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EXPERIMENTAL VERIFICATION OF NEUTRON EMISSION METHOD FOR MEASURING OF FISSILE MATERIAL CONTENT IN SPENT FUEL

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Attya A. Abou-Zaid, Krzysztof Pytel: Experimental Verification of Neutron Emission Method for Measuring of Fissile Material Content in Spent Fuel. A non-destructive method of measurement of fissile nuclides content remained in spent fuel from research reactor is presented. The method, called the neutron emission one, is based on counting of fission neutrons emitted from fissile isotopes : ^{235}U, ^{239}Pu and ^{241}Pu. Fissions are induced mainly by neutrons supplied by the external neutron source. Another effects contribute also to the measured neutron population, e.g. source neutrons penetrating the fuel without being captured and scattered, neutrons from (α,n) reactions and from spontaneous fissions of actinides. Complexity of phenomena occurring within the measurement facility required the detailed numerical simulation and experimental studies prior design of ultimate measurement stand. In the previous paper [1], the results of Monte Carlo simulation on optimisation of measuring stand for neutron emission method were presented. On the basis of those results, the experimental stand for MARIA [2] reactor fuel investigation has been designed and manufactured. The present paper, being the continuation of previous one, contains the description of experimental facility and the results of measurements for the fresh fuel (without burn-up) and the fuel mock-up (without fissile materials). Although some discrepancies were found between Monte Carlo and experimental results, the main conclusions concerning the optimal geometry of measuring facility have been confirmed.

Attya A. Abou-Zaid, Krzysztof Pytel: Eksperymentalna weryfikacja metody mnożenia neutronów do pomiarów zawartości materiałów rozszczepialnych w wypalonym paliwie jądrowym. W pracy przedstawiono wyniki wstępnych pomiarów zawartości izotopów rozszczepialnych w paliwie z reaktora badawczego. Zastosowano metodę emisji neutronów, która polega na rejestrowaniu neutronów emitowanych w wyniku rozszczepień izotopów ^{235}U, ^{239}Pu oraz ^{241}Pu, znajdujących się w paliwie jądrowym. Rozszczepienia te są indukowane neutronami z zewnętrznego źródła. Neutrony rejestrowane w układzie pomiarowym pochodzą nie tylko z rozszczepień lecz również bezpośrednio ze źródła, z reakcji (α,n) bądź z rozszczepień samo-rzutnych. Ze względu na złożoną geometrię układu pomiarowego, przed zaprojektowaniem jego ostatecznej wersji przeprowadzono symulacje komputerowe za pomocą metody Monte Carlo [1], a następnie dokonano eksperymentalnej weryfikacji metody na specjalnie przygotowanym zestawie pomiarowym. W pracy opisano zestaw pomiarowy oraz wyniki pomiarów dla paliwa świeżego (bez wypalenia) i dla makiety elementu paliwowego (bez paliwa). Wyniki pomiarów są na ogół w dobrej zgodności z symulacjami Monte Carlo. Mimo niewielkich różnic, wnioski dotyczące optymalnej geometrii pomiaru, wyprowadzone z symulacji, zostały potwierdzone doświadczalnie.

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1. INTRODUCTION

The Neutron Emission Method (NEM) was already applied to estimate the burn-up of the assemblies of spent fuel rods from nuclear power plants [3,4] as a natural extension of subcritical experiments. Also examination of vertical distribution of fissile materials along the fresh fuel rod was done by means of similar technique [5].

The aim of the method presented in this paper is to determine the fissile isotopes content along the vertical axis of individual spent fuel element. Neutrons emitted from external neutron source are focused on certain slice of fuel element and induce fission in it. Fission rate is proportional to the content of fissile isotopes (^{235}U, ^{239}Pu and ^{241}Pu) within the exposed fuel slice. Fast neutrons originating from fissions are next detected and constitute the output signal from the facility.

Fission neutrons are not the only neutrons contributing to the output signal. The second main component comes from source neutrons penetrating directly the fuel rod without being captured or scattered. The dependence of this component on the fissile materials content is very weak and rather reciprocal than proportional.

There are also another neutron sources that may affect the output signal from neutron detector. These are so called inherent neutron sources i.e. from spontaneous fissions and (α,n) reactions (the last in case of oxide fuels).

Thus, the total count rate of the neutron detector, C_{tot} is equal:

$$C_{tot} = C_{fis} + C_{dir} + C_{inh} \tag{1}$$

where respective count rates are due to:

C_{fis} - induced fissions of fissile materials,

C_{dir} - neutrons coming directly from the neutron source,

C_{inh} - inherent neutron emission.

Among the components of the signal (1) the only C_{dir} is not specific for fissile materials content in the fuel. This value should be subtracted from the total signal C_{tot} to obtain the net signal proportional to the amount of fissile isotopes within investigated fuel.

2. DESCRIPTION OF THE METHOD

The principle of the method is shown in Fig.1. The measuring facility is submerged under water to provide shielding against intensive gamma radiation emitted from the spent fuel. As a neutron source the Pu-Be type one has been. Neutrons escaping the source have rather high energies (of order MeV) and before directing to the fuel should undergo moderation. This is realised by the moderating container having cylindrical shape and filled with water playing a role of moderating medium. Fission cross sections of fissile isotopes have distinctly higher values for thermal neutrons than for epithermal ones.

The neutron source is positioned in a centre of moderating container lined by cadmium sheet. Majority of neutrons reaching container boundary have very low (thermal) energies and they are fully captured by cadmium. The only window, where the neutron may escape from the moderator is the collimator. The collimator is an empty rectangular box and has all the walls shielded with cadmium plates except inlet and outlet windows. The collimator is inserted to the moderating container.

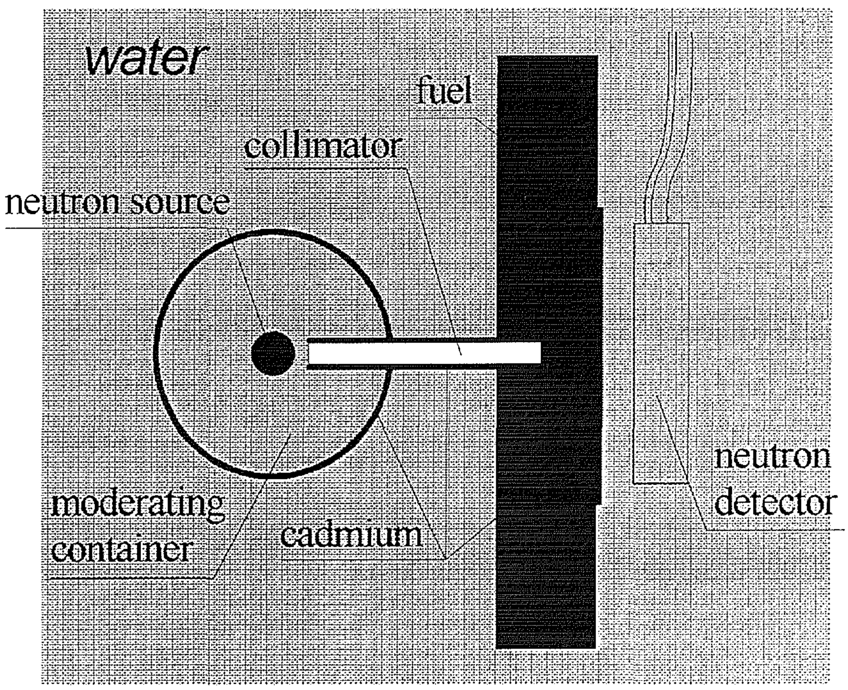

Fig.1 Schematic diagram of the measuring equipment

Distance between the neutron source and inlet window of the collimator has to be chosen to achieve the maximum outward thermal neutron current in the collimator.

Neutrons escape the collimator through the outlet window and intersect the fuel element inducing fissions of fissile nuclei. Fast neutrons emitted during fission undergo thermalisation in water and may be detected by fission chamber applied as a neutron detector. The fission chamber is not strictly adjacent to the fuel surface. Certain distance from the fuel surface guarantees proper moderation of fission neutrons in the water.

Additional neutron absorbing screen (cadmium sheet) can be installed beyond the fuel element opposite to the neutron beam. In this case the amount of non-fission neutrons (e.g. scattered or transmitted from the neutron source) is reduced and the ratio of fission neutrons to all detected neutrons is improved.

3. CONSTRUCTION OF EXPERIMENTAL STAND

Mechanical design

The experimental stand [6] consists of the following major components (see Fig. 2):

- moderating tank with dimensions 300 × 320 mm (pos.1)
- collimator 82 ×20 ×260 mm (pos.2)
- neutron source (Pu-Be) container (pos.3)
- moderating tank base (pos.4)
- rails with a socket for placing the fuel element (pos.5)
- detector base (pos.6).

The stand was placed in a tug enabling to pour it with water playing a role of moderator and shield. All the components of the stand are made of the PA2 aluminium alloy. The moderating tank (pos. 1) is covered with cadmium sheet (1 mm thickness) and situated horizontally along the lateral surface on the moderating tank base (pos. 4). The legs of the base are terminated with blocks which enable a shift of moderating tank by means of a special moderating tank travel device (pos. 8) on rails (pos. 5) forming the base of the whole stand. In lateral walls of the moderating tank there are some holes enabling to fill the moderating tank with water when it is being emerged into the tug.

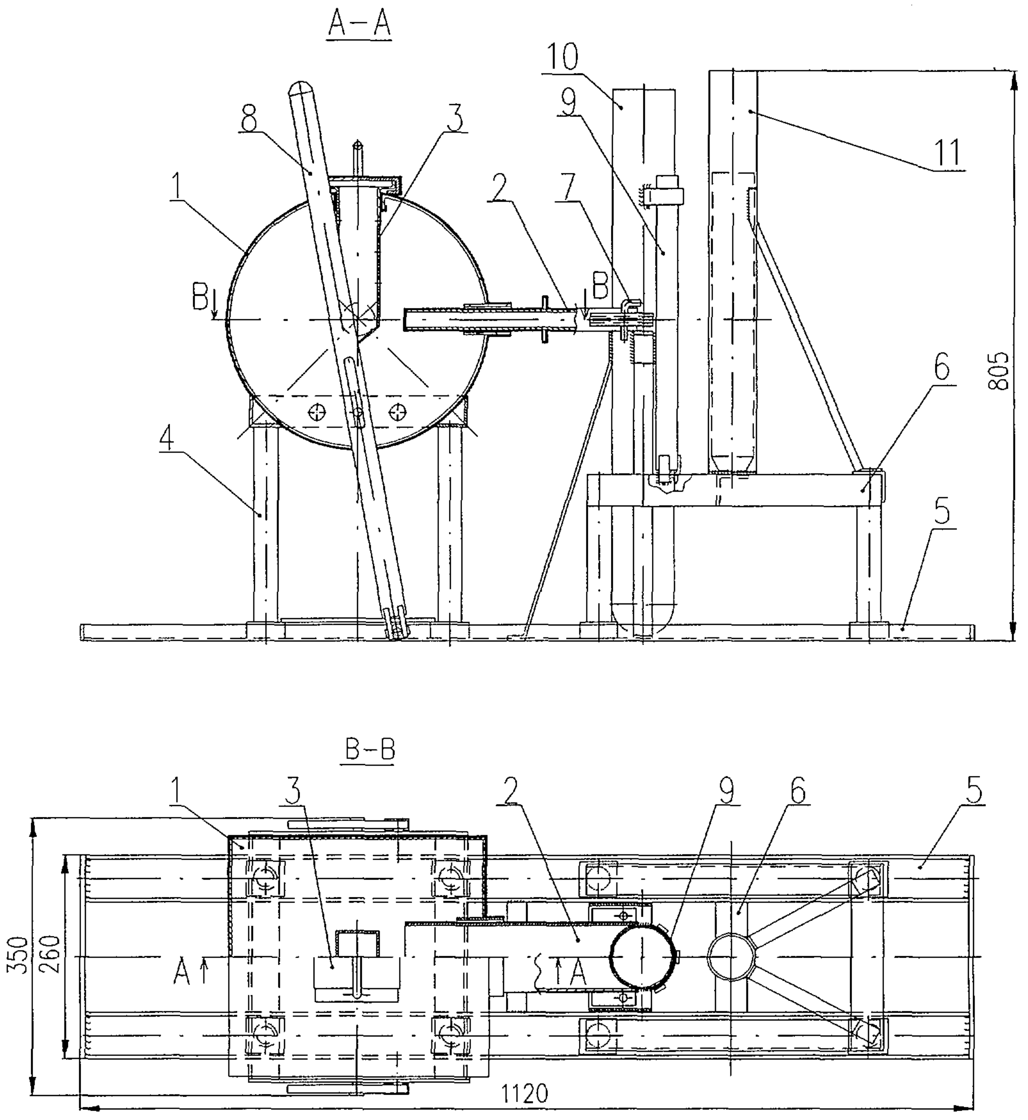

Fig. 2 Mechanical construction of experimental stand.

At the top of the moderating tank there is a rectangular opening (pos. 3) in which is placed a container for Pu-Be neutron source having intensity of $S = 2.07 \cdot 10^7$ n/s (±10%) (pos. 3). This container of a cubicoid shape has a bottom enabling the central positioning of the source. In lateral walls and in the bottom of the neutron source container there are some openings allowing for filling the container with water after inserting it to the moderating tank. Upper walls of the neutron source container are twisted in such a way they form "rails" along which a neutron source container cover can be displaced. The cover is equipped with a lug enabling for opening the container, setting it and removing from the moderating tank. From the top, the container cover is lined by cadmium sheet (1 mm).

The collimator of a rectangular prism box shape (pos 2) with dimensions of 82 × 20 × 260 mm is introduced into the moderating tank through its lateral rectangular hole. Inside the collimator there is no water (moderator) so it must be air tight. All the walls of collimator with exception of the windows are wrapped with the cadmium sheet which is 1 mm thick. One of the collimator windows is of cylindrical shape so it can adhere to the pipe (socket) in which a fuel element is placed (pos. 10). This pipe is immovably fastened to the rails, forming the base for the stand. The collimator is fixed by means of "lug" and a pin (pos. 7) to the socket along with the fuel element, however, it can move in the moderating tank hole. It allows for changing the distance between the source and the fuel element by pushing the moderating tank along the rails. The fuel element socket has a "pocket" into which a cadmium screen can be inserted (pos. 9). This screen absorbs neutrons just after the fuel what could improve the measurement conditions.

The detector base (pos. 6) is set beyond the socket on rails. It is of the form of a tube supported by a structure being able to move towards the socket. The displacement is to be measured by the rule sticked to the rails. It enables choosing an optimal measurement distance. Since the detector applied which is of chamber type of PK 55/RJ 1300/3 KT-2 should not have any contact with water, the detector tube (pos. 11) is air tight and it is sticking out over the water surface in the tug. The efficiency of the applied fission chamber was $\varepsilon = 0.7\,\mathrm{cps/unit\,flux}$.

Description of the neutron flux measuring assembly

Fig.3 below shows the schematic block of the measuring assembly which includes three main parts: fission chamber, pre-amplifier, and measuring channel. The pre-amplifier and measuring channel constitute the so called start up channel. The measuring channel includes main amplifier, discriminator, integrator, low voltage supply, and high voltage supply. In addition to the main three parts, there are also counter and (220/24 V) stabiliser to supply the low voltage supply with 24 volts.

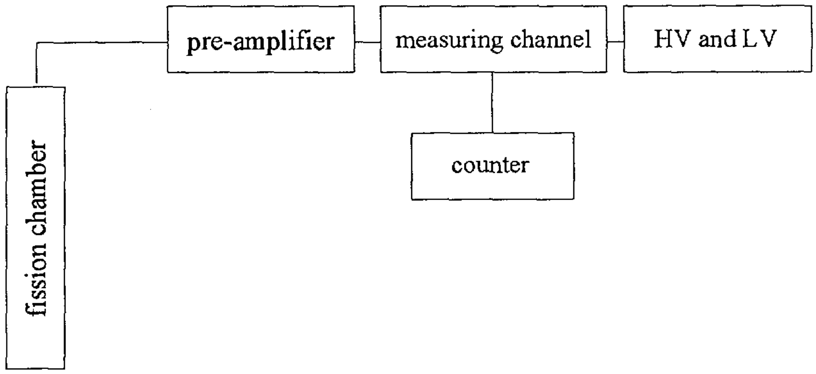

Fig. 3 Schematic block of the measuring assembly

The signal produced in the fission chamber due to thermal neutron is preliminary amplified through the pre-amplifier and next undergo further amplification and shaping by the main amplifier. The output signal of the main amplifier goes to the discriminator unit in which the neutron pulses are separated from that of gamma pulses and noise. Then pulses are directed to the integrator and their intensity is displayed in an analog meter in measuring channel. Simultaneously, pulses are directed to the input of the counter.

4. EXPERIMENTAL RESULTS

Results of measurements were expressed in terms of thermal neutron flux at the surface of neutron detector. Knowing the fission chamber efficiency ε one can calculate the experimental value of thermal neutron flux φ_{exp} corresponding to recorded count rate C_{tot}:

$$\varphi_{exp} = \frac{C_{tot}}{\varepsilon} \tag{2}$$

To make possible comparison between measured and calculated values, the numerical results of Monte Carlo calculations [1] were processed to obtain the neutron fluxes corresponding to the intensity of neutron source applied in the experiment. In the numerical optimisation [1] not the simple fluxes were calculated but so called flux - cross section integrals I_{fc} :

$$I_{fc} = \int_{Area} dA/A \int_{Energy} dE\ \varphi(r,E)\sigma_f(E) \tag{3}$$

where $\varphi(r,E)\,dE\,dA$ is the probability that one neutron emitted from the source intersects the unit area dA located at r and has energy within the range [E,E+dE]. The probability was integrated over the surface and energy with the fission cross section as weighting factor. Such a weighting function makes value of I_{fc} proportional to fission detector response and becomes better optimisation criterion than the neutron flux.

To calculate the neutron flux φ_{cal} having value of flux - cross section integral I_{fc}, the following simple formula was applied:

$$\varphi_{cal} = \frac{I_{fc} \cdot S}{\sigma_f \cdot \eta}$$

where:

$S = 2.07 \cdot 10^7\ n/sec$ - neutron source intensity measured on the source surface,

$\sigma_f = 582\,barn$ - microscopic fission cross section of U-235 for thermal neutrons,

η - probability that a neutron originated within the source volume escape the source surface.

The probability η was introduced to take into account the effect of self - absorption of neutrons in the source volume. Experimental intensity of the neutron source S is defined as a number of neutrons emitted in unit time outwards its surface. On the other hand the probabilities obtained from Monte Carlo calculations refer to one neutron originated within the source. A Monte Carlo simulation of neutron transport within the source volume gave the value of $\eta = 0.766$.

Optimisation of neutron source - collimator distance

To find the optimal distance between the neutron source centre and the collimator, the fission chamber position was fixed and the neutron source was moved starting from the edge of the collimator. There was no fuel and its mock-up in the fuel element socket. The neutron count rate was measured as a function of the source - collimator distance. The neutron flux plotted as a function of the neutron source - collimator distance is shown in Fig. 4 . The results showed that the optimum distance between the neutron source centre and the collimator is equal to 4.2 cm. The maximum value of neutron flux calculated by means of Monte Carlo simulation was found at about 4 cm.

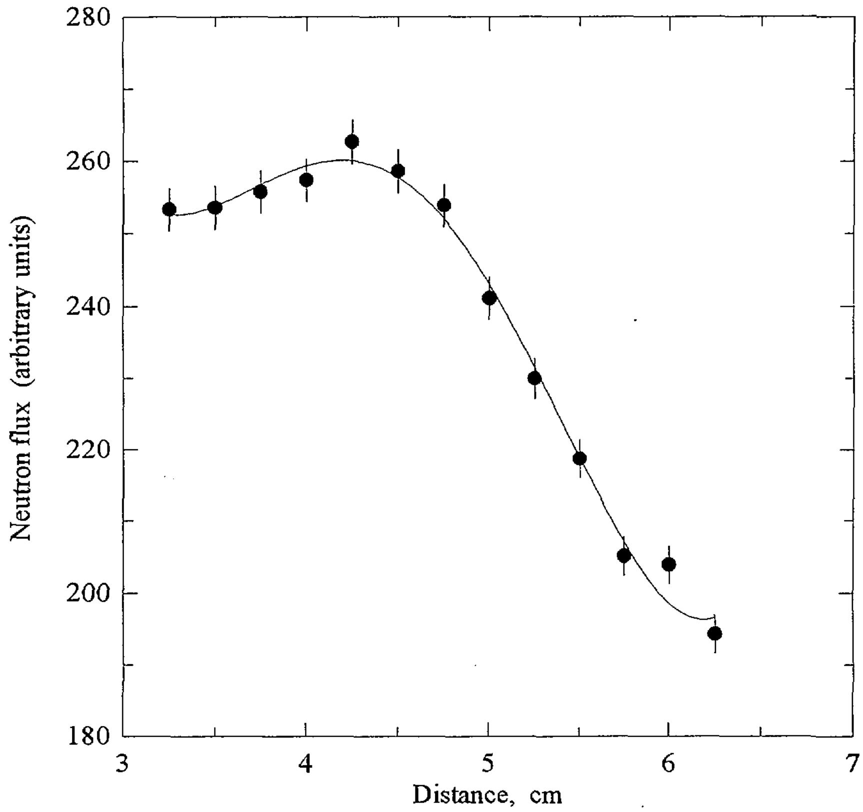

Fig. 4. The neutron flux as a function of the distance between the neutron source and collimator.

Optimisation of fuel element - fission chamber distance

The neutron source has been fixed at an optimal distance 4.2 cm far from the collimator. In this experiment the distance between the fuel element and fission chamber become variable and the neutron count rate was measured as a function of this distance. Four such curves have been collected for:

— fresh fuel element,

— mock-up of the fuel,

— fresh fuel element with cadmium screen,

— mock-up with cadmium screen.

As an optimal criterion the maximum of the difference between the neutron flux for fresh fuel φ_{fuel} and the same flux for mock-up $\varphi_{mock-up}$ has been applied:

$$\Delta\varphi(d) = \varphi_{fuel}(d) - \varphi_{mock-up}(d)$$

where d denotes the distance between fuel element surface and fission chamber. Two curves $\Delta\varphi(d)$ respectively for bare fuel socket and shielded by cadmium are shown in Fig. 5.

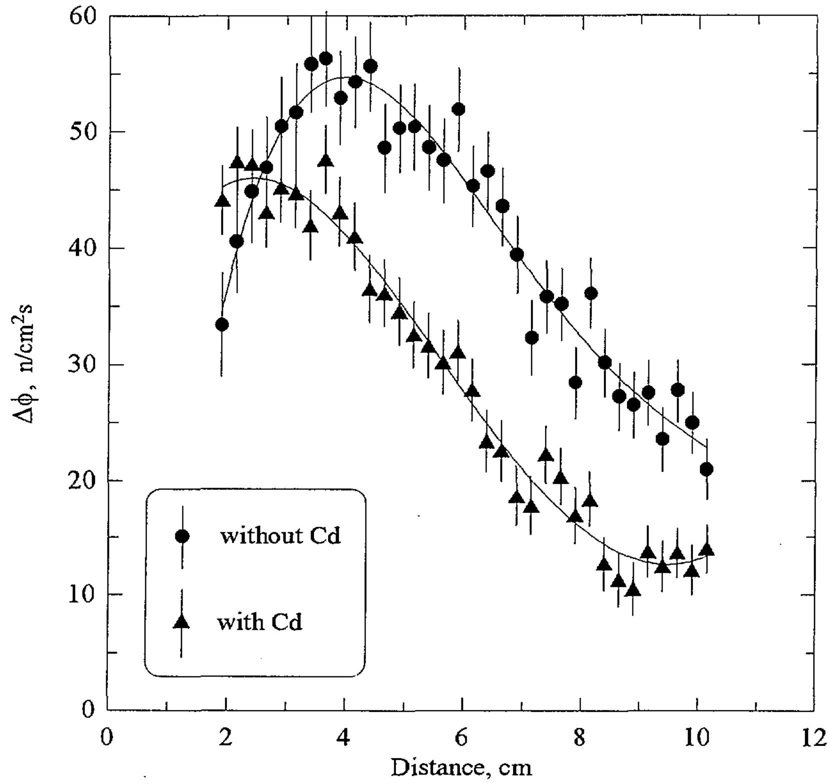

Fig. 5 The difference of the neutron flux for fresh fuel and the same value for its mock-up as a function of the distance from the fuel rod surface.

Optimum distances are obviously different for both cases and equal to 4.0 and 2.2 cm. Application of cadmium screen after the fuel socket turned out to be ineffective. The neutron flux difference between the measurement of fuel and its mock-up being in fact the net signal proportional to the fissile isotopes content within the fuel is higher without cadmium filter. In this case the optimum distance between the fuel socket and the neutron detector is equal 4 cm.

Comparison between experimental and numerical results

The experimental and numerical values of the neutron flux are plotted as a function of distance between fuel element and fission chamber (Figs. 6,7,8 and 9) for four cases:

- fresh fuel,
- mock-up,
- fresh fuel with Cd screen,
- mock-up with Cd screen.

The Monte Carlo results have been plotted together with the error in a fitted curves and the experimental results were plotted as a points with an error bars. The solid line in all the figures represents the MC values of the flux while the dashed lines above and under the solid one represents the respective calculation error.

The results shows a good agreement between the two methods although some discrepancies have been found. The source of these discrepancies comes from the simplification of the geometry simulated by Monte Carlo code i.e in MC calculations all the support parts e.g. fuel element base, fission chamber base, neutron source container were not simulated.

Another source of discrepancies is that the fission chamber was simulated in Monte Carlo code as a point detector whilst the real detector has finite volume and more complicated geometry.

In general one can conclude that MC and experimental results coincide better for the fresh fuel than for its mock-up.

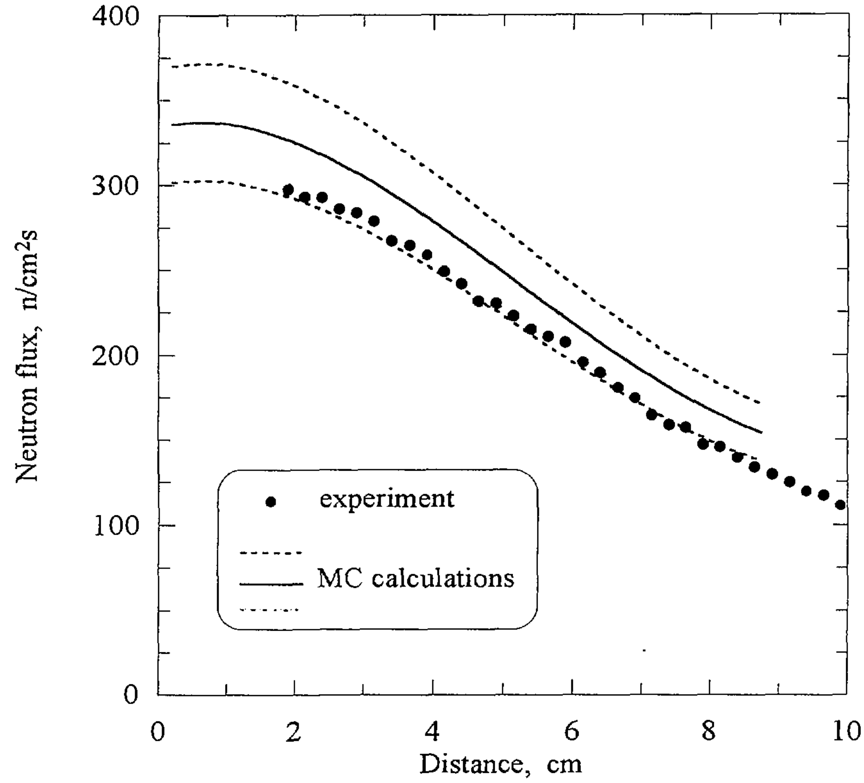

Fig.6. The comparison of MC and experiment for the fresh fuel

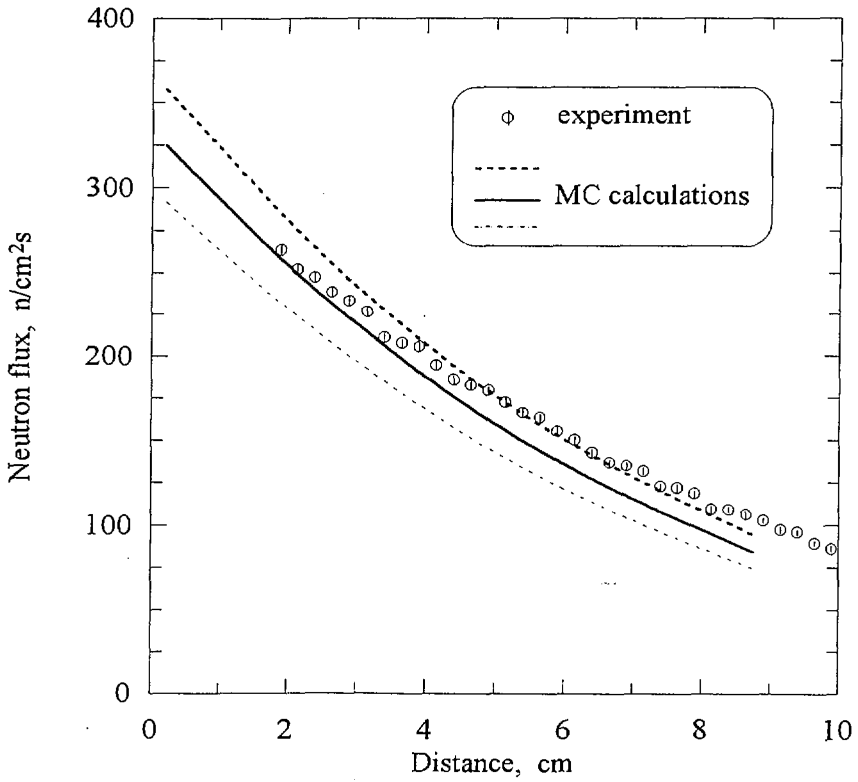

Fig. 7 The comparison of MC and experiment for the mock-up

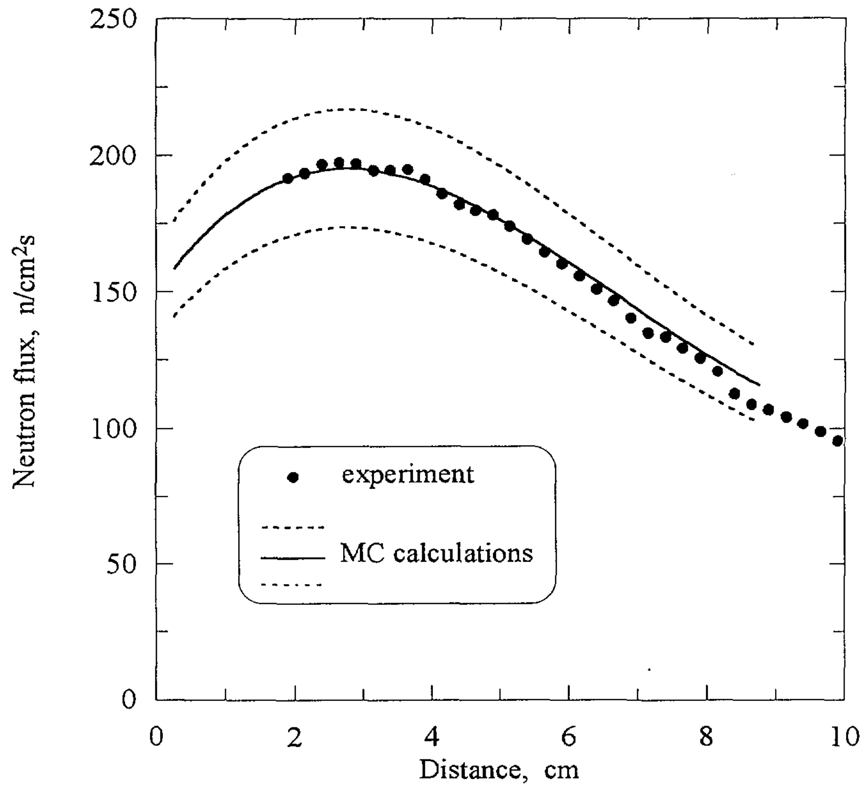

Fig. 8 The comparison of MC and experiment for the fresh fuel and Cd. screen

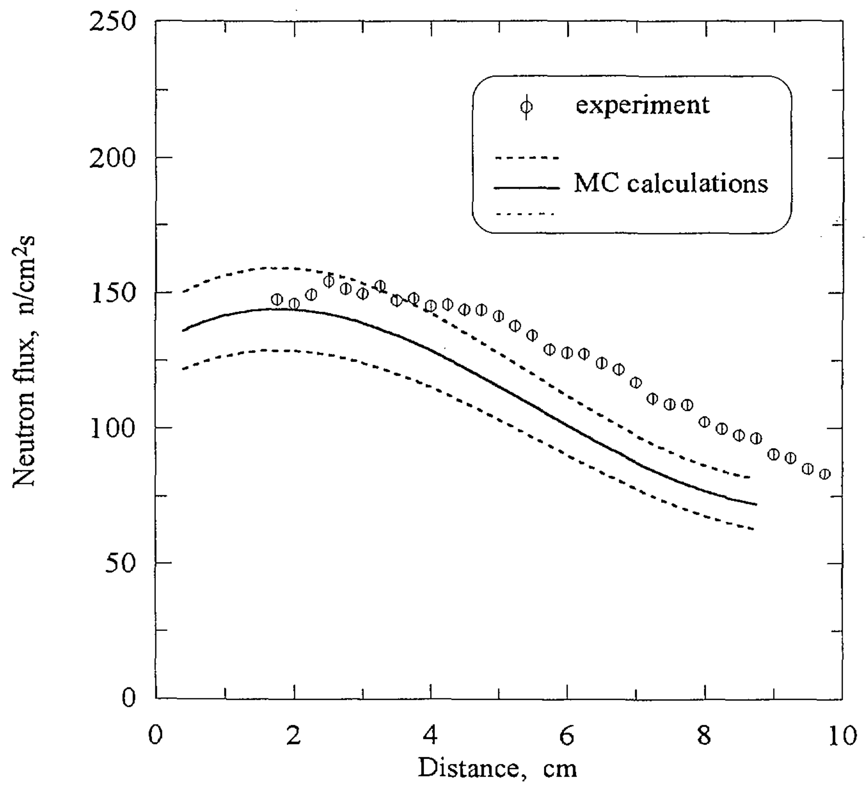

Fig.9. The comparison of MC and experiment for the mock-up and Cd. screen

5. CONCLUSIONS

An experimental stand for measuring the fissile nuclides remained in MARIA spent fuel element has been designed and the measurements for the fresh fuel and its mock-up were performed. It has been confirmed that the measuring signal is dominated by two components: counts due to fission neutrons and neutrons directly transmitted from the source (reported as background). The only first component is proportional to the fissile material content and the background (measured with the mock-up) has to be subtracted from the total signal.

The geometry of the measuring stand was optimised experimentally as well as numerically by Monte Carlo calculations. The measured results showed a good agreement with calculated ones. Thus, it is justified to use Monte Carlo simulations for further development of the neutron emission method. The next important step in application of the neutron emission method is verification of an assumption that the measured neutron flux depends linearly on the spent fuel burn-up.

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