Neutron Monitoring Systems for the Characterisation of Nuclear Fuel and Waste - Methodology and Applications - 12055

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ABSTRACT

The most characteristic behaviour of nuclear fuel or waste contaminated by fission material or isotopes resulting from fissile processes is the emission of neutrons. At the same time because of the high penetration of the material by neutrons, they are an ideal probe for measurement by non-destructive assay.

The detection and data analysis in this case is quite different compared to methods using gamma measuring techniques.

Neutron detection monitors have been in routine operation for a long time, showing their excellent detection capabilities.

INTRODUCTION

During the operation of nuclear power plants nuclear fuel is burned up. At the same time waste is generated which is contaminated by fuel and its fissile products. In general the isotope vector of the contamination is similar to the vector of the burned up fuel.

In addition to the fission there is also a breeding process which produces isotopes like Pu, Cm or Am. These isotopes are not stable and release neutrons and/or alpha particles. The measurement of these isotopes can be used to check the burn up process of the fuel elements or to measure the contamination of waste. They are of high interest because of the long decay times and the hazard potential of the emitted alpha radiation or the chemical properties of the elements itself.

A direct detection of the isotopes by measurement of the emitted alpha particles is not possible because of the strong absorption in the waste matrix. Therefore the emitted neutrons are measured. Part of the neutrons comes from spontaneous or induced fission processes, the other part from (alpha,n) reaction in case of light nuclei like Oxygen being present. The nuclear fuel is normally made of UO2, therefore neutrons from (alpha,n) processes are present [1], [2].

The difference between neutron measurements of burned up fuel elements and of nuclear waste is that in the case of nuclear fuel elements there is a large quantity of neutrons and also a very strong gamma background. Nuclear waste on the other hand has very small numbers of emitted neutrons and low gamma background. Therefore different measurement devices have to be used including different analysis techniques for the measured data. Common to both measurements is the application of passive counting methods, which enables the construction devices that are not too expensive.

Because neutrons are uncharged particles, they can easily penetrate material, but this also makes them difficult to be detected. In general there are two reactions for the neutron detection:

(1) n + He-3 $\rightarrow$ p + H-3 + 765 keV
(2) n + U-235 $\rightarrow$ induced fission process

Both processes are possible only if the neutrons are first slowed down, i.e. moderated to become thermal neutrons. This is the main challenge for the layout of a detection device.

The selection of the neutron detector depends on the gamma background. In case of low background reaction (1) is used and the neutrons are detected by proportional counter tubes filled with He-3.

If the gamma background is high, fission chambers are used. These devices consist of a tube where the inner tube surface is covered with highly enriched U-235. Inside the tube a counting wire is fixed and the tube works similar to a proportional chamber.

If a neutron induces a fission process in U-235, then the event is triggered by the fission products that are charged. Gammas are not able to trigger fission processes and in addition the gammas coming from outside are shielded by the U-235 cover at the wall.

Because fission chambers can contain up to 100g highly enriched U-235, a lot of administrative acts are necessary to receive the admittance to buy, transport and operate the chambers.

In the following monitoring systems are described which are specialised for the measurement of burned up fuel elements and for contaminated waste. The applications demonstrate the wide use of neutron counting technique and the modern refinement of the data analysis.

NEUTRON MONITOR FOR FUEL ELEMENTS (FAMOS)

Neutron monitors have been used to measure the burn up of fuel elements for a long period of time. The measurement principle is to measure the emission of neutrons and gammas coming from the fuel elements. In contrast to the gamma emission, which is roughly proportional to the burn up, the neutron emission changes with the square root of the burn up.

FAMOS monitor consists of a fuel element guidance equipped with two neutron counters and two gamma counters (Geiger Mueller tubes). The detectors are arranged to measure the fuel element from different sides to compensate the inhomogeneity of burned up fuel.

The Geiger Mueller tubes are shielded by a led housing against background gammas from other fuel elements or by reflecting from the walls. The neutron counter consists of fission chambers, which are mounted inside of housings made of plastic material with a high concentration of Hydrogen. The complete measurement system is designed to be water resistant to be able to operate up to 10 m below the water surface in the fuel element storage tank. The detectors are connected with the detector electronic which must be placed outside the water tank to avoid degradation by the intensive neutron and gamma radiation. For the construction special care has been taken on the selection of materials to withstand the high radiation of the burned up fuel element especially if the burn up is 40GWd/MTIHM or higher. The measurement device is shown in Fig.1. The monitor is mounted in the fuel element storage pond and the fuel elements are moved through the central opening of the monitor by the fuel element loading machine. The position of the neutron monitor relative to the fuel element is determined from the loading machine.

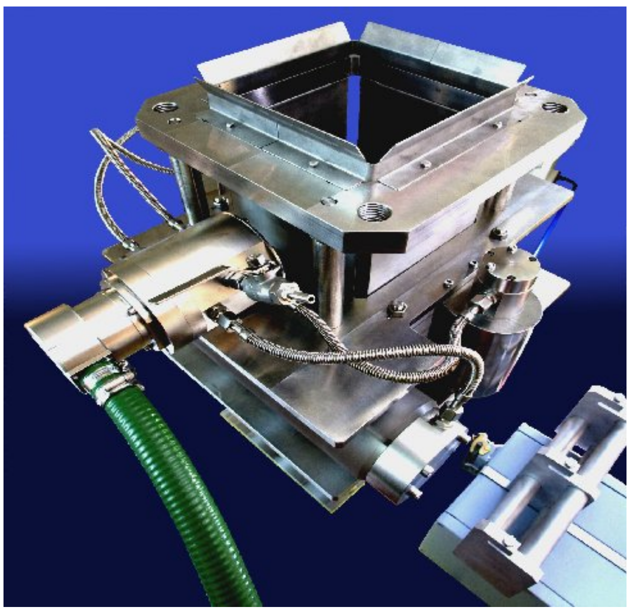

Fig. 1 Neutron Monitor FAMOS

The relation between neutron emission and burn up – as stated above – is only very roughly described by a square root function [3]. In reality it depends from many factors like reactor type, fuel type and enrichment, operation power, duration of burn up and cooling cycles etc. The main purpose of measurements is the determination of the mean burn up value of the fuel elements because this is an important parameter for the transport and storage of the fuel elements. To obtain precise results, burn up programs like ORIGIN or KORIGIN are used and the input parameters according to the data as described in the fuel element card are selected. With the program the gamma and neutron emission for the actual date of the measurement are calculated and compared with the measured count rates.

The results of the analysis depend from the quality of the burn up codes. If a longer cooling time between the last burn up cycle and the measurement is present, than smaller differences of the results of the different burn up codes are expected because isotopes with short decay times disappear over the time.

Comparing the measured data with the calculated values requires a careful calibration of the measurement device. Special care has to be taken on the boron acid concentration in the storage pond, because it influences the neutron emission strongly. The influence is measured by experiments using a water tank and a Cf-252 source. The boron concentration was changed in the range up to 2600 ppm. The count rates as a function of boron concentration were fitted by a polynomial function using a Chi^2 fit.
The Fig. 2 and Fig.3 show some exemplary measurement results:

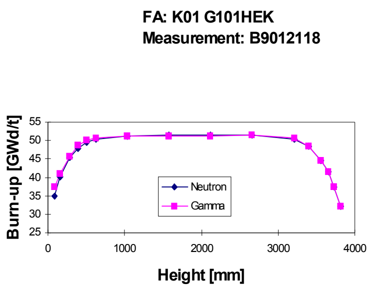

Fig. 2 Axial burn up measurement at a fuel element

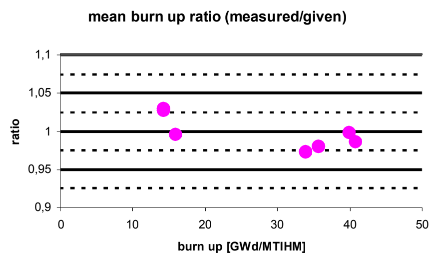

Fig. 3 Measured mean burn up values compared with values from the fuel element data card

NEUTRON MONITOR FOR WASTE (FEMOS)

Waste which contains alpha emitter and/or material with spontaneous fission also emits neutrons. Compared with the fuel elements the neutron emission rate is very low and the gamma background is for most cases in the order up to 1Sv/h. Therefore He-3 filled proportional counter tubes can be used to detect neutrons. If the gamma background is high (i.e. above 1Sv/h), then the sensitivity of the neutron tubes for gammas can be reduced using boron lined tubes. Because of the low emission rate of the neutrons, a careful design of the moderator is required to optimise the detection efficiency and to minimise the loss of neutrons in the moderator or by escaping the measurement arrangement. To avoid the latter, the waste drum must be enclosed by the moderator.

The monitor FEMOS was constructed to measure waste drums with up to 600 l volume [4]. The drum is surrounded by a cylindrical moderator with 46 neutron counter tubes inside. The bottom and the top are also made of moderator material with implemented counter tubes (7 at top and 7 at bottom). The tubes are placed inside the moderator to have an optimal moderation of the neutrons from the waste drum. The moderator material in outside direction has to be thick enough to absorb all neutrons coming from the outside.

In addition to the moderation of the neutron by the moderator, there is also a contribution from the waste matrix which may contain plastics or water. This contribution is not desirable because the influence of the waste matrix on the measurement result should be minimized. The contribution can be reduced by lining the inner side of the moderator with a thin cadmium sheet.

Because of the many parameters that influence the layout the moderator, MCNP [5] calculations have been used to find an optimal arrangement.

The Fig. 4 shows the layout of the FEMOS monitor.

Fig. 4   FEMOS monitor for measuring of waste drums

For a fully automated operation the moderator cylinder can be opened by a pneumatic system and the drum can be transported inside or outside the device using a chain conveyor.

For the detector electronic a special layout was used. Each counting tube has its own high voltage supply, amplifier and window discriminator. The output signals of the discriminators are collected by an LCA board. This board checks every 500 ns the output signals of the discriminators. If there was no signal, then an internal counter is clocked up. Otherwise the status of all 60 counter tubes is stored into an 8 byte structure. In addition the content of the internal counter is stored into 2 additional bytes. All 10 bytes are transferred to the computer memory, the internal counter is cleared and the reading cycles starts after 500 ns again. With the stored data the complete information about the counting events is available. The count rates of each counter tube can be extracted or the correlation of events from different counter tubes can be analysed. With this electronic solution a flexible data analysis can be performed.

The extraction of single count rates allows a quick check of the system. If there are no neutron sources available, it is also possible to activate an internal pulse generator and to feed the pulses into the discriminators of the tubes.

The source distribution inside the matrix can be analysed in different ways: the ratio between the counts of the tubes placed at the bottom part and at the top allows an estimate at which height the main neutron emitters are placed. Comparing the count rates of the tubes placed in the moderator cylinder allows a radial localization of neutron sources. Analysing the correlation between adjacent tubes gives information at which radial distance from the symmetric axis of the drum the main source is placed inside the drum. This information is used to correct absorption effects in the waste matrix. The density of the matrix is calculated from the measured weight of the drum (weighing system is integrated into the bottom plate) and from the volume of the drum. The material of the matrix must be selected by the operator. These parameters are used to select the absorption factor from a data base. The absorption factors were calculated with MCNP beforehand by modelling the complete experimental set up including the drum and its matrix.

From the measured neutron emission rate the activities of the isotopes are calculated. For this step the principal isotopic composition of the neutron sources must be known. For waste coming from nuclear power plants this isotope vector can be assumed to be similar to the isotopic composition of the burned up nuclear fuel. Only isotopes which contribute to the neutron emission rate are of interest, i.e. isotopes which decay by spontaneous fission or by emission of alpha particles. In principle also induced fission effects must be considered. If the content of fissile material is very small, then this effect can be neglected. For the isotope vector the ratio between neutron emission and alpha activity is calculated and the measured neutron emission rate is used to renormalize the vector. With this method it is possible to determine the activity for all isotopes of the vector and to calculate the total amount of fissile material.

The monitor as shown in Fig. 4 is able to detect 10mg Pu in a 200 l drum filled with sand.

NEUTRON MONITOR FOR LEACHED HULLS (CAMOS)

During the reprocessing of burned up fuel elements, it was necessary to determine the remaining fissile materials on the hull segments after the leaching process. For this purpose the monitor CAMOS was developed which measures the neutron multiplication by fissile material. The monitor is operated in an area with extremely high gamma level (up to 60 Sv/h). Therefore fission chambers had to be used for neutron detection. The layout of the monitor is shown in Fig.5.

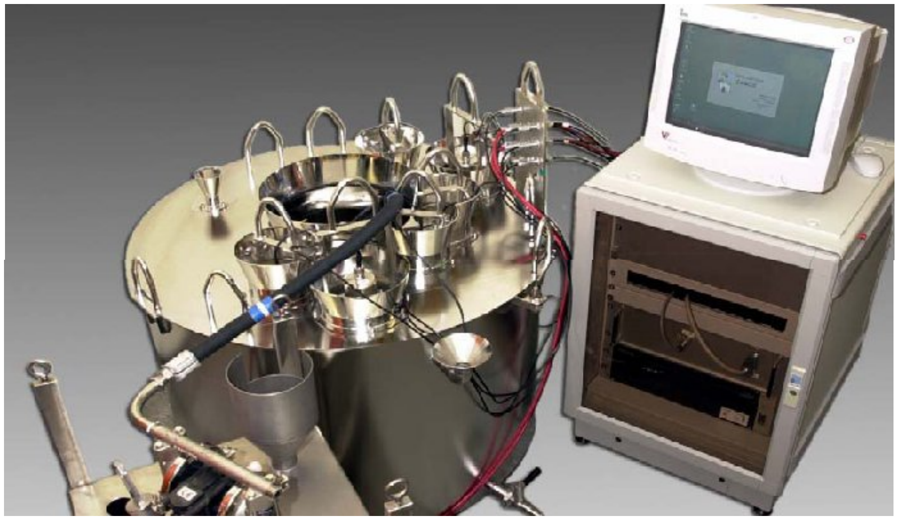

Fig. 5 Layout of the monitor CAMOS

The monitor components are shown as a sketch in Fig.6.

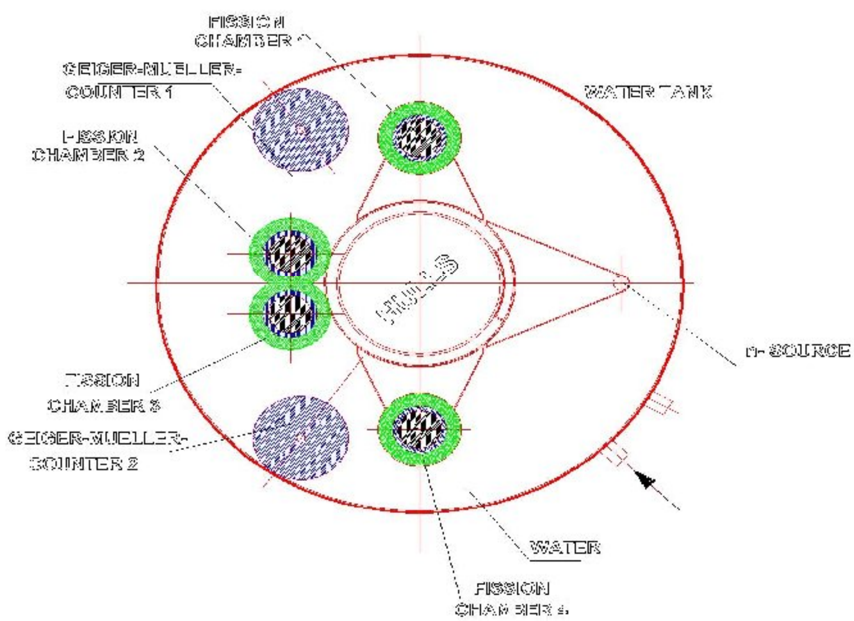

Fig. 6 Sketch of the monitor components for CAMOS

The hulls are lowered with a basket into the water tank through the central opening. A neutron source can be positioned at one side of the monitor. Two fission chambers are placed in forward direction to “see” the neutron source whereas two other chambers are positioned under 90 degree. To check the gamma background level two Geiger-Mueller tubes are placed under 45 degree. By measuring the neutron count rates with and without neutron source, the calculation of the neutron multiplication by induced fission process is possible. With this method the detection of 10mg Pu per litre volume was achieved.

To refine the data analysis the measured neutron multiplication factor is compared with the multiplication factor as calculated with the MCNP code. This method has improved the quality of the results because the matrix effects and characteristics of the experimental arrangement can be taken into account.

CONCLUSION

The neutron monitors designed for different applications have demonstrated their capabilities during daily operation in the field of burned up fuel elements and for nuclear waste with alpha activity. Lately the data analysis was refined and the quality of the results was improved by using MCNP calculations. Last but not least the layout and the calibration of neutron monitors are nowadays unfeasible without support by MCNP simulations.

In the field of non-destructive assay the neutron monitors are undisputed.

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