

**EXAMINATION OF HOT PARTICLES**  
**PhD theses paper**

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## **Abstract**

Micro- and radioanalytical methods were developed for the examination of hot particles originating from the primary circuit of NPP Paks.

Fuel fragments originating from the primary water and from incidentally contaminated water were examined by autoradiography, SEM-EDX and by alpha-spectrometry. According to the measured transuranium ratios the burnup of these particles was estimated. The examination of individual hot particles collected from the primary water enables a more effective investigation of the fuel failures. The study of particles from incidentally damaged fuels shows that high burnup particles from the "rim" layer were also released.

The methods were adopted for examination of hot particles originating from environmental samples.

## **Background of the research**

In nuclear incidents (like power plant accidents, weapon tests) a significant part of the released radioactivity appears bound to material particles having inhomogeneous distribution. These so called hot particles carry information characteristic to the source of their release. From the aspect of the hot particles propagation and dosimetical effect the radiochemical and elemental composition, the size and morphology of the particle is also decisive.

Most of the papers available in the literature deal with the examination and characterization of environmental hot particles, even though in several cases hot particles can be found in samples from power plants as well. Hot particles found in samples from power plants also carry valuable information on their source of release.

Regular examinations on the state of the fuel elements of NPP Paks have started at the Institute of Nuclear Techniques of the Budapest University of Technology and Economics years ago. The state of the fuel elements is studied by analyzing samples obtained from the primary coolant of NPP Paks. The increase of the activity concentration of fission products and transuranium isotopes implies fuel failures. By the analytical examination of the samples using gamma- beta- and alpha spectroscopy and by measuring the activities of iodine, cesium, strontium and transuranium isotopes the number, average burnup, and size of the failures (micro- or macro failure) of the failed fuel elements can be estimated. From fuel elements having macro failures not only volatile fission products, but also micro fuel element fragments can be released into the coolant. By finding and analyzing these fuel element fragments the presence of macro-failure in the power plant can be detected. The characterization and microanalytical examination of these particles from the primary coolant makes it possible to the examination and identification of several failed fuel elements simultaneously. From the isotope-composition and the activity ratios of certain nuclides in the individual particles the physical attributes of the failed fuel element (e.g. burnup) can be estimated, what can be important for the localization and identification of the fuel element in the reactor core.

Unfortunately, examining failed fuel elements might not only be necessary during normal operating conditions. In April 2003 at Unit 2 of NPP Paks a serious incident (level 3 according to the INES scale) took place. The incident happened during a fuel assembly cleaning process resulting in the contamination of cca. 800m<sup>3</sup> of water. Due to a design error the cooling was insufficient during the cleaning process, and thermal shock caused by the subsequent flooding damaged the fuel assemblies, contaminating auxiliary pit, as well as the connected spent fuel storage pool and the reactor pond of Unit 2. In addition to the volatile

fission products ( $^{85}\text{Kr}$ ,  $^{131}\text{I}$ ) non-volatile fission products ( $^{141}\text{Ce}$ ,  $^{144}\text{Ce}$ ,  $^{140}\text{Ba}$ ,  $^{140}\text{La}$ ) and uranium and transuranium isotopes were also released into the coolant. A part of this released activity was in soluble, the other part in particle form.

## **Goals**

My aim was to develop a complex micro- and radioanalytical procedure suitable for examining individual (hot) particles separated from the primary coolant of the units of NPP Paks. I wanted to identify the high-activity particles found in the primary coolant so that after separation I could examine the morphology, elemental and radiochemical composition of the individual particles. I especially focused on the radiochemical and alpha-spectrometric examinations, since I wanted to determine the burnup of the individual particles by measuring the ratios of transuranium isotopes found in the particles. My main aim with the examination of the samples obtained during normal operating conditions (reactor shutdown) was the examination of the possible presence of failed fuel elements in the reactor core.

A further goal was to demonstrate that the complex procedure developed is suitable also for studying hot particles obtained from other sources (environmental samples) as well, so during my work I also wanted to analyze environmental samples if possible.

Because of the April 2003 incident I have adopted the developed particle analyzing procedure not only for hot particles found in environmental samples, but also for particles released in the incident. By characterizing these particles and estimating their burnup I wished to collect information on the fuel elements damaged in the storage pool.

## **Examination methods**

I have developed a procedure for the film autoradiographic inspection of the particles, their localization and their separation for subsequent micro- and radioanalytical examinations. I have thoroughly tested the applicability of a photographic film (Forteapan 100, a black and white film previously not used for autoradiography) for autoradiography and performed calibration measurements. The cooperation with KFKI AEKI enabled the study of clusters of particles by scanning electron microscopy, the examination of the size and morphology of the particles and the determination of their elemental composition (the amount of corrosion products)<sup>1</sup>. The non-destructive examinations were followed by the radioanalytical analysis. I have developed or adapted a method for the alpha-spectrometric examination of individual particles suitable for determining the activity of Pu, Am, Cm, and U isotopes after a radiochemical separation procedure. The results of the alpha-spectrometry had a crucial role, since the activity ratios of the transuranium isotopes are related to the burnup of the fuel elements.

I have successfully applied the developed methods for examining particles obtained by filtering the primary coolant of Unit 3 of NPP Paks and for hot particles found in samples contaminated because of the 2003 incident. I have studied the homogeneity of the distribution of the particles, the change of the activity of solutions and filters, and by determining the burnup of the individual particles I made deductions on the dissolution of the particles.

I have demonstrated that using the methods for analyzing samples from power plants relatively low-activity hot particles found in environmental samples can also be examined and valuable, new information can be obtained even today about hot particles released during the

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<sup>1</sup> I especially thank Pintérmé Csordás Tóth Annának (KFKI AERI) for the scanning electroscopy examinations.

Chernobyl accident, or even about rocks containing high natural radioactivity (e.g. monasite sand) that can be found in certain areas of the environment.

### **New scientific results**

My new results related to my PhD work are summarized in the following theses points:

1. I have developed a complex micro- and radioanalytical procedure for the comprehensive analysis of radioactive particles, that after an optional preliminary separation encompasses the secure fixation of hot particles; the localization of particles (or clusters of particles) using film autoradiography; separation of the area of the samples containing particles, as well as non-destructive (using scanning electron microscopy, direct alpha- and gamma-measurements) and destructive (alpha-spectrometry) measurements. [1], [2], [3], [5], [6], [10], [12], [13], [14]
2. I have developed a reliable, reproducible film autography method for determining the spatial distribution of individual radioactive particles. I have solved the reliable fixation of the particles. During the calibration, I have selected the most suitable film for the autoradiography based on sensitivity measurements and determined its sensitivity to the alpha- and beta-radiation. For the evaluation of the autoradiographic images I have developed an image processing method capable of measuring the blackening of the film, as well as the classification and enumeration of the particles based on their size. [2], [6]
3. Using the above methods, I have examined individual hot particles and particle clusters separated from the primary coolant during normal operating conditions for the more efficient inspection of fuel-failures. I have demonstrated that such samples contain both corrosion elements and transuranium isotopes. I have shown that based on the activity ratios of transuranium isotopes determined by alpha-spectrometry the burnup of the individual hot particles can be estimated. I have demonstrated that the burnups of the particles are different, they differ from both the values estimated from the activity measurements of water samples, both the theoretically calculated burnup values. I have proved that the burnup of the particles is related to the local burnup of the failed fuel elements. [1], [3], [6], [10], [13], [14]
4. I have shown that it is possible to examine hot particles released in the incident at NPP Paks using the methods developed for particles originating from the primary coolant, and have proved that the damaged fuel elements can be characterized based on the examinations. About the particles from the incident I have shown, that:

- The number of particles per unit volume in the water samples related to the incident was higher by 3 magnitudes compared to the samples from normal operating conditions.
- Compared to the samples from normal operation, the particles contained a higher amount of transuranium isotopes, the ratio of corrosion particles was lower, and the activity of Cm isotopes in the particles was exceptionally high. The exceptionally high Cm activities measured in the particles can be explained with the preferred sorption of the Cm.
- The burnup of the particles from the incident in many cases significantly exceeded both the burnup values estimated from water sample measurements and the values expected from theoretical calculations. From these results I have concluded that the particles with outstandingly high burnup values most probably originated from the exceptionally high-burnup rim-layers of the fuel pellets. [1], [3], [7], [9], [10], [11], [12], [13], [14]

5. I have proved that the developed methods are suitable for analyzing not only hot particles originating from power plants, but also particles found in environmental samples. I have shown that antropogenous contamination in particle form can be found also in environmental samples having low average activity. The origin of these particles can be estimated based on the release values of the Chernobyl accident. Using the complex method I have shown that the monazite sand containing natural activity in particle form, the high Th content is coupled with high U content. [2], [4], [5], [8]

### **Utilization of results**

There are many ways to utilize and improve the results, further examination methods could be adapted to examine the particles, the methods currently used could be improved, or they can be adapted to examine other types of particles. The results of the particle analysis can be used as input data for dosimetric calculations, utilized in the evaluation of the general condition of fuel elements, or the study of the 2003 incident. By improving certain examination methods the corrosion procedures of the structure elements in the reactor core could also be studied.

Using the developed particle identification and separation method samples can be made also for other microanalytical measurements, for example synchrotron-beam ( $\mu$ -XANES) analysis for determining the oxidation state of elements (Pu, U) found in the particles. The examinations of a few samples obtained from the incident have already demonstrated the combined applicability of these methods<sup>2</sup>.

The developed autoradiographic method for particle separation could be made more accurate by using a micro-manipulator particle separation system. This would also make the possible synchrotron-beam examinations more economic.

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<sup>2</sup> I especially thank Dr. Szabina Török (KFKI AERI) for the synchrotron-beam examinations.

## **Scientific publications related to the theses points**

### **Journal papers:**

[1] Kerkápoly, N. Vajda, T. Pintér, A.Pintér Csordás: Analysis of Hot Particles Originating from Failed and Damaged Fuels, *Central European Journal of Chemistry* 3 (1), 2005, pp. 106-117

[2] Kerkápoly, N. Vajda, T. Pintér: Film Autoradiography Used for Hot Particle Identification, *Journal of Radioanalytical and Nuclear Chemistry*- accepted

[3] Anikó Kerkápoly, Nóra Vajda, Tamás Pintér: Examination of Hot Particles Originating from Damaged Fuels at Nuclear Power Plant Paks, *International Journal of Nuclear Energy Science and Technology* - accepted

[4] Kerkápoly Anikó, Vajda Nóra, Zagyvai Péter: A csernobili atomerőmű-baleset következtében szennyezett környezeti minták, *Fizikai Szemle*, 2001. augusztus

[5] S. Török, J. Osán, L. Vincze, B. Alföldy, A. Kerkápoly, N. Vajda, C.A. Pérez and G. Falkenberg: Comparison of nuclear and X-ray techniques for actinide analysis of environmental hot particles, *Journal of Analytical Atomic Spectrometry*, 2003, 18, 1202-1209.

### **Conference papers published in full:**

[6] Kerkápoly, N. Vajda, A. Csordás, Z. Hózer, T. Pintér: Fuel failures at Paks NPP - Fuel failure in water reactors: Causes and mitigation, *Proceedings of Technical Meeting held in Bratislava, Slovakia, 17-21 June 2002*

[7] Vajda, Nóra; Molnár, Zsuzsa; Kerkápoly, Anikó; Pintér, Tamás: Radiochemical Control of Damaged Fuel After the Cleaning Tank Incident at Paks Nuclear Power Plant; *Proceedings of Int. Conference on Water Chemistry of Nuclear Power Plants, San Francisco, (11-14 Oct. 2004), CD-rom*

[8] Török Sz., Osán J., Alsecz A., Vincze L., Kerkápoly A., Vajda N.: Környezeti forró részecskék elemzése röntgen-mikroanalitikai módszerekkel - Nukleáris Technikai Szimpózium, 2002.okt.3-4., Budapest (CD-rom)

[9] Vajda N., Molnár Zs., Kerkápoly A., Pintér T., Zagyvai P.: Az üzemanyag szivárgása a paksi tisztítótartályban megsérült fűtőelemekből – II. Nukleáris Technikai Szimpózium, 2003.dec.4-5., Budapest

[10] A. Kerkápoly, Zs. Molnár, D. Tar, N. Vajda, , A.Pintér Csordás, T. Pintér, É. Komlósi: Analysis of Hot Particles Originating from the Primary Circuit of NPP Paks, 6<sup>th</sup> International Seminar on Primary and Secondary Water Chemistry of Nuclear Power Plants, Budapest, 16-19 May, 2005

[11] N. Vajda, Zs. Molnár, A. Kerkápoly, T. Pintér\*: Contamination of the Cooling Circuit due to the Cleaning Tank Incident at NPP Paks, 6<sup>th</sup> International Seminar on Primary and Secondary Water Chemistry of Nuclear Power Plants, Budapest, 16-19 May, 2005

**Conferences (abstracts):**

[12] A. Pintér Csordás, N. Vajda, A. Kerkápoly, T. Pintér: SEM and EDX studies of "hot particles" originated from Paks Nuclear Power Plant, Hungary, Proceedings Vol. 2. Materials Science, University of Antwerpen, Belgium, (August 22-27 2004), pp. 739-740.

**Presentations:**

[13] N. Vajda, A. Kerkápoly, A. Pintér Csordás:  
Examinations of hot particles originating from the primary coolant of a nuclear reactor and from several other sources- Country report, "Radiochemical, Chemical and Physical Characterization of Radioactive Particles in the Environment", Trieszt, (2004)

**Others:**

[14] Kerkápoly A.: Examinations of hot particles originating from the primary coolant of a nuclear reactor and from several other sources, Almanach papers of the Fermi price winners (MNT), Budapest (2005) – accepted