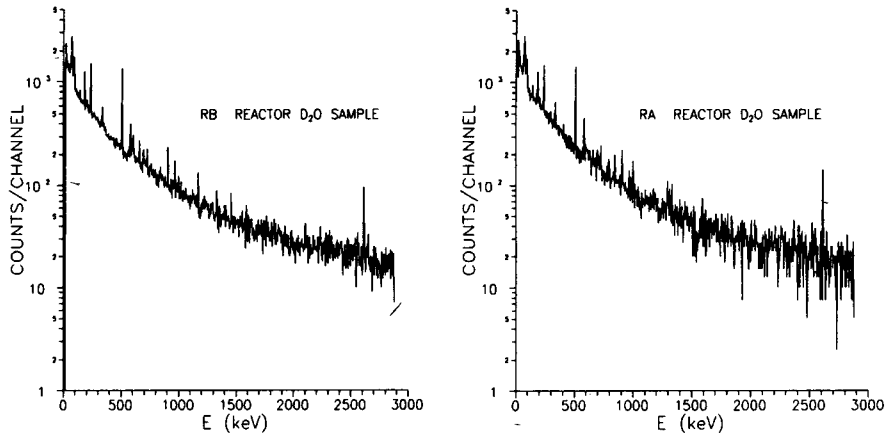
Germanium gamma spectrometer system, used in the experimental room of RB reactor, was energy and efficiency calibrated with several point sources on different distances from the detector axis. These calibrations were verified with four volume gamma radioactive sources in two energy points. These volume calibration sources were made as solutions of KCl in distillate water (with concentrations of 1 g K in 50 ml and 5 g K in 50 ml), and solutions of ^{137}Cs in distillate water with activity of 137.7 ± 5.5 kBq and 275 ± 11 kBq on 1995-03-29. Samples of heavy water from RB and RA reactor and calibrated solutions were made in plastic bottles of 54.0 ± 0.1 ml volume.

RB reactor's heavy water gamma activity analyses were carried out at 15 cm distance from the detector, during 231678 s, with total integral $3{,}7766 \cdot 10^6$ counts, which provides average integral count rate of 16.301 ± 0.009 cps, what is at the gamma background level of the spectrometer. Measurements of gamma activity of the samples of heavy water from RA reactor were carried out for 352449 s, with total integral of $5.6809 \cdot 10^6$ counts, providing average integral count rate of 16.118 ± 0.007 cps, (i.e., at the gamma background level of the spectrometer).

3. Results of measurements

Data from measured gamma spectra were evaluated manually, as well as with several computer codes. Three of used computer codes are commercial ones, GANAAS [1], MicroSAMPO [2] and APOGEE [3] from Canberra Inc., and the fourth one is ANA computer code [4] developed in Laboratory NET. Only representative gamma lines in measured spectra are given. The measured

amplitude gamma spectrums for both samples of heavy water (from RA and RB reactors), obtained at the low-background Ge spectrometer are shown at Figure 1. Results are evaluated at the same measuring time of gamma spectra: 240000 s.



a b

Figure 1. Gamma spectra of heavy water sample from RB reactor (a) and heavy water sample from RA reactor (b)

4. Conclusion

Gamma activity analysis of RB reactor heavy water, including comparations with gamma activity of sample of heavy water (obtained from RA reactor) never used in reactor, shows that its gamma activity is on the gamma background level in "Vinča" Institute, without contamination by fission products.

5. References

[1] GANAAS, Computer Manual Series No.3, IAEA, 1991
[2] MicroSAMPO Version 1.2, User's Manual CISE 511, Canberra Inc., 1988
[3] APOGEE Version 1.3, Gamma Spectrum Analysis Code, Canberra Industries Inc., 1986
[4] Avdić S., Pešić M., "ANA, A Program for Aútomatic Gamma Spectrum Determination with Library of Nuclides Gamma Lines", IBK-NET-Vinča computer codes library, Vinča 1987