Nuclear Energy in Central Europe '99
Portorož, Slovenia, 6 to 9 September, 1999

SI0200016

Siemens Argonaut-Reactor Graz: A method to determine the burn up of fuel elements

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Abstract

The burn up of 234 low enriched (20 % U 235) uranium fuel plates of the Siemens Argonaut Reactor Graz has been determined by means of a High Purity Germanium detector and a NaI(Tl) scintillation detector from the activity of Cs 137. The efficiency of the HPGe detector has been corrected using the Monte Carlo program MCNP Version 4B. The gamma dose rate on the surface of the plates was also obtained by experiment.

1 Introduction

The Siemens Argonaut research Reactor (SAR) [1] ring-cylindrical core contains fuel elements, covered by water, and graphite blocks. These serve together with the water as moderator. The inner container is filled up with a graphite cylinder, the outer container is surrounded by a graphite wall. Both graphite parts act as reflector. Around the reactor core the main part of the biological shield is formed by concrete blocks. Fig. 1 shows the sectional drawing of the SAR.

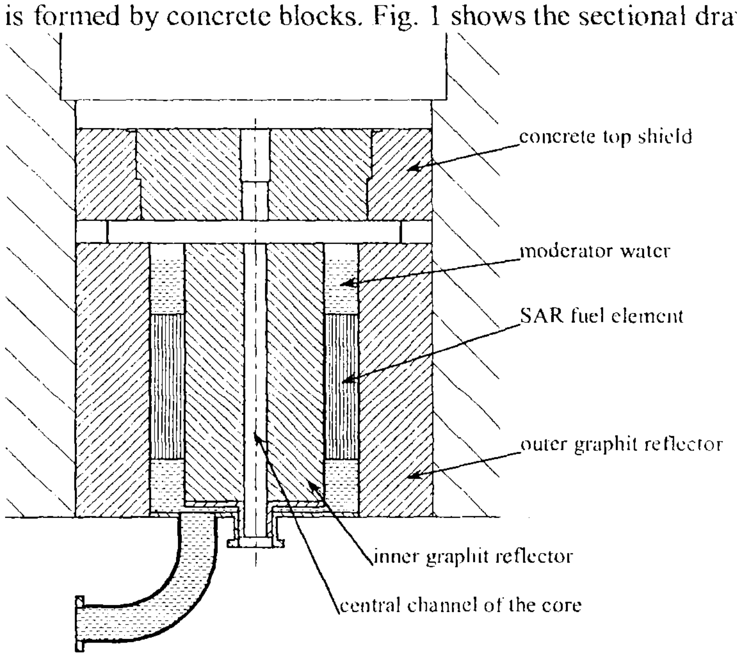

Figure 1. Sectional drawing of the Siemens-Argonaut-Reactor Graz.

From May 1965 to April 1985 the SAR-Graz was driven alternately by an annular core with 234 low enriched (20% U 235) fuel plates and an asymmetrical one-slab loading with 125 high enriched (90% U 235) fuel plates (Fig. 2). Since 1985 all low enriched fuel plates have been located in a dry storage because on 50% of the plates the aluminium cladding was damaged by corrosion. During the reactor operation from 1965-1985 the average reactor power was 1-10 W (10^7-10^8 neutrons/cm² s).

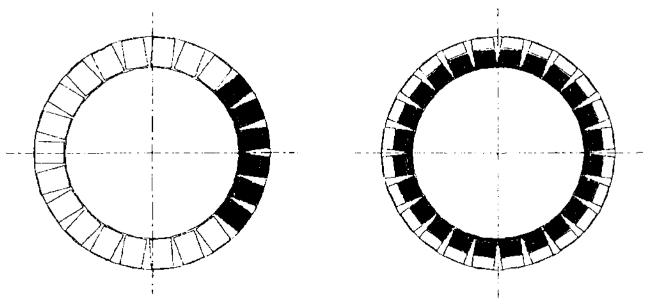

Figure 2. Reactor-core-configurations of the SAR-Graz.

In the coming years the 234 low enriched uranium fuel plates are to be returned to the USA, and therefore it is essential to determine the burn up and the surface-gamma-dose-rate of the fuel plates which will be presented in this work. The long-lived fission product Cs 137 is used for calculation of the burn up and contributes mainly for the dose rate of the fuel plates.

2 Configuration

The fuel matrix of each plate consists of uranium oxide and contains about 124g U_3O_8 with 20.8g U 235. The mean dimensions of the fuel matrix are 600x70x2 mm³ and the total dimensions of each plate are extending to 650x75x3 mm³ which is shown in Fig. 3. The hatched area denotes the fuel matrix.

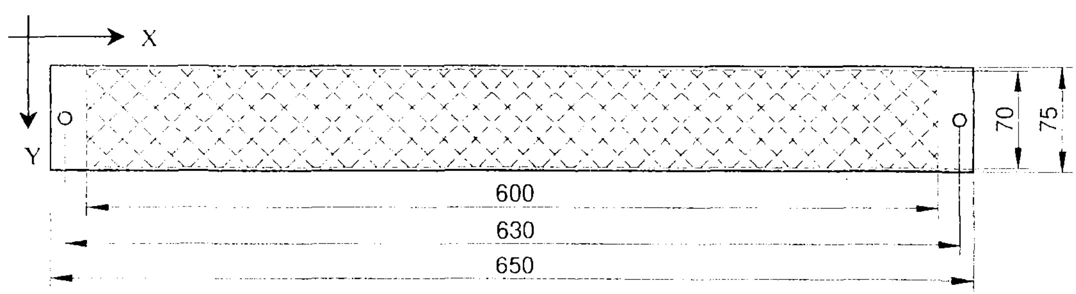

Figure 3. Dimensions of a fuel plate.

The activities of Cs 137 of some fuel plates have been measured by means of a 38/195-High-Purity-Germanium- (HPGe) detector. The peak efficiency calibration was performed using a reference disk source and a homogenous rectangular volume source. The rectangular volume source (dimensions of the fuel plate) was filled with a dry silica-sand from the ÖFPZ-Arsenal which was prepared with Cs 137, Co 57 and Co 60, the reference disk source was prepared by the PTB-Braunschweig, (5.8 cm diameter). The fuel plates and the reference sources were centred 14.5 cm above the HPGe detector end-cap.

As absolute measurements with the HPGe are very time consuming the measurements have been carried out with a 2"x2"-NaI(Tl) scintillation detector.
The relative count rates of all 234 plates were determined only in the central part of the plates by means of the NaI(Tl) scintillation detector. There is a linear correlation between the count rates of the NaI(Tl)- and the HPGe- measurements, therefore the absolute Cs 137 activities for all 234 plates can be derived. It has to be pointed out that the fission product Cs 137 is not distributed homogeneously within the fuel plates. This fact is caused by the known cosine shaped neutron flux density distribution [2], thus, the HPGe-detector-efficiency is to be corrected using a method described in the next paragraph.

3 Calculation of the HPGe-peak-efficiency with MCNP

Another approach to determine the efficiency is the calculation by means of Monte Carlo methods. A Monte Carlo code is simulating the real physical process of energy deposition in the detector crystal. The history of a photon emitted from an arbitrary source which is entering the detector is followed up in the programme until it either escapes from the detector with or without energy loss or until it comes to rest within the detector. In order to simulate the experimental setup of real measurements the calculated energy loss is stored in a corresponding energy channel. In this way a pulse height spectrum is built up from which the peak efficiency can be deduced.
The peak efficiency of the HPGe detector was calculated for photons of Cs 137 (661.6 keV) emitted by the homogenous disk source and the rectangular volume source using the Monte Carlo code MCNP Version 4B [3]. The results of the calculation were compared to experimental data. A cosine distributed standard source was not available for the experiment, therefore the HPGe-peak efficiency is calculated with MCNP. To obtain the shape of the Cs 137 activity distribution some fuel plates were scanned along the x and y-axis by the NaI - scintillation -detector by moving it along a narrow lead collimator slit and observing the counting rate of Cs 137. Relative to the centre of the plate the activity distribution follows a cosine law as to be expected (Fig. 4).

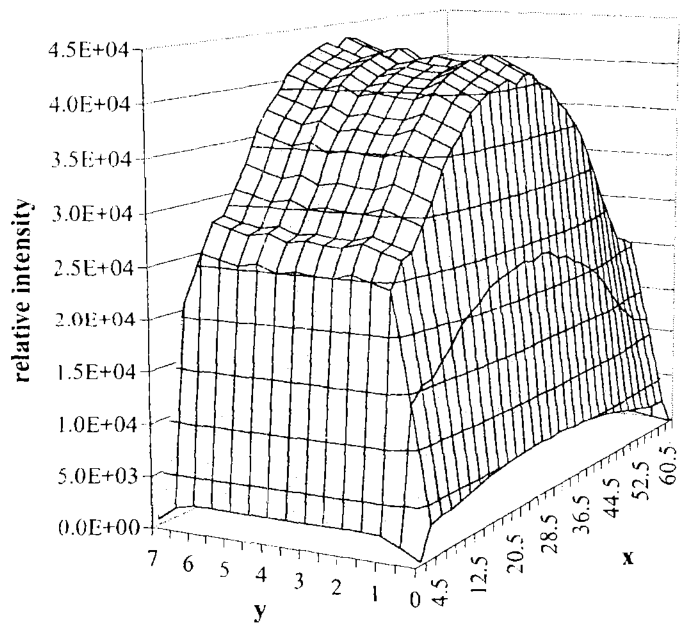

Figure 4. Cs 137 radiation intensity of a fuel plate obtained with a 2"x2" NaI(Tl)-detector; collimator slit 1 cm x 7 cm.

The shape of the cosine distributed volume source of the fuel plate is obtained from the measurements with the NaI detector. An approximation of the measured cosine distribution (Fig. 5) is used to calculate the peak efficiency of the HPGe detector with the Monte Carlo program MCNP.

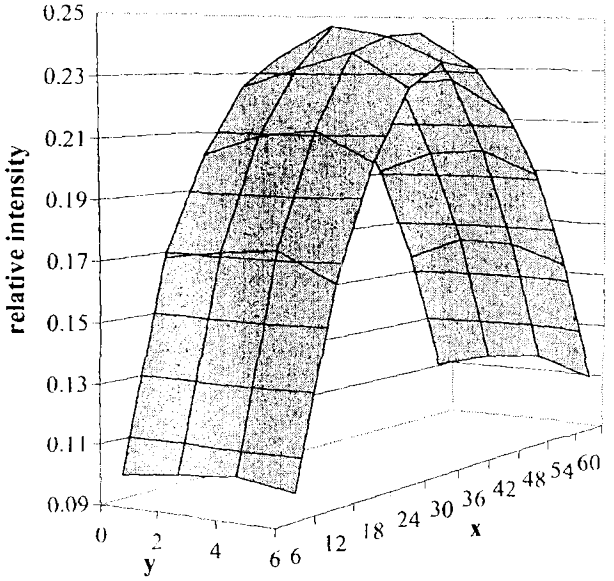

Figure 5. Approximated xy-distribution of the Cs 137 activity in the fuel plate.

4 Results

4.1 Activity determination of Cs 137

Fig. 6. shows the gamma spectrum of a fuel plate within the energy range of 30-1200 keV.

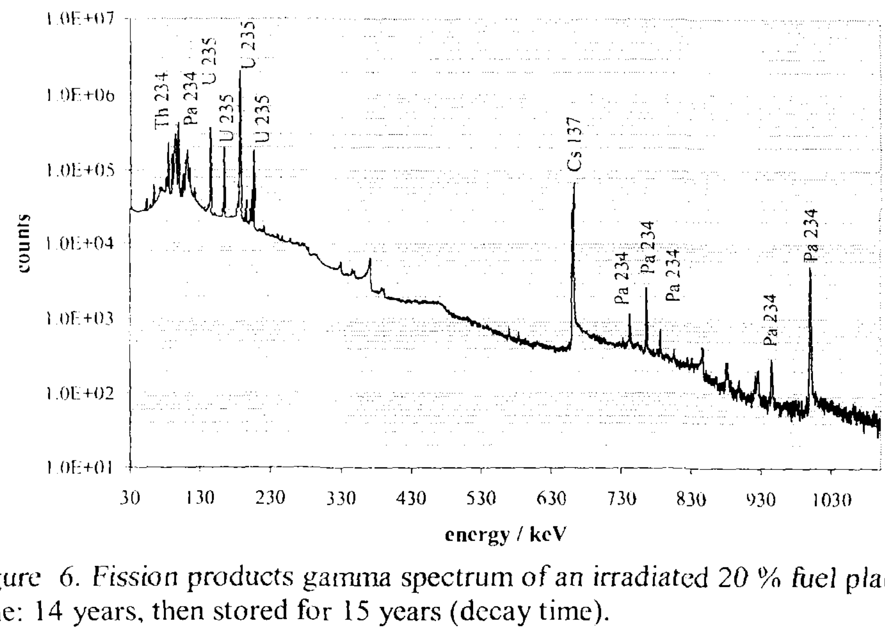

Figure 6. Fission products gamma spectrum of an irradiated 20 % fuel plate. Irradiation time: 14 years, then stored for 15 years (decay time).

With the exception of the fission product Cs 137, the remaining gamma peaks belong exclusively to the decay products of U235 and U238. No Cs 134, Eu 154 and Ru 106 have been observed within a measuring time of about 1 hour.

The results of the MCNP calculation are in good agreement with the experimental results. In general the absolute calculated HPGe-peak-efficiencies are only 8 % higher than the experimental peak efficiencies. The ratios of the peak efficiencies of the rectangular volume source and the disk source are in excellent agreement. Therefore we obtain an efficiency correction factor from the ratio rectangular volume source to cosine distributed volume source from the calculation. The efficiency factor of the rectangular volume source is only 3.3 % lower than the efficiency of the cosine distributed volume source. It is assumed that this correction factor is also valid for the experimental efficiency. The activity of Cs 137 is then corrected for all 234 fuel plates (reference date: April 1999).

A frequency histogram of the fuel plates (Fig. 7) shows that 94.3% are within the range of 2 σ (55.1 –73.0 kBq).

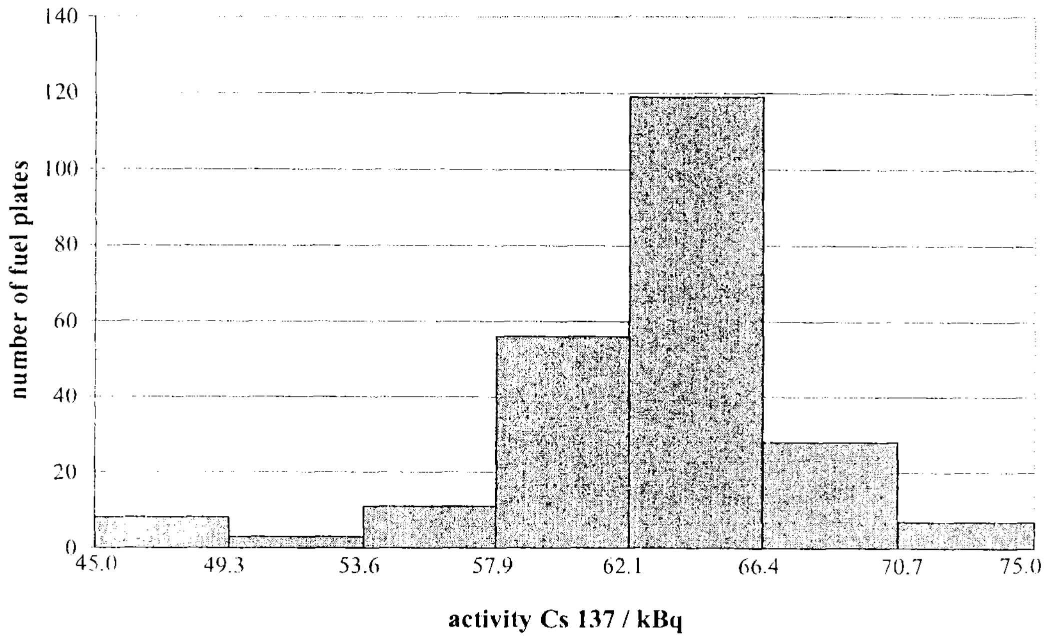

Figure 7. Frequency histogram of the activity of Cs 137 of 232 fuel-plates.

The total activity of 234 plates is (14.9 ± 1.6) MBq. At two plates a Cs 137 activity is obtained which is 3 times higher than the other plates. These two plates are used several times for other experiments.

4.2 The burn up

In this work we use the following burn up definitions [4]:

$$\text{FIMA (Fissions per Initial heavy Metal Atom)} = \frac{N(U235)}{N(U238\ \text{initially})} \cdot 100 \quad [\%]$$

$$\text{FIFA (\underline{F}issions per \underline{I}nitial \underline{F}issile \underline{A}tom)} = \frac{N(U235)}{N(U235\ \text{initially})} \cdot 100 \quad [\%]$$

The mass (in grams) of fissioned U 235 and the burn up of U 235 (FIFA) of a fuel plate are computed by means of the total Cs 137 -activity which was produced during reactor operation from 1965 to 1985. It is our main assumption that there exists a linear correlation between the Cs 137 production and irradiation time. This leads to a FIFA- value of $5.8 \cdot 10^{-6}$ % and a FIMA value of $1.16 \cdot 10^{-6}$ % for all 234 fuel plates, in relation to 4.9 kg U 235 and 24.2 kg U 238 fuel mass. The calculated values of the burn up (FIFA values) of 232 fuel plates are plotted in Figure 8. The circles indicate the end plates which are located at the outer corner of a fuel element. The errors of these calculations are estimated to be about 10 % which is due to the unknown irradiation history and the unknown position of a plate within a fuel element.

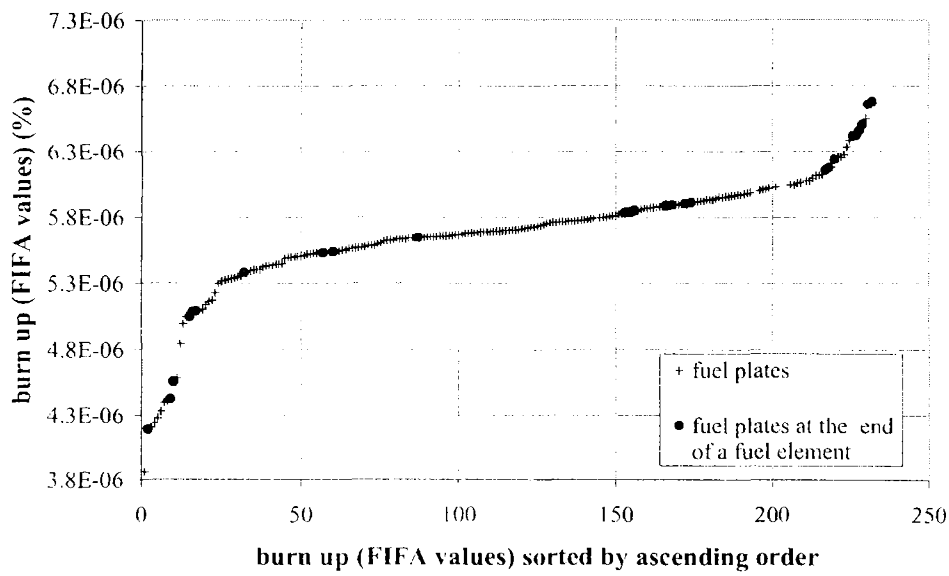

Figure 8. Burn-up distribution (FIFA values) of 232 fuel plates.

4.3 The surface gamma dose rates

The positions of the gamma counter FH40 during measurements on the surface of a fuel element (position 1, 2, 3 and distance d = 1m, position 4, 5) are presented in Figure 9.

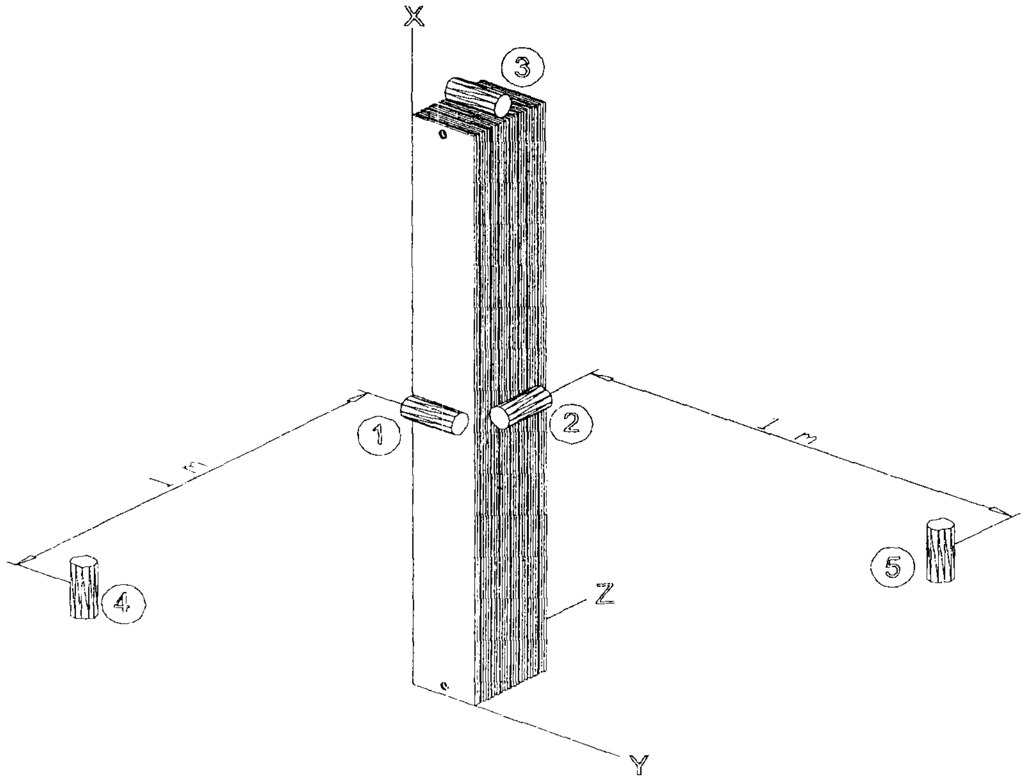

Figure 9. Measuring positions for the gamma dose rate of a fuel element.

The total Cs 137 activities of a fuel element the gamma dose rates of four fuel elements (17 plates per element) at the various positions obtained by with the gamma counter FH40, and the corresponding 2σ uncertainties are listed in Table 1. The gamma dose rates of the four fuel elements and the gamma activities are within 3σ.
The gamma dose rate of one fuel element is only two times greater than the gamma dose rate of one single fuel plate.

Table 1. Total Cs 137 activities of a fuel element (17 plates) and values of the gamma dose rate of several positions on a fuel element.

Gamma dose rate (μSv/h)	**Fuel element number**				
	3	**10**	**14**	**6**	**Mean**
position 1	22.0 ± 0.4	21.0 ± 0.4	23.8 ± 0.4	22.1 ± 0.4	22.2 ± 0.2
position 2	25.5 ± 0.6	24.0 ± 0.4	26.8 ± 0.6	26.5 ± 0.6	25.7 ± 0.2
position 3	5.9 ± 1.2	6.8 ± 1.4	6.6 ± 1.4	5.5 ± 1.2	6.2 ± 0.6
position 4	0.53 ± 0.16	0.46 ± 0.14	0.55 ± 0.16	0.52 ± 0.16	0.52 ± 0.08
position 5	0.50 ± 0.16	0.58 ± 0.18	0.59 ± 0.18	0.46 ± 0.14	0.53 ± 0.08
Cs 137 activity (MBq)	0.95 ± 0.09	1.06 ± 0.11	1.04 ± 0.10	1.19 ± 0.11	1.06 ± 0.10

5 Conclusions

With the Monte Carlo program MCNP the HPGe detector-peak-efficiency has been calculated for a cosine distributed volume source for the homogenous disk source, the rectangular volume source and an inhomogenous rectangular volume source for Cs 137 (E = 661.6 keV) with the *Monte Carlo code MCNP Version 4B* [3].
It is possible to obtain efficiency correction factors for special volume sources by comparing it with experimental efficiencies of standard sources.
The average activity of Cs 137 of a fuel plate of the SAR Graz is 62.8 ± 4.5 kBq.

The mean value of the gamma dose rates on the surface of a fuel element is 22.2 ± 0.2 μSv/h.

ACKNOWLEDGEMENT

The authors appreciate financial support by the Verein zur Förderung der Strahlenforschung, Graz.

The authors would like to thank to Mr G. Kahr for computational assistance.

6 Literature

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