ADDING REALISM TO NUCLEAR MATERIAL DISSOLVING ANALYSIS

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ABSTRACT

Two new criticality modeling approaches have greatly increased the efficiency of dissolver operations in H-Canyon. The first new approach takes credit for the linear, physical distribution of the mass throughout the entire length of the fuel assembly. This distribution of mass is referred to as the linear density. Crediting the linear density of the fuel bundles results in using lower fissile concentrations, which allows higher masses to be charged to the dissolver. Also, this approach takes credit for the fact that only part of the fissile mass is wetted at a time. There are multiple assemblies stacked on top of each other in a bundle. On average, only 50-75% of the mass (the bottom two or three assemblies) is wetted at a time. This means that only 50-75% (depending on operating level) of the mass is moderated and is contributing to the reactivity of the system.

The second new approach takes credit for the progression of the dissolving process. Previously, dissolving analysis looked at a snapshot in time where the same fissile material existed both in the wells and in the bulk solution at the same time. The second new approach models multiple consecutive phases that simulate the fissile material moving from a high concentration in the wells to a low concentration in the bulk solution. This approach is more realistic and allows higher fissile masses to be charged to the dissolver.

Key Words: dissolving, nuclear materials, realism

1 INTRODUCTION: H-CANYON

H-Canyon is the only operating nuclear chemical separations plant remaining in the United States. It is located at the 310-square-mile Savannah River Site in South Carolina and is owned by the U.S. Department of Energy. H-Canyon employs remote operations for most processes, including dissolving, separations cycles, and waste systems.

In the past, H-Canyon's missions have included recovering U-235, Np-237, and Pu-238 for use in defense, space, and commercial nuclear power purposes. Currently, H-Canyon maintains the capacity to safely and efficiently disposition a large inventory of excess nuclear material from across the Department of Energy (DOE) complex and used nuclear fuel from foreign and domestic research reactors.

H-Canyon processes involve large quantities of fissile solutions, including high enriched uranium and weapons grade plutonium, in geometrically unfavorable tanks. Therefore, criticality safety is of the utmost importance. H-Canyon has an extensive criticality safety program, including criticality safety evaluations/double contingency analyses for all processes, monthly criticality assessments/walkdowns, and criticality safety training for all personnel with access to the facility.

1.1 The Dissolving Process

Dissolving is the first step in processing material through H-Canyon. For each new type of material to be dissolved, the criticality safety of the operation must be established. A

Nuclear Criticality Safety Evaluation (NCSE) must be developed that provides Criticality Safety Limits (CSLs) and controls to ensure all normal and credible abnormal conditions remain subcritical.

There are two dissolvers in use in H-Canyon. One is a 12-foot-diameter, 8-foot-tall, vertical, cylindrical tank. The other is an 8-foot-diameter, 8-foot-tall, vertical, cylindrical tank. The dissolving process utilizes nitric acid as the solvent with various catalysts, if needed. Nitric acid, at the beginning of the dissolving process, is usually in the 4-6 Molar range.

An insert is placed in the dissolver pot to provide some control over geometry during the dissolution process. The insert is a basket-like device with ten long, cylindrical, vertical wells. Each well is 6 inches in diameter, ~18 feet long, and has hundreds of holes to allow for flow of nitric acid in and nitric-acid-fissile solutions out into the bulk solution. Figure 1 shows several views of the 10-well insert.

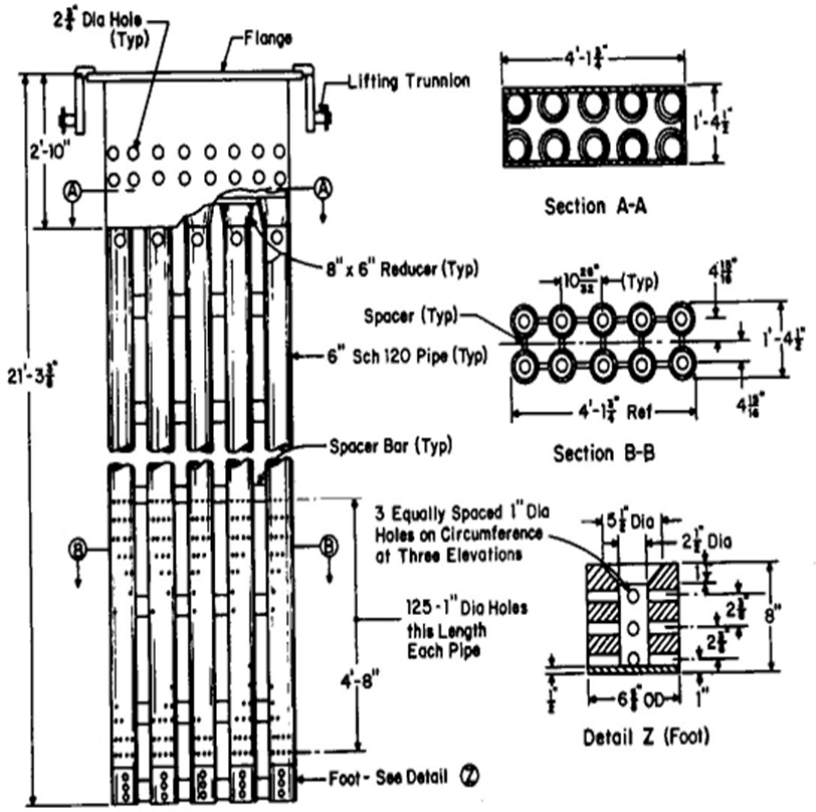


Figure 1 10-Well Insert

To charge the dissolver with nuclear material, a long, thin, vertical metal tube (called a bundle) is pre-loaded with nuclear material and lowered into each well in the insert. The bundles may be constructed of stainless steel if they are to be re-used, or the bundle may be constructed of aluminum if it is to be dissolved along with the nuclear material. A plug may be used to block various wells if the criticality safety evaluation determines that loading all ten wells is not safe for that particular material.

2 HISTORICAL APPROACH

There have been two major improvements recently in the criticality analysis of dissolving nuclear materials in H-Canyon. The first improvement is the development of the linear density method, and the second is the evaluation of various phases of dissolution. To fully appreciate the improvements, it is necessary to understand how these analyses have historically been performed.

In the past, criticality safety analyses for H-Canyon have largely ignored the laws of physics and chemistry for the sake of conservatism. For example, it was assumed that all the material charged to a well instantaneously dissolved and stayed inside the well at an arbitrary concentration of 150 g U/L. Also, even though it was assumed that the fissile material never left the well, it was assumed that the bulk solution in the dissolver pot instantaneously existed at an arbitrary concentration of 4.0 g U-235/L, which was established as the CSL. These assumptions ignored the laws of chemistry and physics because, in order for the dissolving process to occur, nitric acid molecules must move into the well from the bulk solution, break down the uranium metal by creating uranium-nitrate molecules, and then move back into the bulk solution to allow for fresh acid to move in to continue the dissolving process. Clearly, the uranium must move out of the wells and into the bulk solution if the dissolving process is to achieve its purpose.

Using this historical approach also double-counted the uranium mass in the dissolver and essentially modeled an upset condition (over-batch) as the normal condition. Obviously, the same atom of uranium cannot exist in two places at once. It must be either in the well or in the bulk solution. Also, if the bulk solution is already at the CSL, then there should not be additional material charged to the wells, as was modeled. That constitutes an upset condition, not the normal condition. The new dissolving phases model addresses this concern and is given in detail in Section 4.

In addition, to support the fissile concentration in the well at 150 g U/L, it was assumed that all the material existed (in solution) in the bottom portion of a well, with a varying height to accommodate the mass that was to be charged to each well. To illustrate this, assume that the volume inside the well per cm height of the well is 0.1825 L/cm. At the arbitrary 150 g U/L, the mass per cm height of the well is 27.375 g U/cm. Therefore, if 1.0 kg U was to be modeled in each well, the height of the 150 g U/L solution in each well would be 36.53 cm (1,000 g / 27.375 g/cm = 36.53 cm). This would make sense if the material to be modeled was in pieces of unknown shape or size that would collect in the bottom portion of the well. However, for the Used Nuclear Fuel (UNF) that has been analyzed, most of it is long, thin fuel assemblies that are well characterized. This leads to the first improvement that has been made – the linear density method.

3 LINEAR DENSITY

The linear density method takes credit for the physical distribution of the fissile material throughout the length of the fuel assembly. Before the linear density method is fully explained, it is important to understand the used fuel material that was modeled.

3.1 Used Fuel Assemblies

The used nuclear fuel (UNF), also referred to as spent nuclear fuel, that has been evaluated for dissolution in H-Canyon is an aluminum-based fuel. The fuel meat is typically a uranium-aluminum alloy with aluminum cladding and aluminum structural material. It often contains high enriched uranium (93.5 wt. % U-235 at the beginning of life) in fuel

plates that can be straight, slanted, or curved. The fuel meat in a typical assembly is about 60 cm (~2 ft) long. The fuel was irradiated in domestic or foreign research reactors, but the fuel originated in the United States.

The fuel assemblies are placed end-to-end inside an aluminum bundle at the L-Area basin, where they are currently stored at SRS, to be shipped to H-Canyon and loaded into the dissolver. The bundles are about 11 ft. long and 5" diameter, so 4 or 5 assemblies will fit into one bundle.

The University of Missouri Research Reactor (MURR) is the fuel with the highest U-235 loading of the used fuel stored in L-Area. It has an active fuel length of about 60 cm and contains about 775 g U-235 per assembly. Figure 2 shows a MURR fuel assembly.

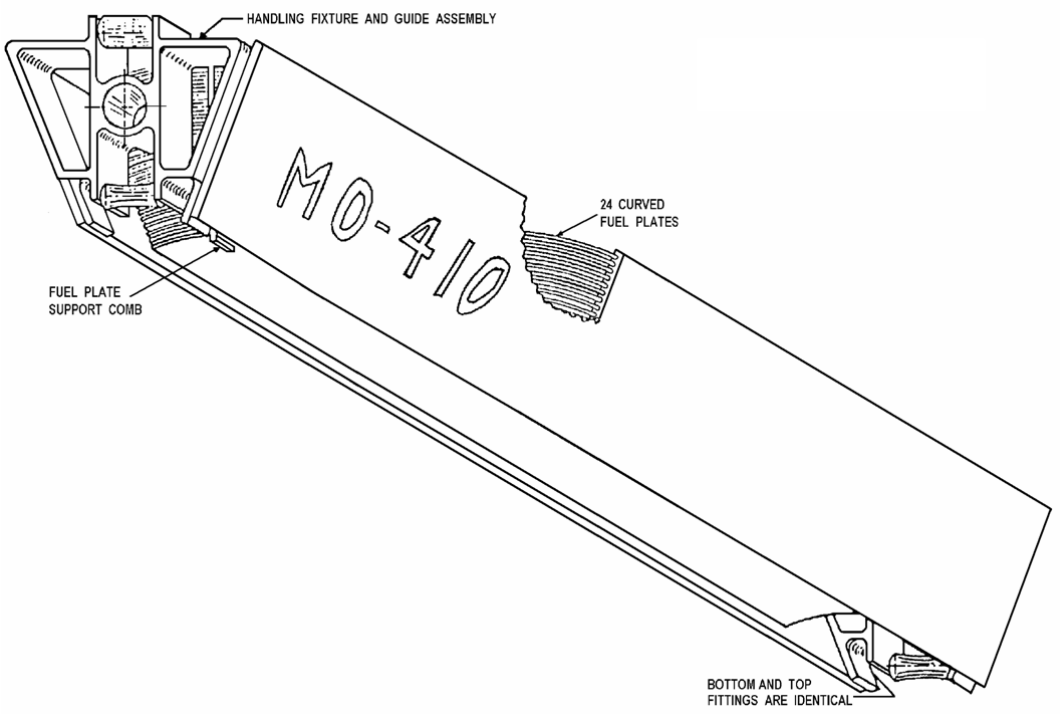


Figure 2 MURR Fuel Assembly

3.2 Calculation of Linear Density

Linear density is a straightforward parameter used to take credit for the physical distribution of the fissile mass throughout the length of the fuel assembly. For MURR fuel, the linear density is easily calculated. The linear density is 12.9 g U-235/cm [775 g U-235/60 cm = 12.9 g U-235/cm (393.7 g/ft)]. Using the value of 0.1825 L/cm for the volume inside the well per unit height of the well, 12.9 g/cm equates to a concentration of 70.8 g U-235/L (12.9 g/cm / 0.1825 L/cm = 70.8 g/L).

When this value is compared to the historical approach (150 g U/L), it is easy to see how over-conservative the historical approach was. How could the material exist at 150 g U/L when there is only 70.8 grams available per liter of solution inside the well? This question was the impetus for developing the linear density method.

Although it is simple to calculate the linear density of MURR or any other fuel stored in L-Area, the criticality analysis sought to find a maximum safe concentration in all ten wells.

This maximum safe concentration could be equated to a maximum safe linear density to which the candidate fuels could be compared.

SCALE 5/KENO VI [1] was used to model the dissolver with fissile material charged to all 10 wells with varying concentrations. For example, 58.8 g U-235/L was modeled in each of the ten wells, and k_{eff} was determined. Then 74.9 g U-235/L was modeled in each of the ten wells. The concentration was increased until k_{eff} exceeded k_{safe} (based on the validation of the code in comparison to experiments). 4.0 g U-235/L was also modeled in the bulk solution. The maximum safe concentration in the wells was found to be 81.8 g U-235/L [2], which corresponds to a linear density of 14.2 g U-235/cm (432 g U-235/ft). As discussed above, the linear density of MURR is 12.9 g U-235/cm. This indicates that MURR fuel, and therefore almost all fuels in L-Area, can safely be charged to the dissolver in all ten wells.

3.3 Remaining Conservatisms

Although excess conservatism was removed in order to model a more realistic scenario, there is still sufficient conservatism present. The first conservatism still present is the assumption that the fuel instantaneously exists in solution in the wells. In reality, it takes a long time (on the order of 12-24 hours) for the fuel to fully dissolve.

For this analysis, the material that was evaluated for dissolution is used fuel that has some amount of burnup. However, there was no credit taken for burnup. The U-235 mass and enrichment were assumed to be at their beginning-of-life values. Also, although the maximum expected beginning-of-life enrichment was 93.5 wt. % U-235, a more conservative 95 wt. % U-235 was modeled for all cases.

Another important conservatism that remains is the assumed corrosion of the wells. It is assumed that the inner diameter of each well has been increased by a half inch. This increases the diameter by about 9%, which has a significant effect on reducing neutron leakage and increasing k_{eff}.

4 DISSOLVING PHASES

The method of analyzing the dissolving process in phases helped to address another unnecessary conservatism mentioned in Section 2 above. The dissolving phases model was developed for an analysis of dissolving enriched uranium – plutonium (EU-Pu). This material is in a dissolvable can, which is placed inside a reusable bundle.

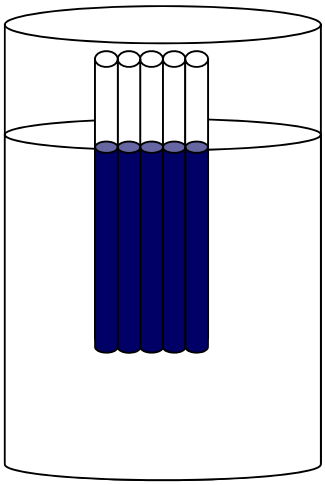
4.1 Definition of Dissolving Phases

If one considers the actual dissolving process, the concept of dissolving phases naturally follows. At the beginning, what is referred to as Phase 0, there is clean nitric acid solution as the bulk solution in the dissolver. There is also fissile material in the ten wells. Initially, this material is in dry, discreet, solid pieces. However, acid quickly dissolves the outer canister that holds the fissile material. In a move to maintain sufficient conservatism, the fissile material is modeled as being instantaneously in solution inside the wells. As discussed in Section 3, if the material configuration is well defined (such as with used nuclear fuel), we can limit the maximum concentration modeled in the wells. However, many nuclear materials that H-Canyon has the capacity to process do not have well-defined configurations. The size, shape, and mass of the pieces to be dissolved may be irregular and/or classified. Therefore, no credit is taken for the physical distribution of mass in the wells. Because credit cannot be taken for well-characterized geometry (like for the used fuel discussed in Section 3), a maximum concentration of 150 g/L is used in the wells, and the height of solution is

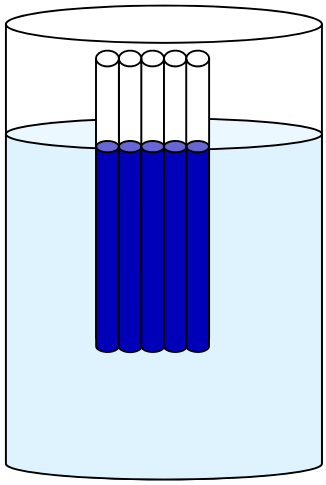
varied to account for varying mass. For example, if 3 kg was to be modeled in each well, there would be a higher 150 g U/L-solution height than if 2 kg was to be modeled in each well. Also, if a certain amount of mass would not fit in the 150 g U/L-solution at the maximum dissolver solution height, then the remaining mass would be modeled as a metal sphere at the bottom of the well.

Once the dissolving process begins, fissile material slowly moves from inside the wells into the bulk solution. To model this, the course of this process is divided into eleven phases – Phase 0 through Phase 10. If fewer phases had been chosen, a maximum peak in k_{eff} may not have been identified. If more phases had been chosen, it would unnecessarily add computational time. At Phase 0, all the fissile material is in solution at 150 g U/L, and clean, nitric acid solution is used for the bulk material. At Phase 1, 10% of the fissile material is assumed to have moved out of the wells and into the bulk solution. Since we keep the concentration in the wells at a conservative, constant 150 g/L, this loss of mass in the wells is modeled as a lower 150 g/L-solution height inside the well. Also, the mass that is lost from the well is assumed to go into the bulk solution, thereby increasing the fissile concentration in the bulk solution. The change in bulk fissile concentration is calculated by dividing 10% of the initial mass by the minimum volume expected in the dissolver. The 10% of the initial mass is the amount that is assumed to have left the wells in Phase 1. Therefore, the snapshot for Phase 1 models less fissile material in the wells and a slightly higher fissile concentration in the bulk solution than in Phase 0.

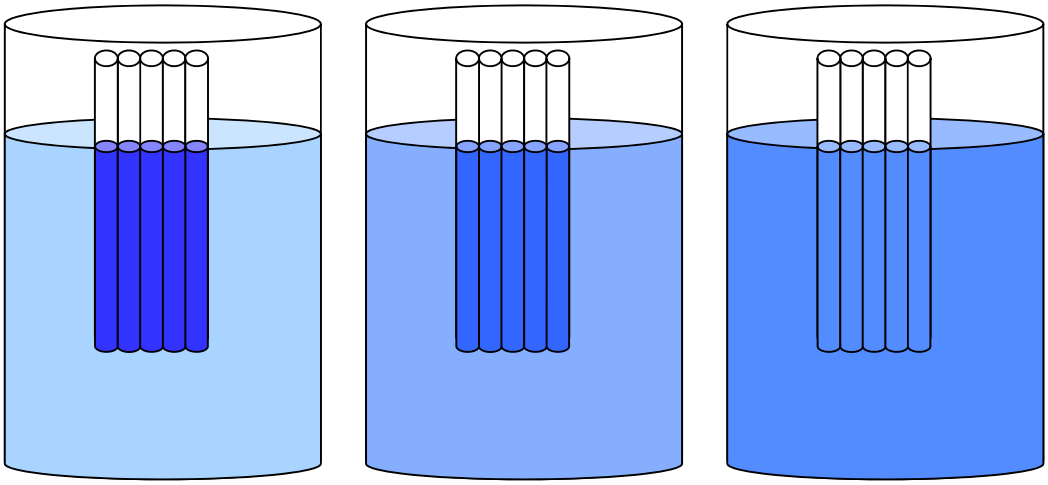
Similarly, in Phase 2, 20% of the initial fissile material is assumed to have moved out of the wells and into the bulk solution. The bulk fissile concentration is expected to increase proportionately. Phases 3 through 9 follow suit until ultimately Phase 10 is reached. At Phase 10, 100% of the fissile material charged to the dissolver is modeled as being in the bulk solution; there is no high-concentration fissile material remaining in the wells. To illustrate the concept of dissolving phases, a figure was created as if there were only five phases (Phase 0 through Phase 4). This is shown in Figure 3. However, for this NCSE, eleven phases were used (Phase 0 through Phase 10).



Phase 0



Phase 1



Phase 2 Phase 3 Phase 4

Figure 3 Dissolving Phases

4.2 Change in k_{eff} Over Dissolving Phases

A significant change in k_{eff} is noticed if k_{eff} is graphed over the dissolving phases. Typically, k_{eff} increases from Phases 0 through 5 or 6. This is to be expected since the bulk solution (which acts as a reflector to the material in the wells) is increasing in fissile solution. Also, as material moves out into the bulk solution, it is better moderated, and k_{eff} increases. Once the maximum k_{eff} is reached around Phase 5 or 6, k_{eff} drops significantly. This also makes sense because, after Phase 5, the majority of the fissile mass is located in the bulk solution at a low concentration. At Phase 10, all the fissile material is at a low, subcritical concentration in the bulk solution, so it has the lowest k_{eff} of all the phases.

The criticality safety evaluation that first utilized the dissolving phases was written for the purpose of establishing the safety of dissolving EU-Pu metal. This material may contain a significant amount of both high enriched uranium and weapons-grade plutonium. The fact that there were two fissile isotopes of concern greatly complicated the analysis. Because the two fissile isotopes have different fission spectrums and different fission threshold energies, the change in k_{eff} over the dissolving phases was different depending on the ratio of U-235 to Pu-239. Figure 4 shows the change in k_{eff} over the dissolving phases for 4.5 kg U-235 and various amounts of Pu-239.

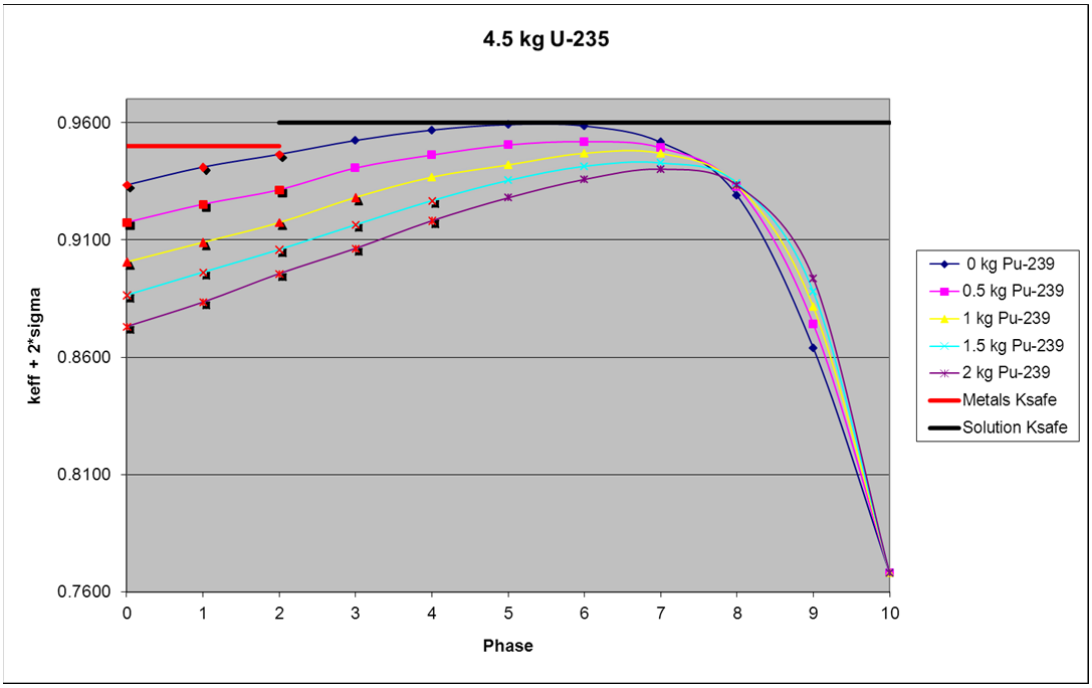


Figure 4 Change in k_{eff} over Dissolving Phases

In general, when the uranium mass exceeds the plutonium mass, k_{eff} goes down when more plutonium is added. This phenomenon is due to two factors having to do with the neutron energy spectrum of the system and how it changes when more plutonium is added. Figure 5 shows the average energy group at which fission occurred. The energy groups range from 0 (being very fast, like just after fission) to 238 (being very slow, like after being moderated completely). Therefore, the higher the energy group, the slower the neutron was travelling before it caused a fission. The lower the energy group, the faster the neutron was travelling. The average energy groups shown in Figure 5 are also a good indication of the average energy of the neutrons in the entire system. So, as the energy groups go down when plutonium is added, the average energy of the neutrons in the system is going up. This is referred to as a harder, or faster, neutron spectrum.

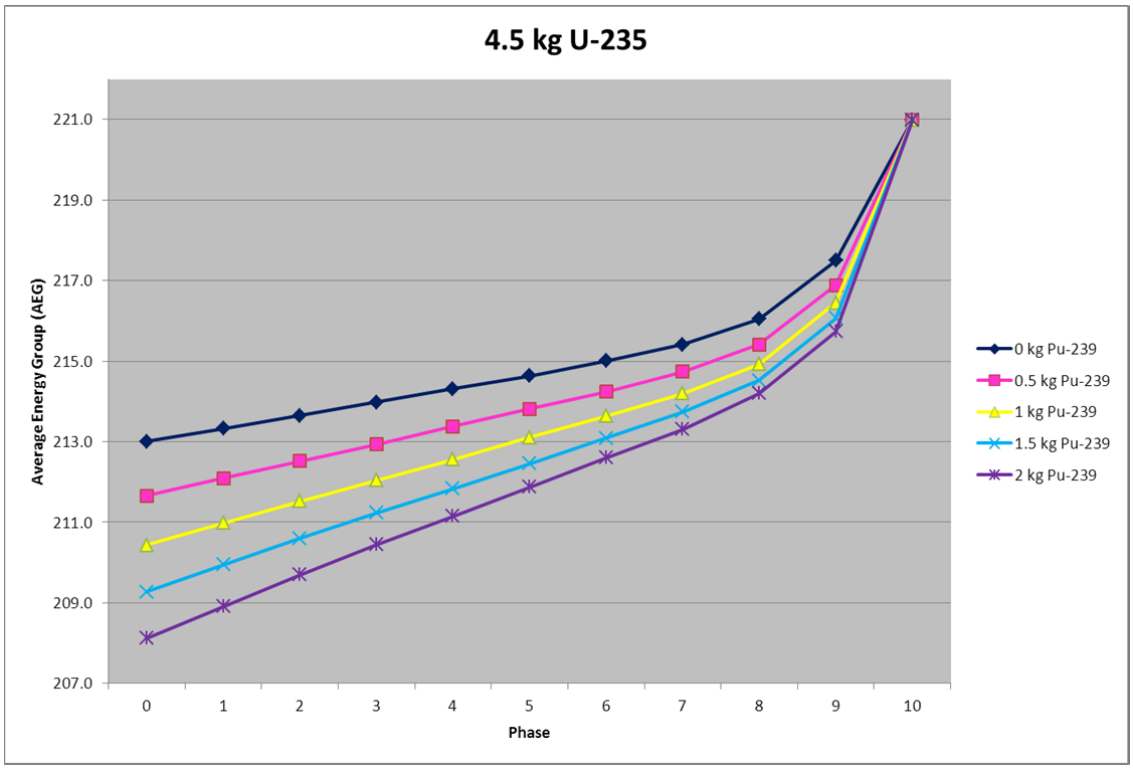


Figure 5 Average Energy Group

The consequence of a faster spectrum is best understood by examining the cross section graph for these nuclides. The cross section is directly related to the probability of a certain reaction (fission or absorption) occurring. Figure 6 shows the fission cross section for U-235 and Pu-239 and the absorption cross section for Pu-240. The U-238 cross sections are not included because they are too low to have any significant effect on the system. It is important to note that neutrons move across the graph *from right to left* (at first, a neutron's energy is very high, then it decreases as it interacts with moderating material).

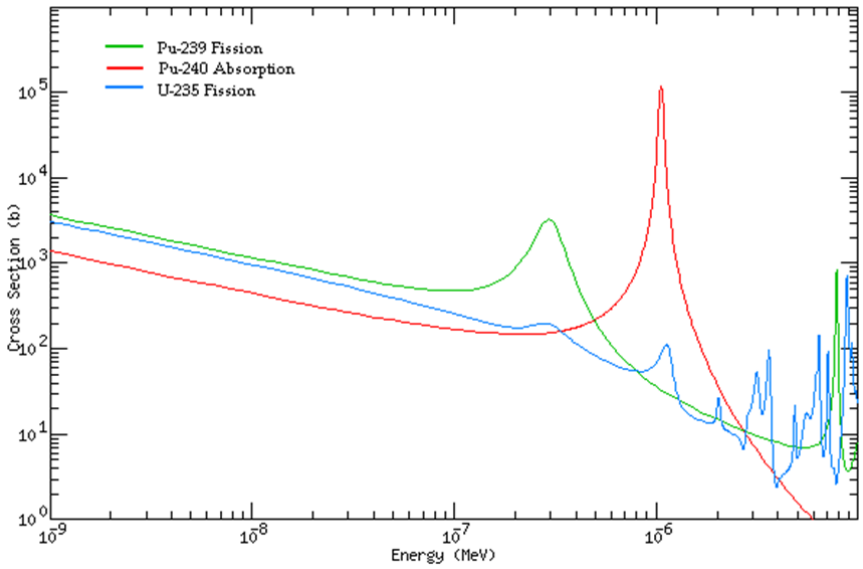


Figure 6 Cross Sections [3]

As plutonium is added to a uranium system, the neutron spectrum gets harder (shifts to the right), and two things occur. First, the Pu-240 absorption peak has a large effect. As the neutrons are slowing down, they encounter this large absorption peak at around 1 eV before they are allowed to reach favorable fission energies. Second, there is a large Pu-239 fission peak at around 0.3 eV. The neutrons that make it past the Pu-240 absorption peak are then likely to cause a Pu-239 fission. This means fewer neutrons are available to make it to the thermal energies needed for U-235, which accounts for the majority of the mass. Therefore, k_{eff} goes down. In systems that are predominately plutonium, this effect is not seen.

4.3 Remaining Conservatisms

Similar to the linear density method, there are plenty of conservatisms remaining in the models that utilize dissolving phases. The same assumptions of the material instantaneously existing in solution and the well inner diameter corroding a half inch are also present in the dissolving phases model.

Also, since part of this method models changes in fissile concentration in the bulk solution, the assumed volume is very important. The largest change in concentration is achieved when the dissolver is at its lowest operable volume. A conservatively low volume of 5,000 liters was assumed for the concentration changes. However, H-Canyon will normally operate their small dissolver at around 7,000 liters. The larger dissolver will operate at even higher volumes.

The molarity of the acid in the dissolver is assumed to be zero in the SCALE models. As molarity is increased, the number of hydrogen atoms decreases. Assuming zero molarity is conservative because, inside the wells where there is higher fissile concentration and relative lack of hydrogen atoms, increasing molarity causes a decrease in k_{eff}. In reality, the dissolver will be operated at 4-6 molar nitric acid, and k_{eff} will be lower than modeled.

Also similar to the linear density method, the isotopic distributions are modeled conservatively. For the plutonium in these models, more Pu-239 and less Pu-240 are modeled than what actually exists.

5 RESULTS

Both of these new methods resulted in an increase in the criticality safety limits (CSLs) compared to the historical approach. The increase in limits has a direct savings for the facility in time and money.

5.1 Linear Density Results

Using the historical approach, only 1.0 kg U-235 could be loaded in each of the ten wells in the dissolver insert. However, the new linear density method allows up to 3.4 kg U-235 to be loaded into each of the ten wells. As mentioned previously, MURR fuel is considered the bounding fuel and consists of a significant portion of all the fuels available for processing. Using the historical approach, if MURR was to be charged to the dissolver, only six wells could be loaded. With the new linear density method, MURR can be loaded in all ten wells.

There are also other constraints that affect the projected savings from the linear density method. The cask used to ship the fuel assemblies may only hold up to nine bundles per shipment. This means that all ten wells would not be utilized; one would remain empty. So, instead of charging six bundles of MURR, the operators may now charge 9 bundles, thereby increasing efficiency by 50%. The dissolution of MURR, and similar high-U-235-loaded fuels that would benefit from the linear density method, was estimated to take one and a half

years. However, with the linear density method, it would now only take one year, thereby saving six months worth of time. Six months of processing time in H-Canyon is equivalent to ~$75 million saved.

5.2 Dissolving Phases Results

Using the historical approach, only 3.4 kg U-235 and 1.0 kg Pu-239 could be loaded in up to five of the ten wells in the dissolver insert. However, the new dissolving phases method allows up to 4.7 kg U-235 and 2.0 kg Pu-239 to be loaded into up to five of the ten wells. This is a significant increase in the CSLs.

This campaign was originally scheduled to run 48 weeks. However, using the dissolving phases method in the criticality safety evaluation shortens the run time to 36 weeks. Also, since the material will be able to be processed in fewer dissolver batches, less cold chemicals will be needed and less waste will be produced. All of these savings equate to $39 million.

6 CONCLUSIONS

Historically, excess conservatisms have been added on to criticality safety analyses in the hopes of reducing risk. However, there is a risk in modeling unrealistic conditions, because one cannot know what the real margin is between safe and unsafe operations. It also unnecessarily restricts operations. It is imperative to model more realistic conditions when possible. The result is a sufficiently conservative analysis that allows the facility to operate more efficiently and save time and money. It also results in a higher level of confidence and awareness of the true margin between safe and unsafe conditions.

7 REFERENCES

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