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DEVELOPMENT OF AN EFFICIENT AND ACCURATE ANALYSIS APPROACH FOR RADIOISOTOPE PRODUCTION IN SAFARI-1

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Abstract. The Radiation and Reactor Theory Section of the South African Nuclear Energy Corporation SOC Ltd (Necsa) is developing a core simulator, OSCAR-4, for the analysis of material test reactors and to provide nuclear engineering analysis services to the SAFARI-1 reactor and NTP Radioisotopes SOC Ltd, which is located within Necsa. The analysis services include reactor reload verification and in-core and ex-core irradiation analyses for safety assessment and irradiation planning for radioisotope production.

This work presents our calculation system and tools used to facilitate the production of radioisotopes and their ability to predict experimental measurements. In particular, we present the results from the comparison of calculation prediction and measured activities for the production of the yttrium-90 radioisotope.

The calculated results obtained for sixty three different irradiation configurations show an under-prediction of the measured activities 9.1 % with a variance of 5.4 %.

1 INTRODUCTION

The South African Nuclear Energy Corporation SOC Ltd (Necsa) owns and operates the SAFARI-1 research reactor. The reactor is used for several scientific applications which range from in-core to ex-core irradiation. The reactor analysis and other nuclear engineering analyses for the application of the reactor are extensively supported by the Radiation and Reactor Theory (RRT) Section of Necsa.

The SAFARI-1 reactor is a multi-purpose, 20 MW plate-type material testing reactor and is used as a neutron source for neutron radiography, radioisotope production and silicon doping. Radioisotopes that are produced in fixed in-core locations, experience irradiation under various geometric configurations and core depletion states. These factors affect the neutron flux distribution in the irradiated sample, and need to be taken into account in order to perform accurate activity predictions. In order to serve these modelling requirements and to increase the fidelity of the calculations, we developed an approach that allows us to accurately and efficiently model different sample configurations that may occur in the irradiation rig. This approach consists of two contributions; the first is the development of a tool to transfer detailed atom densities from OSCAR-4 to the MCNP input, and the second is the automation of the modelling of sample configurations in MCNP.

This paper describes RRT's calculation codes, models and analyses methodology for supporting in-core irradiation analysis in SAFAR-1. In particular, the methodology is applied to the production of yttrium-90 (^{90}Y) under sixty three different irradiation configurations. The calculated results obtained show an under-prediction of the measured activities 9.1 % with a variance of 5.4 %.

Section 2 gives a brief description of SAFARI-1. Sections 3 and 4 describe the deterministic and Monte Carlo codes that are used for the analysis of SAFARI-1. Section 5 presents the software programs that were developed for the purpose of constructing accurate, reliable and efficient analysis models, which are an integral part of the calculation methodology. Section 6 presents the application of the methodology for the purpose of radioisotope production; which serves as a first performance test of the calculation scheme. Section 7 presents the results for the analysis performed in this work. Section 8 presents the conclusions and future work.

2 DESCRIPTION OF SAFARI-1

The SAFARI-1 reactor is a multi-purpose tank-in-pool type material testing reactor (MTR), which is located at Necsa. The nominal power of the reactor is 20 MW. The reactor core is contained inside the reactor vessel, which is inside the reactor pool. The reactor vessel is immersed in light water which serves as coolant, moderator and shielding. In the configuration analyzed in this paper, the core contains 26 fuel elements and 6 control rod elements. Apart from the routine molybdenum production, several isotopes are produced through neutron irradiation in the reactor. In this paper we analyze the particular irradiation of yttrium oxide (Y_2O_3) for the production of yttrium-90 (^{90}Y).

3 SAFARI-1 ANALYSIS CODES

Several computer codes are used in the RRT Section to support the operation of SAFARI-1, the main ones are the OSCAR-4 code system and MCNP. The OSCAR-4 (OSCAR) code system is used for reactor reload design and core-follow analysis. On the other hand, MCNP is used to obtain accurate flux solutions in the irradiated samples where detailed geometric modelling is required. For a given burnup state, the isotopic composition of the core is transferred from OSCAR to MCNP using a dedicated tool.

The OSCAR code system contains a three-dimensional, multigroup, nodal diffusion code, MGRAC, which performs the calculations in a six energy group structure for homogeneous nodes. During core depletion analysis, MGRAC tracks the depletion history of each fuel element in the reactor core (Prinsloo, 2012).

The energy group collapsing and spatial homogenization of the materials for the core calculations are performed with the HEADE transport code. HEADE is a lattice physics code that uses a collision probability transport solver with a response matrix formalism to solve the two-dimensional fine-group transport problem for a given assembly type.

For detailed transport calculations where the diffusion theory is not applicable, the Monte Carlo code MCNP, version 5.1.51, is used (X5 Team, 1987). MCNP is a general-purpose Monte Carlo N-Particle transport code that is used in RRT for neutron, photon and coupled neutron/photon transport. MCNP's general geometry modelling capability and the use of pointwise cross-sections are among its main features that makes it so applicable for the analysis of complex problems.

RRT has developed a MCNP model for SAFARI-1 which includes the reactor core, as well as the ex-core irradiation facilities. It contains detailed modelling of all the assemblies available in SAFARI-1 and has been designed in order to make it easy to implement variance reduction techniques outside the reactor core.

A dedicated computer tool was developed to transfer the core isotopic composition from OSCAR to MCNP, i.e. OASYS2MCNP, in order to use the correct core depletion state in the MCNP model. To implement the wide variety of arrangements for sample irradiation in the MCNP model in a relatively user friendly way, the LOAD_IPR (Load Isotope Production Rig) code was developed. Both of these software tools will be described in more detail in Section 5.

4 MCNP MODEL

The MCNP model for SAFARI-1 includes the reactor core, the core box, the reactor tank and the beam tubes. Inside the reactor tank is a grid plate with a rectangular arrangement of 8×9 positions where different assembly types can be loaded as shown in Figure 1. In this figure, the beam tubes and the large facility nozzle are shown as void, but they are mostly flooded with water during reactor operation. Figure 1 also shows the core configuration grid. Note that positions D6 and F6 contain the Isotope Production Rigs (IPRs) where the samples analyzed in this paper were positioned.

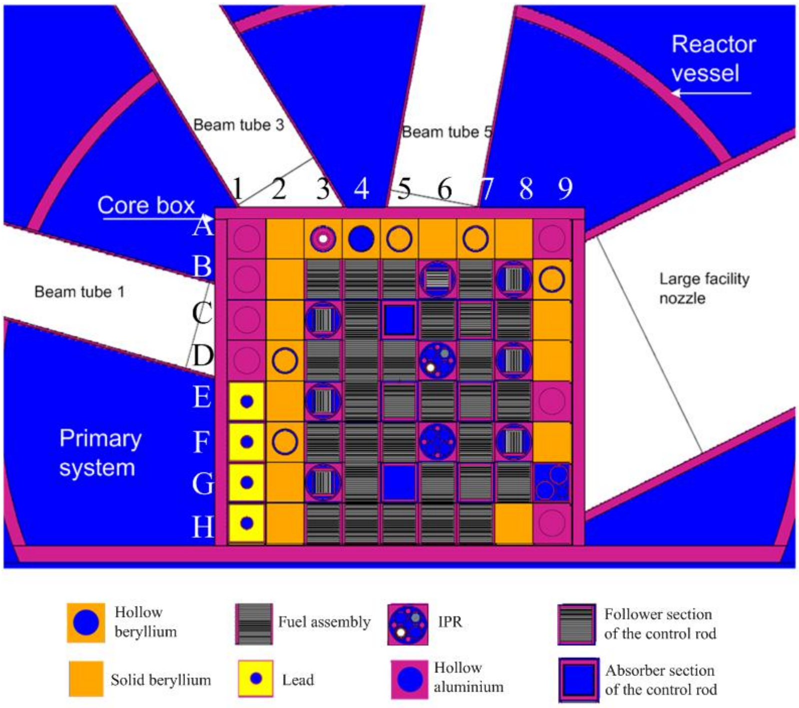

Figure 1: XY view of the core and beam tubes

Figure 2 shows a planar view of the model with the fuel, control rod elements and control rod fuel follower, at the active regions of these components. The fuel elements used in SAFARI-1 are MTR type fuel with 19 plates each. The fuel plates consist of a Uranium-Silicide-Aluminium (U_3Si_2-Al) powder dispersed core, enclosed in an aluminium-alloy cladding. The active length of each fuel element in the MCNP model is discretized into axial regions in order to be able to use the same axial burnup distribution as used in the OSCAR system.

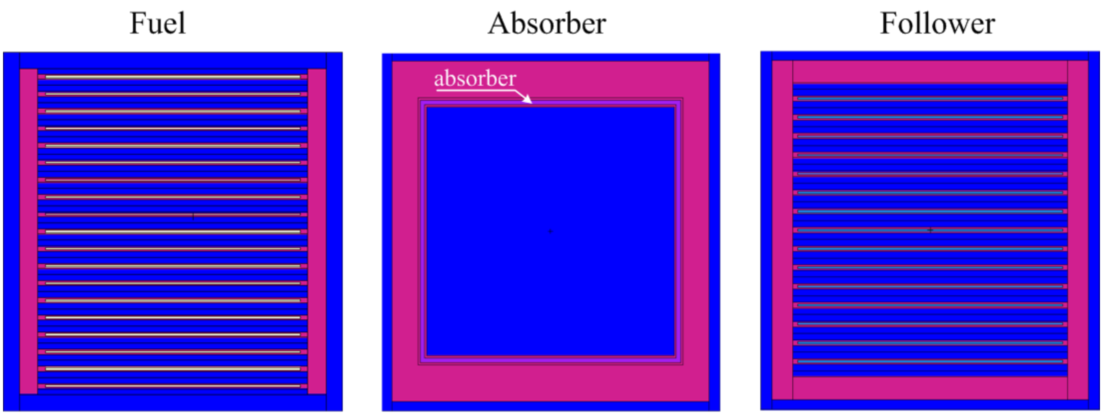

Figure 2: View of the fuel element, absorber and fuel follower

The control rod assemblies consist of an upper absorber section and a lower fuel section connected through a rigid aluminium coupling mechanism. The absorbing section, shown in Figure 2, consists of an aluminium box that contains a cadmium layer as absorber. The fuel section, also called the control rod follower, is similar to the fuel elements but is constructed inside an aluminium box and contains only 15 plates.

The reactivity control system contains six identical control rods that are located in positions

C5, E5, G5, C7, E7 and G7. Five control rods (C5, G5, C7, E7 and G7) comprise the control rod bank and are always at the same axial position at any given time. The control in E5 is the regulating rod and moves independently of the control rod bank to account for any reactivity fluctuations that result from the removal or insertion of molybdenum assemblies or IPRs.

As shown in Figure 3, when a control rod (CR) is fully inserted, the absorber section extends along the core, and when it is fully withdrawn the active region of the follower coincides with that of the fuel elements.

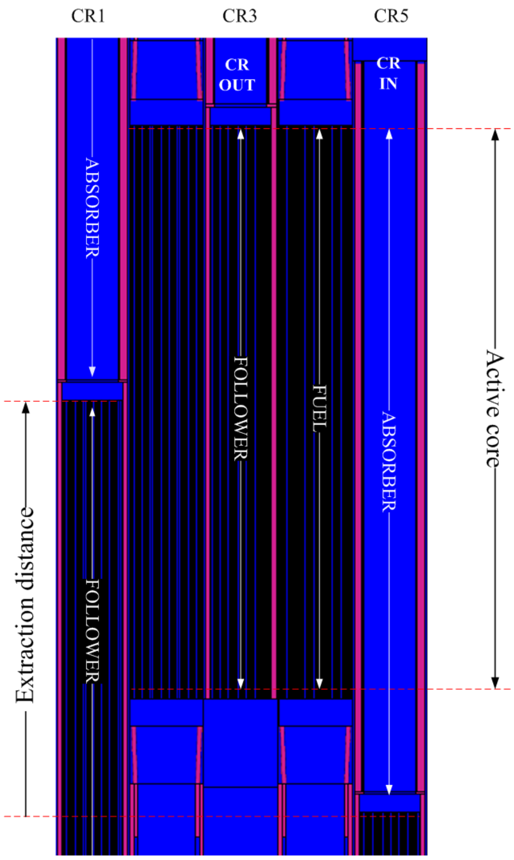

Figure 3: Illustration of possible axial control rod positioning

The IPR positions in SAFARI-1 consist of positions D6 and F6 where samples can be irradiated. In the analyses performed in this work, a four-column target holder as shown in Figure 4, is used to position the samples in cylindrical sample holders (holders not shown) in the IPR.

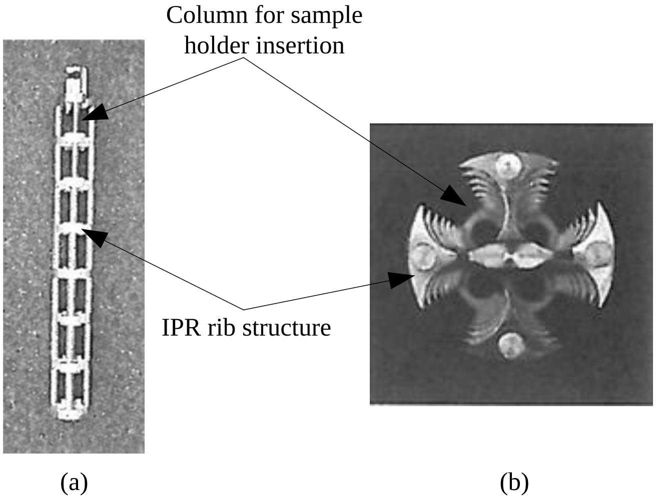

Figure 4: (a) Side view of IPR, (b) Top view of IPR

The MCNP model of the IPR is shown in Figure 5. The model of the experimental column configuration for the sample irradiations considered in this work is shown in Figure 5(a). Figure 5(b) is a horizontal section through the model showing the four IPR columns and their associated designation; i.e. 1, 2, 3 and 4. Figure 5(c) shows the modelling of the sample geometry contained in Figure 5(a).

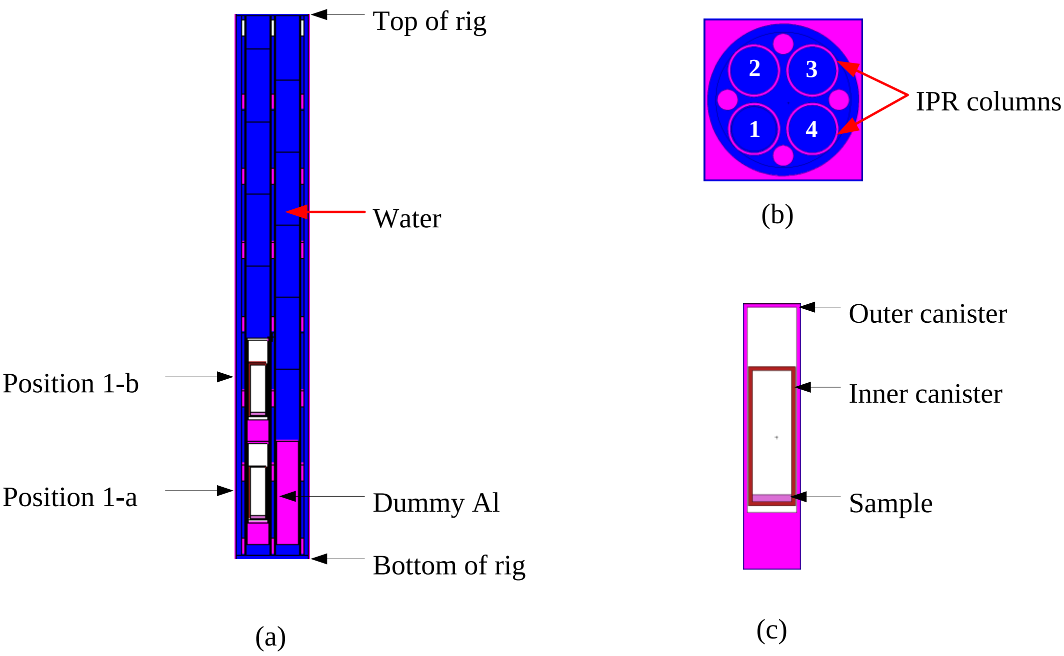

Figure 5: MCNP model of IPR and samples

5 CALCULATION TOOLS

In order to ensure the accurate, reliable and efficient construction of the MCNP model, two model automation tools were developed. The first is the tool to transfer the OSCAR core depletion state (fuel and follower isotopic number densities) to the MCNP model. The second tool is used to load the IPR irradiation columns with an accurate representation of the sample arrangement and control rod positions. These two software tools are described in the remainder of this section.

5.1 OASYS2MCNP

In principle, in order to achieve accurate analyses in SAFARI-1, it is important to use a good representation of the reactor depletion state which is applicable to the conditions of irradiation. In order to transfer specific core isotopic number densities from OSCAR to MCNP, we developed the OASYS2MCNP program. This tool reads the number densities from the OSCAR output files at predefined time steps and transfers the data to MCNP.

OASYS2MCNP de-homogenizes the nodal densities to densities per fuel plate, creates the same axial discretization used in OSCAR into the MCNP model, and renames the isotopes according to the libraries to be used in MCNP. This process is shown in Step 1 of Figure 6.

The lattice calculations are performed in OSCAR with the HEADE code, which uses the 103 burnable isotopes available in the 172 group JEF2.2 library for the fuel and control rod followers. For each burnup step, the microscopic cross sections of the 37 isotopes listed in Table 1 are transferred to the six-group library used by MGRAC. The 66 lumped isotopes are stored in the 6-group library through macroscopic cross sections, Σ_L, which are used by MGRAC to perform the core calculations.

Actinides		Fission Products	
^{234}U	^{241}Pu	^{135}I	^{147}Nd
^{235}U	^{242}Pu	^{135}Xe	^{148}Nd
^{236}U	^{241}Am	^{141}Ce	^{147}Pm
^{237}U	^{243}Am	^{142}Ce	^{148}Pm
^{238}U	^{242}Cm	^{144}Ce	^{148m}Pm
^{237}Np	^{243}Cm	^{143}Pr	^{149}Pm
^{239}Np	^{244}Cm	^{143}Nd	^{147}Sm
^{238}Pu	^{245}Cm	^{144}Nd	^{148}Sm
^{239}Pu		^{145}Nd	^{149}Sm
^{240}Pu		^{146}Nd	

Table 1: Isotopes contained in the profile history of OSCAR-4

For a given core state and for each fuel material region, the number densities of the 37 isotopes listed in Table 1 are transferred to MCNP together with an Equivalent Boron Content (EBC) that represents the 66 lumped isotopes that are not explicitly taken into account. The EBC is the number density that corresponds to the macroscopic cross section of Boron-10 (^{10}B) whose average absorption reaction rate is equal to that of the lumped materials represented by Σ_L:

$$N_{10}\,\sigma_{10}\sum_{g=1}^{6}\phi_g \equiv \sum_{g=1}^{6}\Sigma_{Lg}\,\phi_g \tag{1}$$

In this equation, N_{10} is the EBC, ϕ_g and Σ_{Lg} are the scalar flux and the macroscopic cross section of the lumped isotopes in group g, which were obtained in a diffusion calculation for a given fuel material. The summation is over the six energy groups used by MGRAC. The one-group microscopic absorption cross section of ^{10}B, σ_{10}, has been obtained by collapsing the ^{10}B cross section in the 172 group JEF2.2 library used by HEADE with the spectra obtained with MGRAC at different typical burnup values.

In summary, the MCNP model described in this report uses the 37 isotopes listed in Table 1 for the meat of the fuel and control rod follower together with an EBC that replaces the lumped isotopes.

5.2 Irradiation rig loading tool: LOAD_IPR

LOAD_IPR was developed to ensure the rapid and accurate construction of the detailed sample configuration in the MCNP analysis model. This automation was achieved through the use of predefined irradiation samples in the MCNP model (i.e. an MCNP base model) and the development of the rig loading program known as LOAD_IPR.

Step 2 in Figure 6 shows the function of LOAD_IPR. It takes as input the upstream MCNP model in which the core number densities have been updated to match the core depletion state from OASYS2MCNP and adds the sample irradiation configuration to the model. That is, it adds the explicit loading configuration for both the irradiation rigs (in the D6 and F6 core positions) and a representative control bank position to the model. The rig loading configuration is obtained from NTP (NTP Radioisotopes SOC Ltd, a subsidiary company of Necsa), while the control bank movement information during the irradiation period is obtained by processing data from the SAFARI-1 plant computer.

The output of LOAD_IPR is an MCNP model with a representative isotopic core composition, the actual rig loading configuration and a representative control rod bank position. LOAD_IPR also constructs the necessary MCNP flux and reaction rate tallies according to the sample configurations in the irradiation rigs.

The final MCNP model, in principle, is an accurate representation of the core state for a particular analysis. The application of the final state dependent model is not confined to a particular calculation, but may be modified by the user to either add tallies or to serve as a basis for performing other analyses.

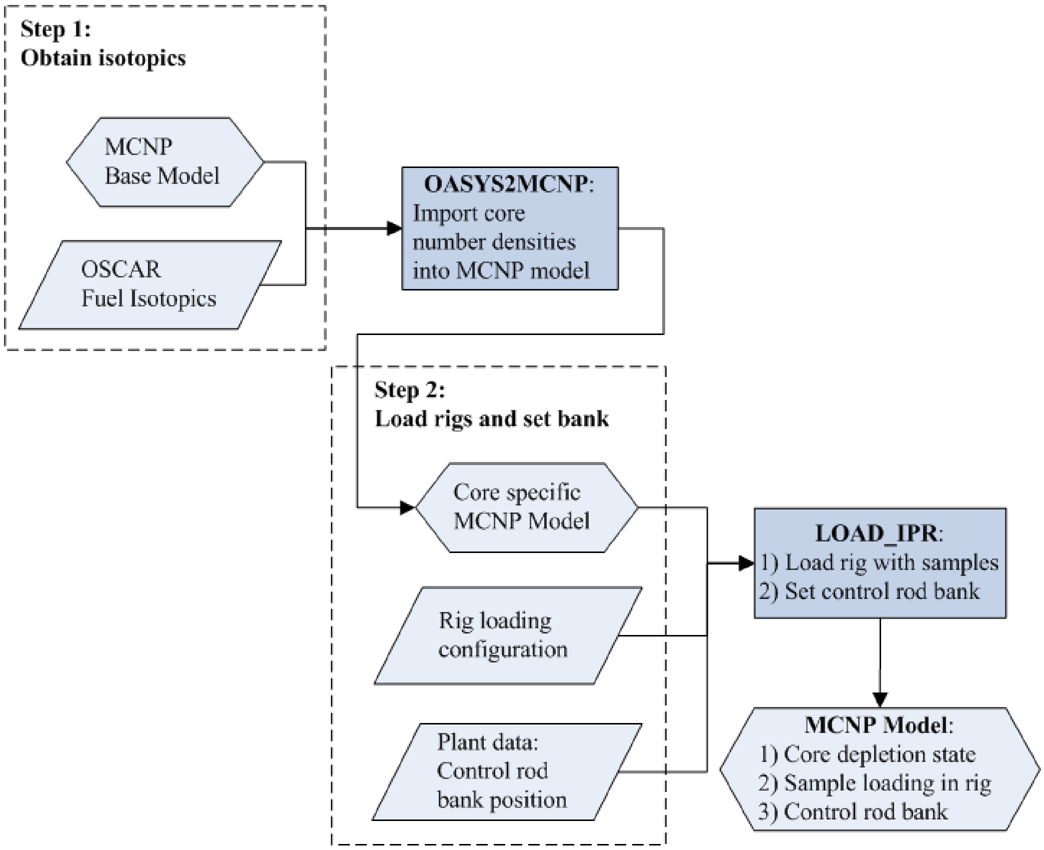

Figure 6: MCNP model automation

6 CALCULATIONS

This section describes the testing of the OSCAR/MCNP calculation approach for the purpose of radioisotope production.

Aligned with the calculations reported in (Ball, 2003), the results presented in this section can be considered as part of the validation exercise for the computational tools and modelling of irradiation samples.

A brief description of the reactor operation during isotope production is given. This is followed by a description of the experimental data set. Section 6.3 describes the calculation model that was used in the analysis.

6.1 Reactor operation during irradiation

Research reactors are typically designed as multi-purpose machines for accommodating various reactor experiments and irradiation applications. Although the operation of SAFARI-1 has become routine in terms of the fuel cycle strategy, the operation pertaining to sample irradiation is still quite dynamic. That is, the four columns in the irradiation rig can accommodate arbitrary sample loading configurations. In addition, individual columns can also be removed during the cycle to either remove- or re-insert samples into the desired positions. Figure 7 shows possible column sample loading configurations that may occur in the IPR columns. All of these configurations can be generated using the LOAD_IPR rig loading program, allowing for rapid and reliable model construction.

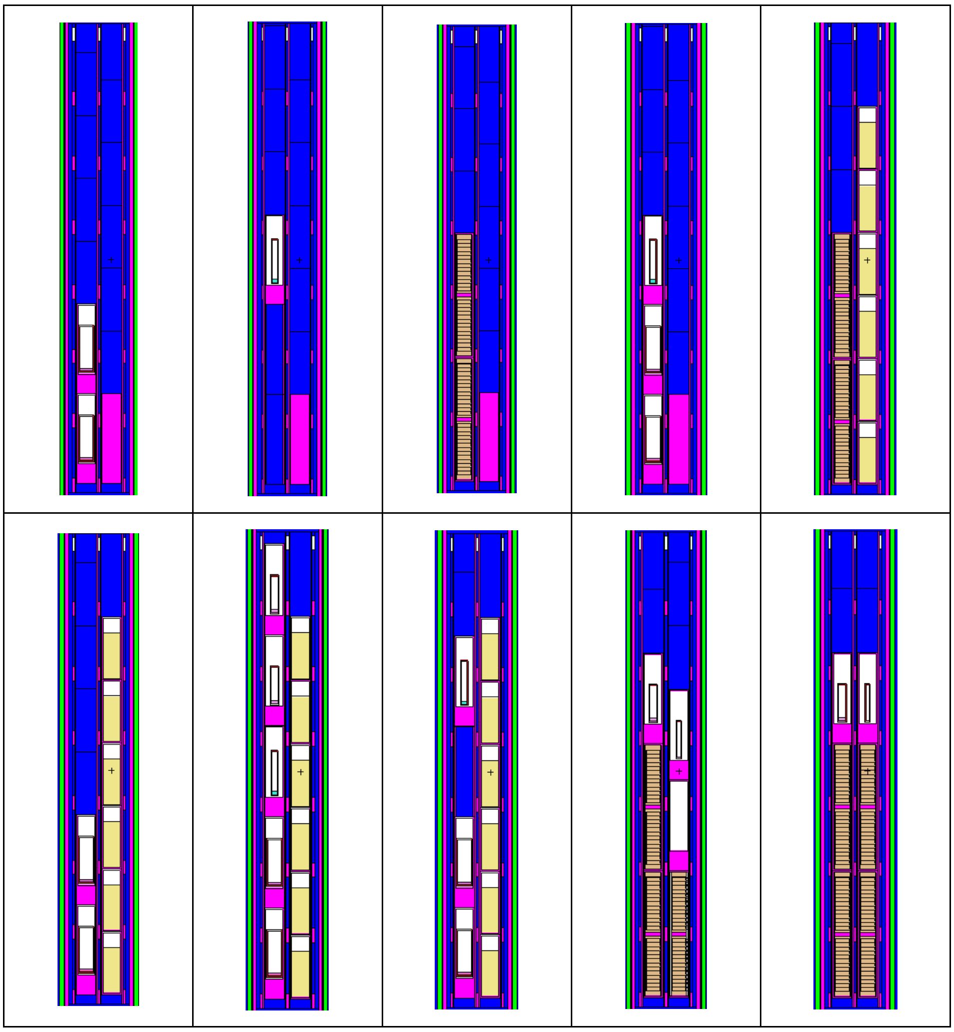

Figure 7: Vertical section through the IPRs showing different sample configurations in water

6.2 Experimental data

In order to validate the calculation methodology presented in this work, calculations were compared to a series of measured activities obtained for the production of ^{90}Y. The data set consists of 33 irradiations of yttrium oxide (Y_2O_3) samples that were irradiated over a period of one year during 2013. Each irradiation case contained two samples per irradiation, giving a total of 66 experimental data points.

The locations for the sample irradiations considered in this paper are as shown in Figure 5(a). Note that while the Y_2O_3 samples were always loaded in the 1-a and 1-b positions, other sample loading configurations above the Y_2O_3 and in neighbouring columns included the loading variations as shown in Figure 7.

6.3 Calculation model

As a first approximation the major variables that were considered of importance for the accurate modelling of the experiment were:

i) Experimental information – sample composition, sample mass, irradiation- and decay time; and measurement uncertainty;

ii) Irradiation rig sample configurations;

iii) The control rod bank position during irradiation; and

iv) The use of the correct core depletion state.

The experimental information in i) was obtained from NTP with a maximum measurement uncertainty of 1 %. The sample loading configurations were also obtained from NTP and could be modelled just as accurately in MCNP by using the LOAD_IPR tool.

The control bank positions were obtained from the SAFARI-1 reactor plant data at intervals of 4.2 seconds. The core depletion state is given by the cycle used in the irradiation, and the corresponding isotopic composition of the fuel is transferred from OSCAR to the MCNP model.

The present OSCAR/MCNP methodology allows for a detailed time-dependent modelling during an irradiation. However, the detailed application of this methodology is impractical since it would involve many MCNP calculations; therefore we developed an approximate modelling approach by making some approximations which are described below.

In the MCNP model, all the control rods were positioned at the same level which was defined as the average extraction of the rods during the irradiation. Figure 8 shows the control rod bank (5 rods) and regulating rod (1 rod) positions given by plant data, and the average control rod bank position (6 rods) that was implemented in the MCNP model. The assumption of an average bank was motivated by the fact that the average bank travels only about 3 cm during a 60 hour irradiation period. The all rods out position correspond to an extraction of 74.79 cm.

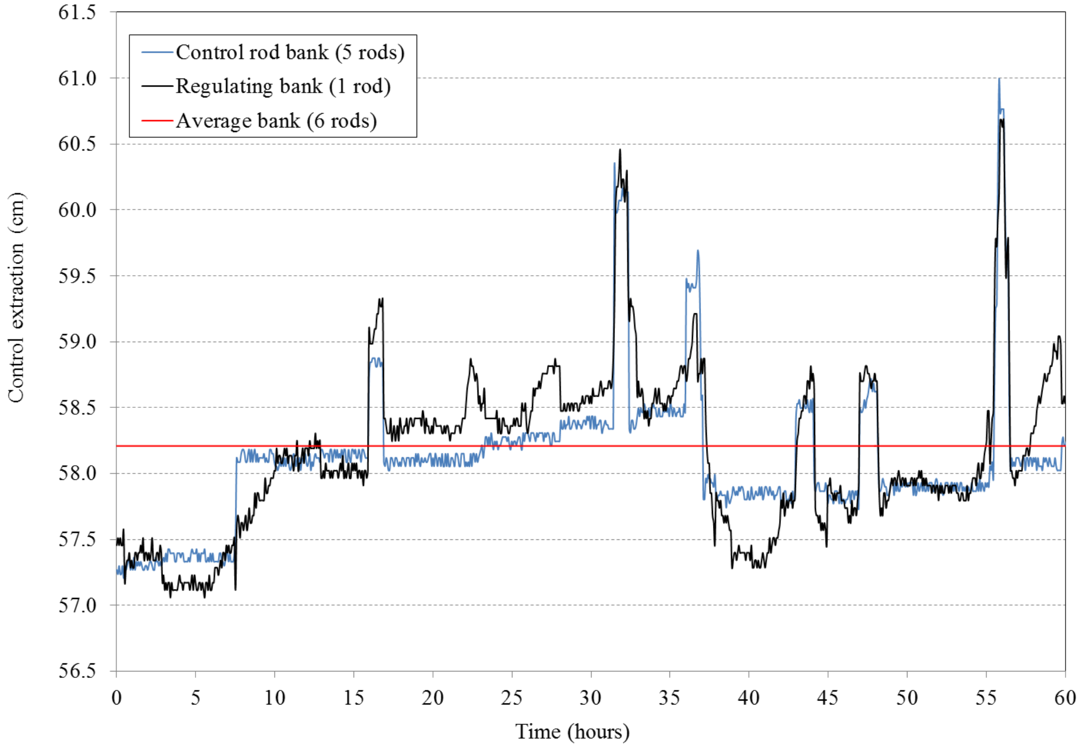

Figure 8: Example of control rod bank movement during irradiation

Note, that the average control rod bank position will not give a k_{eff} equal to 1 as it should be for operating conditions. Instead, for all the irradiations analyzed in this work, the core reactivity predictions ranged from +200 to -1900 pcm.

What is important in this work however, is the effect of the control rod bank position on the axial flux profile; we assume that the simple average bank position is accurate enough in this sense. To support this assumption, we verified that for the irradiations modelled in this work, the standard deviation of the average control rod position is about 0.5 cm, which affects the predicted sample activities by about 1 %.

It is important to note that we did not have all the details pertaining to the insertion and removal of samples and molybdenum rigs during the operation. From varying control rod position information in Figure 8, it is evident that the core configuration is not constant during the irradiation time, as we assumed. Not taking these effects into account in the analysis, could have a significant effect on the results.

Regarding the core depletion state, a representative core cycle was used to perform all the calculations in this work. This approximation is based on the knowledge that the SAFARI-1 fuel strategy is fixed, that the cycle length is fairly constant and that the samples were irradiated at fixed times during the cycle. That is, a cycle is typically 4.5 weeks long and the samples were typically irradiated on a weekly basis, about 4 days after reactor startup.

Figure 9 shows a representation of reactor operation and sample irradiation. The figure shows the 4 day delay period after startup, which is followed by irradiation weeks 1 to 4 (blue, green, grey and yellow respectively), with the sample irradiation periods shown in red.

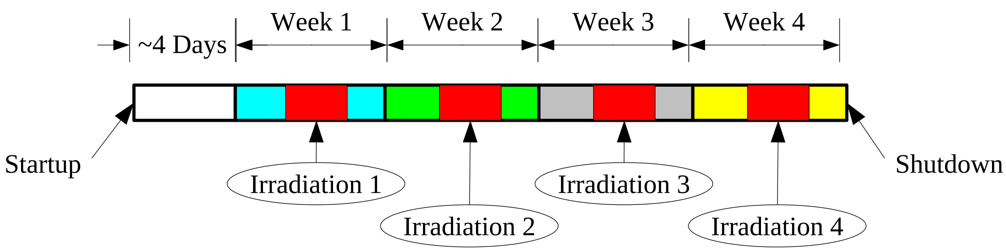

Figure 9: Schematic representation of typical sample irradiation during the depletion cycle

The core cycle selected as a generic basis to perform the calculations, was Cycle 8 of 2013 (C2013-8). The main reasons for its selection were that the length of C2013-8 contains the four weeks with a four day delay, and that no scram occurred. The core composition states were then extracted for the four weeks at about midweek when the samples were typically irradiated. These depletion states correspond to the irradiation periods (red) shown in Figure 9.

In summary, the MCNP model that was thus used to model a given irradiation period contained a representative core isotopic composition given by C2013-8, which is dependent on the week of irradiation. The control bank position was taken as the average control rod bank obtained from plant data over the irradiation period.

The series of MCNP models that were constructed for each of the 33 cases were then executed and the total neutron scalar flux, ϕ, and the ^{89}Y capture cross-section, σ_{cap}, were computed for each sample. In this way, the activity for each sample was then calculated using Eq. (2)

$$A^{90}(t_{Decay}) = N^{89}\phi\sigma_{cap}\left(1-e^{-\lambda t_{Irrad}}\right)e^{-\lambda t_{Decay}}, \tag{2}$$

where:

$A^{90}(t_{Decay})$ is the ^{90}Y activity after decay time t_{Decay};
N^{89} is the number of yttrium-89 (^{89}Y) atoms present;
λ is the decay constant of Y90; and
t_{Irrad} is the irradiation time.

7 RESULTS AND ANALYSIS

This section contains a summary of the major results from this work. Section 7.1 presents the results for the MCNP flux solution in the irradiation rig, while the Section 7.2 presents the comparison of calculated and measured activities.

7.1 Neutron flux in irradiation rig

Figure 10 shows a typical unperturbed (blue curve) and perturbed (red curve) neutron flux profile in the Column 1 of the irradiation rig. The control rod insertion depth relative to the rig and axial flux distribution are also illustrated.

One notices that the unperturbed flux shows several axial flux depressions, which result from the structural ribs in the rig design. Comparing the unperturbed with the perturbed flux profile, one sees that the perturbation in the column due to the sample is fairly local. The thermal flux depression in the perturbed case is due to the replacement of the moderator by the void inside the sample canisters. The calculated average unperturbed thermal flux in positions 1-a and 1-b are in the region of about $3\times10^{+14}$ to $3.5\times10^{+14}$ n.cm^{-2}.s^{-1} respectively.

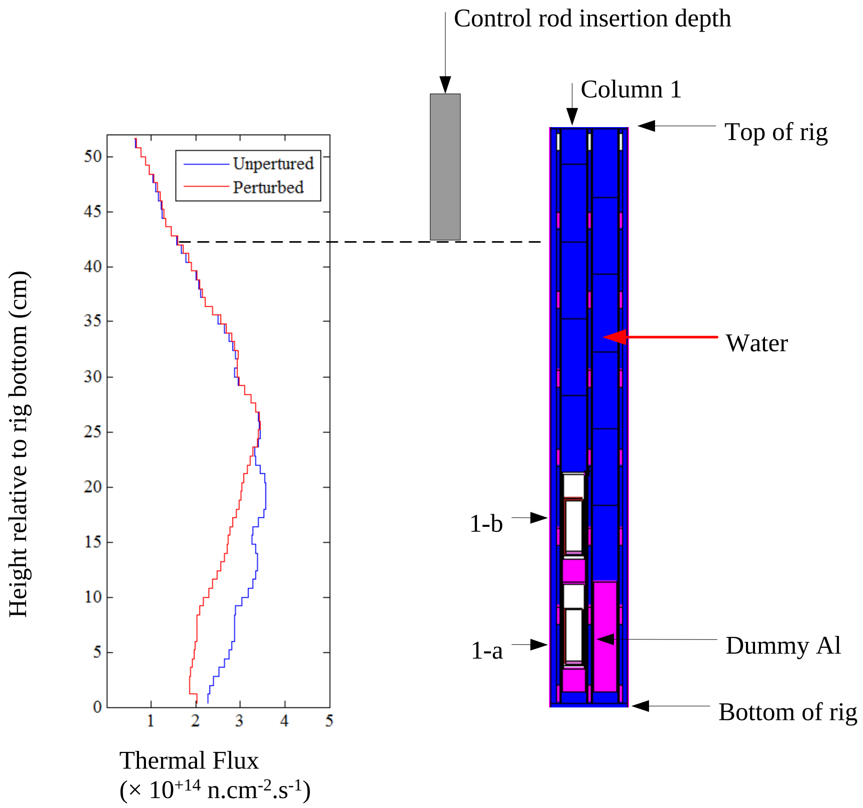

Figure 10: Example of thermal flux suppression in column 1

7.2 Comparison between calculation and experiment

Figure 11 shows the comparison between calculation and experimental ^{90}Y activity results, (C/E - 1) expressed in percent, for the 1-a (red) and 1-b (blue) irradiation positions. Note that the irradiations corresponding to the data points 55 to 62 were performed during cycle C2013-8, the cycle that was used as the core depletion basis for the calculations reported in this paper.

The results clearly show a general under-prediction of the measured activity in both positions, with a maximum under-prediction of -20 %.

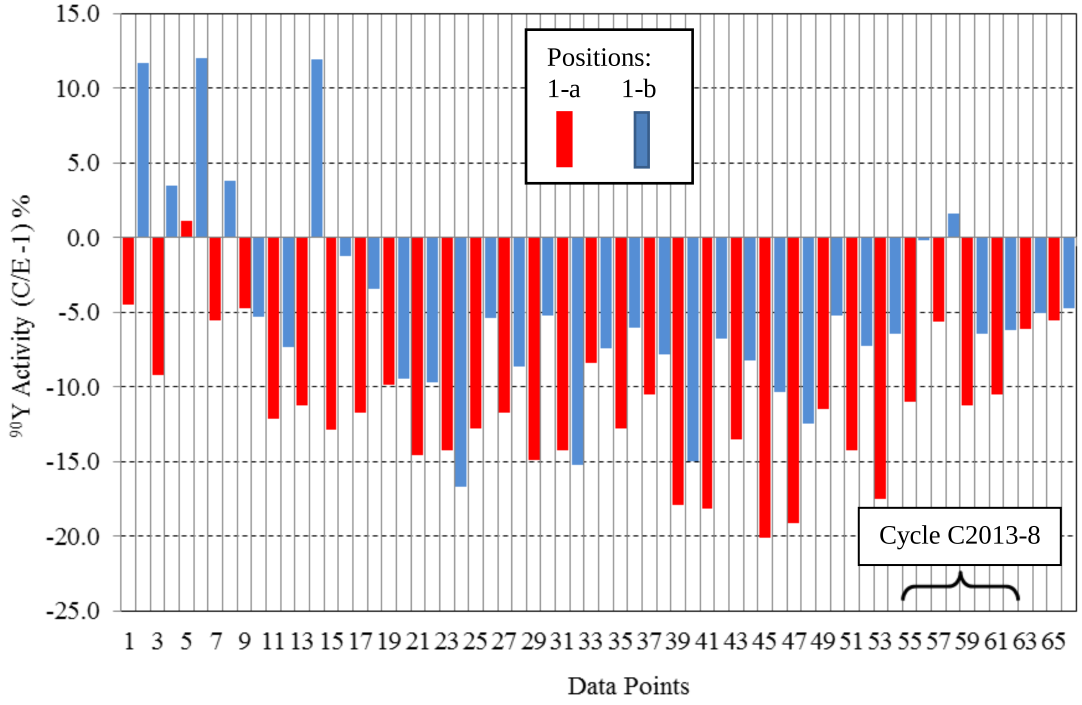

Figure 11: ^{90}Y activity comparison between calculation and experiment [(C/E - 1) %]

Figure 12 shows a Gaussian fit for the observed difference in activity for the entire data set. The average under-prediction from the fit is 9.1 % with a standard deviation of 5.4 %. Three of the 66 data points (2, 6 and 14) analyzed were not considered in this analysis since they show C/E ratios that are very far from the behaviour shown in Figure 11.

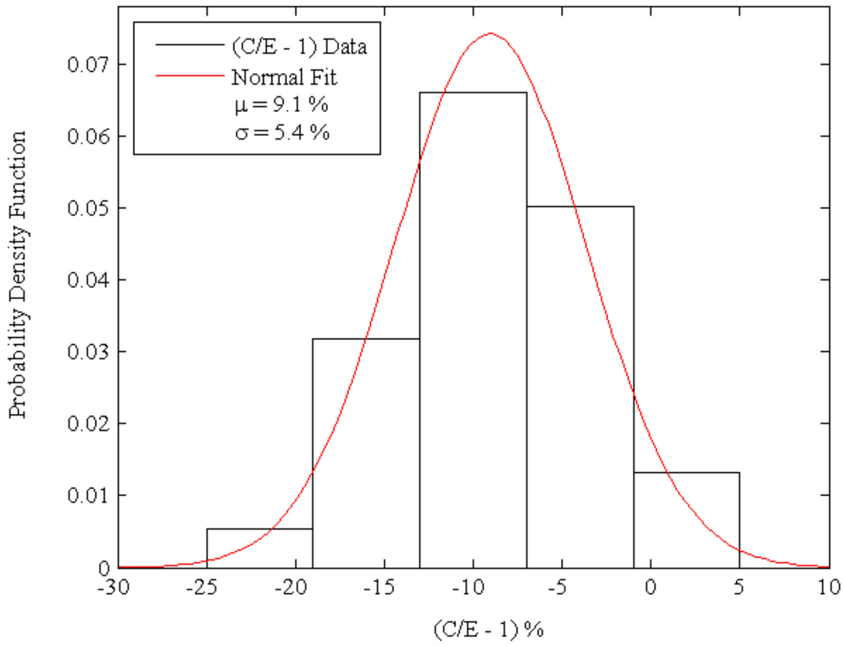

Figure 12: Gaussian fit to ^{90}Y activity difference in position 1-a

The fact that the experimental results are consistently under-predicted indicates that there may be a consistent effect which causes the bias in the calculated results. Note however that these results do not reflect the true capability of the OSCAR/MCNP calculation methodology, but is a measure of the performance of the assumptions that were made in the calculation approach, which can certainly be improved.

8 CONCLUSION AND FUTURE WORK

In this work we presented the calculation tools and methods for irradiation analyses at the SAFARI-1 reactor. The calculation methodology consists of the reactor core simulator OSCAR-4 and MCNP. OSCAR-4 is used to obtain the detailed core depletion state (core isotopic composition), which is passed to MCNP for a detailed particle transport calculation. The methodology is supported by a set of software tools which facilitate the transfer of core isotopic and the detailed loading configuration of in-core irradiation rigs.

A calculation approach for radioisotope production, which is based on the OSCAR/MCNP methodology, has been developed and tested with the measured activity data that was obtained for the production of the ^{90}Y radioisotope. Sixty three data points were analyzed and the results were compared to the experiment.

Comparison between the calculation and experiment shows that the calculation approach applied in this work, results in an under-prediction of activities by (9.1 ± 5.4) %, 68 % of the time. Considering the diverse operations of the reactor, i.e. with various sample loading configurations in the rigs and sample insertion and removal during the cycle, the calculation approaches developed in this work, performed reasonably well as a first test.

The predicted results however show a systematic behaviour for which the exact cause is not immediately apparent. The three major sources of calculation inaccuracy are thought to be the specific core depletion state, the modelling of the control rods and the detailed core operation conditions during sample irradiation.

From data points 55 to 62 in Figure 11, in which the calculations for the irradiations correspond to the core depletion state, reasonable C/E ratios were obtained. These results indicate that the use of the cycle specific core depletion state in the calculation may improve the accuracy of the prediction; work which will be undertaken in future. In addition, future work will include sensitivity analyses on control rod position and sample loading configuration.

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