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Reactor Physics Modeling of Spent Research Reactor Fuel for Technical Nuclear Forensics

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ABSTRACT

Technical nuclear forensics (TNF) refers to the collection, analysis and evaluation of pre- and post-detonation radiological or nuclear materials, devices, and/or debris. TNF is an integral component, complementing traditional forensics and investigative work, to help enable the attribution of discovered radiological or nuclear material. Research is needed to improve the capabilities of TNF. One research area of interest is determining the isotopic signatures of research reactors. Research reactors are a potential source of both radiological and nuclear material. Research reactors are often the least safeguarded type of reactor; they vary greatly in size, fuel type, enrichment, power, and burn-up. Many research reactors are fueled with highly-enriched uranium (HEU), up to ~93% ^{235}U, which could potentially be used as weapons material. All of them have significant amounts of radiological material with which a radioactive dispersal device (RDD) could be built. Therefore, the ability to attribute if material originated from or was produced in a specific research reactor is an important tool in providing for the security of the United States. Currently there are approximately 237 operating research reactors worldwide, another 12 are in temporary shutdown and 224 research reactors are reported as shut down. Little is currently known about the isotopic signatures of spent research reactor fuel. An effort is underway at Savannah River National Laboratory (SRNL) to analyze spent research reactor fuel to determine these signatures. Computer models, using reactor physics codes, are being compared to the measured analytes in the spent fuel. This allows for improving the reactor physics codes in modeling research reactors for the purpose of nuclear forensics. Currently the Oak Ridge Research reactor (ORR) is being modeled and fuel samples are being analyzed for comparison. Samples of an ORR spent fuel assembly were taken by SRNL for analytical and radiochemical analysis. The fuel assembly was modeled using MONTEBURNS(MCNP5/ ORIGEN2.2) and MCNPX/CINDER90. The results from the models have been compared to each other and to the measured data.

INTRODUCTION

Technical Nuclear Forensics (TNF) is comprised of the technical means by which nuclear material is characterized and interpreted. These may suggest or exclude possible sources of origin when attempting to attribute a nuclear device, designs, or material to its origin (1). One potential source of nuclear material is research reactors. Research reactors are typically located at facilities that have minimum security. The developing world has 25% of the world's research reactors (2). Research reactors have dynamic operations varying in power, fuel type, moderator, coolant, etc… The enrichment in fuel ranges from natural to <93% enriched ^{235}U. These characteristics are attractive to some terrorist groups and possibly state or sub-state organizations.

Isotopic composition of spent fuel or signature from a research reactor is needed for TNF to make a determination of likely sources of interdicted material. An effort is underway at Savannah River National Laboratory (SRNL) to produce a database of research reactor signatures. The database captures two types of signatures measured and digital signatures. Digital signatures are necessary for reactors where measured data is unobtainable. However, to determine how accurately current reactor physics codes are for calculating isotopic composition of spent research reactor fuel for the purpose of nuclear forensics is currently unknown. Therefore, as part of the signature effort at SRNL, each spent fuel assembly that is measured is also modeled and the results compared to one another.

The research reactor selected for this work is the Oak Ridge Research reactor (ORR). Construction was finished in 1958 and decommissioned in 1987, for most of its operating life it ran using highly enriched uranium (3). Initially designed for 20 MW_{th}, regulatory approval for 30 MW_{th} was obtained shortly after operations had begun (3). The ORR used materials test reactor (MTR) type fuel. Table 1 lists the specifications for the ORR fuel assembly which had samples taken for analysis at SRNL. The fuel assembly has 54% burn-up and was originally enriched to 93.14% ^{235}U. As reported to SRS by the operator the burned fuel assembly contained 132 g ^{235}U and 177 g of total U. This gives the fuel an enrichment of 74.58% ^{235}U which could be obtained through reprocessing. Though the assembly has been cooling for almost 20 plus years the contact dose from gamma rays was calculated to be 4846 R/hr with a dose at 1 m of 73 R/hr. The dose was calculated using ORIGEN-S for the source term and MCNP5 to determine the dose in air at a certain distance from the fuel assembly.

Assumption/Premise	Value
Assembly ID Number	T-397
Time at Power	108.9 days
Burn-up	121 MWd/Assembly
Average Assembly Power	~1.11 MW_{th}
Burn-up Percentage	54 %
Fuel Meat Material	U_3O_8 alloyed with Al
Cladding Material	Al
Moderator	H_2O
Fuel Meat Thickness	0.0508 cm
Plate Thickness	0.1638 cm (2 outer) & 0.1275 cm (17 inner)
Active Fuel Length	59.8043 cm (2 outer) & 59.7839 (17 inner)
Reactor Cycle	239.6 day cycle, 21.78 days at power and 217.82 cooling days
Assembly Decay Time	8799.82 days
Number of Plates	19
Initial ^{235}U	285 grams
Initial U_{tot}	306 grams
Final ^{235}U	132 grams (reported value)
Final U_{tot}	177 grams (reported value)

Table 1 Specifications of the ORR fuel assembly which had samples taken from it.

MONTEBURNS and MCNPX were the two codes used to model the ORR fuel assembly. MONTEBURNS uses MCNP5 for transport calculations and used ORIGEN2.2 for depletion calculations. MCNPX uses CINDER90 for depletion and decay calculations which are available via the BURN card in the MCNPX input file. ORIGEN2.2 employs matrix exponential method to solve for burn-up while MCNPX uses Markov chains (4).

An infinite lattice model of the fuel assembly was constructed in MCNP (Figure 1 left side). Due to the curvature of the plates and the MCNP constraint that a periodic surface can only be located on flat plane, the assembly was "split" in the middle to retain the proper pitch between assemblies Figure 1 right side). The benefit to an infinite lattice model is it is a relatively simple model compared to a full core model. However, it is unable to take into account reflecting or absorbing materials which may occupy the space around a fuel element. With this understanding it is useful to know how well an infinite lattice model performs in calculating burn-up.

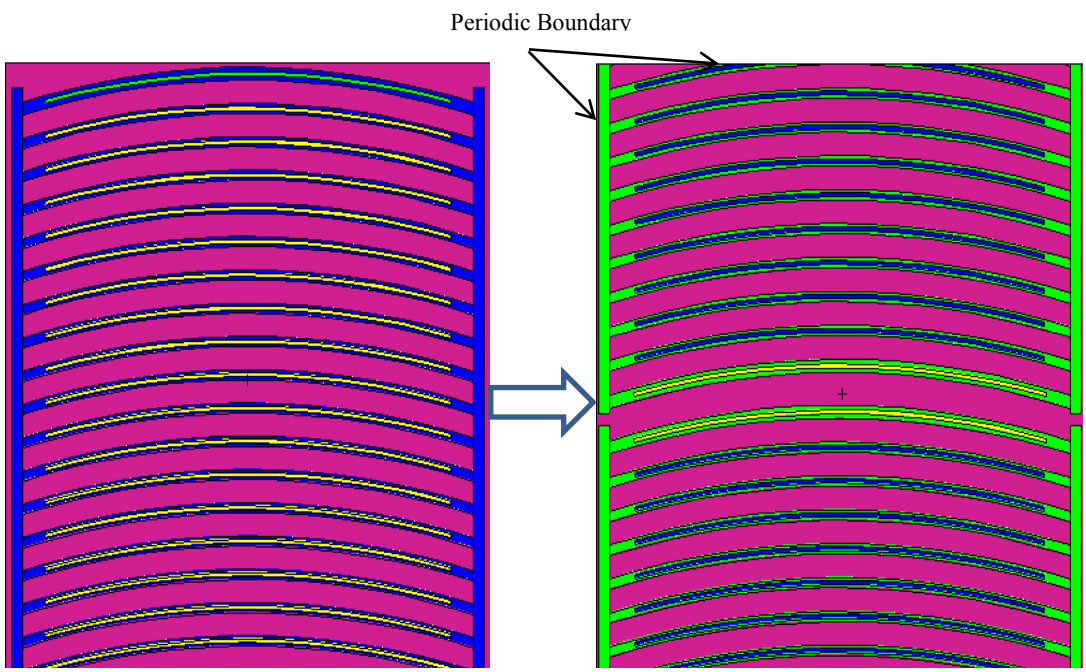

Figure 1 Left: Model of ORR fuel assembly. Right: Model split in two pieces to preserve geometry in the infinite lattice model.

DISCUSSION

K-effective for the system was calculated by both codes at each time step. The initial calculations were based on the following time steps: 5 steps of 4.356 days for a total of 21.78 days and decay of 217.82 days between, this was repeated 5 times for a total of 108.9 days at power, with a final decay step of 8799.82 days. Figure 2 has a plot of the k_{eff} values at each burn-step the decay time between burn-steps (217.82 days) was omitted from the plot. There is good agreement between MCNPX and MONTEBURNS on k_{eff}, most of the difference between the codes is due to when k_{eff} is

calculated. MCNPX calculation is approximately 2 days after the MONTEBURNS calculation. This is why the MONTEBURNS data points show a larger increase in k_{eff} after ^{135}Xe decays away during each decay step between the 21.78 day cycles.

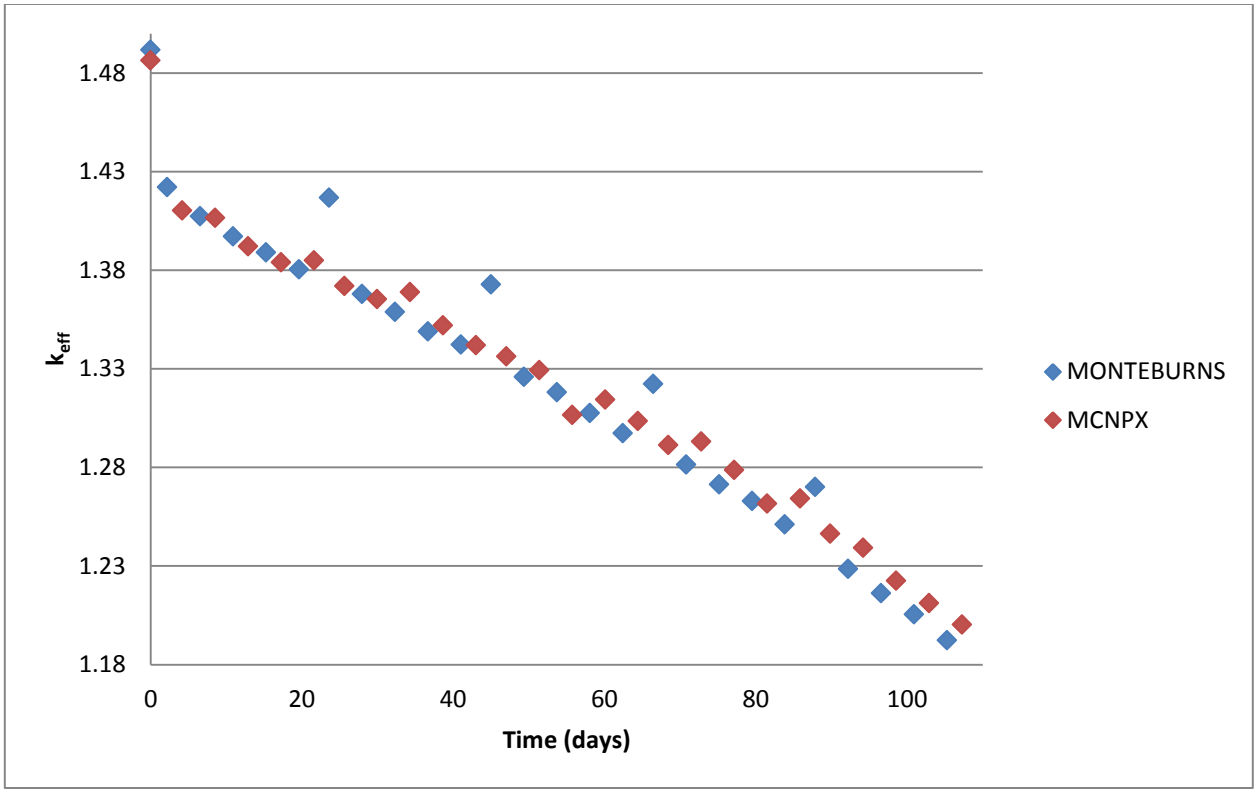

Figure 2 Plot of k_{eff} using both codes. The plot shows good agreement between the codes. Note that there is a 2 day offset between when each code calculates k_{eff} .

Since the specific details of the reactor cycle that the fuel assembly experienced is not known, the MONTEBURNS model was tweaked to do a straight burn without a decay step between each burn step. The two codes agree well with each other on the amount of uranium and plutonium in the fuel assembly at the end of life. A comparison of the codes over each time step shows good agreement for plutonium isotopes (see Figure 3 and Figure 4). MB 2 in Figure 4 was a straight burn with no intermediate decay steps. Removing the intermediate decay steps did not significantly change the calculated values for MONTEBURNS. Figure 4 has a bar chart of different uranium and plutonium isotope masses as calculated by each model. The two MONTEBURNS and the MCNPX model agreed with each other over uranium and plutonium isotopics. They also agreed fairly well with the measured values for uranium. However, the models did not agree with the measured plutonium data. One possibility for the difference between plutonium data is that small changes in the beginning uranium isotopics could mean large changes in some of the plutonium isotopics. This is because ORR has HEU fuel, therefore there is a significant amount of ^{234}U in the fuel and the amount, if any, of ^{236}U that was in the fresh fuel is unknown. Changing amount of these two isotopes leads to significant changes of ^{238}U. This assembly has 306 grams of starting and 285 grams of ^{235}U leaving only 21 grams for ^{234}U, ^{236}U, and ^{238}U. The current assumption for starting concentration of ^{234}U is 3 grams, which changes the remaining grams by ~ 14%. Figure 4 shows

that there is about ~10% difference between the measured value of ^{234}U and the calculated value. Currently work is being done to determine better starting values for ^{234}U, ^{236}U, and ^{238}U.

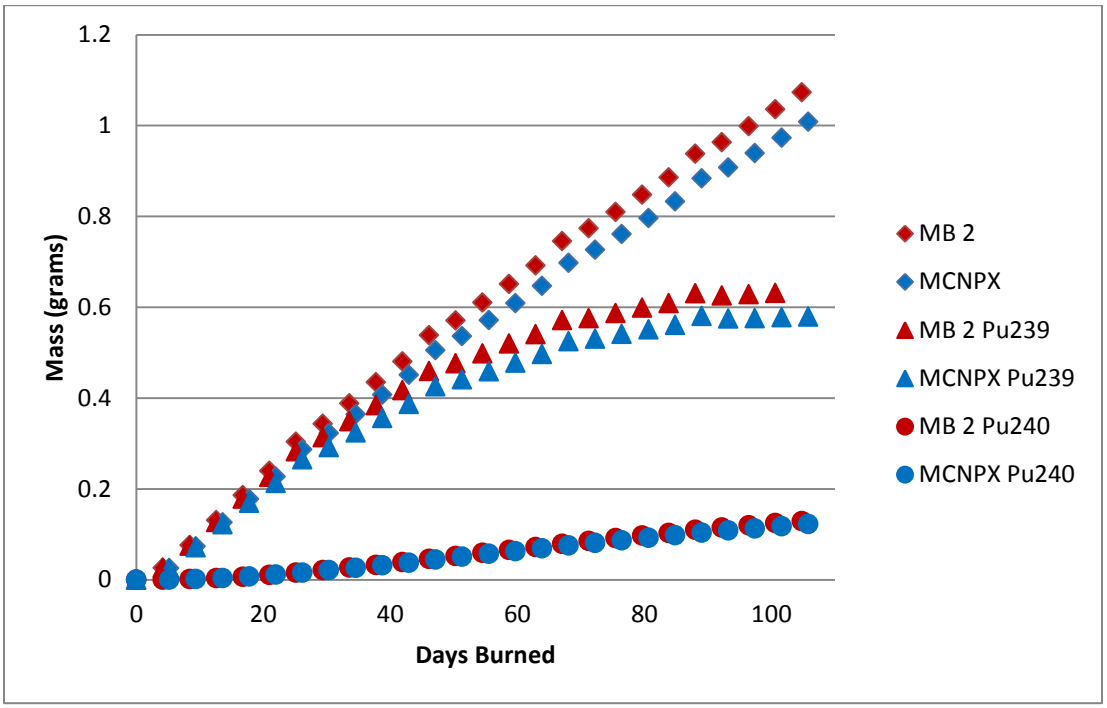

Figure 3 Plot of Total Pu, 239Pu, and 240Pu. The plot shows good agreement between the two codes.

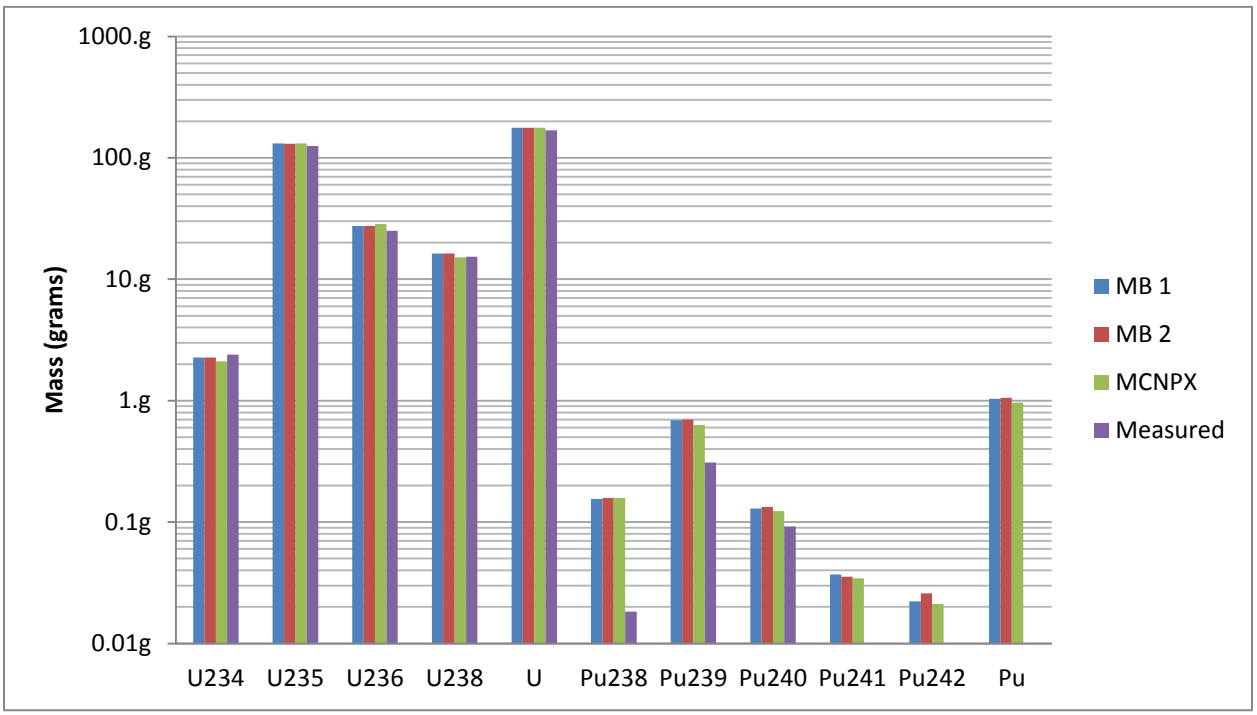

Figure 4 Calculated uranium and plutonium isotopics for the fuel assembly. MB 1 is the mass for the MONTEBURNS model with decay steps and MB 2 is the MONTEBURNS model with no decay steps. The purple bars represent the measured data for uranium isotopics and ^{238}Pu, ^{239}Pu, and ^{240}Pu.

The spent fuel assembly was sampled inside Shielded Cells at SRNL. Due to legacy contamination issues, a sealed isolation box was constructed. The isolation box was kept under a slight positive pressure and had modified gloves which accommodated the manipulators. This allowed the operators to drill into fuel assembly and then dissolve the samples into a solution. Since the samples were taken under such conditions there is an un-quantified uncertainty in the samples due to working in such an environment. Once the sample material was dissolved, three solutions were prepared. Samples of each of the three solutions were bagged and then removed from both the isolation box and Shielded Cells. It was these samples which were measured to determine the composition of the spent fuel.

The heavy elements, above atomic number 82, were measured using an enclosed inductively coupled plasma-mass spectrometer (ICP-MS). An aliquot of each solution was diluted further and then injected into the ICP-MS for analysis; ISO 17025 QA protocols were followed. The ICP-MS measures mass to charge ratio, therefore some isotopes will be counted together (i.e. m/z = 137 could potentially contain ^{137}Cs and ^{137}Ba).

Elemental results were primarily determined by ICP-Atomic Emission Spectrometry (AES); again an aliquot of the submitted solution was further diluted and injected into the plasma of an enclosed instrument following ISO 17025 QA protocols. Trace elements which exceeded the minimum detectable activity (MDA) are reported. Results for the ICP-AES are shown in Figure 6.

Radiometric analyses completed to date include ^{90}Sr, ^{99}Tc, ^{137}Cs, ^{154}Eu, and ^{238}Pu. ^{90}Sr and ^{99}Tc were analyzed by chemical separation and liquid scintillation counting. ^{137}Cs and ^{154}Eu were determined by gamma spectrometry on the whole sample. The Pu was separated by TTA extraction

prior to plating and alpha counting. All analyses except ^{99}Tc are ISO 17025 accredited. Results are shown in Figure 6.

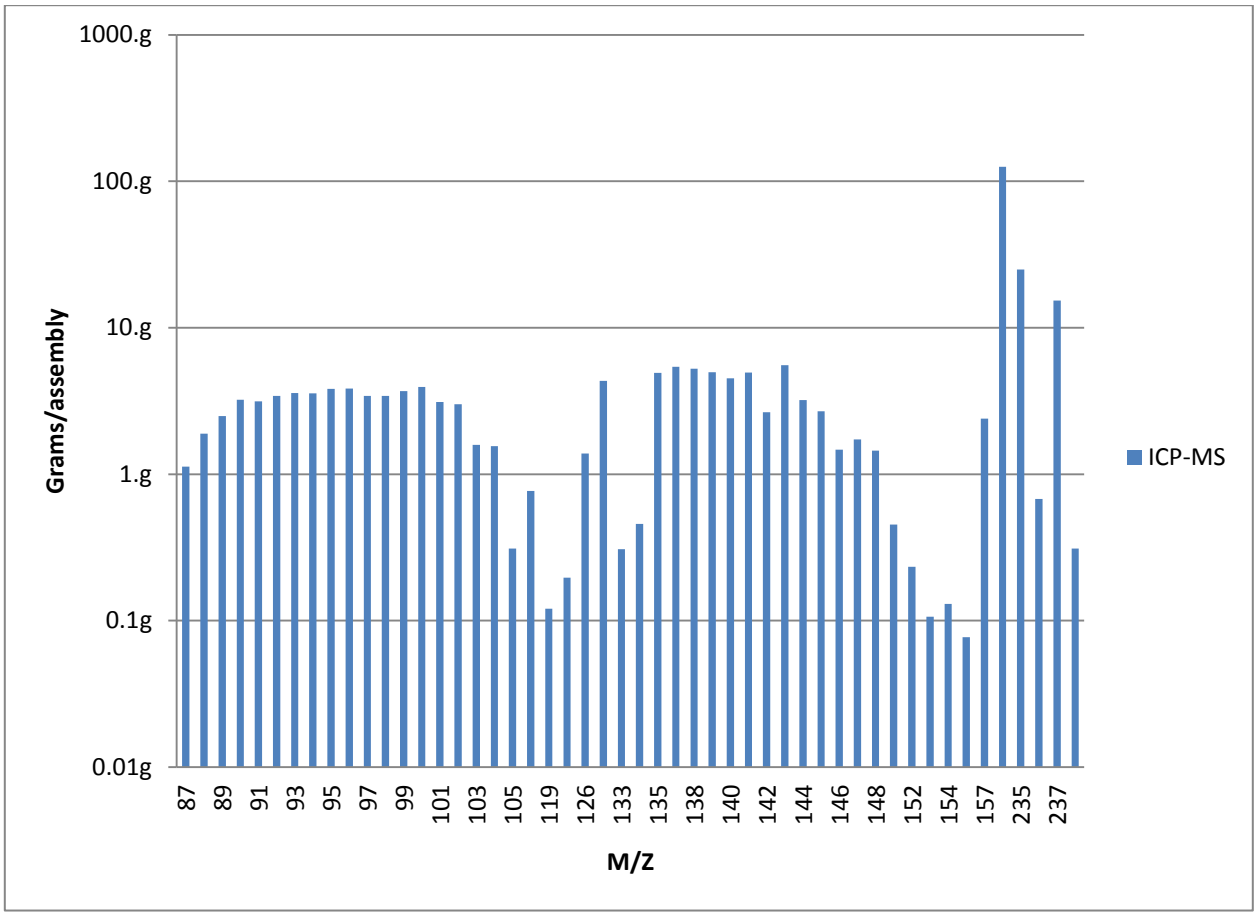

Figure 5 Log plot of the averaged grams per assembly at each mass per charge (m/z). The numbers on the x-axis are the m/z, and not mass numbers. Therefore, 137 contains a measurement for isotopes ^{137}Ba and ^{137}Cs.

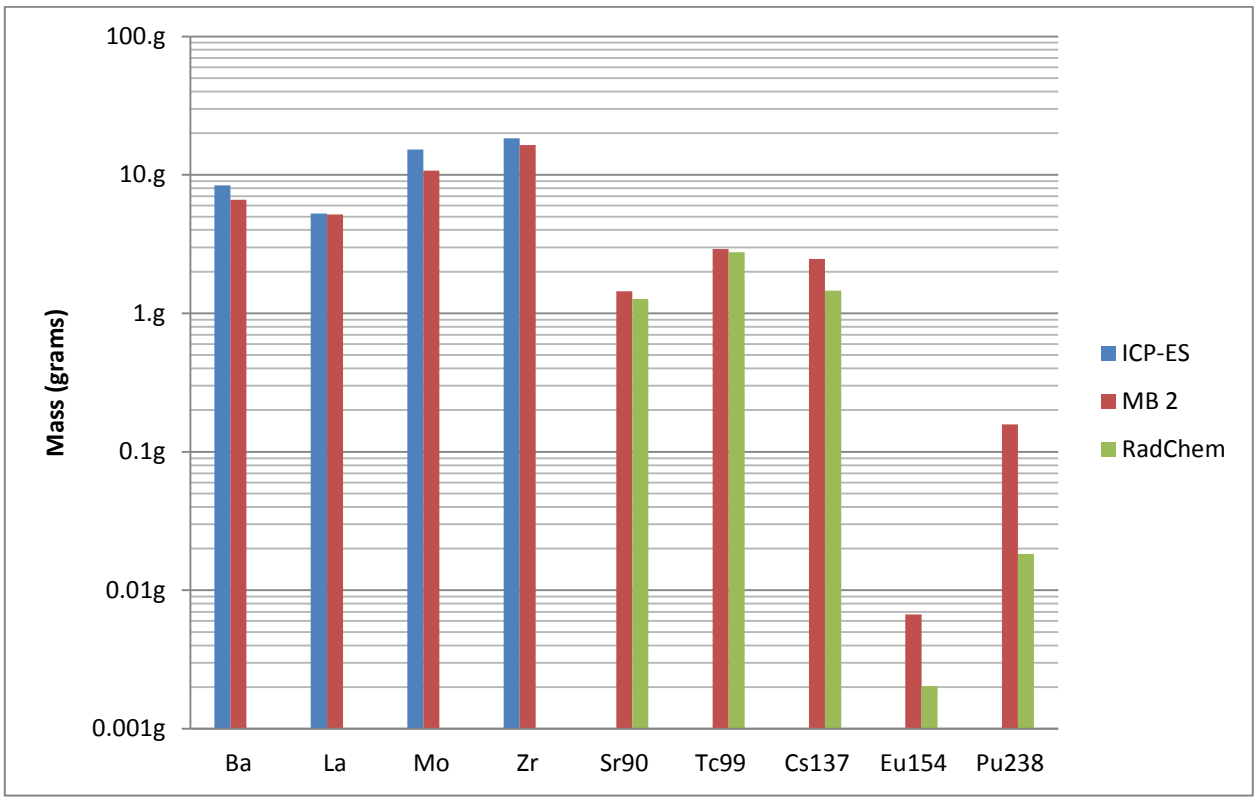

Figure 6 A few ICP-AES and counting measurements (RadChem) results with the values calculated by MONTEBURNS.

The comparison between MONTEBURNS and the measured data shown in Figure 6 indicates that the model does not perform as well for these elements or isotopes. The reason for poorer performance is not known, however, it is postulated that it could be due to modeling using and infinite lattice. Another potential difference is that ORR was a research reactor and had many experimental locations in which different targets were placed, some of which could potentially be highly absorbing or reflecting, these targets could affect isotope production in the fuel. This will affect the isotopic composition of the spent fuel assembly. There is uncertainty in the measurements which contributes to the differences between the models and the measured values. There is uncertainty introduced due to collecting samples inside Shielded Cells with manipulators. Also, each instrument contains a certain amount of uncertainty. The uncertainty due to the difficulty in working in Shielded Cells is hard to quantify.

CONCLUSION

Currently, an infinite lattice model of an ORR spent fuel assembly is able to calculate isotopics that agree fairly well with measured isotopics, especially for uranium and plutonium. These results indicate that an infinite lattice model of a spent fuel assembly for the ORR is a good starting point to modeling the burn-up in the research reactor. Furthermore, whether the model accounts for downtimes in the reactors operating cycle or just burns to the correct burn-up does not affect the values produced by the model. This is important for nuclear forensics work as often times specific reactor operations will not be known. Therefore, not having to know the up and down times an assembly experienced during its lifetime is advantageous. This may not be the case for all isotopes, such as strong absorbers or short lived isotopes and should not be used as a general rule. Another

finding is that when dealing with HEU fueled reactors, the starting values of ^{234}U and ^{236}U are important. Unfortunately, it is difficult to track down what were the exact starting amounts of ^{234}U and ^{236}U in a fresh fuel assembly. Further research should look into methods of solving this issue. Future work will involve expanding the models to a full core model if possible. Also, more measurements should be made with different techniques to single out certain isotopes for comparison.

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