ASSESSMENT OF CONDITIONS OF THE SPENT NUCLEAR FUEL STORED IN THE STAINLESS STEEL CHANNEL-HOLDERS

M. Pešić, O. Šotić, S. Cupać, T. Maksin, N. Dašić
Centre for Nuclear Technologies and Research - NTI
VINČA Institute of Nuclear Sciences, P. O. Box 522, 11001 Belgrade, Serbia and Montenegro

ABSTRACT

The IAEA technical co-operation project "Safe Removal of Spent Fuel of the Vinca RA Research Reactor" is carried out at the Vinča Institute of Nuclear Sciences, Belgrade, Serbia and Montenegro, since January 2003. Present activities will provide up-to-date information on the conditions of the spent nuclear fuel, stored in the stainless steel channel-holders ('chekhols') and on the water quality in the storage basins. Water samples taken out from the chekhols and the basins are measured to determine their activity and chemical parameters. Until September 2003, about 1/3 of the chekhols containing spent fuel elements with initial enrichment of 2% and 80% of uranium were inspected. High activity of Cs-137 was found in several water samples taken out from chekhols. All water samples show very high electrical conductivity, while those taken from the basins show the presence of chlorides and aluminium ions, too. Information on established procedures and measuring results are given in this paper. The obtained results, so far, show that the spent nuclear fuel elements are leaking in about 10 % of chekhols.

1. Introduction

The 6.5 MW heavy water moderated and cooled research reactor RA (Figure 1) was bought as a turn-in key project from the ex-USSR in 1956. It started operation in December 1959, with 2% low enriched uranium (LEU) metal fuel elements of the TVR-S shape, produced in the "Elekhtrostal" factory near Moscow, Russia. The reactor operated with LEU fuel until 1976, when new, 80% high-enriched uranium (HEU) dioxide TVR-S fuel elements were purchased from the "Novosibirsk Chemical Concentration Plant", Novosibirsk, Russia. Both fuel elements have the same geometry, shape and similar mass of U-235 nuclide (7.25 g – 7.7 g). After several years of operation with both types of fuel elements in the 'mixed' core, the RA research reactor switched to operation with HEU fuel elements only in 1981. The RA reactor was temporarly shut down in 1984 for refurbishment and reconstruction. However, due to technical and mainly political reasons, the reactor has never been restarted again, but stayed in the extended shutdown condition for 18 years [1]. Finally, in July 2002, the Government of the Republic of Serbia has made a decision, based on economical reasons and operational demands, to shut down the RA reactor permanently and to decommission it in the near future [2]. The first step towards decommissioning of the RA research reactor is the safe and reliable disposal of spent fuel. These activities, recognized as very important by international community and by the Serbian government too, led to set up of the 'Green Vinča' (Vinča Institute Nuclear Decommissioning – VIND) Program [3-4]. Activities of the VIND Program have been supported by the US non-governmental organization Nuclear Treat Initiative (NTI) and by the IAEA.

Figure 1. The 6.5 MW heavy water research reactor RA at the Vinča Institute

Figure 2. The TVR-S LEU and HEU fuel elements of the RA reactor

2. TVR-S fuel element

The TVR-S fuel element is an annular cylinder, with 3.72 cm outer diameter, consisting of a tube with 2 mm thick fissionable material and 1 mm thick inner and outer aluminium cladding. The fuel layer has the total length of 10.0 cm. An inner aluminium tube ("expeller") serves as a coolant flow adjuster. At bothe ends of the fuel element, an aluminium ‘star-shaped’ element (3 mm thick) is fixed to expeller, so the total length of the fuel element is 11.3 cm. During reactor operation 8030 fuel elements were used. Fuel elements with uranium metal having initial enrichment of 2 % ^{235}U were used until 1979, and afterwards, the fuel elements with uranium dioxide in aluminium matrix, having initial enrichment of 80 % ^{235}U were used. Full material compositions of the fuel elements are described in Ref. [5]. Aluminium, used in the fuel elements, is known in Russia as the SAV-1 alloy (0.985 weight fraction is aluminium, with very low contents of neutron high-absorbing impurities, e.g., boron or cadmium). Fuel channels of the RA research reactor are formed using 11 such fuel elements placed one above another in a 5.5 m long aluminium tube. Between 40 and 82 fuel channels could be used in the RA reactor core in a regular square lattice with a pitch of 13 cm. Heavy water is used for neutron moderation, and as the top and the bottom axial reflector and as the primary coolant, too.

3. Burn up data

The total of 6656 LEU metal fuel elements have been irradiated in the RA reactor core during its operation from 1959 to 1979. Maximum fuel burn up for 2% enriched metal uranium fuel is about 12 GW days per ton of uranium (GWd/tU), while average fuel burn up for all LEU fuel elements in the reactor RA is 18.3 MW per ton of uranium. The total of 480 HEU spent fuel elements have been left in the drained RA reactor core, since 1984, while 894 irradiated fuel elements of HEU are stored in the stainless steel containers in the spent fuel storage pool. Maximum burn up of HEU fuel elements in the RA core is less than 65 GWd/tU, i.e., the change of the content of ^{235}U nuclide is less than 10 %. The total activity of all spent LEU and HEU fuel elements stored in the spent fuel storage pool is now estimated at 3800 TBq, according to the fuel irradiation history in the RA reactor core and cooling time in the spent fuel storage pool. It was shown that one third of this activity comes from uranium and transuranium nuclides, while about 99 % of two thirds of the total activity originates from ^{137}Cs and ^{90}Sr nuclides (^{85}Kr nuclide contribution is 1 % only).

4. Spent fuel storage at the RA research reactor

Temporary spent fuel storage pool, 6.5 m deep, in the basement of the RA reactor building, consists of four connected basins, having thick concrete walls covered by 1 cm thick stainless steel plates. It is filled with approximately 200 m^3 of stagnant ordinary water. The total of 304 channel-type stainless steel holders (‘chekhols’) are used as the original fuel containers. Each chekhol stores reactor fuel channel with 11 spent fuel elements. In order to increase the storage capacity, since no plans for spent fuel removal have been envisaged, beginning in the second half of 1960’s, the spent fuel elements were gradually taken out from chekhols and repacked into 30 specially manufactured aluminium barrels. Each aluminium barrel contains 30 aluminium tubes that could receive up to 6 spent fuel elements per tube. These aluminium barrels were placed in two layers in the annex to the basin No. 4. Both the barrels and the chekhols were filled with demineralised water and hermetically closed.

According to the original ex-USSR design, the RA reactor spent fuel storage had no system for the pool water purification. Monitoring and maintaining the water radiochemical parameters in the pool were not regulary conducted, and were even considered unnecessary since the pool water was not in the direct contact with the spent fuel. During the years, a lot of sludge has been accumulated at the bottom of the pool. A serious concern about conditions of the spent fuel storage pool was expressed in 1994, when possibility of the fuel leakage inside the containers and eventual release of fission products from the containers, was pointed out. Water samples were taken in 1996 from 28 chekhols. Increased ^{137}Cs specific activity in some of these samples was an indication that some spent fuel elements were leaking. This activity was in a range from 10 Bq/mL to 400 Bq/mL in most of the chekhols, reaching up to several hundreds kBq/mL ^{137}Cs in one chekhol with normally disposed fuel. Very high activity of ^{137}Cs about 50 MBq/mL, was measured in the chechol containing the fuel element that failed during reactor operation.

Figure 3. Spent fuel storage of the RA reactor

5. Examination of conditions inside the stainless steel channel-holders

Within the IAEA technical co-operation project "Safe Removal of Spent Fuel of the RA Research Reactor," carried out in the Vinča Institute of Nuclear Sciences, Belgrade, Serbia and Montenegro since January 2003, several activities are initiated to provide up-to-date information on the conditions of the spent nuclear fuel stored in all stainless steel channel-holders ('chekhols') and on the water quality in the storage basins. Main task was to take out water samples, both from the chekhols and the basins and to measure their activity and chemical parameters. In order to fulfil this task several preparatory activities had to be worked out:

- Rough estimate of gaseous fission products release inside the chekhols containing spent fuel with 2% initial enrichment. Such a release from spent fuel with 80% initial enrichment may be neglected due to dispersed type of fuel;
- Establishing general procedures for conducting activities of the project;
- Assembling of the operating team and introducing team members with the scope of the project and activities planned (including the training in conducting specific operations);
- Installation of stationary gamma-ray control system and movable aerosol measuring monitor system in the spent fuel storage.

Figure 4. Stainless steel channel-holders ('chekhols') in the RA storage pool

Estimation of possible gaseous fission product release inside the chekhols was necessary for establishing adequate protective measures during opening and contents verification operations of these holders. The worst condition assumed, was the case that all spent fuel elements with initial enrichment of 2% inside one chekhols are leaking [6]. As expected, the possible accumulation of ^{85}Kr and ^{3}H required additional protective measures to be applied.

The first step in verifying the location and contents of each chekhol was to gather existing data on the current situation. Those data, although not quite accurate, represent the basis in performing this activity. All the basins and transport channels were visually checked and the following general data about chekhols are recorded:

Total number of holders : 304

- in basins No. 1 : 63 (1 added from the basin No. 4)
- in basins No. 2 : 103
- in basins No. 3 : 103
- in transport channel : 35 (28 in the reactor room, 7 in the spent fuel storage)

According to existing documents, contents of the checkhols is expected to be the following one:

- empty holders: 34
- holders with various components (without fuel): 22
- holders with 2% enriched fuel elements only: 123
- holders with 80% enriched fuel elements only: 50
- holders with mixed 2% and 80% fuel elements: 75

Before starting to verify the chekhols, adequate technical and safety measures and procedures were established. Also, the purchase and manufacture of necessary equipment, measuring and control devices, handling tools and protective clothes, has bean worked out. All these activities can be summarized as:

- Providing the document on technical procedures and instructions for the set of operations planned for examination of the chekhols,
- Providing the document on safety measures in the reactor room and in the spent fuel storage room for the chekhols handling and examination,

- Determination of additional safety measures when handling the chekhols having spent fuel with 2% initial enrichment using an adequate mobile sucking device connected to the existing special ventilation system in the RA reactor building and using an instrument for continuous monitoring of radioactive gases leaving the rooms,
- Preparing existing devices for tritium detection and for continuous on-line monitoring of α and β particles in the air originated from spent nuclear fuel,
- Manufacturing simple devices for determining contents of the chekhols ("depth meter") and for taking out the sample of water from the chekhol,
- Purchase of necessary laboratory supplies for water samples handling, and also protective clothes, shoes covers and consumable materials.

Adequate tools have been prepared, because of verification of chekhols' contents could be performed only indirectly. The "depth meter" consists of a small metal bar connected by a special hook to a standard tape-meter. Inserting the bar into the chekhol it is possible to measure the height of an object placed inside it. A 200 ml laboratory pump with 7 m long plastic hose has been prepared for taking out water samples from the chekhols. The end of the hose has a nozzle with lead ball preventing the leakage of water from the hose, after being sucked in by the pump. Few sets of these devices were assembled and when a high contamination of the bottom parts of these devices occurred, the new set of devices has been applied.

The calculations have shown that only ^{85}Kr and ^{3}H gasses may be present if fuel elements in the chekhols were leaking. But, due to possibility of existence of other volatile elements in the water inside the chekhols, a provisional movable plastic shield (to be placed above the top of the chekhols) was constructed and connected to the existing special ventilation system of the RA reactor building via separate vacuum cleaner device equipped with filtration unit. The control instrument for continuous on-line measurements of air contamination has been placed in the storage area nearby channel-holders to be verified.

At first, only 56 chekhols without spent fuel (according to previous records) were examined and it was a good training for the operating personnel to get a proper feeling and experience in conducting envisaged operations. Although the obtained data for the chekhols with the fuel elements inside show no discrepancies with previous documents up to now, the complete picture of the real situation can be obtained only after all the chekhols will be verified.

For the measurements of radioactivity of water samples, and for the measurements of their chemical composition, the quantity of about 50 mL of water was taken out. The easiest way to determine the leakage of the spent fuel inside the chekhols is to measure the activity of ^{137}Cs in the water sample. Since the primary coolant circuit was severely contaminated by ^{60}Co during the first three years of reactor operation, this activity was measured, too. The first information on chemical properties of water inside the chekhols is its pH value and specific electrical conductivity. Taking into account corrosion processes of aluminium cladding and the previous results of chemical analyses of the water samples taken from several chekhols (performed in 1996), it was decided to determine the presence of: chlorides, sulphates, aluminium and iron ions in the samples of water to be taken out from chekhols.

Figure 5. Preparation for taking out the water sample from the chekhol

Results of the chekhols verification up to now [7] have shown that out of 56 chekhols without spent fuel, 15 of them were completely dry and 20 of them were inaccessible for water sucking (the plastic hose could not pass to the lower parts of chekhols). Twenty-one water samples were taken and measured. Out of 83 chekhols containing spent fuel, water samples were taken from 63 ones, while 20 of them were inaccessible. The measurements of radioactivity of water samples were performed at the Centre NTI, while the measurements of their chemical parameters were performed at the Laboratory for Radioisotope Production. Firstly, preparatory activities for these measurements were realised comprising:

- Preparing measuring system based on NaI crystal, calibrated for energy and efficiency of gamma-rays detection,
- Preparing gamma-ray standard volume sources of ^{137}Cs and ^{60}Co nuclides in adequate dishes,
- Preparing measuring devices for pH and electrical conductivity determination,
- Preparing analytical measuring techniques for chemical contents determination.

Results show that radioactivity of water in chekhols has values in following ranges:

^{60}Co : 2.5 Bq/mL - 707 Bq/mL (120 ± 40 Bq/mL on average)
^{137}Cs : 5 Bq/mL – 850 kBq/mL (80 ± 20 Bq/mL on average).

Chemical parameters given by pH and specific electrical conductivity show following values:

pH : 2.4 - 8.1 (5-7 on avarage)
El. cond.: 21 µS/cm - 21000 µS/cm (400 µS/cm on average).

Chemical components concentration analysis of the water samples has been done for two samples only, for chekhols without spent fuel. Out of 21 such samples, all having very low radioactivities, the two were chosen for chemical components analysis and the only detectable component were iron ions, though in very small quantities (< 0.05 mg/L). Among 84 water samples taken from chekhols, 8 of them show very high activities of ^{137}Cs (25 kBq/mL – 850 kBq/mL), obviously due to the spent fuel leakage inside them. During the opening of the chekhols (not all of them had covers, and none of them wos hermetically closed), the gamma-ray dose rate at the top of them has been measured in the range from 4 μSv/h to 25 μSv/h. Water samples from the basin No. 4 and from the transport channel inside the spent fuel storage have been taken each month. So far, neither changes in radioactivity of this water, nor in chemical components concentration have been detected. The measured values of radioactivity of water samples are in the range of 97 Bq/mL to 106 Bq/mL for ^{137}Cs, while ^{60}Co is not detected.

Specific electrical conductivity of the water is in the range (510 - 570) μS/cm and pH in the range 7.5 – 8.2. Chemical components concentration of the water samples shows the following constituents: chlorides (69 – 72) mg/L, sulphate ions (31 – 37) mg/L, iron ions less than 0.05 mg/L while aluminium ions were not detected.

Instead of conclusion

Final assessment of conditions of the spent nuclear fuel stored in the stainless steel channel-holders will be given after verification of all chekhols and measurements. Results obtained up to now show that the spent nuclear fuel elements are leaking in about 10 % of chekhols.

Acknowledgement

This work was supported in part by the Ministry of Science, Technologies and Development of the Republic of Serbia through the Project no. 1958 "Transport of particles in fission and fusion systems" and by the NTI through the IAEA TCP YUG/4/029 "Safe Removal of Spent Fuel of the Vinca RA Research Reactor."

References

[1] M. Pešić, V. Kolundžija, V. Ljubenov, S. Cupać: "Causes of Extended Shutdown State of "RA" Research Reactor in "Vinča" Institute", IAEA TCP RER/9/058: *Safety Review of Research Reactor Facilities*, Regional Workshop on *Extended Shutdown and Decommissioning of Research Reactors*, pp. 001-011, Riga, Latvia (26-30 November 2001).

[2] M. Pešić, K. Subotić, O. Šotić, I. Plećaš, V. Ljubenov, A. Perić: "Vinča Nuclear Decommissioning Program", Proc. of *XLVI Yugoslav Conf. ETRAN*, invited paper, Vol. **IV**, pp. 21-30, Banja Vrućica, Teslić, Republika Srpska (3 - 6 June 2002)

[3] V. Ljubenov, M. Pešić, V. Kolundžija: "RA Research Reactor in Vinča Institute – Decommissioning Project," *USDOE/CNCAN Joint Decommissioning Workshop*, Power Point Presentation, pp. 1-23, CD ROM, Bucharest, Romania (May 05-09, 2003)

[4] M. Pešić, K. Subotić, O. Šotić, I. Plećaš, V. Ljubenov, A. Perić, M. Milošević: "Yugoslav Spent Nuclear Fuel Management Program and International Perspectives," *International Conference on Irradiated Nuclear Fuel Management 2002, INFM2002: Russian New Initiatives*, Proc., pp. 105-110, Moscow, Russia (September 8-12, 2002

[5] M. Pešić: "Criticality Safety of LEU and HEU Fuel Elements in Heavy Water Lattices," Nuclear Science and Engineering, Vol. **145**, No. 2, pp. 000-000 (October 2003**)**

[6] M. Milošević: "Summary of Calculation of One Spent Fuel Reactor RA Element Activity, " Centre NTI Internal Memo, Vinča (January 2003)

[7] O. Šotić, M. Pešić, S. Cupać, T. Maksin, N. Dašić, M. Jevremović, et al., "IAEA Project YUG 4028-92292A Preliminary Analyses of the Feasibility of Spent Fuel Management", Progres Report (December 2002 – July 2003) Vinča (July 31, 2003)