

DESIGN OF A SUBCRITICAL AQUEOUS TARGET SYSTEM FOR MEDICAL ISOTOPE PRODUCTION

An Undergraduate Research Scholars Thesis

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ABSTRACT

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The United States consumes almost half of all medical isotopes produced worldwide, and relies on foreign sources for nearly its entire supply. These isotopes are produced in nuclear reactors which are very costly to construct. A domestic supply may be realized if research reactors at universities and national laboratories can be enhanced with isotope production capabilities. This research is dedicated to the design of an aqueous target system that can be appended to existing research reactors for this purpose. The design aims to combine attributes of solid target irradiation by conventional reactors and in-solution production by aqueous homogeneous reactors in order to realize some of the benefits of each method. The benefits for the former include using existing reactors as the external neutron source hence reducing the investment capital significantly. The benefits for aqueous homogeneous reactors are numerous and include higher efficiency, substantial reduction in waste, lower fuel cost, and reduced isotope separation complexity. Utilizing a flowing fuel design will enable continuous isotope separation and more efficient heat removal, as well as eliminate some of the complications that have plagued solution fueled reactors in the past

such as power oscillations and fuel precipitation. The aqueous target system described in this thesis is designed for the Annular Core Research Reactor (ACRR) at Sandia National Laboratories. The system is optimized for ^{99}Mo production, as this is the medical isotope in highest demand and used in a majority of all medical diagnostic procedures excluding x-ray imaging. The optimized production rate is calculated to be $3044 \text{ Ci}_{6\text{-day}}$ per week which accounts for 50.7% of domestic consumption.

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CHAPTER I

INTRODUCTION

This report is the second stage of the design of an aqueous target system for medical isotope production. The first stage proposed a reactor named the Flowing Fuel Aqueous Homogeneous Reactor (FFAHR) that was different from previous AHR designs in that the fuel flowed through the core allowing for more efficient heat removal and continuous isotope production. The flowing of the fuel was also intended to eliminate some of the technical deficiencies plaguing previous AHR designs such as power oscillations, fuel precipitation, and high temperature corrosion. The low power density of the first design made the ^{99}Mo production insufficient, and hence the need for a new design was realized.

The second stage of this design utilizes many features of the first design. The primary difference between the two systems is that the aqueous target will be subcritical by itself, and externally driven by a research reactor. The reactor chosen as the neutron source is the Annular Core Research Reactor (ACRR) at Sandia National Laboratories, although the current design should be able to be adapted for other research reactors as well. The reason for choosing the ACRR is purely one of familiarity and convenience.

The FFAHR was intended to produce five of the most commonly used radioisotopes in the medical industry. These radioisotopes are ^{99}Mo , ^{133}Xe , ^{89}Sr , ^{90}Y , and ^{131}I . The intent was to produce these isotopes at a rate that would make it a financially viable and competitive option when compared to the research reactors that currently produce the global supply of these isotopes. Of the

five isotopes listed above, ^{99}Mo is by far in the highest demand, and the insufficient production of this isotope by the FFAHR made it unlikely to compete with foreign sources.

It should be remembered that bigger is not always better in this case. Ideally, ^{99}Mo would be produced in small amounts in hundreds of facilities across the world so that an unplanned outage of a large scale production facility would not cause widespread shortages. The difficulty in realizing this is that nuclear reactors are extremely expensive, and small scale production facilities simply do not pay the bills. In addition, the licensing process in the United States makes it nearly impossible to physically realize any new reactor design. This is the reason that the current design is proposed as a target system that could be adapted to existing research reactors.

I.A. Need for Domestic Production of Radioisotopes

This section describes the use of radioisotopes in the medical industry as well as an overview of the current supply and demand of these isotopes. The need for radioisotope production in the United States in particular is emphasized due to its dependence on foreign supplies and its susceptibilities due to this dependence.

I.A.1. Radioisotopes Used in the Medical Industry

The medical industry uses radioisotopes for a variety of procedures. These procedures can be split into two categories. The first of these is diagnostic imaging which typically utilizes the long range and monoenergetic properties of gamma decay. [1] The second category is the treatment of various types of cancer through the short range of high energy beta particles to irradiate tumors without delivering a dose to the whole body. The most prominent isotopes used in the medical

industry today, which can be produced with significant yields from a nuclear reactor, are ^{99}Mo , ^{133}Xe , ^{89}Sr , ^{90}Y , and ^{131}I . [2]

Other isotopes used in the medical industry include positron emitting radioisotopes for use in positron emission tomography (PET). These are low Z isotopes that are typically produced by accelerators and have little to no fission yields. These isotopes include ^{11}C , ^{13}N , ^{15}O , and ^{18}F . Accelerators can also be used to produce the high Z isotopes that are typically produced in reactors, but with much less efficiency and very low yields.

I.A.1.1. ^{99}Mo

The primary use of ^{99}Mo is to produce $^{99\text{m}}\text{Tc}$. ^{99}Mo decays by beta emission with a 65.94 hr half life into $^{99\text{m}}\text{Tc}$. [3] $^{99\text{m}}\text{Tc}$ then decays with a half life of 6.01 hr by internal conversion while emitting a 143 keV gamma particle. [4] The primary use of $^{99\text{m}}\text{Tc}$ is in medical diagnostic imaging. The $^{99\text{m}}\text{Tc}$ can be injected into the body with a pharmaceutical, which transports it to the region of the body that is to be imaged. The low energy gamma ray is then detected giving an image of the region that the $^{99\text{m}}\text{Tc}$ has populated. Such images of the lungs and skeleton can be seen in Figure I.1.

$^{99\text{m}}\text{Tc}$ has a wide range of imaging capabilities that is only limited by the effectiveness of the pharmaceutical that transports it to the site for imaging. In addition to the lungs and skeleton shown in Figure I.1, $^{99\text{m}}\text{Tc}$ can be used to image the vascular system, intestines, brain, heart, thyroid, and liver among other organs. $^{99\text{m}}\text{Tc}$ is commonly used to detect stress fractures that do not show up clearly in X-ray images.

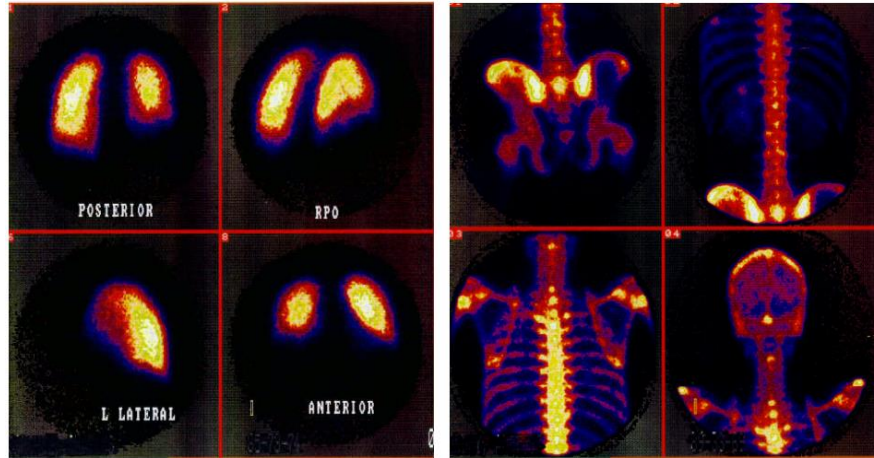


Figure I.1. Images taken with ^{99m}Tc of the lungs (left grid) and of the skeleton (right grid). Figure courtesy of Parma. [4]

Because of the extremely short half life of ^{99m}Tc , it would be inconvenient to try to sell this isotope in pure form. Instead, hospitals typically purchase ^{99}Mo and use a device called a ^{99m}Tc generator that separates the ^{99m}Tc from the ^{99}Mo as it becomes available. Over 80% of diagnostic imaging procedures, excluding X-ray imaging, use ^{99m}Tc . [5]

I.A.1.2. ^{133}Xe

The primary use of ^{133}Xe is for lung ventilation studies known as lung perfusion. ^{133}Xe decays by beta emission with a 5.243 day half life, while also emitting low energy gamma particles. [3] Since ^{133}Xe exists in a gaseous state at room temperature, the radioactive gas can be inhaled by the patient and scintillation cameras can be used to detect various amounts of radioactivity in different regions of the lungs. This procedure allows the physician to assess how well the lungs are functioning by detecting pulmonary embolisms, determining how much blood is flowing to the

lungs, and determining which areas of the lungs are capable of ventilation. [6] A lung scan using ^{133}Xe can be seen in Figure I.2.

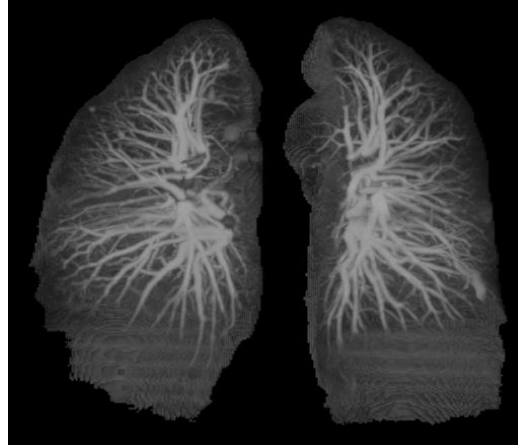


Figure I.2. Lung perfusion scan using ^{133}Xe . Figure courtesy of Johnson. [7]

I.A.1.3. ^{89}Sr

Various forms of cancer including prostate, breast, and lung cancer are accompanied by painful bone metastases. ^{89}Sr is a beta emitting radionuclide with a half life of 50.53 days. [3] ^{89}Sr is found to localize in areas of active bone formation when consumed by the patient. While ^{89}Sr is not a cure for any type of cancer, it has been found to significantly relieve the pain of bone metastases. The mechanism of this pain relief is unknown. One proposed mechanism is that the beta radiation stops the tumor from releasing pain producing enzymes. [8] Pain relief can occur in as few as 7 days after treatment and last for as long as 6 months.

I.A.1.4. ^{90}Y

Historically, liver cancer has been one of the most resistant cancers to chemotherapy. ^{90}Y is another beta emitting radioisotope with a half-life of 64 hr. [3] Treatment of liver cancer has recently utilized ^{90}Y in microsphere beads that are injected into the tumor where the short range of the beta radiation kills cancer cells without reaching un-affected portions of the body. [9] Although the dose administered cannot safely be high enough to cure the cancer, the treatment has been shown to increase the lifetime and the quality of life for patients suffering from liver cancer. In addition, ^{90}Y has been used for its beta radiation to sever pain-transmitting nerves in the spinal cord as well as to treat inflamed joints and rheumatoid arthritis. [10] It's only deficiency with respect to other beta emitters is that it is not attracted to a specific region of the body and hence must be injected with a very fine needle to the site of the tumor.

I.A.1.5. ^{131}I

The use of ^{131}I has been primarily as a treatment of thyroid cancer. ^{131}I is a beta emitting radioisotope with a half life of 8.02 days. [3] The treatment mechanism is similar to that of liver cancer using ^{90}Y , yet much more efficient. The thyroid absorbs iodine in order to produce the hormones that regulate the body's metabolism. In patients with thyroid cancer, it is often the case that cancer cells will also absorb iodine from the blood stream, and if the iodine is radioactive in the form of ^{131}I , the beta radiation emitted will help to kill these cancer cells. [11] This treatment can be as simple as taking a ^{131}I pill because the thyroid can be made to absorb nearly 100% of the iodine in the bloodstream by starving the thyroid of iodine for weeks before treatment.

I.A.2. *Global Supply and Demand*

The need for domestic production of the isotopes mentioned above cannot be overstated. The United States may be even more dependent upon foreign sources for these isotopes than it is on oil. Nearly the entire global supply of these isotopes is produced by only five reactors worldwide. [5] In addition, none of these reactors are in the United States and all five of them are over 40 years old and can be expected to be decommissioned within the next decade. All five of these reactors are also subsidized by their respective governments. Two more reactors have recently entered the market in Australia and Poland, but their contribution to the global supply cannot yet be determined. These reactors are listed in Table I.1 below.

Table I.1. List of current radioisotope producing reactors which produce nearly the entire global supply of high Z isotopes used in the medical industry. Table courtesy of the IAEA. [5]

Existing research reactors used by large-scale producers						
Country	Name	Thermal power, MW	Thermal neutron flux, n/s/cm ²	Target type	Maximum annual operation, days ³	Typical share of production %
Canada	NRU	135	4.0e14	HEU	315	40
Netherlands	HFR	45	2.7e14	HEU	290	30
Belgium	BR-2	100	1.0e15	HEU	115	10-15
South Africa	Safari-1	20	2.4e14	HEU	315	10-15
France	OSIRIS	70	1.7e14	HEU	220	5-8
<i>Australia</i>	<i>OPAL</i>	<i>20</i>	<i>3.0e14</i>	<i>LEU</i>	<i>340</i>	<i>yet to enter market</i>
<i>Poland</i>	<i>MARIA</i>	<i>30</i>	<i>3.5e14</i>	<i>HEU (for Covidien)</i>	<i>200</i>	<i>not known</i>

Relying on foreign sources for these isotopes means longer travel times between the production facility and the hospitals that uses them. With a half life of 66 hr for ^{99}Mo , this reduces the amount of product received for a given purchase price. More importantly however, relying on foreign sources which each provide such a large fraction of the global supply makes the United States highly susceptible to unplanned or extended outages. A clear example of such an outage occurred recently at the NRU facility in Canada from May 2009 to July 2010. [5] During this time, the other four reactors struggled to meet the demands of the market. At any given time during this period in the United States, 20-70% of all diagnostic imaging procedures were either cancelled or postponed. [5] From these experiences, it seems that the best course of action would be to build a series of reactors with limited isotope production that would supply isotopes to smaller geographical regions. In this case, the market would not be as significantly affected during unplanned outages of a single reactor.

Currently, the United States uses the same amount of ^{99}Mo as the rest of the world combined. The domestic consumption is estimated at 6,000 $\text{Ci}_{6\text{-day}}$ per week (a $\text{Ci}_{6\text{-day}}$ is the activity in Ci, 6 days after it leaves the producer's facilities). This consumption is expected to grow at a rate of 5-10% per year. [4] The estimated revenue generated by the five reactors listed above from the United States alone is \$150 million per year from just ^{99}Mo . [4]

I.B. Current Production Method

Designing a reactor for radioisotope production necessitates an understanding of the current methods of production so that various figures of merit can be developed for comparison. This section will focus on the advantages and disadvantages of the current production methods so that the incentive for a new design can be clearly understood.

I.B.1. Solid Target Irradiation

Nearly the entire global supply of radioisotopes for use in the medical industry is produced by solid target irradiation. With this production method, a solid target made of a neutron absorbing material is placed in a neutron beam or near the core of a nuclear reactor. The material is chosen based on the isotopes to be produced. For medical isotope production, the targets are made of highly enriched uranium (HEU) which is over 90% ^{235}U . [2] The ^{235}U atoms fission upon absorbing a neutron and the resulting distribution of isotopes are neutron rich and radioactive. Included in this distribution are the five isotopes mentioned in section I.A.1.

Particle accelerators have been used to produce neutron beams by spallation in which protons are accelerated into a material which absorbs the protons and subsequently emits neutrons. Particle accelerators and cyclotrons are typically used to produce short-lived, low Z isotopes, such as ^{11}C , ^{13}N , ^{15}O , and ^{18}F , which are not typically produced from the fission of uranium, but can be used for positron emission tomography. Unfortunately, they do not efficiently produce ^{99}Mo or other high Z isotopes at quantities suitable to match the medical industry's demand. In addition, the specific activity of the product is simply too low to compete with the product attained by irradiation in a nuclear reactor. One nuclear reactor using HEU can produce the same amount of ^{99}Mo as 160 accelerators. [12]

For this reason, it is no surprise that nuclear reactors are used to produce nearly the entire global supply of high Z isotopes for the medical industry. As previously mentioned, this supply is produced almost entirely by only five nuclear reactors. There are smaller reactors capable of producing these isotopes, but they do so infrequently and in small amounts, or are used primarily as research reactors. HEU is used in the large scale production of medical isotopes because the higher

concentration of ^{235}U allows for a larger production rate. Although the use of HEU creates a large supply of ^{99}Mo , it is criticized for the larger risks associated with nuclear security and proliferation concerns. This has caused regulatory bodies such as the IAEA to call for medical isotope producing reactors that use low enriched uranium (LEU) targets. [13]

I.B.2. Inefficiencies

For medical isotope production by a nuclear reactor, solid target irradiation is a rather inefficient process. This is due in part to the high power density ratios between the fuel of the reactor and the HEU target. It is not unusual to have these ratios be as high as 100:1. [2] With such a large ratio, effectively 99% of the neutrons that could be used if the target were inside the core are wasted. It is expected that one might ask why the targets are not placed inside the core. This is because the core is typically designed to be in steady state operation for months or even years at a time to maximize the burnup of its fuel. The targets on the other hand are typically only irradiated for time scales on the order of days before the activity of the desired isotope has reached its saturation value. By placing the target in the vicinity of the core, the target can be irradiated without contributing to the chain reaction and subsequently be removed while the reactor is still in operation.

After the target is irradiated, it is dissolved in solution which allows for isotope separation by a series of sorbents and chemical treatment. Once the separation stage is complete, the remaining fuel in solution is stored as high level waste (HLW). It is not economically attractive to attempt to form this solution into a new solid target because this process would be more expensive than the already costly process of fabricating fresh solid targets. [2]

The isotopes of interest for the medical industry typically have half-lives on the order of days or weeks. The half life of the most profitable medical isotope, ^{99}Mo , is only slightly below 66 hours. For this reason, it is desirable to separate the isotopes as quickly as possible so that the product does not decay entirely before it can be sold. Any unnecessary step in this separation process is costly. As will be seen in section I.C, the dissolution of the fuel in solution is unnecessary if the target exists in this form throughout its irradiation.

I.B.3. *Benefits*

There are benefits to the solid target irradiation method using either a nuclear reactor or an accelerator. The reactors that typically perform solid target irradiation are either pool type reactors, which make it easy to place samples near the core, or they have beam ports that allow for irradiation in a dry environment. An example of such a reactor is the TRIGA nuclear reactor at the Nuclear Science Center at Texas A&M University. In fact, the “I” in TRIGA stands for isotope production.

The use of such reactors allows for great flexibility. For instance, many of these reactors have multiple beam ports that can be used for neutron scattering experiments, neutron tomography, or other forms of research. In addition, these reactors utilize typical heterogeneous lattice cores similar to those found in the power industry. For this reason, reactors such as the TRIGA reactor are commonly used for training senior nuclear engineering students or beginning nuclear reactor operators.

A similar argument holds in favor of solid target irradiation by an accelerator or cyclotron. Accelerators can be used in a number of research areas when they are not being used to irradiate solid HEU targets or other neutron absorbing materials. Accelerators are often used in materials

science to deliver large doses of radiation damage in short time intervals to determine the change in the properties of new and innovative materials in a radiation field. In addition, accelerators can easily be turned on or off, have little security or proliferation risks, and may be scaled to match the local demand if used for isotope production.

I.B.4. Attempts and Motivation for Using LEU

The technology of utilizing LEU targets for use in reactors is still under development. There are small scale-producers in both Argentina and Australia that use LEU for isotope production. [13] These facilities have shown that using a lower enrichment increases the purity of the radioisotopes and their yield per unit power. It also allows for the recovery of isotopes other than ^{99}Mo such as ^{131}I . Although the use of LEU seems to be more beneficial because it decreases proliferation risks, increases the variety of isotopes produced, and has a higher purity of ^{99}Mo ; it is not economically favorable. This method decreases the amount of ^{99}Mo that companies can supply, significantly decreasing the profit margins. Unless there is government funding for the use of these targets, there will be little incentive for facilities convert to LEU.

I.C. Overview of Solution Fueled Reactors

Recent research into new and innovative methods of radioisotope production has revived interest in the aqueous homogeneous reactor (AHR), which was considered as one of the very first nuclear reactor designs. The FFAHR is a variation of this type of reactor. It is therefore necessary to review AHR's and their benefits for isotope production. It is also necessary to understand the shortcomings of these reactors in order to understand why changes need to be made in their design. This section focuses on these topics and presents various examples of AHR's throughout history and currently in operation.

I.C.1. History and Technical Deficiencies

The thought to design a nuclear reactor whose fuel is mixed with its moderator is not exactly unfamiliar. Such designs were considered as early as 1940 for use in the Manhattan Project, and interest has waxed and waned ever since. The motivation for such a design is clear when you consider the cost of solid fuel fabrication that is necessary for typical heterogeneous reactors. This inconvenience is doubled if the end goal is to extract fission products from the fuel; in which case the spent fuel would need to be dissolved in solution to chemically separate the isotopes of interest. This was indeed the case in the early 1940's when the purpose of a nuclear reactor was to produce plutonium. For this reason, it can be understood why chemical engineers of the time were so vocal in support of a reactor whose fuel existed in a liquid form that could be chemically treated for isotope separation, and then reinserted into the reactor.

AHR's utilize various forms of uranium compounds that are soluble in water. In its simplest form, an AHR is simply a container of this solution of the right size and shape to achieve criticality. The earliest examples of AHR's utilized a mixture of uranium oxide (U_3O_8) and heavy water. [14] The cost associated with the predicted amount of heavy water required was the eventual downfall of such early designs. Fortunately, the absorption cross section for deuterium at the time was highly over estimated and the amount of heavy water required was much less than predicted. Further reductions in moderator requirements would be made when enriched uranium became available, making it possible to eliminate the need for heavy water altogether.

Even with the technical advancements made into the mid 1940's, the prominent issue plaguing AHR's was the corrosive properties of the various fuel solutions. This continues to be a challenge today. In the 1940's and 1950's, Los Alamos National Laboratory (LANL) conducted re-

search into the use of AHR's for power production with five experimental reactors known as LOPO, HYPO, SUPO, LAPRE-1, and LAPRE-2. LOPO was a low power reactor which utilized uranyl sulfate fuel, while HYPO and SUPO had higher power outputs and utilized uranyl nitrate fuel. LAPRE-1 and LAPRE-2 utilized uranium oxide mixed in high concentration phosphoric acid. While each of these reactors was able to run for extended periods of time, each one also exhibited ample corrosion rates at high temperatures. [14] Similar reactors were built at Oak Ridge National Laboratory (ORNL) with the same results.

The most recent, and by far the most successful AHR experiment, is the ARGUS reactor at the Kurchatov Institute in Russia. This reactor has been in operation at 20 kW since 1981 and has demonstrated the ability to produce ^{99}Mo and ^{89}Sr for use in the medical industry with extraordinary efficiency. ARGUS has been able to avoid excessive corrosion due to extensive cooling systems and by operating at a lower power density of 0.9 kW/L. This leads to a lower operating temperature than the initial U.S. experiments. ARGUS utilizes a uranyl sulfate solution with a concentration of 73 g U / L and an enrichment of 90% ^{235}U . [15]

Another complication that arose in the design of AHR's was the potential for power oscillations due to several contributing factors. The first of these factors was the bubble formation caused by radiolysis. Even with the temperature of the fuel solution well below its boiling point, radiolysis of water in a radiation field can cause the water molecules to dissociate into hydrogen and oxygen gas, which give the appearance of fuel solution boiling. The constant changing of both the local density and the shape of the critical mass due to bubble evolution at the surface of the fuel can cause oscillations in the reactivity and hence the power. [2] Fortunately, these oscillations are damped and can be worked around, however a truly steady state can never be reached with an AHR in which the fuel is stagnant.

The second contributing factor to the power oscillations was the precipitation of the fuel out of solution. If the hydrogen and oxygen gas is not recombined and reinserted into the system, or if additional water is not added to make up for the lost hydrogen and oxygen gas, the pH of the system may stray from its ideal value of slightly above unity. If the pH rises above 3 or drops below 1, the fuel will precipitate out of solution causing local rises in fissile atom density. [2] The control of the pH is thus an important factor in avoiding power oscillations. It was for these reasons that AHR's were not chosen as the initial design for power producing nuclear reactors.

I.C.2. Benefits for Isotope Production

While AHR's may not be suitable for power production due to their limited power density, ARGUS has shown their potential for use in radioisotope production. When compared to the current solid target irradiation production method described in section I.B, the benefits of AHR's for radioisotope production are numerous.

The most striking benefit of AHR's for radioisotope production is their efficiency due to high neutron economy. As mentioned previously, it is not unusual for solid target irradiation systems to have power density ratios between the fuel and the target as high as 100:1. [2] Even with the limited power density of AHR's, the fuel and the target are one and the same, so that the power density can theoretically be comparable to that in the target of solid target irradiation systems while only producing a hundredth of the total power.

Another benefit of AHR's is the reduction in radioactive waste. In solid target irradiation systems, the irradiated target is dissolved in an acid solution to separate the isotopes of interest. What remains is stored as waste due to its high radioactivity. The fuel of an AHR is already in the form of an acid solution, so that once the isotopes are retrieved from the fuel, it can be rein-

serted into the reactor and used repeatedly until the reactor is decommissioned. This significantly reduces the amount of waste produced for a given amount of isotope production.

The targets used in solid target irradiation are also very costly to fabricate. This is in part due to the high enrichment which is normally above 90% ^{235}U . [2] The solid target fabrication step is obviously eliminated by using a liquid fuel in which the uranium dioxide is simply mixed with an acid solution and diluted to the desired concentration.

Another advantage of liquid fueled reactors is their limitless burnup. Heterogeneous reactors are limited in burnup due to radiation damage of fuel elements or loss of reactivity. [14] AHR's could conceivably run for 40 years on the same fuel loading and burn actinides since the only radiation damage to the fuel is radiolysis, which is reversible. In addition, there is no need to shut down an AHR for isotope separation if a separation loop is included into the design so that isotopes are separated continuously as the reactor is running. If it were not for the corrosion issue that necessitates the replacement of reactor components on a regular basis, an AHR could run uninterrupted until the excess reactivity of the initial fuel loading is used up.

The separation of gaseous fission products such as ^{133}Xe is also much more efficient in an AHR where an off-gas extraction line can be designed to remove these isotopes from the reactor while it is running. [2] These isotopes may be released from solid irradiated targets prior to dissolution in acid for isotope recovery.

AHR's are also attractive from a safety perspective due to their large negative temperature coefficients of reactivity. This is due to the reduction in the density at elevated temperatures which reduces the fissile atom density and hence the criticality. The temperature coefficients of reactivity are typically more negative than those of a heterogeneous reactor. In early studies at

LANL, the criticality was able to be controlled simply by adjusting the fuel concentration and relying on this temperature dependent density change without the use of any control rods. [14] Finally, and perhaps most importantly, AHR's are expected to cost almost an order of magnitude less than their research reactor brethren which currently carry out the solid target irradiation. [2]

It is not surprising that with all of these benefits over the current radioisotope production method, interest in AHR design has peaked recently. Babcock & Wilcox has recently proposed a design for an AHR which it calls the Medical Isotope Production System (MIPS). [16] The MIPS is a 200 kW unit that uses LEU that is less than 20% ^{235}U , whereas most of the previously designed AHR's have used HEU. A diagram of MIPS is shown in Figure I.3.

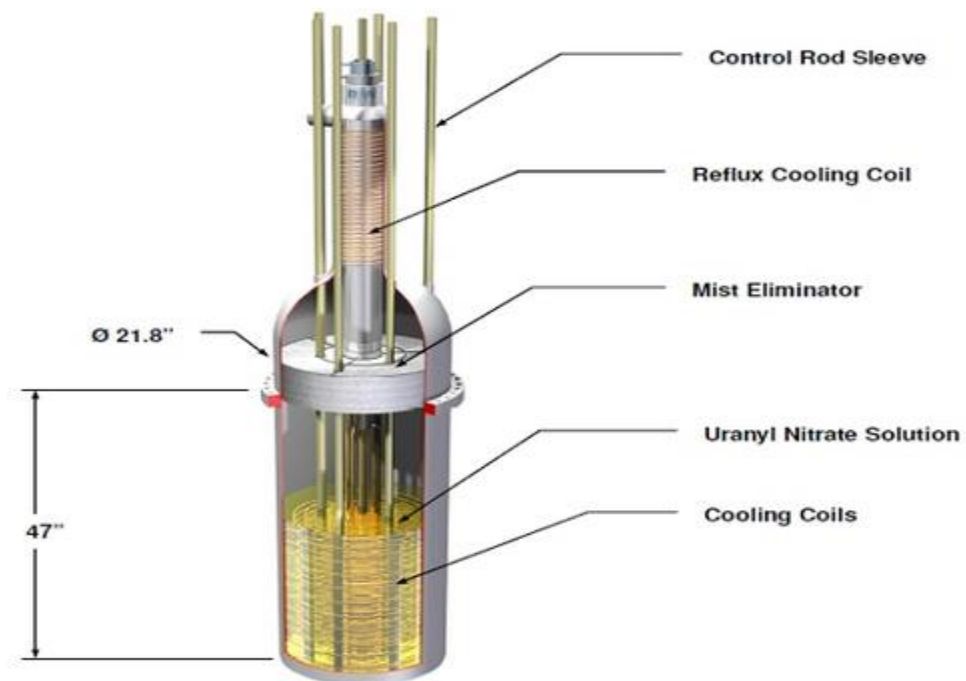


Figure I.3. Diagram of the MIPS design by Babcock & Wilcox. Figure Courtesy of Babcock and Wilcox. [16]

I.C.3. Necessary Improvements

Although the benefits of using an AHR for radioisotope production are abundant, the complications associated with AHR's cannot be denied. Especially with the stringent regulations for reactor licensing in the United States, extensive work must be done to improve upon the current AHR designs.

The corrosion issue in particular must be addressed. This can be accomplished by using materials that have strong resistance to corrosion such as zirconium and titanium alloys. Zircaloy-4 in particular has been shown to have a corrosion rate of less than 0.1 mm per year when in contact with uranyl sulfate at 250 °C. [14] Recent designs have limited the corrosion by limiting the operating temperature that the reactor materials are exposed to.

The potential power oscillations must also be confronted. This may be achieved by mixing the fuel to even out local density fluctuations or by reducing the surface area of the fuel that is not in contact with a solid surface to reduce the effect of surface perturbations by bubble evolution. In addition, systems must be in place to carefully control the pH of the solution either by adding fuel or water, or re-combining the gases that result from radiolysis. This is especially important for systems that utilize uranyl nitrate fuel since the nitrate is susceptible to radiolysis in the same way that water is. [2]

The MIPS design by Babcock and Wilcox is intended to use LEU. This is a very attractive feature for future AHR designs, both from a cost perspective as well as a nuclear non-proliferation perspective. This is bound to be a necessity for any AHR seeking licensing in the United States. In fact, the National Nuclear Security Administration recently awarded NorthStar Medical Radi-

oisotopes, LLC, with two cooperative agreements of \$2.8 million in federal support to establish a domestic production of ^{99}Mo using LEU. [17]

I.D. Description of Proposed Design

This section focuses on the current design as well as the previous FFAHR design. The FFAHR was intended to improve upon traditional AHR deficiencies primarily by the flowing of the fuel through the active core. The isotope production is directly dependent upon the power density, which is limited by the rate at which heat can be removed from the aqueous fuel. From the design of the FFAHR, it became apparent that a reactor using an aqueous solution as both its fuel and target would be far too limited by these constraints. It is for this reason that a hybrid system was proposed to utilize some of the benefits of both a liquid target system and the heterogeneous lattice configuration of conventional nuclear reactors.

I.D.1. The FFAHR

The design of the FFAHR is not very different from the traditional AHR. The primary difference is that in the FFAHR, the fuel solution flows through various regions of the reactor system which is shown in Figure I.4. During a shutdown period, all of the fuel solution resides in the mixing vessel which is kept subcritical by permanently placed absorber rods. During operation, the fuel is pumped upward from the mixing vessel through the active core region where fission reactions take place and isotopes are produced. While in the active core, the temperature of the fuel will rise and after the solution leaves the core, it must be cooled back down to room temperature. To accomplish this, the fuel leaving the active core passes through a heat exchanger. After passing through the heat exchanger, half of the fuel is fed back to the mixing vessel and the other half is transported to the isotope separation system where the various desired isotopes can be separated

from solution. The chemically treated fuel is then returned to the mixing vessel where it is once again pumped upward through the active core. The cycle repeats itself indefinitely until the reactor is shutdown, in which case, all fuel is once again safely stored in the mixing vessel.

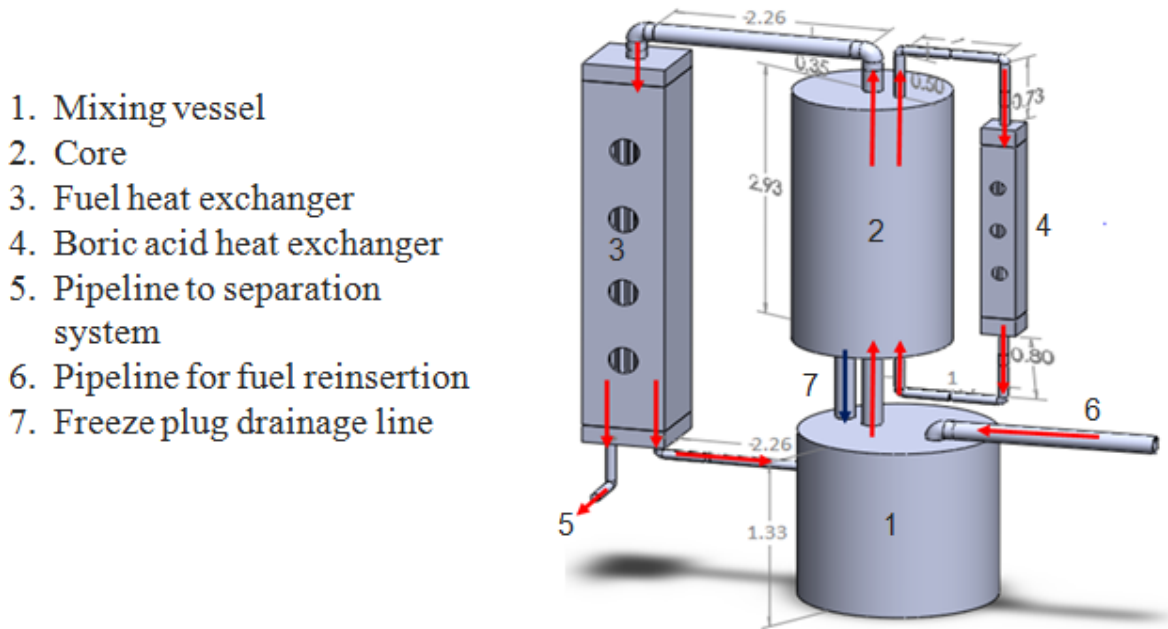


Figure I.4. Conceptual drawing of the FFAHR.

Despite the fact that the fuel is cooled back down to room temperature after leaving the active core, measures must be taken to keep the fuel below its boiling point within the core if the fuel is to reside in this region for an extended period of time. In addition, a control mechanism must be in place to raise and lower the power level. This is accomplished by flowing boric acid through pipes that run through the active core. The boric acid serves as both a heat removal system and a control system. The boric acid flows quickly through the interior pipes removing heat from the

fuel solution and is cooled back down outside of the core in the boric acid heat exchanger. The boric acid also serves as a control mechanism through the large neutron absorption cross section of ^{10}B . In this way, the concentration of the boric acid can be changed to control the criticality of the active core and allow for power level changes. In addition to the boric acid, boron carbide control rods in the core can be raised and lowered for large changes in the reactivity.

The primary benefit of the flowing fuel lies in the fact that the active core no longer has a perturb-able surface as in stagnant AHR designs. There is no exposed fuel surface anywhere within the active core or the pipelines during steady state operation; the only surface is located within the mixing vessel where the fuel is kept subcritical. In this way the power can be maintained at a steady, predictable level without having to adjust control rods to account for changes in the shape of the critical volume, as would have to be done in a stagnant fuel AHR subject to surface perturbations.

The fuel solution of the FFAHR is aqueous uranyl sulfate ($\text{UO}_2\text{SO}_4 + \text{H}_2\text{O}$). Uranyl sulfate is highly soluble in water, but its solubility is extremely sensitive to the pH of the solution. The pH should be kept close to unity, and should not exceed a value of three or else the precipitation of uranium and several fission products will become severe. [2]

Changes in the pH level of the solution are due mostly to radiolysis; the water molecules dissociate into hydrogen and oxygen gases. Fortunately, the recombination of these gases to form water is a well-understood existing technology. The hydrogen and oxygen can be separated from the fission product gases in the gaseous isotope separation system which collects the gaseous fission products from the top of the mixing vessel. The gases are then sent through a recombination process in which they will be re-combined to form water, which is then inserted back into the mix-

ing vessel. By keeping the water concentration nearly constant, the pH will remain within a safe range around the target value of unity.

In addition to keeping the pH balanced, precipitation of the uranyl salt out of solution can be discouraged by constantly agitating the fuel solution. This is accomplished in part by the continuous movement of the fuel throughout the reactor system, and implemented further within the mixing vessel.

Solution fuels are notorious for being highly corrosive to standard structural materials. Although corrosion damage is unavoidable, it can be limited to an extent by operating the system at low temperatures where corrosion rates are slower. In stagnant fuel AHRs heat removal is often accomplished by running coolant lines through the fuel solution. Overall, this is an effective way to keep the fuel temperatures below a set limit, but it allows for the formation of hot spots within the core away from the coolant pipe lines. This can lead to changes in fuel density and therefore uneven and inefficient fuel burn up.

The flowing fuel design achieves a uniform cooling of the fuel which will limit the corrosion rates while also reducing excessive hot spot formation within the fuel. The fuel enters the active core at room temperature and exits at a maximum temperature of 75°C. At these low temperatures the corrosion rates are at a minimum, thus maximizing the lifetime of the structural materials. By keeping each cycle of the fuel through the active core thermodynamically identical there will be no need to worry about fluctuations in the fuel properties due to changes in the temperature profile over the lifetime of the reactor.

An added benefit of the flowing fuel design is the ease of isotope separation. Using solution fuels generally simplifies the isotope separation process compared to using solid target fuel be-

cause the dissolving of the solid target is avoided all together. The isotope yields per kW are also higher because the fuel is the target in a solution reactor, whereas in a solid fuel reactor the fuel and the target are separate components and the majority of the neutron population is used to fission the fuel and not the targets.

The flowing fuel design not only carries the benefits associated with liquid fueled reactors, but also allows for online isotope separation. Typical stagnant AHR's operate for a fixed amount of time, are shut down, drained, and the fuel is sent to the separation facilities all at once. Once the isotopes have been separated, the fuel is added back to the reactor and it is restarted. In the flowing fuel design, a portion of the fuel solution goes directly to the separation facilities after exiting the heat exchanger, while the rest goes back into the mixing vessel. Once the desired isotopes have been extracted the fuel is sent back into the reactor system. Continuously extracting the fuel solution to separate out the desired medical isotopes maximizes the yields by limiting the decay time between activation and isotope separation.

The unfortunate conclusion of the FFAHR design was that the ^{99}Mo production rate was determined to be a fraction of a percent of the current U.S. consumption. As previously mentioned, this production rate was limited by heat removal from the fuel. The heat removal could be made more efficient by increasing the flow rate of either the fuel solution or active coolant through the core; however, increasing the flow rate of the fuel leads to undesired safety issues. Increasing the fuel flow rate decreases the fraction of delayed neutrons that are emitted within the core making the reactivity more difficult to control. The flow rate of the FFAHR was chosen so that over 99.9% of delayed neutrons were emitted within the core, justifying a quasi-static assumption for the criticality calculation. The fraction of delayed neutrons emitted within the core drops off sharply with increasing core flow rate as shown in Figure I.5.

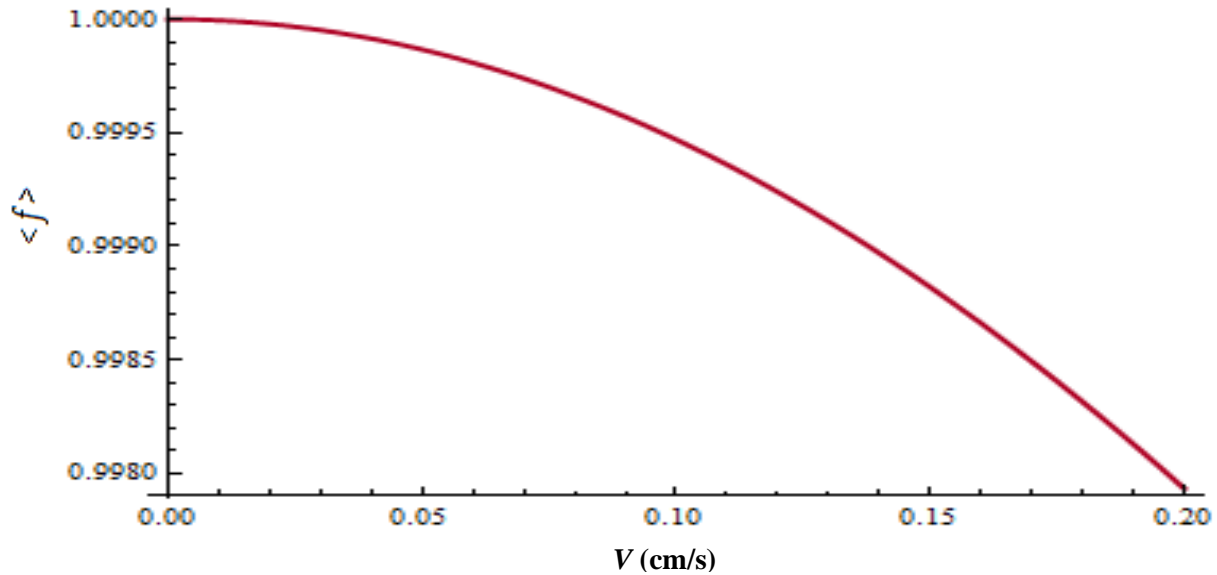


Figure I.5. Fraction of delayed neutrons emitted in the FFAHR core as a function of velocity.

If the solution reactor were designed to be subcritical and coupled to a neutron source with a control system already in place, the importance of these escaping delayed neutrons would be highly diminished. This would allow for a higher, variable fuel flow rate and hence more efficient heat removal and a more flexible isotope separation scheme. This is the motivation behind the design presented in this report which will be referred to as the Aqueous Target System (ATS).

I.D.2. *The ATS*

The ATS design is intended to act as a hybrid of the current solid target irradiation method and the FFAHR. As a first approach, imagine that the solid targets in the current production method were replaced by aqueous solution targets to utilize the waste reduction benefit of traditional AHR's. This benefit may be overwhelmed by the difficulty of target cladding corrosion at high

temperatures that has traditionally plagued all aqueous fuel reactors. In addition, the solution in these targets would eventually boil leading to pressurization and fuel precipitation issues as previously mentioned. Now imagine that the targets exist as pipes that run through the active core so that the liquid target can now flow through the core as in the FFAHR. The primary difference from the FFAHR is that the solution is no longer required to achieve criticality by itself. While this reduces the benefit of high neutron economy that is inherent in AHR designs, it allows for a variable fuel flow rate and more heat removal without sacrificing safety in operation, since the delayed neutrons emitted in the solution are no longer essential to reactivity control.

The reactor chosen as the neutron source for the preliminary ATS design is the Annular Core Research Reactor (ACRR) at Sandia National Laboratories. This reactor is chosen because of its central irradiation cavity in which a flowing aqueous target can be easily placed. Although the current design is focused on using the ACRR as the external neutron source, there is absolutely no reason why the ATS could not be coupled to a more conventional and accessible research reactor such as the TRIGA reactor at the Nuclear Science Center at Texas A&M University. The symmetry of the ACRR core simply allows for a more uniform flux distribution in the cavity where the ATS will be placed. Details such as material compositions and geometry specifications will be presented in Chapter II, but the one sentence description of the ATS is a series of pipes that run upward through the central irradiation cavity of the ACRR carrying a uranyl sulfate fuel, while the rest of the cavity is filled with a coolant flowing in the opposite direction for heat removal. To extend this idea to other research reactors, it may be feasible to simply replace selected fuel rods with pipes that can carry the solution target. For details about the ACRR, one should consult Sandia Report 2006-3067 titled *MCNP/MCNPX Model of the Annular Core Research Reactor* by DePriest *et al.* [18]

CHAPTER II

METHODS

This chapter describes the methods and codes used in the preliminary design of the ATS. The design of a nuclear reactor or target system is an extraordinarily complicated process that involves analyses in multiple disciplines including nuclear engineering, mechanical engineering, chemical engineering, and of course, economics. It would be foolish to claim that any analysis performed by a single individual over the course of a two semester research project could adequately include every aspect of a full design analysis. It is with this in mind that the chapter begins by defining the scope of the ATS design presented in this report.

II.A. Scope

While the FFAHR design was unsuccessful in its attempt to supply a reasonable fraction of the domestic consumption of ^{99}Mo , the scope of that research was quite broad. Nearly every major aspect of a nuclear reactor design process was considered in that analysis. [19] The scope of the FFAHR design included:

- ❖ Neutronics
- ❖ Thermal hydraulics
- ❖ Radiation shielding
- ❖ Isotope separation
- ❖ Environmental analysis
- ❖ Nuclear non-proliferation analysis
- ❖ Economic analysis

The FFAHR design was the result of a four person, two-semester long research project. The preliminary evaluation of the ATS will involve a slightly more limited scope, focusing instead on the major physics concerns in reactor design: neutronics and thermal hydraulics.

II.B. Neutronics

The modern nuclear engineer is blessed to have access to a plethora of computer codes that can calculate key parameters in the design of a nuclear reactor or target system. A good starting point to any research in reactor design is choosing the appropriate codes to use based on their capabilities and the researcher's experience level. For the neutronics analysis of the ATS, most of the calculations will be performed with Serpent 2 (Beta).

Serpent 2 is a Monte Carlo neutron transport code that simulates individual neutron histories in a three dimensional model to calculate parameters such as criticality, neutron fluxes, reaction rates, and burnup. As the number of histories increases, the statistical error in the calculation of these parameters decreases significantly. This allows the use of parallel computing to simulate a large number of histories in a relatively short amount of time to reduce the statistical error to an acceptable level. Details including code comparisons and benchmark studies for Serpent 2 can be found at the developer's website. [20]

Prior to using a code as robust as Serpent 2, a model of the target system must be constructed involving all material compositions and geometries. As the construction of this model may be extremely complicated, it is not desirable to try to get everything right on the first shot. This was a mistake made in the design of the FFAHR. The problem with that approach was that an exorbitant amount of time went into creating the model, which left no time to adjust it when the results were not close to what was desired. Thus, the first step must be to approximate the most im-

portant results with a preliminary model, to determine if they are in the desired range, and then to refine the model to perfection. The most important result for the ATS design is the production rate of ^{99}Mo .

The effective strategy to accomplish this estimation will be to add the solution target to the ACRR irradiation cavity and calculate the ^{99}Mo production rate for various target geometries and compositions. Once a configuration is found that both produces a substantial amount of ^{99}Mo and is thermodynamically viable, necessary changes to the design of the reactor can be made. These changes might include the addition of control rods or the removal of fuel rods to counteract the addition of a significant amount of fissile material in the irradiation cavity. Other changes such as the composition of the solid fuel may be necessary from a licensing and economic standpoint. However, these changes have a minimal effect on the ^{99}Mo production rate when compared to the target geometry and composition.

As the model is adjusted after a target geometry and composition are found, many calculations must be performed to insure the feasibility of such as design. These calculations include the reactivity of the reactor/target system, the flux profile in the target, the production rates of other important nuclides, and the lifetime of the fuel loading of the ACRR when run at constant power for isotope production.

The reactivity of the reactor/target system is a standard output of the Serpent 2 simulation. It must be considered because the control mechanisms of the ACRR should not be changed significantly by adding the target system. This may require the addition of control rods in the irradiation cavity and reactor lattice to allow the reactor to achieve criticality at its current control rod heights. What this means is that the addition of burnable poisons in the irradiation cavity and in

the reactor lattice should bring the reactivity of the combined system close to the reactivity of the reactor with a vacant irradiation cavity without adjusting the ACRR control rod heights significantly.

The flux profile in the target solution can be obtained by adding detectors to the Serpent 2 model. The primary purpose of the flux profile is to determine the heat generation rate for use in the thermal hydraulics analysis. Heat generation is directly proportional to the neutron scalar flux and is used to determine the temperature of the target solution exiting the irradiation cavity. As mentioned in Chapter 1, this temperature must be kept below 100°C to avoid excessive corrosion.

The production rates of nuclides aside from ^{99}Mo are important for a variety of reasons. The first of these is that other nuclides such as ^{133}Xe , ^{89}Sr , ^{90}Y , and ^{131}I are profitable as well and there exist known separation processes for these nuclides. The second reason is that some alpha emitting nuclides are undesirable to have in-solution at high quantities. This is because their presence reduces the purity of the deliverable ^{99}Mo as well as the separation efficiency. [2] Other nuclides such as ^{135}Xe and ^{149}Sm have high neutron cross sections and act as neutron poisons. Their equilibrium concentrations in the target solution will slightly reduce the production rate, and hence must be considered.

The final neutronics consideration is the lifetime of the ACRR fuel. The ACRR is currently operated in pulse mode and is rarely run at constant power. In this mode, the fuel lifetime is so long that it is rarely considered. If the reactor is operated at constant power continuously however, the fuel will eventually be consumed and the reactor/target reactivity will drop below the level to allow continued operation. This requires replacement of the solid fuel elements on a regular ba-

sis. The rate of replacement is anticipated to be on the order of one year, but a better estimation can be obtained via a burnup calculation using Serpent 2.

II.C. Thermal Hydraulics

The primary goal of the thermal hydraulics analysis will be to determine the flow rate and inlet temperature of the coolant required to keep the target solution outlet temperature below 100 °C. To perform these calculations, the code ABAQUS will be used. ABAQUS is a finite element solver used to solve the fluid flow and heat transfer equations for a three dimensional meshed geometry. The model built using ABAQUS will be identical to that built in Serpent. The most important inputs for the ABAQUS model will be the space dependent heat generation rate that will be superimposed over the mesh geometry and the target inlet velocity.

One benefit to using active cooling in the irradiation cavity to cool the target is that the ACRR can function in its normal passive cooling mode. This will effectively separate the thermal hydraulics analysis of the reactor and the target. This allows for a simple calculation of the power level in the target because the Serpent 2 model can be normalized so that the power level in the solid fuel of the ACRR can remain below the steady state power rating of 4 MW for the ACRR.

Additional thermal hydraulics considerations would be the design of the exterior coolant and target heat exchangers. As the target flows through the core, the temperature will rise to the outlet temperature and an external heat exchanger will be required to bring the temperature back down to the inlet temperature prior to re-entry. The same considerations must be treated for the coolant so that each cycle through the core is thermodynamically identical.

CHAPTER III

RESULTS

This chapter will discuss the Serpent 2 and ABAQUS models in depth as well as the results of various calculations performed using these models as they are refined. Prior to presenting the model of the ATS, it may be helpful to review the ACRR as it currently exists. The layout of the ACRR can be seen in Figure III.1.

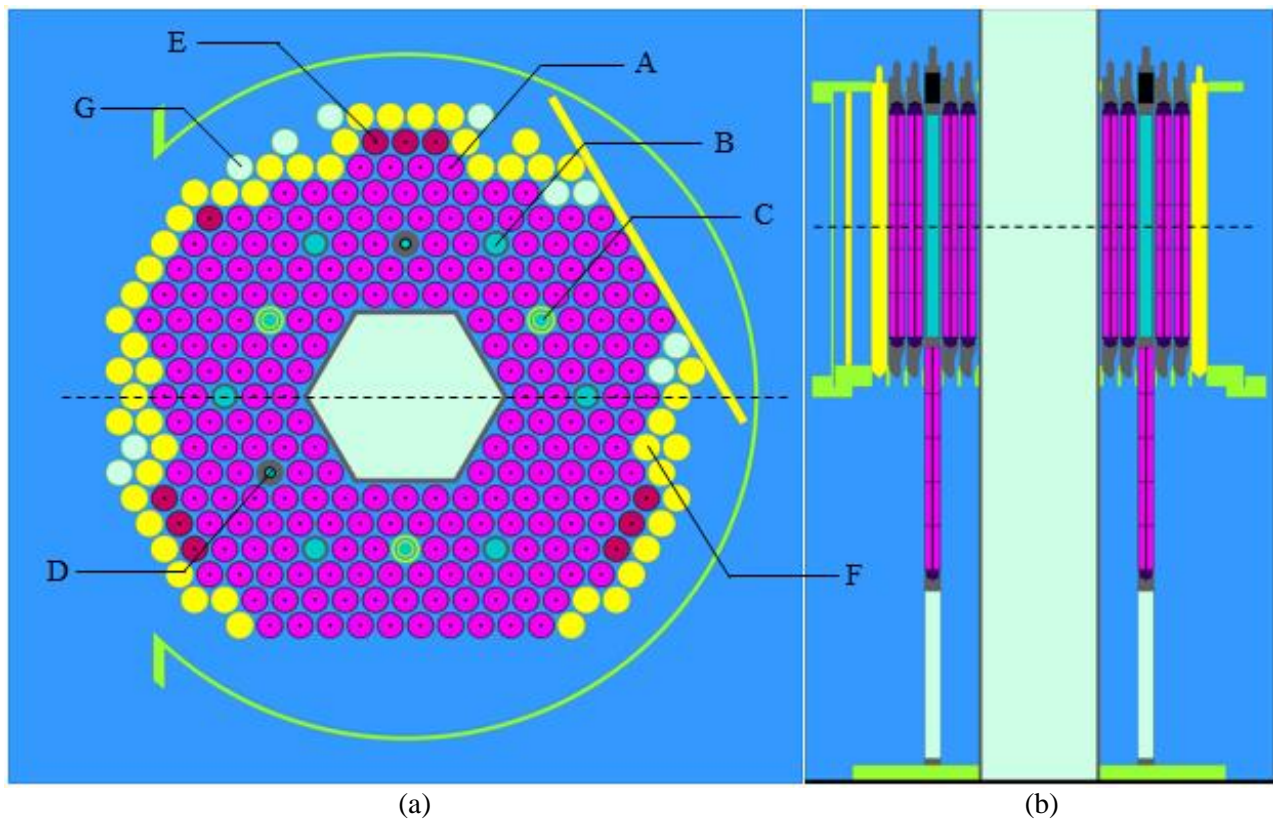


Figure III.1. (a) Top view of the ACRR core lattice. (b) Side view of the ACRR core lattice. Each view is to be interpreted as a slice through the core along the dotted line of the opposite view.

The reactor core as seen in Figure III.1 is submerged in a pool of water where the fuel elements are cooled by natural convection. The labeled components in Figure III.1 are discussed briefly below; however, the more interested reader is invited to read SAND Report 2006-3067 for a more detailed description. [18]

A: Standard fuel elements. These elements are composed of uranium dioxide/beryllium oxide pellets that are stacked in niobium cups as seen in Figure III.1 (b). This fuel material is 21.5 wt. % UO₂ and 35% enriched in ²³⁵U. The density is 3.3447 g/cm³.

B: Fuel followed control rod. This type of control rod is common in research reactors. As the control rod is removed, the reactivity of the reactor increases not only due to the removal of the neutron absorbing material, but also due to the addition of fuel. There are six of these in the ACRR lattice and they are inserted or removed together to approach criticality. The absorber material in the upper half of the control rod is boron carbide.

C: Fuel followed safety rod. These rods are nearly identical to the fuel followed control rod with the exception that they have a smaller diameter and are housed in a control rod guide tube to facilitate quick movement. These rods are fully removed during operation and are fully inserted in the event of a transient rod being stuck in the removed position during a pulse.

D: Void followed transient rod. These rods consist of an upper absorber region followed by a lower void region. They are ejected quickly to cause the reactor to pulse. These rods are also housed in guide tubes to facilitate quick movement.

E: 90% fuel elements. These fuel elements are identical to the standard fuel elements with the exception that their density is 90% of the density of the standard fuel elements.

F: *Nickel reflector elements*. These rods are designed to have the same exterior dimensions as the fuel rods. They are entirely composed of nickel which acts as a neutron reflector to harden the neutron energy spectrum. Also seen in the upper right hand side of Figure III.1 (a) is a nickel plate that is used to cover the neutron radiography window when not in use.

G: *Experiment tubes/water elements*. These positions in the lattice can contain either air filled or water filled nickel elements, or even experiment tubes used to irradiate samples. They are modelled as air filled nickel elements in Figure III.1.

A primary goal of the ATS design is to add the target to the central hexagonal irradiation cavity without changing the operation of the reactor significantly. This involves two significant calculations. The first of these is that the power in any individual pin with the target in the cavity cannot exceed the power of the highest power pin in the vacant cavity case. This must be the case so that the reactor external to the irradiation cavity can continue to be cooled by natural convection of the pool water. Of course, the power in any individual pin is directly related to the total power produced by all of the standard fuel elements. The power profile in the lattice is expected to change significantly with the addition of the solution target to the cavity. The total power level must then be adjusted, to keep the maximum pin power below its value in the vacant case. The power profile calculated using the Serpent 2 model of the unaltered ACRR is shown in Figure III.2. This power profile assumes a total power in the standard fuel elements of 3 MW. Although the maximum steady state power of the ACRR is cited as 4 MW [21], such operation is rare, and a power of 3 MW was chosen for safety concerns as well as to extend the lifetime of the solid fuel elements of the lattice. Assuming a power level lower than the maximum power level will also allow for an additional degree of freedom in case this parameter needs to be raised in further analysis.

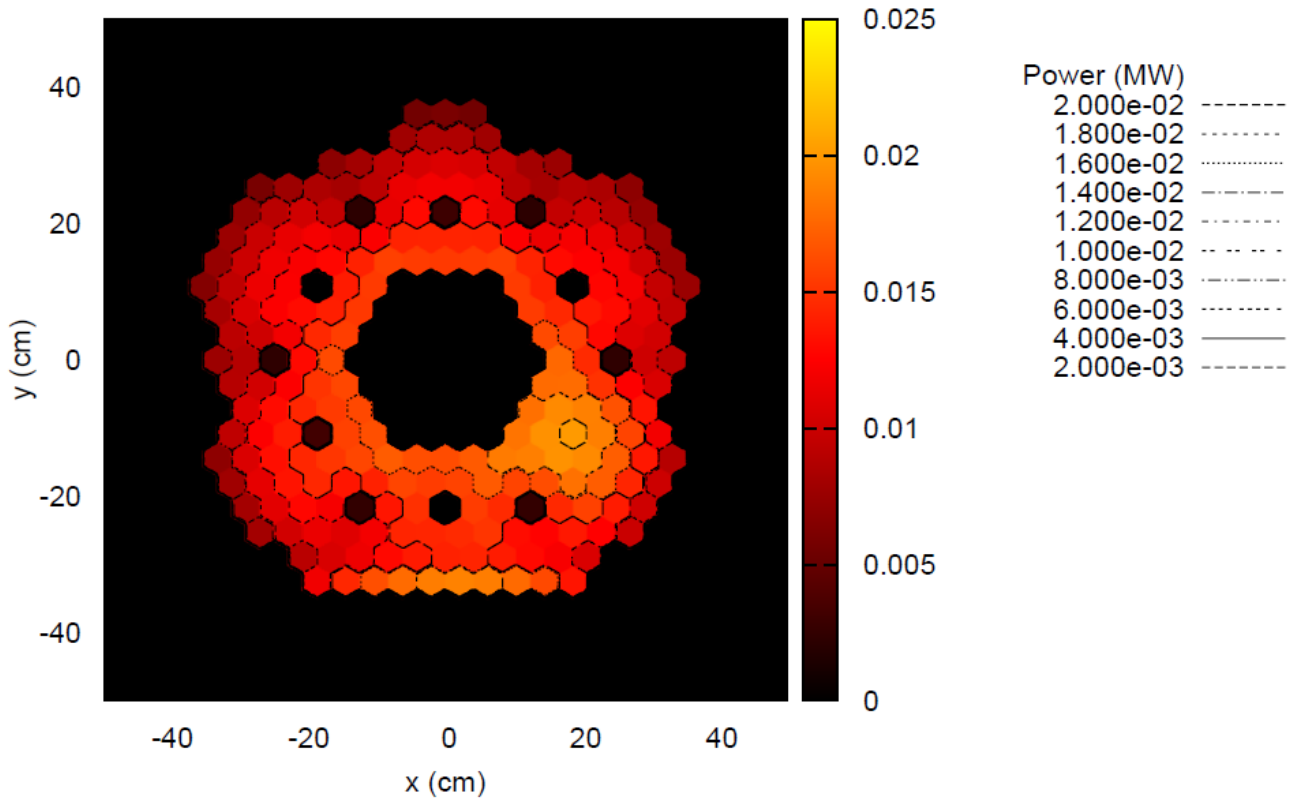


Figure III.2. Power profile of the unaltered ACRR at a steady state power level of 3 MW in the standard fuel elements.

The second calculation that must be performed is an integral rod worth analysis. This involves moving the control rod bank throughout its full range and calculating the criticality of the reactor at each step. The control rods are shown fully inserted in Figure III.1. When the control rods are fully removed, their fuel portion is at the same height as the standard fuel elements. The integral rod worth curve calculated using Serpent 2 for the unaltered ACRR is shown in Figure III.3. The criticality for the ACRR at cold shutdown is calculated to be 0.926, which corresponds to a reactivity of more than ten dollars below critical. The reason for this is that the ACRR is a research reactor designed to allow positive reactivity insertions to the irradiation cavity. For the ATS, it

will be sufficient to have a cold shutdown criticality of roughly 2 dollars below critical with the solution target in the cavity, which is double the requirement set by the NRC in NUREG-1537.

[22]

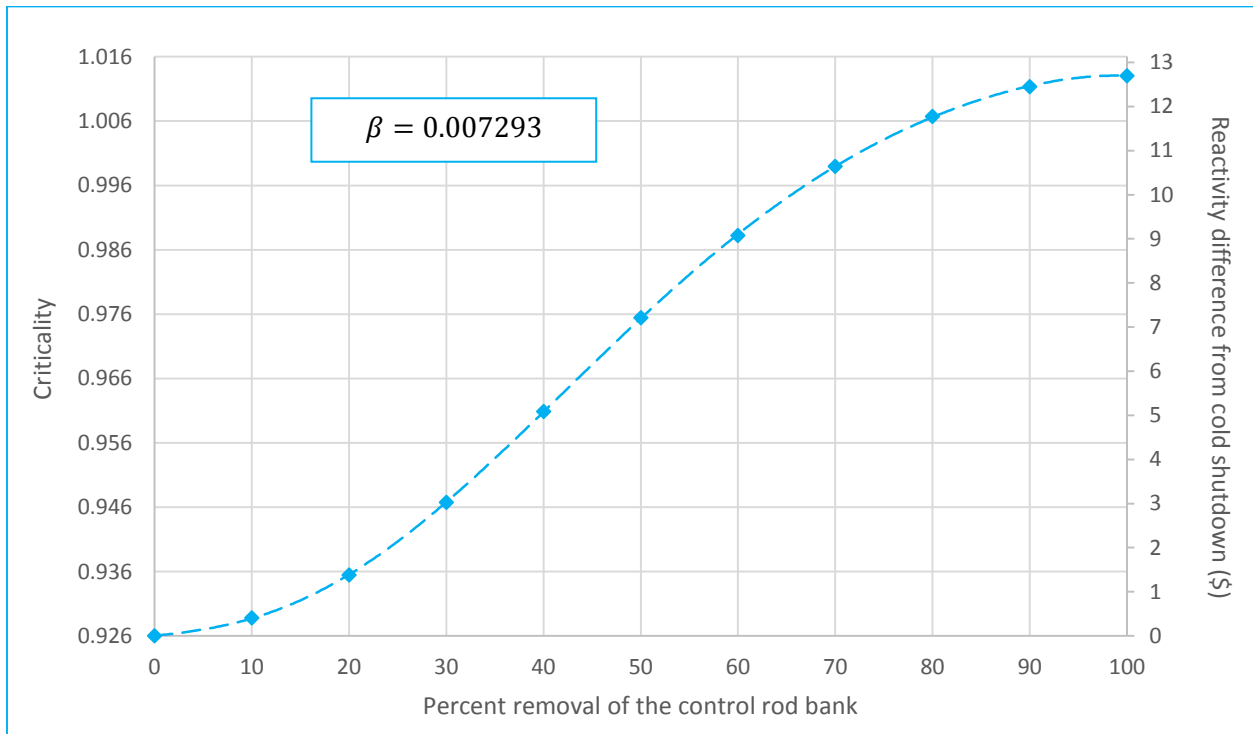


Figure III.3. Integral rod worth curve for the control rod bank of the unaltered ACRR.

It is encouraging that the calculations performed using Serpent 2 match those performed by De-Priest *et al* using MCNP very closely. [18] For the ATS, the reactivity worth of the control rod bank is expected to be greater than the already large \$12.7 for the unaltered ACRR. This is because the transient rods are of no use for a non-pulsing reactor and will instead be replaced with fuel followed control rods.

III.A. Addition of the Solution Target

In order to get a rough estimate of the production rate for various target geometries and compositions, the first step is to add the solution target to the cavity in various configurations. Once the various configurations are modelled, the total production rate of ^{99}Mo can be determined from the total fission rate in the solution target. This fission rate can be easily calculated through the use of detectors in Serpent 2. The lattice of solution pipes is chosen such that it is self-similar to the lattice of solid fuel rods in the ACRR. This can be seen in Figure III.4.

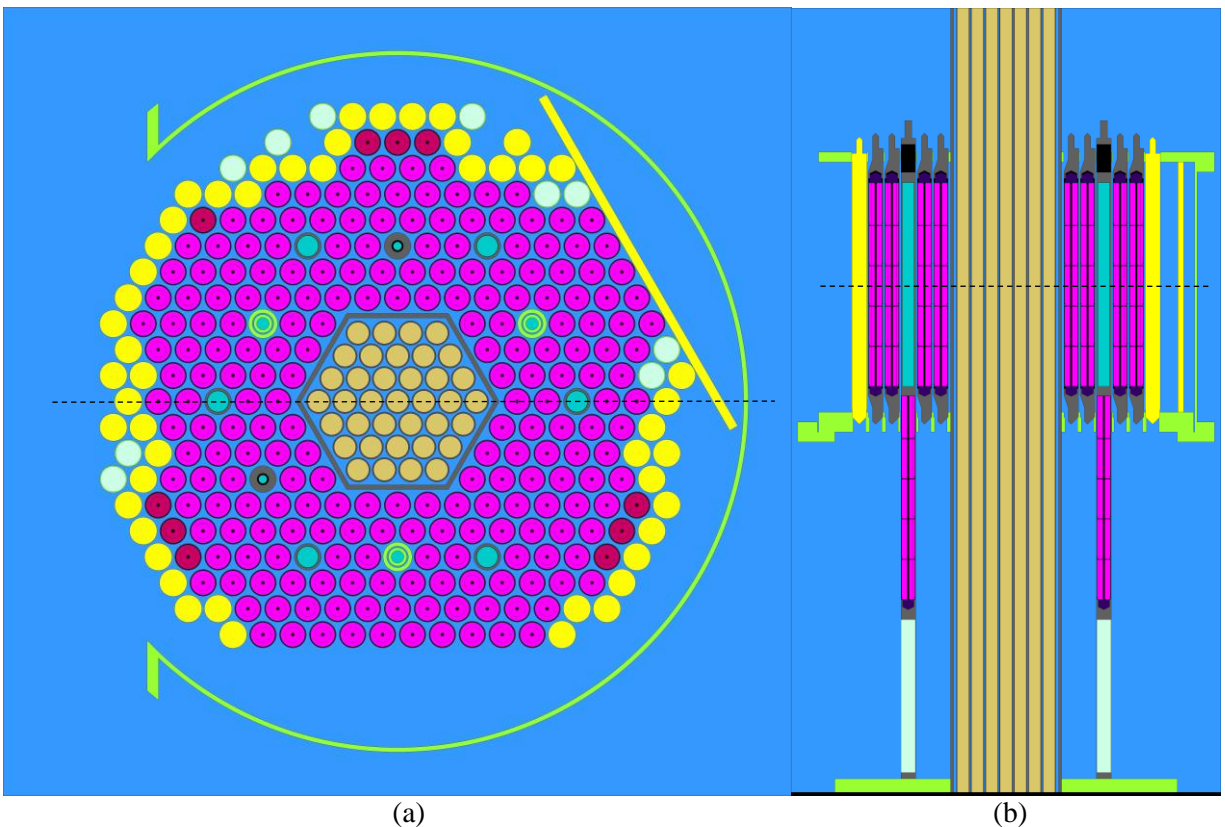


Figure III.4. (a) Top view of the ACRR and ATS core lattice. (b) Side view of the ACRR and ATS core lattice. Each view is to be interpreted as a slice through the core along the dotted line of the opposite view. The pipe inner radius shown here is 1.5 cm.

There are two properties that can be easily changed at this point that will directly affect the ^{99}Mo production rate; these are the solution pipe radius and the concentration of the solution in g U/L. The enrichment is fixed at 20% ^{235}U in order to classify the target as LEU. The solution concentration has an added constraint in that it affects the separation efficiency of ^{99}Mo for most pass through sorbent extractors. Values of 100 g U/L to 200 g U/L can be found in the literature to produce the highest efficiencies of separation depending upon the sorbent and the nature of the compound. [14] [23]

For the ATS, the solution chosen is uranyl sulfate. There is significant debate among experts over whether the sulfate or nitrate is best for the purpose of ^{99}Mo production. Uranyl sulfate was chosen over uranyl nitrate because of its favorable properties in a radiation field. While it is true that nearly any compound will dissociate if irradiated with a large enough intensity of highly energetic neutrons, the sulfate is much more stable against dissociation. The nitrate on the other hand will undergo radiolysis at a rate comparable to water. [2] This radiolysis produces nitrogen and nitrogen oxide gases that are difficult to separate from other gases in order to reinsert them back into the solution. This makes it more difficult to control the pH of the solution in order to avoid uranium precipitation, which could potentially lead to power oscillations.

A range of radii and concentrations are considered for the initial analysis. The inner radius of the solution pipe is varied from 1.2 cm to 1.5 cm in steps of 1 mm. A smaller radius allows for more coolant in addition to less heat production. This may turn out to be beneficial if it is found that the solution outlet temperature cannot be kept below 100 °C in all cases. In each case, the thickness of the pipe wall is 2 mm. The concentration is then varied from 150 g U/L to 180 g U/L in steps of 10 g U/L. The ^{99}Mo production rate and the criticality with all rods completely inserted for each of the 16 combinations of the above radii and concentrations are given in Table III.1.

Table III.1. Pre-separation ^{99}Mo production rates and criticalities for each of the 16 combinations of radius and concentration initially considered for the original ACRR core.

Pipe inner radius (cm)	Concentration (g U/L)	^{99}Mo production rate ($\text{Ci}_{6\text{-day}}$ per week)	^{99}Mo production rate relative standard deviation (%)	Criticality	Criticality standard deviation
1.2	150	8205	0.00038	0.95668	0.000089
1.2	160	8747	0.00038	0.96116	0.000088
1.2	170	9290	0.00038	0.96557	0.000088
1.2	180	9802	0.00037	0.96986	0.000087
1.3	150	9999	0.00037	0.97110	0.000086
1.3	160	10650	0.00037	0.97630	0.000086
1.3	170	11261	0.00037	0.98171	0.000085
1.3	180	11848	0.00036	0.98652	0.000085
1.4	150	11930	0.00036	0.98656	0.000084
1.4	160	12662	0.00036	0.99282	0.000084
1.4	170	13367	0.00035	0.99843	0.000084
1.4	180	14044	0.00036	1.00423	0.000083
1.5	150	13944	0.00035	1.00268	0.000083
1.5	160	14757	0.00035	1.00920	0.000082
1.5	170	15509	0.00035	1.01565	0.000081
1.5	180	16207	0.00034	1.02139	0.000081

While each of the production rates above are significantly higher than the 6000 $\text{Ci}_{6\text{-day}}$ per week domestic consumption rate, it must be remembered that these production rates do not consider the efficiency of the separation process. After the solution passes through the irradiation cavity, it must pass through the external heat exchanger and the separation loop, and the efficiency of this separation loop is dependent upon many factors. These factors include the contact surface area of the solution and the sorbent, the uptake of the sorbent at the given concentration and temperature, and the time that the solution is in contact with the sorbent. The contact surface area can be

increased by diverging the flow of solution into many small pipes with the sorbent painted on their inner walls. The temperature can be controlled by the placement of the separation loop with respect to the external heat exchangers. The contact time is controlled by the flow rate of the solution as well as the length of the sorbent pipes. As ^{99}Mo is radioactive, there will also be a loss in efficiency due to decay as the time spent in the heat exchangers and separation loop increases.

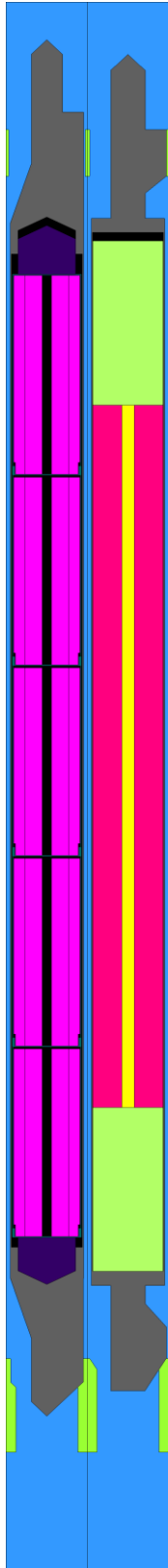
Design of the separation loop is not within the scope of this report; however, its importance cannot be diminished. Design of this process deserves its own detailed research project in the discipline of chemical engineering. For the purposes of this report, a total separation efficiency will be assumed, and it will also be assumed that external heat exchangers exist that can bring the temperature of the solution back down to room temperature before re-entry into the irradiation cavity. Luckily, there is a wealth of research that has been performed on various separation processes and heat exchanger design is a very well understood field of study. One such process reported by Dale *et al* claims that the separation efficiency using Titania sorbents can be greater than 90% at concentrations from 150 g U/L to 300 g U/L. [24] Another such study shows that the uptake of Thermoxid sorbents can be even higher than that of Titania sorbents at certain concentrations. [25] These studies were likely performed using batch irradiation, and although the processes should be identical to the current case, an extremely conservative total separation efficiency of 50% will be assumed to take into account any added inefficiencies that may come with continuous separation.

Even with the conservative separation efficiency assumed, the lowest production rate reported in Table III.1 constitutes almost 70% of the domestic consumption. These preliminary results are encouraging, although they are expected to decrease slightly as core modifications are made and thermodynamic constraints are applied.

III.B. ACRR Core Modifications

As mentioned in Chapter II, several modifications must be made to the ACRR core to facilitate economic steady state production. The first and most prominent is the replacement of the fuel elements. The standard ACRR fuel elements were specially made to facilitate pulsing operation. Continuous operation at 3 MW would likely deplete this fuel, which was extremely expensive to make and is the only fuel of its kind, within a year. Replacing this fuel with a cheaper alternative that classifies as LEU is expected to decrease the criticality of the reactor significantly. If this decrease is such that it makes it impossible to reach two dollars of negative reactivity with the control rods fully inserted, several options exist to increase it. These include replacing the nickel reflector elements, which are just as unnecessary as the specialized $\text{UO}_2\text{-BeO}$ fuel for the purpose of ^{99}Mo production, with more fuel elements. If this still does not meet the minimum criticality, fuel compositions can be adjusted.

The fuel type chosen for the modified ACRR core is standard TRIGA fuel which has the same physical dimensions as the standard ACRR fuel. In fact, the ACRR fuel was designed this way because its predecessor, the Annular Core Pulse Reactor (ACPR), used standard TRIGA fuel and the ACRR was designed to use the existing grid plates and irradiation cavity. A comparison of the fuel elements can be seen in Figure III.5. In addition to the ease with which this fuel can be accommodated in the ACRR is the fact that most research reactors at universities across the country use TRIGA fuel, making the ATS design more feasible to be implemented in existing research reactors. Another factor in this decision is the availability of TRIGA fuel. Using a fuel type that may be more beneficial from a neutronics standpoint, but that is not currently in production, would lead to an increase in fuel costs as this fuel type would need to be manufactured specifically for the purpose intended.



As seen in Figure III.5, although both the ACRR and TRIGA fuel will fit into the ACRR lattice, there are some significant differences between the internals of the fuel elements. Both fuels utilize reflectors on the top and bottom. The graphite TRIGA reflectors are much larger than the BeO ACRR reflectors, leading to a significantly shorter active fuel length. There is also a significant difference in the ^{235}U enrichment; the TRIGA fuel is enriched to 20% while the ACRR fuel is enriched to 35%, which classifies as HEU. In addition, the weight percent of uranium is higher in the ACRR fuel than in the TRIGA fuel; the ACRR fuel is 18.95 wt. % uranium, while the TRIGA fuel is only 11.53 wt. % uranium. On the bright side, TRIGA fuel is available in a variety of weight percents. For instance, the TRIGA fuel at the Nuclear Science Center at Texas A&M University is 30 wt. %. The value of 11.53 wt. % was chosen because this was the composition of the TRIGA fuel that was used in the ACPR.

Other modifications that should be made to the ACRR core before continuing are the replacement of the transient rods and the establishment of symmetry. As mentioned previously, the ATS will have no need for transient rods, and the safety rods may as well be upgraded to fuel rods. Fuel followed control rods that use TRIGA fuel in their follower region are common in research reactors at universities. While the ACRR displays a great deal of symmetry, it is not perfect symmetry. The reasons for the broken symmetries, such as the flat edge to allow coupling to the external FREC II core, are not present in the ATS design. Some

Figure III.5. Comparison of ACRR and TRIGA fuel geometries.

liberty is then taken to establish full symmetry as seen in Figure III.6. In Figure III.6, the fuel rods and safety rods have been replaced with TRIGA rods and the control rods and transient rods have been replaced with TRIGA control rods. In addition all nickel reflector elements and experiment tubes have been removed. Finally, fuel elements were shuffled around until perfect symmetry across angles of 120° was established. The results of the ^{99}Mo production rate and criticality calculations are shown in Table III.2.

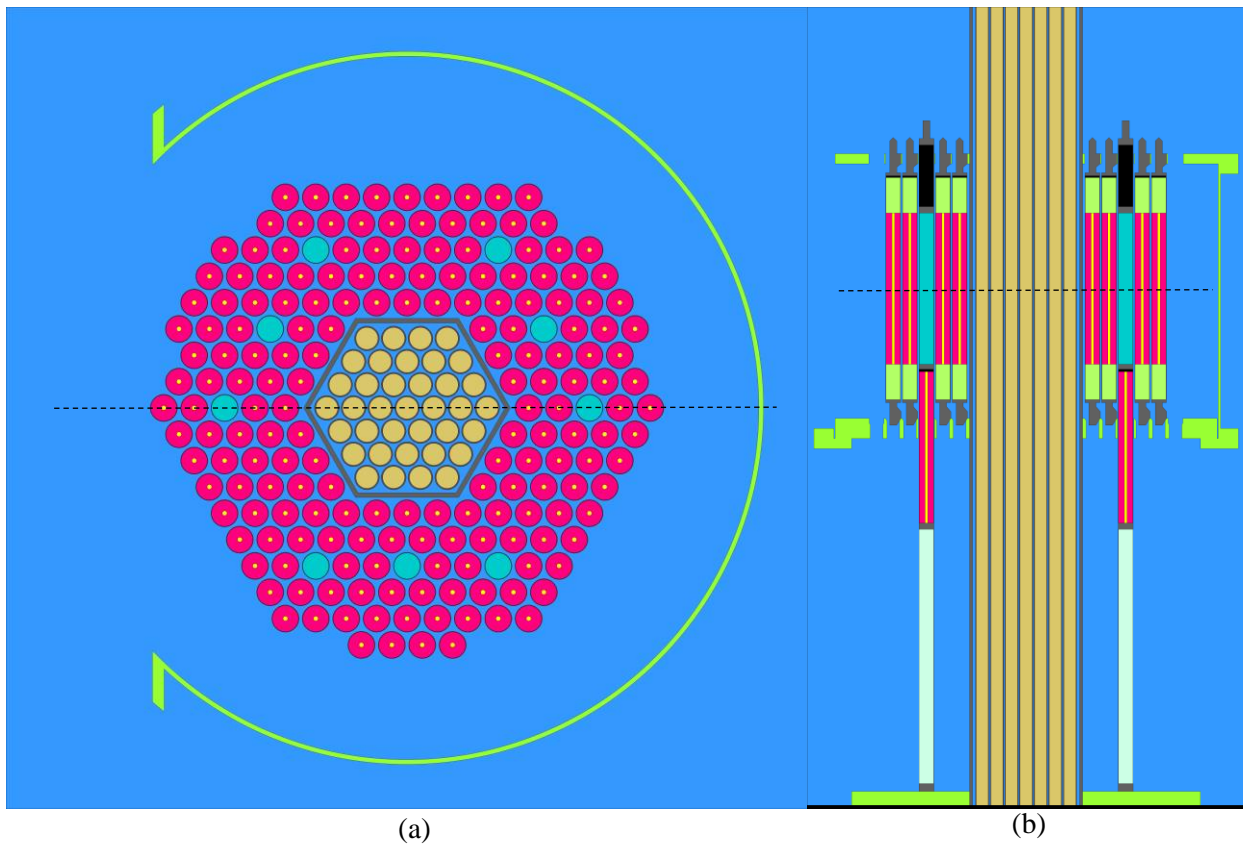


Figure III.6. (a) Top view of the modified ACRR and ATS core lattice. (b) Side view of the modified ACRR and ATS core lattice. Each view is to be interpreted as a slice through the core along the dotted line of the opposite view. The pipe inner radius shown here is 1.5 cm.

Table III.2. Pre-separation ⁹⁹Mo production rates and criticalities for each of the 16 combinations of radius and concentration initially considered for the modified ACRR core.

Pipe inner radius (cm)	Concentration (g U/L)	Mo-99 production rate (Ci _{6-day} per week)	Mo-99 production rate relative standard deviation (%)	Criticality	Criticality standard deviation
1.2	150	5612	0.00042	0.96812	0.000075
1.2	160	6088	0.00042	0.97144	0.000074
1.2	170	6566	0.00042	0.97481	0.000074
1.2	180	7045	0.00041	0.97834	0.000074
1.3	150	6864	0.00041	0.97761	0.000074
1.3	160	7452	0.00041	0.98160	0.000073
1.3	170	8037	0.0004	0.98575	0.000072
1.3	180	8627	0.0004	0.99009	0.000071
1.4	150	8245	0.0004	0.98802	0.000072
1.4	160	8950	0.00039	0.99281	0.000072
1.4	170	9659	0.00039	0.99755	0.000071
1.4	180	10365	0.00039	1.00237	0.000071
1.5	150	9747	0.00039	0.99904	0.000071
1.5	160	10572	0.00038	1.00454	0.000071
1.5	170	11381	0.00038	1.01005	0.000070
1.5	180	12180	0.00038	1.01540	0.000070

III.C. Active Target Cooling

For each case considered in the neutronics analysis, it must be confirmed that the solution can be kept below its boiling point at reasonable flow rates. In order to confirm this, a model of the irradiation cavity is constructed in ABAQUS as shown in Figure III.7. The maximum target velocity is set at 25 cm/s and the inlet temperature of the solution at the bottom of the ACRR core is set at 25 °C. The inlet temperature of the coolant at the top of the ACRR core is set at 20 °C. All that is left to perform the calculation is the volumetric heat generation rate as a function of position within the solution.

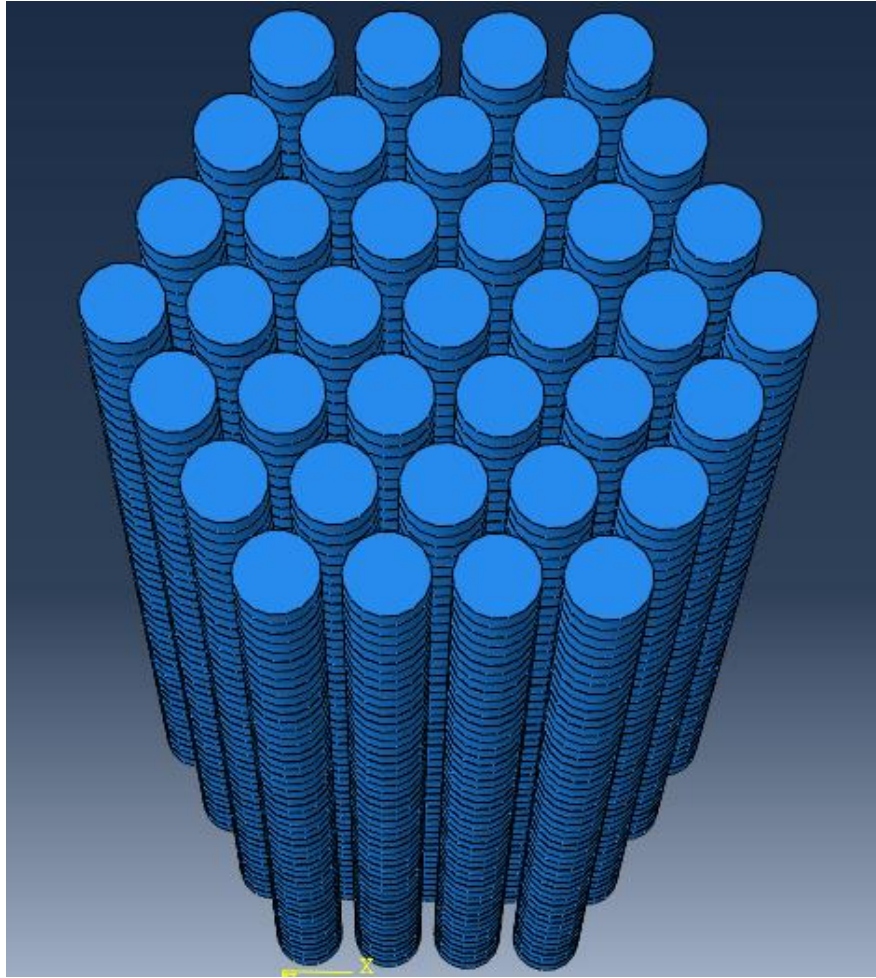


Figure III.7. ABAQUS model of the irradiation cavity showing the solution pipes discretized in the vertical direction.

The volumetric heat generation rate is obtained through the use of mesh detectors in Serpent 2 which calculates the power in each discrete section of each pipe in the cavity. This power level is then divided by the volume of each discrete section to give a volumetric heat generation rate that ABAQUS uses to calculate the outlet temperature and temperature profile of the target as well as the coolant. Such a temperature profile can be seen in Figure III.8.

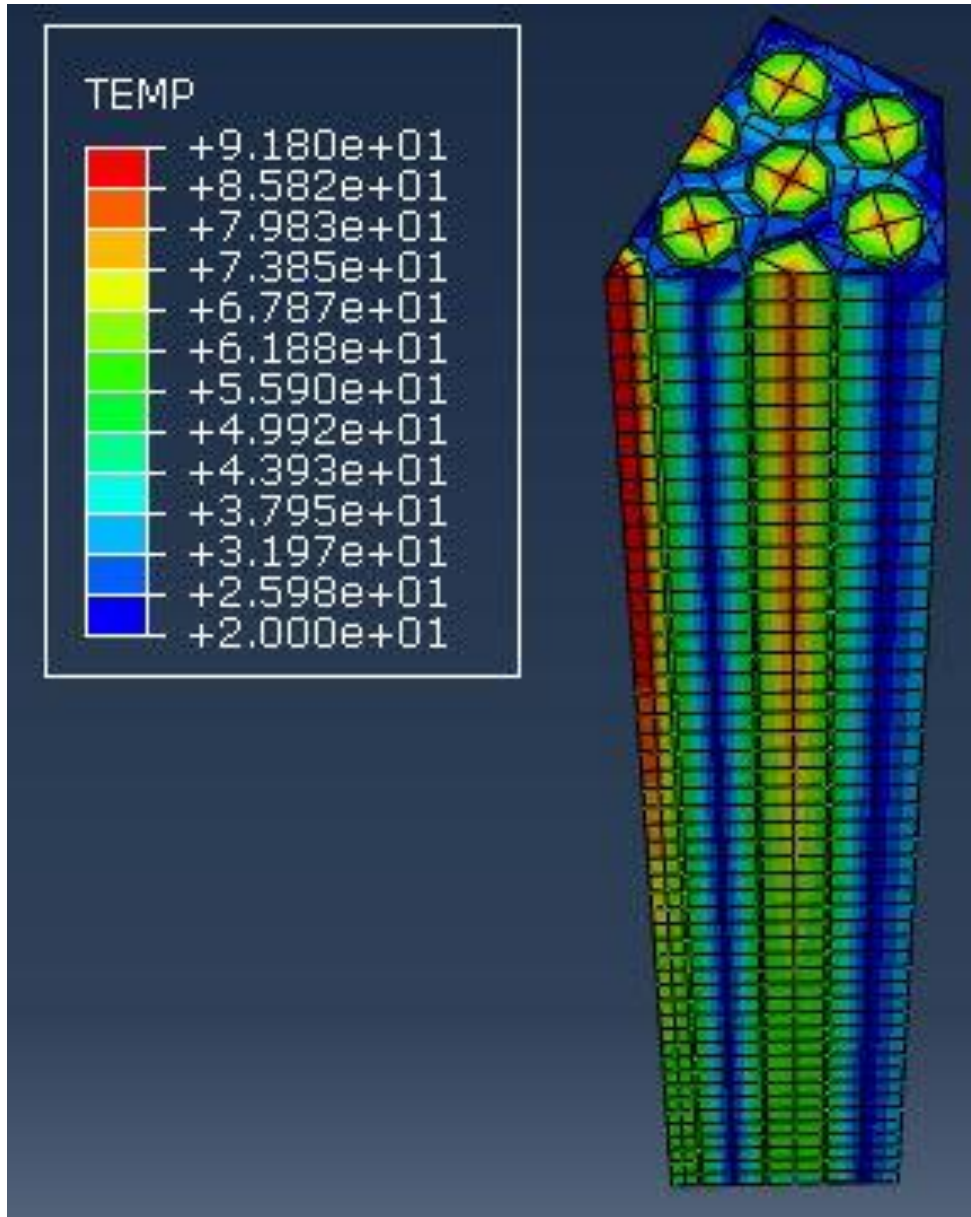


Figure III.8. Temperature profile of a sixth of the irradiation cavity for the 1.4 cm inner radius and 180 g U/L concentration case.

The case shown in Figure III.8 can be considered a worst case scenario in that it is the first case to exceed a criticality of unity with all of the control rods fully inserted, and therefore produces

the more ⁹⁹Mo, and therefore more heat than any of the neutronicly viable cases. Even in this worst case scenario, it is shown that with a target velocity of 25 cm/s, the outlet temperature of the target can remain below the 100 °C limit imposed at the start of the design. Table III.3 gives the outlet temperatures of all cases except for those with inner radius of 1.5 cm, as these were already shown not to be neutronicly viable. The maximum power per solid fuel element also remained below the 0.025 MW limit imposed in order to maintain passive convective cooling by the pool water in all cases considered.

Table III.3. Outlet temperatures of the target solution for neutronicly viable cases.

Pipe inner radius (cm)	Concentration (g U/L)	Peak Temperature (°C)
1.2	150	82.25
1.2	160	84.36
1.2	170	86.45
1.2	180	88.54
1.3	150	83.72
1.3	160	85.81
1.3	170	88.03
1.3	180	90.30
1.4	150	84.90
1.4	160	87.15
1.4	170	89.49
1.4	180	91.80

CHAPTER IV

CONCLUSION

Of the sixteen combinations of pipe radii and solution concentrations considered, the lowest ^{99}Mo production rate, assuming a 50% separation efficiency, is 2806 $\text{Ci}_{6\text{-day}}$ per week, which constitutes 46.8% of the domestic consumption rate. The highest production rate is 6090 $\text{Ci}_{6\text{-day}}$ per week, which is over 100% of the domestic consumption rate. It is not desirable however, to have a single reactor producing the entire U.S. supply because an unplanned outage could cause severe shortages nationwide. Instead, the best plan would be to have several reactors in operation, each producing a fraction of the domestic supply, so that if one needs to be shut down for any reason, the impact to the market will be less severe. Unfortunately, limiting the fraction of the domestic supply produced by a single reactor will also dissuade companies from investing in this technology because decreasing the production rate will decrease the profits without significantly decreasing the capital and operating costs. For this reason, an inner pipe radius of 1.2 cm and a concentration of 160 g U/L is suggested, which could produce 50.7% of the domestic consumption rate, is recommended. Fortunately, there are at least 10 research reactors nationwide at universities alone, that are capable of employing a system similar to the one presented here, including the TRIGA reactor at the Nuclear Science Center of Texas A&M University.

While the research presented in this report is incomplete in that it does not consider fundamental design issues such as isotope extraction and economic analyses, it is certainly a good starting point for proving that such a system is indeed possible. As the current isotope producing reactors are decommissioned, proposals such as this one will be necessary in order to realize a domestic supply. Further research on this design would include a calculation of the TRIGA fuel lifetime as

well as a calculation of the concentrations of other isotopes of interest in the solution target as a function of time after startup. At a certain point, the solution target may need to be re-processed to remove some isotopes that may damage the purity of the ^{99}Mo product.

REFERENCES

- [1] R. M. Mayo, Introduction to Nuclear Concepts for Engineers, La Grange Park, IL: American Nuclear Society, 1998.
- [2] International Atomic Energy Agency, "Homogeneous Aqueous Solution Nuclear Reactors for the Production of Mo-99 and Other Short Lived Radioisotopes," IAEA, Vienna, 2008.
- [3] Knolls Atomic Power Laboratory, Nuclides and Isotopes: Chart of the Nuclides, Bechtel Marine Propulsion Corporation: New York, NY, 2010.
- [4] E. Parma, "The Supply of the Medical Radioisotope Tc-99m/Mo-99: Recent Shortages Call for Action in Developing a Domestic Production Capability," Sandia National Laboratories, Albuquerque, NM, 2009.
- [5] International Atomic Energy Agency, "Production and Supply of Molybdenum-99," IAEA, Vienna, 2010.
- [6] Nordion, "Our Products and Services," 2013. [Online]. Available: http://www.nordion.com/our_products/medical_isotopes_xenon133.asp. [Accessed 20 October 2013].
- [7] T. R. Johnson, "Case: Assessment of Normal Lung Perfusion with Flash Scanning," 2010. [Online]. Available: <http://www.dsct.com/index.php/case-assessment-of-normal-lung-perfusion-with-flash-scanning/>. [Accessed 13 August 2013].
- [8] AETNA, "Radiopharmaceuticals Metastron (Strontium-89) and Samarium-153 (Quadramet) for Metastatic Bone Pain," 2012. [Online]. Available: http://www.aetna.com/cpb/medical/data/300_399/0361.html. [Accessed 28 September 2013].
- [9] D. Brauser, "Yttrium-90 Microspheres May Increase Survival for Patients with Liver Cancer," *MedScape Today*, 2010.
- [10] D. Stewart, "Yttrium," 2012. [Online]. Available: <http://www.chemicool.com/elements/yttrium.html>. [Accessed 13 September 2013].
- [11] C. K. & S. Mandel, "Radioactive Iodine (I-131) Therapy for Thyroid Cancer," 2011. [Online]. Available: <http://www.oncolink.org/types/article1.cfm?c=268&id=9630#.UUy5FxyyCSp>. [Accessed

16 September 2013].

- [12] Nuclear Energy Agency, "The Supply of Medical Radioisotopes: Review of Potential Molybdenum-99/Technetium-99m Production Technologies," OECD, Paris, 2010.
- [13] International Atomic Energy Agency, "Non-HEU Production Technologies for Molybdenum-99 and Technetium-99m," IAEA, Vienna, 2013.
- [14] J. A. Lane, *Fluid Fuel Reactors*, Boston: Addison-Wesley Publishing Company, 1958.
- [15] V. A. Pavshook, "Effective Method of Mo-99 and Sr-89 Production Using Liquid Fuel Reactor," IAEA, Moscow, 2007.
- [16] E. Reynolds, "Medical Isotope Production System," B&W Technical Group, Barberton, OH, 2007.
- [17] National Nuclear Security Administration, "NNSA Awards Agreement to Establish Non-HEU-Based Production of Molybdenum-99 in US," Washington D.C., 2012.
- [18] P. J. C. E. J. P. K. R. DePriest, "MCNP/MCNPX Model of the Annular Core Research Reactor," Sandia National Laboratories, Albuquerque, NM, 2006.
- [19] K. H. A. A. N. G. R. Vega, "Final Design Report: Flowing Fuel Aqueous Homogeneous Reactor for Medical Isotope Production," Nuclear Engineering Department, Texas A&M University, College Station, TX, 2013.
- [20] J. Leppanen, "Serpent: a Continuous-energy Monte Carlo Reactor Physics Burnup Calculation Code," VTT Technical Research Centre of Finland, 4 August 2013. [Online]. Available: <http://montecarlo.vtt.fi/index.htm>. [Accessed 23 January 2014].
- [21] R. L. C. M. K. B. M. R. G. J. J. Dahl, "Documented Safety Analysis (DSA) for the Annular Core Research Facility (ACRRF)," Sandia National Laboratories, Albuquerque, NM, 2013.
- [22] Office of Nuclear Reactor Regulation, "Guidelines for Preparing and Reviewing Applications for the Licensing of Non-Power Reactors," United States Nuclear Regulatory Commission, Washington, DC, 1996.
- [23] A. J. Youker, "Separation, Purification, and Clean-Up Developments for MIPS and SHINE," Argonne National Laboratory, Lemont, IL, 2011.
- [24] D. D. M. G. K. J. C. K. I. M. S. R. G. S. G.E. Dale, "99Mo Separation from High-Concentration Irradiated Uranium Nitrate and Uranium Sulfate Solutions," *Industrial &*

Engineering Chemistry Research, vol. 51, no. 40, pp. 13319-13322, 2012.

- [25] S. A. K. Q. G. V. A.J. Bakel, "Thermoxid Sorbents for the Separation and Purification of ^{99}Mo ," in *26th International Meeting on Reduced Enrichment for Research and Test Reactors*, Vienna, Austria, 2004.