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ADVANCED METHODS OF RADIATION DETECTION FOR THE SAFETY OPERATION OF NUCLEAR POWER PLANTS

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Abstract

Applied nuclear physics assists the development of contemporary methods to control the radioactive contamination in the primary circuit of power reactors. We collected experience in the in-situ gamma-spectrometry by the regular measurements on units of type VVER-440/213 for two decades at Paks Nuclear Power Plant. Special techniques were developed for the successful completion of the Restart and Recovery projects after the incident at Paks in 2003: under-water gamma-spectrometry, in-situ alpha-spectrometry, utilization of high-temperature semiconductor detectors.

I. Introduction

A long term and mutually fruitful cooperation between Paks Nuclear Power Plant (NPP) and the Institute of Experimental Physics was established in 1985. The first mission was carried out after the second shut-down of reactor unit 1. All the other three blocks were analyzed from their first operation cycle. The investigations resulted in important data for the conditions of water chemistry, corrosion/erosion processes and hermeticity of the uranium fuel elements by gamma-spectrometry of the main piping system, ion exchange columns and steam generators (heat exchangers) [1].

These continuous sets of measurements are outstanding in the world for VVER-440 reactors as well as for other pressurized water systems giving coherent information on the reactor operation. It enables to follow the effects of water chemistry on the material transport in the primary loops determining the life time of the main construction elements. These measurements are also important for the continuous power upgrade and the near-future licence prolongation. Similar demands required three missions to Biblis-A NPP in Germany.

Regular measurements, performed after the yearly shut-down of the reactor units, have been recently integrated into the normal maintenance work. However, the nuclear methods are also able to produce important information in non-expected circumstances of reactor operation, like the serious incidence at Paks in 2003. Such cases ask for new techniques developed and tailored to the special problems in a very short period of time. Based on the experiences in the fundamental and applied nuclear research as well as utilizing the workshops our institute could react immediately. The success in solving very wide range of problems would have not been achieved without the cooperation between the spin-off company Quantechnologies Research and Development Kft and the University of Debrecen for-establishing the Laboratory of Nuclear Safety and Techniques. Utilizing their own resources and state grants the company performed a flexible procurement policy making possible the fast construction and implementation of new devices in 2003, 2005 and 2007 thus assisting the Restart and Recovery Projects at Paks NPP.

II. Sources of radioactivity in the primary circuit of pressurized water reactors

Reactor vessel, primary loops and other piping, ion exchange columns and heat exchangers are in close contact with a high temperature (∼300 C), high pressure (∼125 bar) moderator/cooling water which has boric-acid in a concentration of some 10 g/kg. Although the water chemistry is tailored so as to decrease the solubility of the stainless steel components by increasing the pH-value over 7, the corrosion processes always take place. High speed turbulent water flow (Reynolds-number well over 10000) together with some particulates in the liquide phase may produce erosion. Atoms

liberated from the surfaces are flushed into the reactor zone where artificial radioactivity will be the result of the neutron irradiation. Such nuclei are then moved throughout the primary tract. Lost materials from malfunction of any devices and/or personnel faults can be also sources of radioactivity. Dehermetization of the fuel assemblies by any small cracks on the surface of the Zr-Nb alloyed tubes containing uranium/transuranium and fission products results in radioactivity of the cooling water, too.

During shut-down of the reactor the temperature decreases and these materials may precipitate to the wall of the tubes forming magnetite or similar layers. The final result of all these processes is a gamma-radiation field around the primary circuit which may give high dose rate causing danger of radiation. The corrosion layers on the fuel tubes may decrease the water flow rate, resulting in lower heat energy transport to the steam generators, which is dangerous because of the possible dehermetization and melting of the fuel.

A chemical cleaning procedure was necessary at Paks in 2003 to make many fuel assemblies free of the magnetite layer built up due to some earlier questionable decisions in the water chemistry and routine maintenance. Design and construction errors of the foreign company, lack of real supervision at the licence procedure in Hungary, malfunctions, further human faults resulted in a serious incidence by insufficient cooling of the cassettes. They ruptured and the radioactive materials contaminated the technical pool where the system worked. Radioactivity disseminated later to the cooling pool as well as to two primary loops of unit 2. (The reactor itself remained intact from the event, therefore it might be restarted later when the appropriate parts were decontaminated.)

III. Nuclear methods to determine radioactive contamination

Precise measurement of the radiation fields gives important data for the reactor operation, quality of water chemistry and maintenance [1]. The main features of the non-destructive determination of the isotope and spatial distributions for radioactive nuclides at an NPP are:

a) measurements on site: “in-situ” spectrometry;

b) high counting rate in the detectors;

c) high background radiation field for the equipment;

d) radiation hazard for the personnel;

e) complex geometry of the sources;

f) hard accessible locations inside the containment box of the reactor.

Since the measurements are generally performed about two weeks after shut down, medium and long half life nuclides may be detected by gamma-spectrometry: ^{51}Cr, ^{59}Fe, ^{58}Co, ^{60}Co, ^{54}Mn, ^{91}Y, ^{110m}Ag, ^{95}Nb, ^{95}Zr, ^{103}Ru, ^{106}Ru/Rh, ^{122}Sb, ^{124}Sb, ^{125}Sb, ^{131}I, ^{133}I, ^{134}Cs, ^{136}Cs, ^{137}Cs, ^{140}Ba/La, ^{141}Ce, ^{144}Ce/Pr, ^{148m}Pm, ^{154}Eu, ^{155}Eu, ^{235}U, ^{238}U.

Gamma-dosimetry was used to account for the local radiation circumstances. Alpha-spectrometry is applied to observe long-lived uranium and transuranium isotopes on contaminated surfaces: ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm. Specific activity refers to a surface or volume of the extended sources. It is calculated from the measured data using the complex efficiency determined by experiments and geometrical calculations.

III.1. Regular measurements with gamma-spectrometers [2]

Six loops transport heat from the fuel to the steam generators in VVER-440/213 type reactors. Their main piping of 50 cm in diameter stainless steel with a wall thickness of 3,5 cm are investigated. Four points of a loop are analysed: two spots before the steam generators (hot leg) and two spots after the generators (cold leg). Ion exchange columns for cleaning the moderator/cooling water are mapped in 10 points along the vertical axis. Horizontal distribution of nuclides on the steam generators is determined at 17 locations.

We apply, in-situ gamma-spectrometry using liquide nitrogen cooled High Purity Ge detectors (HPGe) with active volume of 10 - 100 cm^3 depending on the radiation conditions. PC controlled digital signal processor is connected to the detector preamplifier by a 100 m long cable set. An appropriate lead shield/collimator system restricts the radiation field to a well defined solid angle. This ensamble faces the primary circuit elements from outside. We use paired measurement sequence with open and closed collimator to account for the radiation background.

III.2. Utilization of very high efficiency gamma-detector

High energy but low intensity gamma-lines from dense absorbing material can be detected with extremely high efficiency detectors. Such a measurement was performed at three steam generators of unit 2 to analyse Ce/Pr-144 nuclide by its 2186 keV line. A 100 cm^3 HPGe detector system of 4 crystals, so called "clover", was utilized in the add-back mode [3]. The efficiency at that given photon energy was more than 6 times higher than that of a normal crystal of the same dimensions. Since the transuranium elements behave chemically similar to the Ce-atoms, their correlation made it possible to determine nuclides which have gamma-radiations of low energy and intensity.

III.3. Under-water gamma-spectrometry

During the Restart and Recovery Projects at Paks the decontamination of the surfaces around reactor unit 2 required the determination of specific activity of fission products and uranium as well as transuranium isotopes in advance. While contamination in the cooling pond might have been determined by the activity of dry surfaces of fresh fuel assemblies picked up from water with a crane, the analysis of the walls of the technical pool should have been measured by gamma-spectrometry under water at depth of 0 to –7 m. This task was solved by two stainless steel barrels containing a 100 cm^3 and a 10 cm^3 HPGe detector, neutron counters, GM-tubes, temperature and water sensors. Lead shield and collimator served the precise determination of the location. To avoid effects from other surface activities and the surrounding radioactive water a mobile lead brick made it possible to close and open the window in front of the detectors. Thus the usual paired measurements were available to enhance the reliability of the measurements like in the regular analysis. Since existing equipment in the literature do not apply such collimator [4,5], our solution is quite unique at NPPs. Data transfer and window control are made through a water tight plastic tube containing the necessary cables. Vapor from the liquide nitrogen was exhausted also by this tube. At high altitudes the larger detector of 100 cm^3 was used while at a depth of –7 m (close to the highly radioactive broken fuel) only the smaller device of 10 cm^3 was able to survive the

strong gamma-radiation field. Based on our results the decontamination was successfully performed.

III.4. Gamma-spectrometry with CdZnTe semiconductor detector

There were many places to decontaminate where the accessibility of the surfaces was very limited. A small detector with room temperature operation in a water tight container fitted best the requirements. Since this time (4 years after the incident) the gamma-spectrum became less complex, the energy resolution was not so important as before. A high temperature semiconductor detector of CdZnTe with Coplanar Grid arrangement was utilized [6]. This state-of-art device has a rather large volume of 2.25 cm^3 being on the top in its class [7]. Full width at half maximum of peak around 700 keV is about 17 keV which is quite acceptable for the present purposes (and much better than for a scintillation counter). It was the first occasion when this type of detector was applied for scanning the walls of a reactor technical pool. This method was applied to show fragments of uranium tablets after the excavation of the fuel assemblies and before the final decontamination. Immediate data evaluation was carried out on the spot to select tablets from other contaminated materials.

III.5. Scintillation gamma-ray detector

After the broken cassettes have been taken out a conventional NaI(Tl) scintillation spectrometer was also used as an additional tool for high sensitivity detection of gamma-radiation in the final analysis of the contaminated technical pool.

III.6. In-situ alpha-spectrometry of contaminated surfaces

Dry surfaces of fuel assemblies as well as container and pool walls were analyzed by in-situ alpha-spectrometry instead of sample taking methods of radiochemical laboratories. Energy loss of alpha-particles in air required the utilization of vacuum pump by which the detector container was fixed to

the surfaces, too. The surface inequalities and contamination layers made the spectrum peaks wide. Since the alpha-emitters can contaminate the detector surface itself, we applied a specially passivated Si-detector with a window that can be regularly cleaned [8]. Electronics was similar to that used for gamma-spectrometry. Such a direct surface alpha-spectrometry was not applied earlier in power plant circumstances.

IV. Results and discussions

Some examples of the measurement results at Paks and their conclusions are discussed below.

IV.1. Yearly measurements

Specific surface activity of the isotopes are analyzed as a function of the measurement location and time [2]. As an example Fig.1. shows an interesting set of measurements: ^{59}Fe-activity is extremely high in points 4 at loops III., IV. and V. for a period of 5 years. These loops are by-passed to the feed the water system at points 4 by auxiliary tubes. It was finally concluded that the water was not sufficiently filtered against oxigen which resulted in an extra high corrosion. This effect was observed by in-situ gamma-spectrometry 3 years earlier than the visual inspection could detect it.

The spatial distribution of the specific activity is also interesting at all loops: points 1 and 2 have lower contamination than points 3 and 4 (see Fig.1). The last seems to be the highest for all loops. This is a general feature caused by the temperature difference among the locations. Points 1 and 2 are in the "hot leg" while those of 3 and 4 are in the "cold leg" of the loops. During the years this "normal" behavior may change in time and from unit to unit. These and other observations make it possible to draw conclusions about the quality of the water chemistry and its effects on the reactor operation.

Lost material event was observed at Unit 2 in 1998 by the ^{59}Fe and other corrosion product activity distributions. Fig. 2. clearly shows a well defined increase in the contamination which was dispersed at all loops and

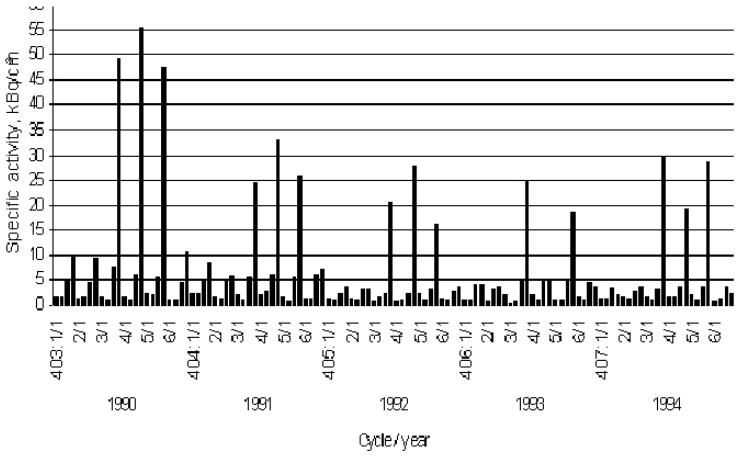

Figure 1: Specific activity of ^{59}Fe at Paks Unit 4 as a function of reactor cycle, year 1/1 ... 6/4: primary circuit points in Loop/location notation. "405": Unit 4, cycle 5.

points. This effect was also observed later by other independent methods.

Radioactive nuclides are produced by different reactions on identical or different targets of corrosion materials. Comparison of the activity of appropriate short and long half-life isotopes makes it possible to estimate the possible operation period of the reactor when the contamination occured/changed.

IV.2. Incidence situations

Results of the under-water gamma-spectrometry are displayed in Fig. 3 for fission product ^{144}Ce. Partly similar spatial dependences were achieved for many other nuclides. Wall of the technical pool was scanned down to a depth –7 m and in three polar positions. Non-isotrop angular and depth distributions are caused by the asymmetric flow of the cooling water around the cleaning tank containing the broken fuel assemblies [9].

Inspection of the cleaning tank and technical pool after removal of the fuel residues was carried out by the CdZnTe gamma-detector. Fragments of the fuel tablets (sometimes in powder form) were found in some regions with

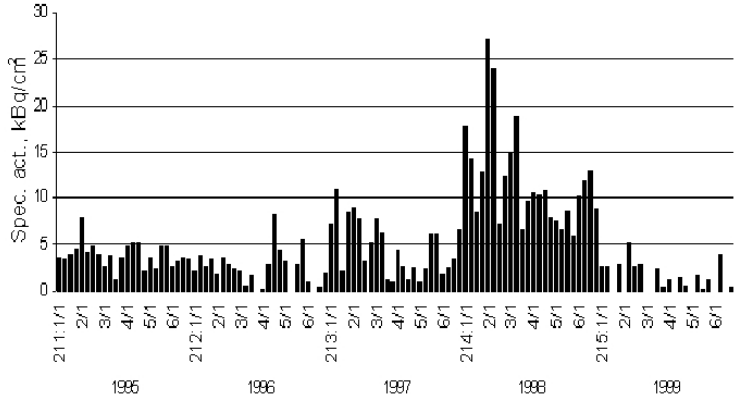

Figure 2: Specific activity distribution of ^{59}Fe around the loop points as a function of time for Unit 2 1/1 ... 6/4: primary circuit points in Loop/location notation. "212": Unit 2, cycle 12..

high amount as it is seen in Fig. 4. Planning of the final decontamination was assisted by these nuclear measurements which have been performed parallel with visual observations using a TV-camera.

V. Conclusions

Sophisticated nuclear methods, based on commercially available equipment, were devised and applied for safety purposes in nuclear power plants. This technology can also be used in other fields of nuclear industry: fuel fabrication and reprocessing, production and use of artificial radioactive sources in medicine, agriculture and industry. Environmental investigations including normally occuring and technologically enhanced radioactive materials (oil industry, water sources, phosphate and other mining), waste management, illegal nuclear material transport may be the main fields of interest in the future applications.

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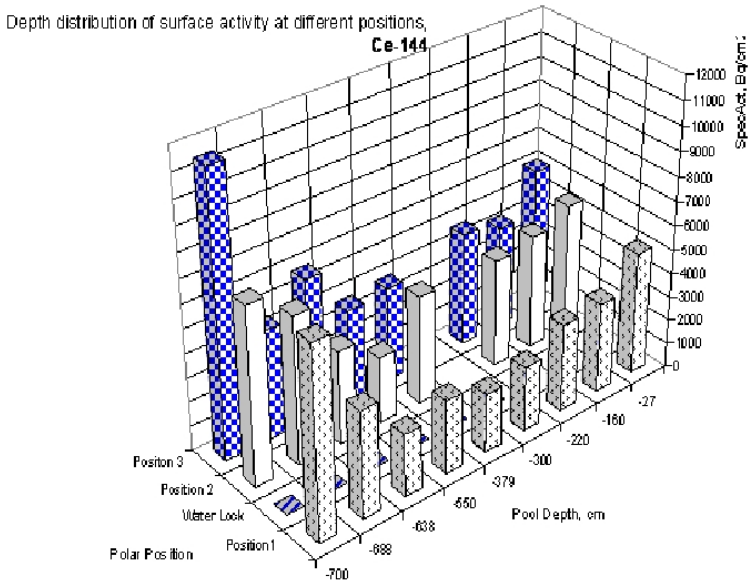

Figure 3: Spatial distribution of the ^{144}Ce specific activity in the technical pool containing the cleaning tank with broken fuel assemblies Under-water measurements as a function of depth and angle.

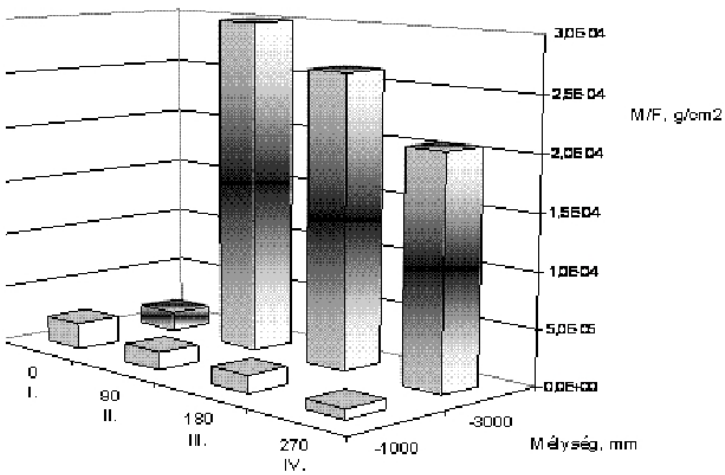

Figure 4: Uranium distribution in the technical pool after the removal of the broken fuel. Measurements in two depths (-1000 and -3000 mm) and four polar angles (0° - 360°).

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