

COMPARISON OF ^{233}U PRODUCTION BY THORIUM IRRADIATION IN HEAVY WATER
REACTOR AND ACCELERATOR-DRIVEN SUBCRITICAL PILE

A Thesis

by

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Submitted to the Office of Graduate and Professional Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of
MASTER OF SCIENCE

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May 2020

Major Subject: Nuclear Engineering

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ABSTRACT

The purpose of this research was to compare the viability of producing uranium-233 (^{233}U) using two oft-overlooked methods of fertile isotope transmutation: a heavy water moderated reactor and an accelerator-driven system. This type of investigation can be helpful in evaluating not only the capabilities available to a potential proliferator, but also indicate to regulators possible vulnerabilities that exist within the current safeguards regime. The impetus behind the selection of the two production methods was their ability to avoid many of the most heavily safeguarded methods and materials, while ^{233}U was chosen due to the global prevalence of thorium.

A scoping analysis was performed to identify benchmarks to judge the effectiveness of each transmutation method. These benchmarks were based off of several pragmatic critical masses that were found using Monte Carlo neutron transport simulations. These critical masses had variation in material properties and geometric configurations, which established a range of material production quantities to compare between. A Monte Carlo N-Particle (MCNP) model of the National Research Experimental (NRX) reactor was used to determine the optimal place to irradiate thorium targets. That same model was then modified to include the thorium targets, and fuel burnup calculations were conducted to determine the mass of ^{233}U that could be produced in 2 years. It was found the NRX reactor was capable of producing 2.79 kg in two years, with a contamination of 10.1 ppm ^{232}U . Next, a model of an accelerator-driven system (ADS) was created using characteristics found in open literature. The optimal placement of thorium irradiation targets was found, and a two year fuel burnup calculation was conducted. The ADS was capable of producing 5.76 kg by the end of the two year period, although this large advantage in material production quantity was primarily because more thorium was able to be loaded around the neutron spallation source of the ADS than in the J-rod annulus of the NRX reactor. The material produced via the ADS also had no ^{232}U contamination.

While there were several factors that would impede a proliferator that were not included in this analysis, such as material losses from milling, metal conversion, target fabrication, and post-

irradiation isotope separation, this analysis presents some of the ways that a determined state could take advantage of portions of nuclear science that the safeguards community is not focusing heavily on. The current IAEA material safeguards significant quantity (SQ) for thorium is 20 tons, well above the amount of thorium needed to produce a critical mass worth of ^{233}U using both these production methods. One current factor that helps marginally protect against these production methods is that both methods require certain items that are closely monitored due to their status as dual-use technology items.

ACKNOWLEDGMENTS

I would first like to recognize my committee members, Dr. Ross and Dr. Tsvetkov, for their valuable insights, as well as their kindness. I also would like to sincerely thank my committee chair and mentor, Dr. Chirayath, for all of the knowledge and skills he has helped me develop. This endeavor would not have been possible without the support of my family and the unending encouragement they give me. I would also like to thank the many wonderful friends I have made at this fine university, the ones who were always there when I needed them most.

CONTRIBUTORS AND FUNDING SOURCES

Contributors

This work was supported by a thesis committee consisting of Professor Sunil S. Chirayath and Pavel V. Tsvetkov of the Department of Nuclear Engineering and Professor Andrew L. Ross of the Department of International Affairs.

The initial idea for this project was conceived by Professor Sunil S. Chirayath of the Department of Nuclear Engineering.

All other work conducted for the thesis was completed by the student independently.

Funding Sources

The financial support for this work came from the discretionary funding of Professor Sunil S. Chirayath of the Department of Nuclear Engineering.

NOMENCLATURE

ADS	Accelerator-driven system
BWR	Boiling water reactor
η	Neutron reproduction factor
FBR	Fast breeder reactor
HEU	High enriched uranium
HTGR	High-temperature gas-cooled reactor
IAEA	International Atomic Energy Agency
JAEA	Japan Atomic Energy Agency
J-PARC	Japan Proton Accelerator Research Complex
JPCA	Japan Prime Candidate Alloy
LBE	Lead-bismuth eutectic
LEU	Low enriched uranium
LFTR	Liquid fluoride thorium reactor
LWBR	Light water breeder reactor
LWR	Light water reactor
MCNP	Monte Carlo N-Particle
MSBR	Molten-salt breeder reactor
MSRE	Molten-salt reactor experiment
MTR	Material test reactor

MYRRHA	Multi-purpose hYbrid Research Reactor for High-tech Applications
NRX	National Research Experimental
NS-FFAG	Non-scaling fixed field alternating gradient
ppm	parts per million
SQ	Significant quantity
²³² Th	Thorium-232
²³² U	Uranium-232
²³³ U	Uranium-233
²³⁵ U	Uranium-235
²³⁹ Pu	Plutonium-239
WC	Tungsten Carbide

TABLE OF CONTENTS

	Page
ABSTRACT	ii
ACKNOWLEDGMENTS	iv
CONTRIBUTORS AND FUNDING SOURCES	v
NOMENCLATURE	vi
TABLE OF CONTENTS	viii
LIST OF FIGURES	ix
LIST OF TABLES.....	x
1. INTRODUCTION.....	1
1.1 Objective.....	1
1.2 Background.....	1
1.3 Previous Work.....	4
1.4 Methodology	4
2. FISSILE MATERIAL REQUIREMENTS	11
3. HEAVY WATER MODERATED REACTOR	13
3.1 NRX Reactor History	13
3.2 Technical Specifications	14
3.3 NRX Modelling	14
3.4 NRX Fuel Burnup Calculations	19
4. ACCELERATOR-DRIVEN SUBCRITICAL REACTOR	24
4.1 Introduction to ADS	24
4.2 State of ADS Development	24
4.3 ADS Modelling	25
4.4 Target Burnup Calculations.....	29
5. SUMMARY AND CONCLUSIONS.....	34
REFERENCES	38

LIST OF FIGURES

FIGURE	Page
1.1 Geographic locations of estimated sizeable thorium deposits. [1].....	2
1.2 Gamma exposure rates for 1 kg highly contaminated (100 ppm ²³² U) ²³³ U as loose pour powder in canister with 0.5 mm thick steel walls.[2]	8
1.3 Dose rate at 0.5 m from various 5 kg spheres of ²³³ U and ²³⁹ Pu with different levels of ²³² U contamination and Pu isotopic compositions respectively.[3]	9
3.1 Fuel pin and calandria tube geometric configuration.[4]	15
3.2 Full core cross section of a generic NRX.[4]	16
3.3 Neutron Flux in the central thimble location calculated using MCNP6.	17
3.4 Neutron Flux in the J-rod annulus calculated using MCNP6.....	18
3.5 Rendering of MCNP6 model with J-rod annulus fueled (red color), in the XY view. .	19
3.6 Rendering of MCNP6 model with J-rod annulus fueled (red color), in the XZ view. .	20
3.7 ²³³ U production within the NRX over the 2 year burn period calculated using MCNP6. 22	22
3.8 ²³³ U contamination ingrowth within the NRX over the 2 year burn period calculated using MCNP6.....	23
4.1 Detailed schematic of the JAEA ADS[5]	26
4.2 RZ view of the JAEA ADS with dimensions.[6]	27
4.3 Detailed schematic of the beam window, in which the thickness transitions from 0.2 cm at the top to 0.3 cm in the cylindrical portion.[6].....	28
4.4 A survey of neutron flux spectra calculated using MCNP6 at different radial values away from the LBE target center.	30
4.5 XY view of the MCNP6 model of the ADS, with light blue, dark blue, yellow, and red representing the LBE, JPCA vessel, graphite, and thorium target rods respectively. 31	31
4.6 XZ view of the MCNP6 model of the ADS.	32
4.7 ²³³ U production within the ADS over the 2 year burn period calculated using MCNP6. 33	33

LIST OF TABLES

TABLE	Page
1.1 Survey of research efforts to incorporate thorium and ^{233}U into the nuclear fuel cycle. [7].....	5
1.2 IAEA SQ values. [8]	6
1.3 Study of dose rate and working limitation for 0.5 m away from a 5 kg sphere of ^{233}U one year after separation.[3]	8
2.1 Critical masses for unreflected spheres with varying multiplying factors on the density.....	11
2.2 Critical masses for unreflected spheres with varying multiplying factors on the density.....	12
3.1 Key dimensions of a typical NRX reactor.[4].....	15
4.1 Key dimensions of the proposed JAEA ADS. [9] [10] [11].....	26
4.2 JPCA composition. [5].....	27
5.1 A comparison of Th loading and ^{233}U production quantities with established benchmark quantities.	34
5.2 A comparison of key system parameters as well as ^{233}U produced normalized for Th loading	35

1. INTRODUCTION

1.1 Objective

The objective of this project was to assess the feasibility of producing uranium-233 (^{233}U) in two different ways. An analysis of the amount of ^{233}U production in these two ways can be used to inform strategies needed to curb potential clandestine ^{233}U production methods. These ^{233}U production pathways were assessed using Monte Carlo neutron transport and fuel burnup simulations on the basis of practicality and timeliness.

1.2 Background

Thorium—despite its nuclear applications and prevalence in nature—is often an afterthought in the field of nuclear nonproliferation. It is a difficult and arduous technical process to glean weapons usable material from both natural thorium and uranium, yet much more attention is given to ensuring uranium is not covertly enriched than guarding against thorium transmutation. Thorium is one of the most abundant heavy elements in the earth’s crust and is estimated to be 3 to 4 times more abundant than uranium. [12] While the majority of uranium deposits are relatively concentrated geographically, sizeable thorium deposits exist in many countries, including some that have not yet developed nuclear weapons, namely Brazil, Turkey, Australia, and Egypt. [13] This can be seen in Fig. 1.1.

The most salient reason thorium, a fertile nuclide, is of interest from a safeguards perspective is its role as a precursor in the production of ^{233}U , a fissile nuclide. If ^{232}Th is bombarded with neutrons, then it can produce ^{233}U via the production chain depicted in Eq. 1.1. This breeding method is similar to plutonium production from ^{238}U . When the two fertile materials are compared, ^{233}U has a much higher thermal neutron absorption cross section (7.4 barns compared to 2.4 barns) while maintaining a comparably fast neutron cross section. [14] This means that the ^{232}Th - ^{233}U cycle can be effectively utilized in systems with either energy spectra and have a higher conversion rate.

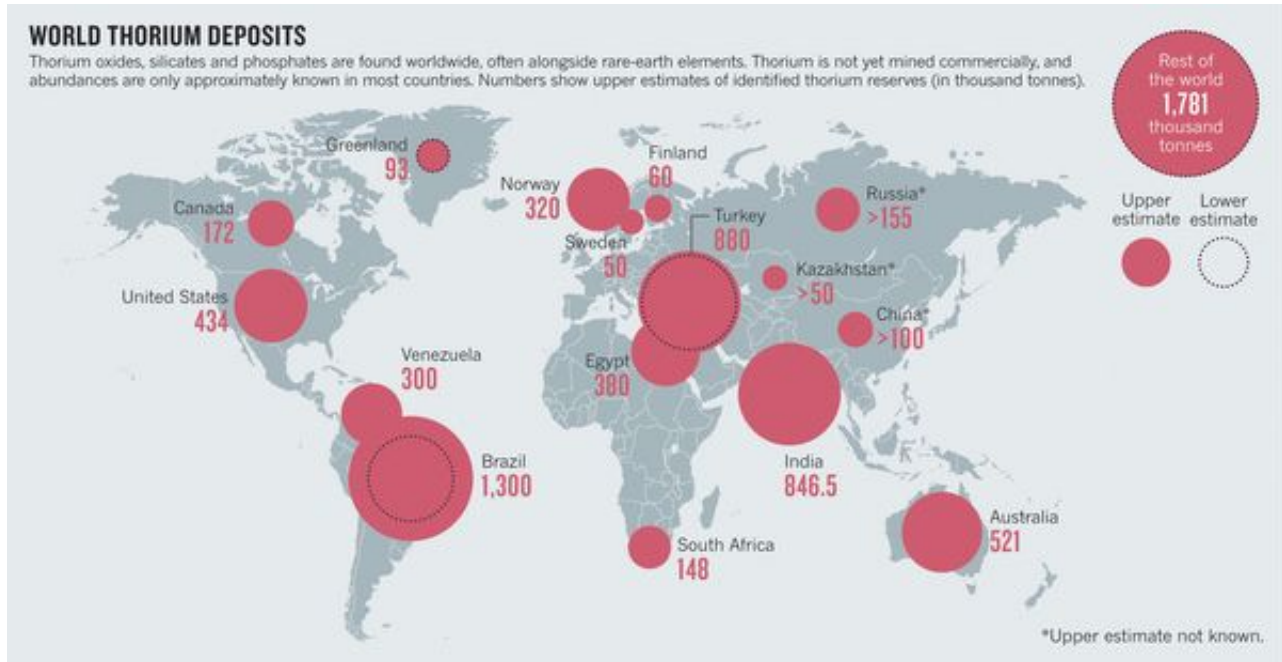
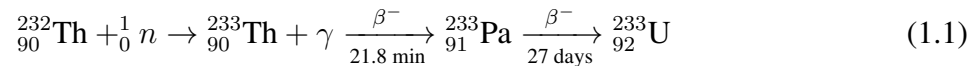


Figure 1.1: Geographic locations of estimated sizeable thorium deposits. [1]



${}^{233}\text{U}$ is a fissile isotope that can be used in the same ways as the more prevalent ${}^{235}\text{U}$ and ${}^{239}\text{Pu}$. In a few metrics of fissile nuclide performance, ${}^{233}\text{U}$ outperforms the other two nuclides, most notably in thermal η (neutrons produced per neutron absorbed). This higher thermal η allows the ${}^{232}\text{Th}$ - ${}^{233}\text{U}$ breeding process to occur even in thermal systems, unlike the ${}^{238}\text{U}$ - ${}^{239}\text{Pu}$ breeding process which can only operate efficiently in a system with a fast neutron spectrum. Another reason that ${}^{233}\text{U}$ production can occur in a thermal spectrum while ${}^{239}\text{Pu}$ cannot is the fact that the thermal neutron absorption cross section of ${}^{232}\text{Th}$ is higher than that of ${}^{235}\text{U}$. These two nuclear properties lend flexibility to the design of a ${}^{233}\text{U}$ breeder reactor as it can be either spectrum, unlike a ${}^{239}\text{Pu}$ breeder which requires a fast spectrum system. In regards to η in a fast neutron environment, ${}^{233}\text{U}$ performs better than ${}^{235}\text{U}$, with an η of 2.5 versus the 2.3 of ${}^{235}\text{U}$. [15] Another advantage

^{233}U possesses a lower thermal capture cross section than the other fissile nuclides, which means during production less fissile material will be lost to higher transmutations. When ^{233}U is produced in a thermal spectrum system, the cannibalization of the ^{233}U by (n,2n) reactions is also minimized. ^{233}U is a more attractive material to produce a nuclear weapon compared to ^{235}U . This is because of ^{233}U 's more favorable fast neutron fission cross section (about 1.5 times the size for a neutron at 2 MeV) as well as the fact that more neutrons are produced per fission in ^{233}U than ^{235}U . [16] This means less material is required to form a critical mass. The United States, Soviet Union, and India have previously tested weapons with cores either fully or partially composed of ^{233}U . [17] In examining the ideal spectrum for the production of fissile materials via neutron bombardment, there are several factors to consider, namely: which neutron energy ranges have the most favorable fertile isotope absorption cross sections and how much of the fissile isotope will be cannibalized by further neutron capture and (n,2n) reactions. The neutron absorption cross sections of ^{232}Th combined with the need to minimize (n,2n) reactions leads to the conclusion that the ideal ^{233}U production would occur in a thermal spectrum.

^{233}U also has been investigated for commercial uses, which in turn has influenced much of the research conducted on how to safeguard against its usage in a nuclear weapon. One vehicle for thorium usage in power production was using thoria (ThO_2) fuel in not only conventional light water reactors (LWR), but also in experimental reactors, such as the high-temperature gas-cooled reactor (HTGR). The thoria, when exposed to the neutron flux of the ^{235}U powered reactors, would eventually produce ^{233}U . This would allow reactors to be operated with less initial fuel, as it would breed additional fuel during the course of operation. The United States was a leader in this area of research, going as far as using thoria fuel in two HTGRs and two LWRs. Two similar projects were developed, one by Germany and one by the United Kingdom, with collaboration from several Euratom members. Another method to incorporate ^{232}Th and ^{233}U into a nuclear fuel cycle is to use them in molten salt reactor concepts first examined by researchers at Oak Ridge National Laboratory beginning in the 1960s. The molten-salt reactor experiment (MSRE) was the first nuclear system to be fueled by ^{233}U , and while it did not actually use any thorium during

operation, it provides the technical basis for the liquid fluoride thorium reactor (LFTR) concept. [18] The LFTR concept is presently being developed in several countries. The third and most thoroughly developed effort to utilize ^{232}Th and ^{233}U in a nuclear fuel cycle has come from India. Near the inception of its nuclear program, India had sought to leverage its large thorium reserves into power production capabilities. Dr. Homi Bhabha, the first head of India's nuclear program, made the intention known to the world in 1954 when he laid out India's three stage nuclear power program, which included thorium based systems as the third stage. [17] As a result, India has made the most efforts in developing reactors capable of using ^{232}Th to breed ^{233}U fuel. [19] India currently operates the only nuclear reactor fueled entirely by ^{233}U , the KAMINI research reactor. [20] A brief collection of the previous projects to utilize thorium and ^{233}U for power production can be found in Table 1.1.

1.3 Previous Work

There have been efforts in the past to evaluate the proliferation risk of thorium and ^{233}U , but these were done almost entirely through the lens of their incorporation into the fuel cycle. Much of the previous work done was meant to assuage the fears of the global community that, in a thorium fuel cycle, proliferation would be an easy task. Most literature detailing safeguards approaches aims to safeguard thorium and ^{233}U in either thorium based fuel or a molten salt reactor. [21].

While there have been efforts to detail safeguarding requirements of a full thorium fuel cycle, little work can be found in open literature that addresses a scenario where a would-be proliferator attempts to use thorium and facilities that are not a part of a thorium fuel cycle to achieve nefarious ends. As such, there exists some novelty to conducting research of this nature.

1.4 Methodology

This research effort was to evaluate methods of procuring ^{233}U . To accomplish this, isotopes and facilities typically overlooked in nuclear nonproliferation research were utilized. An investigation of how ^{233}U can be produced via neutron bombardment of ^{232}Th at facilities with potentially little to no safeguards was used to demonstrate the merits of such a methodology. A demonstration

Name	Location	Reactor Type	Fuel	Period of Operation
Peach Bottom	USA	HTGR	Th- ²³⁵ U Oxide	1966-1972
Fort St. Vrain	USA	HTGR	Th- ²³⁵ U Dicarbide	1976-1989
MSRE	USA	MSBR	²³³ U Molten Salt	1964-1969
Shippingport	USA	LWBR	Th- ²³³ U Oxide	1977-1982
Indian Point	USA	PWR	Th- ²³³ U Oxide	1962-1980
Borax IV and Elk River	USA	BWR	Th- ²³³ U Oxide	1963-1968
AVR	Germany	HTGR	Th- ²³⁵ U Oxide & Dicarbide	1967-1988
THTR	Germany	HTGR	Th- ²³⁵ U Oxide & Dicarbide	1985-1989
Lingen	Germany	BWR	Th-Pu Oxide	1973
Dragon	UK	HTGR	Th- ²³⁵ U Dicarbide	1966-1973
KAMINI	India	MTR	²³³ U Metal	1996-
CIRUS	India	MTR	Th metal & oxide in J-rod	1960-2010
Dhruva	India	MTR	ThO ₂ in J-rod	1985-
FBTR	India	FBR	ThO ₂ Blanket	1985-

Table 1.1: Survey of research efforts to incorporate thorium and ²³³U into the nuclear fuel cycle. [7]

like this can be used to find what kinds of materials and facilities should be monitored to meet the current International Atomic Energy Agency (IAEA) timely detection goals of special nuclear materials and whether the currently defined significant quantity (SQ) for thorium is appropriate. An SQ is the amount of nuclear material for which the possible ability to produce a nuclear weapon cannot be discounted. This is the metric that the IAEA uses to determine the effectiveness of its various material safeguards programs around the world, which can be seen in the stated objectives of IAEA safeguards which are, "the timely detection of diversion of significant quantities of nu-

clear material from peaceful nuclear activities to the manufacture of nuclear weapons . . . and deterrence of such diversion by the risk of early detection." This objective necessitates that the IAEA define two parameters: timeliness goals and SQ thresholds. The SQ values can be found in Table 1.2. The timeliness detection goals places an imperative on the IAEA to discover the diversion of an SQ of material within a certain time period. This is one month for unirradiated direct use material, three months for irradiated direct use material, or one year for indirect use material.

Material	Classification	Quantity
Pu	Direct use	8 kg
²³³ U	Direct use	8 kg
HEU	Direct use	25 kg ²³⁵ U
LEU	Indirect use	75 kg ²³⁵ U
Natural uranium	Indirect use	10 t
Depleted uranium	Indirect use	20 t
Th	Indirect use	20 t

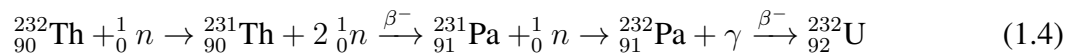
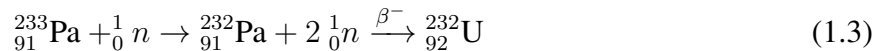
Table 1.2: IAEA SQ values. [8]

The first step in this methodology was to establish the baseline for fissile material requirements for ²³³U. This was done using the Monte Carlo N-Particle (MCNP) transport code to find the critical mass of ²³³U in a variety of configurations.

After establishing how much ²³³U is required, the parameters of the neutron irradiation scenario were established. The hypothetical proliferator in this scenario is a state with some, but certainly not abundant, nuclear resources and that places significant importance on ensuring that the weapons development is covert. It has unfettered access to thorium and the ability to mill, convert, and fabricate the thorium into metal rods. It is also assumed that the state would have

the resources to chemically separate the ^{233}U produced from the thorium fuel. In order to avoid IAEA scrutiny, the ^{232}Th transmutation must not be conducted in a commercial nuclear power plant. Additionally, the transmutation cannot depend on enriched uranium or plutonium to produce the required neutron flux, as the state would more easily be able to divert those resources into a weapons program. Under these constraints, two facilities that could be utilized would be a natural uranium fueled research reactor system or a subcritical accelerator driven system.

The two facilities to be investigated were modelled in MCNP. Using neutron flux data, the optimal thorium target placements were selected. Next, depletion/fuel burnup calculations were conducted to find the time required to produce the necessary ^{233}U mass. Some parameters for the facilities include using less than a SQ of thorium, and the required fissile material should be produced as quickly as possible. Another parameter is to ensure that the level of ^{232}U contamination is not too high so that that the ^{233}U could still be used in an explosive device. There are several strong radiation emitters that exist in the ^{232}U decay chain, in particular the 2.6 MeV gamma that is produced when ^{208}Tl decays. [22] This fact, when combined with the low half-life of ^{232}U (72 years), means if too much ^{232}U is present, then the material becomes too dangerous to handle.[23] ^{232}U is produced by several (n,2n) reactions that can occur during the thorium transmutation process. This thorium transmutation process can be seen in Eq. 1.2, 1.3, and 1.4. The radiation hazards that occur when working with ^{233}U that has ^{232}U contamination Fig. 1.2 and 1.3. Table 1.3 shows the occupational limits of working with ^{232}U if following the US radiation worker dose limit of 5 rem/yr.



To model a natural uranium reactor, a design based on the National Research Experimental (NRX) reactor in Chalk River, Canada was used. The NRX uses heavy water to moderate neu-

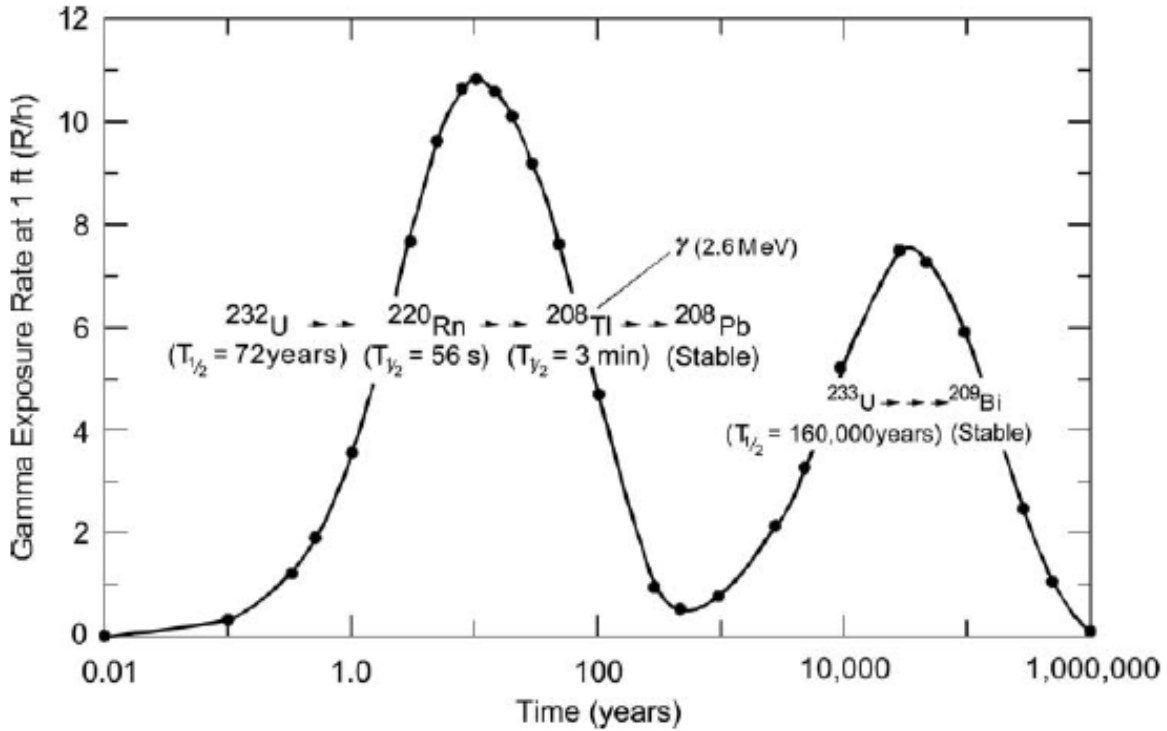


Figure 1.2: Gamma exposure rates for 1 kg highly contaminated (100 ppm ^{232}U) ^{233}U as loose pour powder in canister with 0.5 mm thick steel walls.[2]

^{232}U contamination (ppm)	Dose Rate (rem/hr)	Contact Hour
1	0.013	380
5	0.059	80
100	1.27	4
10,000	127	0.04

Table 1.3: Study of dose rate and working limitation for 0.5 m away from a 5 kg sphere of ^{233}U one year after separation.[3]

trons and has no power production capability. This is a reactor design that has been successfully emulated in many countries including India, Pakistan, Israel, and Taiwan. [24] [25] It is feasible for a nation to produce a "home grown" variant of this reactor that would be outside of the IAEA

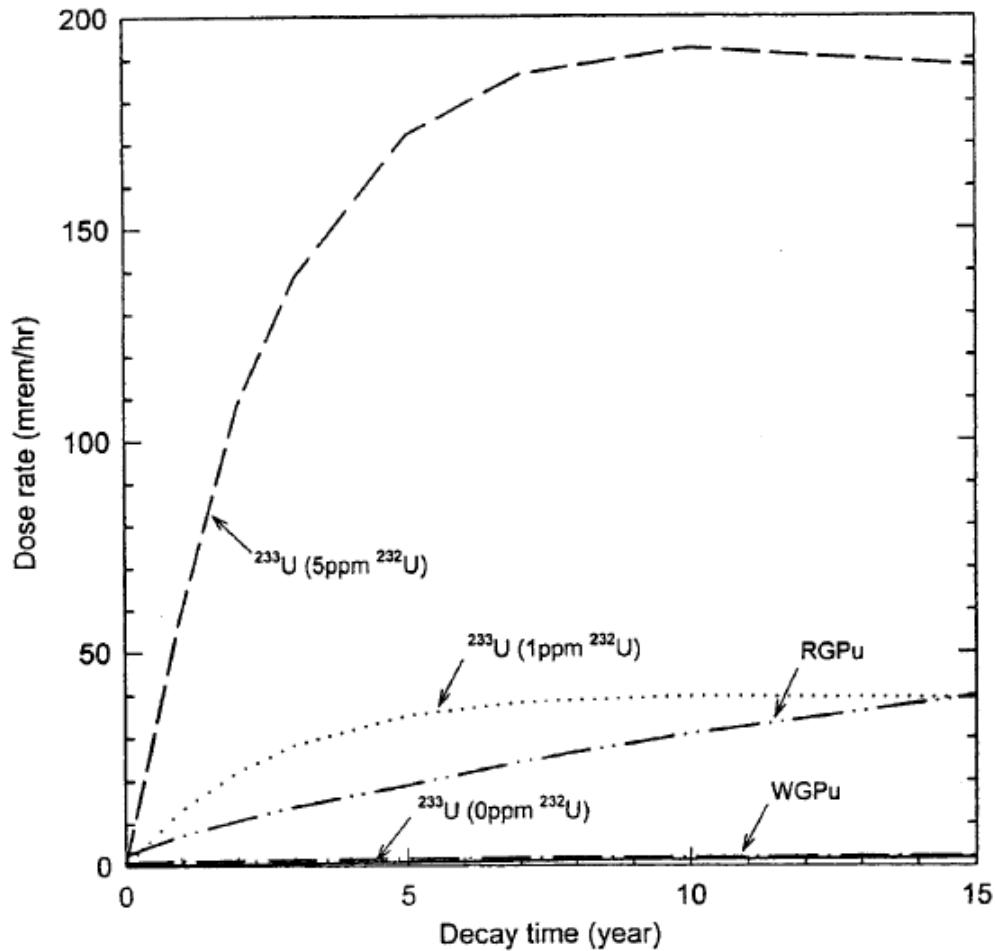


Figure 1.3: Dose rate at 0.5 m from various 5 kg spheres of ^{233}U and ^{239}Pu with different levels of ^{232}U contamination and Pu isotopic compositions respectively.[3]

safeguards regime.

The hypothetical accelerator driven system was modelled using a design that utilizes a high energy proton beam impinging on a lead bismuth eutectic target in order to produce spallation neutrons. As no accelerator-driven systems presently operate, this design was largely based off of currently available literature. Like the NRX reactor, this facility would also not be subject to IAEA safeguards. After modelling the ^{233}U production for these two facilities, safeguards recommendations will be made. This MS Thesis was completed as follows:

1. Perform a scoping analysis to quantify how much ^{233}U is required for a pragmatic critical

mass. This will act as a baseline used to inform the selection of and to evaluate the feasibility of different fissile material acquisition pathways.

2. Select facility types that could act as fissile material acquisition pathways. These should adhere to the parameters of the scenario and be capable of transmuting thorium in a timely manner.
3. Use simulations to determine optimal thorium placement in selected facilities. Perform depletion calculations to determine the fissile material production capability of the heavy water research reactor and the accelerator-driven subcritical system facilities.
4. Provide evaluation and suggestions for improving safeguarding efforts on the types of facilities and materials in question.

2. FISSILE MATERIAL REQUIREMENTS

The first task was to set a benchmark to judge the different ^{233}U production methods. The metric by which the production methods would be judged was decided to be the quantity of ^{233}U required for a state to produce a nuclear weapon. That quantity could be judged in several different ways, for example, the IAEA SQ value of 8 kg could be used. Another could be the IAEA nuclear material categorization system to inform a state's physical protection requirements (in that case for ^{233}U : 2 kg, 0.5 to 2 kg, and 0.015 to 0.5 kg for category I, II, and III respectively). [26] The bare critical mass of a sphere is another potential benchmark. In the case of this analysis, a series of benchmarks was gathered by using MCNP6 to simulate different critical configurations of ^{233}U . This, coupled with a facility's production capabilities, will indicate how easy it would be for a state to misuse that facility. The first set of configurations tested were unreflected critical spheres at varying compression factors of ^{233}U metal's natural density of $19.1 \frac{\text{g}}{\text{cm}^3}$. The k value that was achieved was not the usual 1, but rather 1.0041 which corresponds to a prompt critical mass of ^{233}U . This means that the chain reaction does not rely on delayed neutrons to continue. This k value satisfies the condition of being greater than the delayed neutron fraction β plus one. For a reaction driven by fast neutrons as these criticality calculations are, the β value for ^{233}U is equal to 0.0041. The unreflected critical masses can be found in Table 2.1. Next, a series of benchmarks was

Compression Factor	Critical Mass (kg)
Uncompressed	15.17
2	3.78
2.5	2.83
3	1.7

Table 2.1: Critical masses for unreflected spheres with varying multiplying factors on the density.

found for reflected critical spheres of varying levels of compression. The reflector material used was tungsten-carbide (WC), which is a material that has been used as a neutron reflector dating back to some of the earliest nuclear criticality experiments. The WC reflector was compressed at the same rate as the ^{233}U core. To ensure maximum effectiveness of the reflector, the thickness was incremented until the configuration received no more reactivity gains before the criticality search began in earnest. Table 2.2 shows the reflected critical masses found through this analysis. Both the unreflected and reflected critical masses served as guides to judge the fissile material production capabilities.

Compression Factor	Critical Mass (kg)	Reflector thickness (cm)
Uncompressed	5.51	10
2	1.28	8.5
2.5	0.65	6.4
3	0.58	8.76

Table 2.2: Critical masses for unreflected spheres with varying multiplying factors on the density.

3. HEAVY WATER MODERATED REACTOR

The first method of ^{233}U production to evaluate was the use of a reactor that utilizes heavy water as a moderator and natural uranium as fuel. This type of facility was selected due to this facility's ability to produce a substantial neutron flux without access to any enriched uranium or plutonium. This type of facility is also desirable to a proliferator, because a country could feasibly possess the technical ability and materials to produce a "home-grown" variant of one of these systems. The model produced for this system was based on the NRX reactor located at the Chalk River Laboratory in Canada.

3.1 NRX Reactor History

The Chalk River Laboratory and the NRX reactor were born out of war-time collaboration between Canada, the United States, and the United Kingdom. While many of the famous physicists of the Manhattan Project hailed from the two future nuclear weapons states, much of the nuclear material was provided by Canada. With sizeable uranium reserves, Canada proved a critical collaborator in the effort to produce the world's first atomic weapon, and many early nuclear physics discoveries took place at Chalk River Laboratory and its predecessor in Montreal. Some of the earliest studies of ^{233}U and its properties occurred at Chalk River. After the war, Canada leveraged their nuclear physics expertise into producing nuclear power. The NRX reactor first achieved criticality in 1947. At the time, the reactor was the most powerful in the world with a thermal power of 20 MW. The power level was later increased to 42 MW. Aside from a brief period in the 1950s to cleanup after an accident, the NRX reactor operated until the 1993. During its operational period, the NRX reactor was used to perform research on neutron scattering, isotope production, and materials to be used in nuclear power applications. The NRX reactor also served as a model for CIRUS, a research reactor in India that was used to accelerate both the Indian nuclear power and nuclear weapons programs. The NRX reactor design was also the predecessor for many similar reactor systems such as the Pakistani Khushab reactor series, the Israeli Research Reactor-2 (IRR-2)

at the Shimon Peres Negev Nuclear Research Center, the since decommissioned Taiwan research reactor, the never completed IR-40 in Iran and the never completed Iraqi Osirak reactor.[27] [24] [25] [28] The design has even been scaled up in power capability for the Dhruva reactor in India.

3.2 Technical Specifications

The NRX reactor is fueled by natural uranium, moderated by heavy water, and cooled with light water. The usage of heavy water as a moderator as opposed to light water allows for a much higher flux and power density, as heavy water has a lower neutron absorption cross section. Heavy water has advantages over graphite as a moderator for safety reasons and has a higher lethargy per collision value (ξ). The fuel rods are clad in aluminum and contain 54 kg of natural uranium. 192 fuel rods arranged in a hexagonal lattice make up a full NRX core which is placed in an aluminum vessel called a calandria. The fuel positions within the calandria are separated by aluminum tubes. The calandria is surrounded by a large graphite reflector. The use of two different liquids to moderate and cool necessitated a creative design in which the fuel rods are placed within a smaller aluminum tube that pumps light water coolant through the core. This coolant tube is then placed within the larger calandria tubes that are filled with air. The calandria tubes are surrounded by the heavy water moderator. The fuel geometry can be seen in Fig. 3.1.

The NRX reactor has two locations to irradiate samples, the central thimble and the J-rod annulus. The central thimble is an empty calandria tube in the center of the reactor core, while the J-rod annulus is a gap within the graphite reflector. The NRX reactor type layout can be seen in Fig. 3.2. Dimensions of a typical NRX reactor are detailed in Table 3.1.

3.3 NRX Modelling

The model utilized to complete the ^{233}U production analysis was of a generalized NRX reactor, created within MCNP6. This model was generic, meaning it did not account for the peculiarities of any real world variant or the material impurities that would be found in any of the real NRX-style reactors. Because of the symmetry that exists in the NRX reactor, only one sixth of the reactor needed to be modelled in MCNP6. This was done to reduce computational costs. As a result the

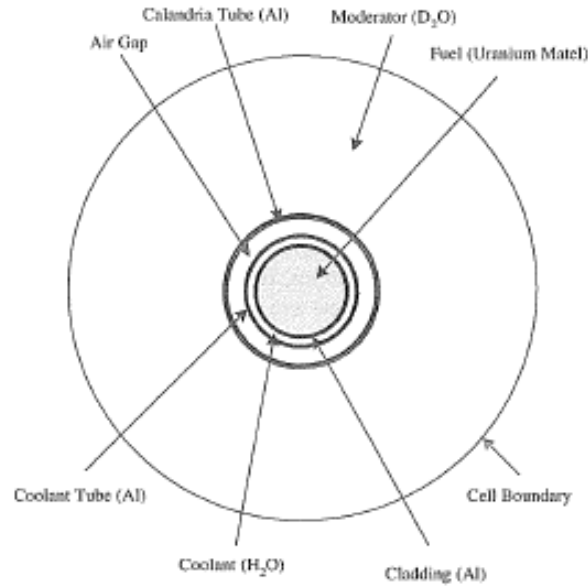


Figure 3.1: Fuel pin and calandria tube geometric configuration.[4]

Dimension	Value (cm)
Fuel Length	306
Hexagonal Pitch	17.3
Fuel Radius	1.73
Fuel Clad Outer Radius	1.74
Coolant Channel Width	0.28
Coolant Tube Outer Radius	2.11
Air Gap Width	0.65
Calandria Tube Outer Radius	3.02
Calandria Vessel Inner Radius	133.35
Calandria Vessel Thickness	0.64
First Graphite Reflector Inner Radius	137.8
First Graphite Reflector Outer Radius	160.55
J-Rod Annulus Width	6.45
Second Graphite Reflector Inner Radius	167.0
Second Graphite Reflector Outer Radius	229.2

Table 3.1: Key dimensions of a typical NRX reactor.[4].

core was divided into a 60 degree section containing 32 fuel rods, one guide tube location, and one sixth of the central thimble. The model had reflective boundary conditions on the 0 degree

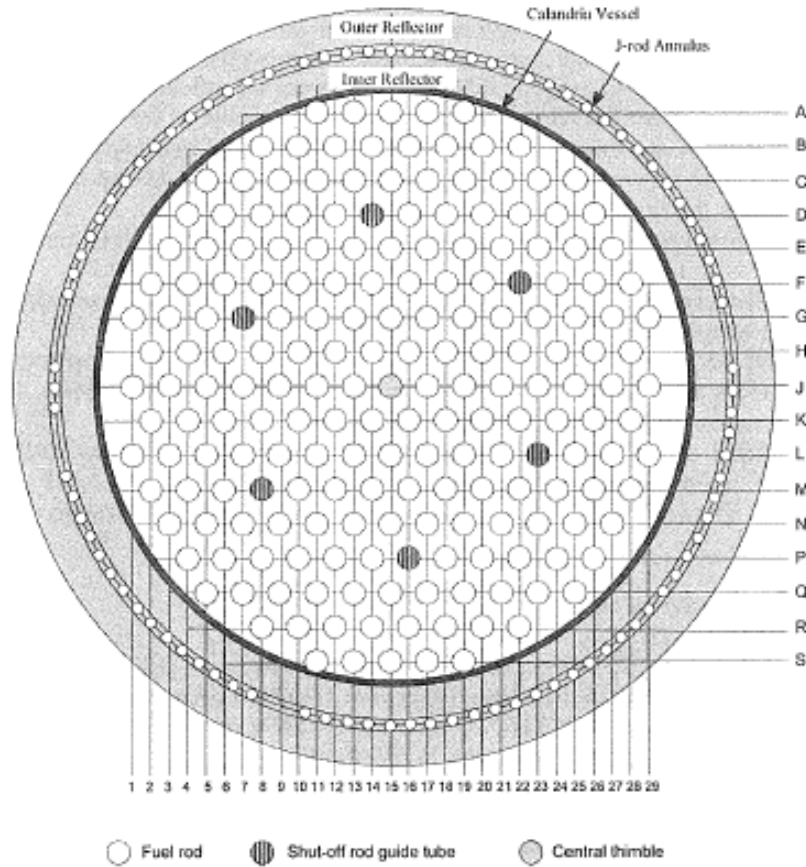


Figure 3.2: Full core cross section of a generic NRX.[4]

and 60 degree planes that divided the reactor. This reduction in the size of the simulation was particularly useful for performing fuel burnup calculations. The NRX reactor was modelled with a power of $40 \text{ MW}_{\text{th}}$. The first modelling consideration was the optimal location and configuration to use to put the thorium into the reactor. The three potential locations for irradiation targets were within fuel locations, the central thimble and the J-rod annulus. The fuel locations were ruled out as this would negatively impact the operational performance of the system, as the targets would introduce negative reactivity. Neutron flux spectra were found for both remaining locations within the reactor and compared. The two spectra obtained by performing volumetric tallies within the central thimble and J-rod annulus cells in MCNP6 can be seen in Fig. 3.3 and Fig. 3.4. While the central thimble offered a higher flux, the space is rather limited and, for this reason, the J-rod

annulus was selected as the irradiation location. The loss in transmutation speed from having a less intense neutron flux would be compensated for by the ability to irradiate more material at one time. Another benefit of this irradiation location is that the thorium does not have as significant an effect on the reactor neutronics in the J-rod annulus as it would in the central thimble location.

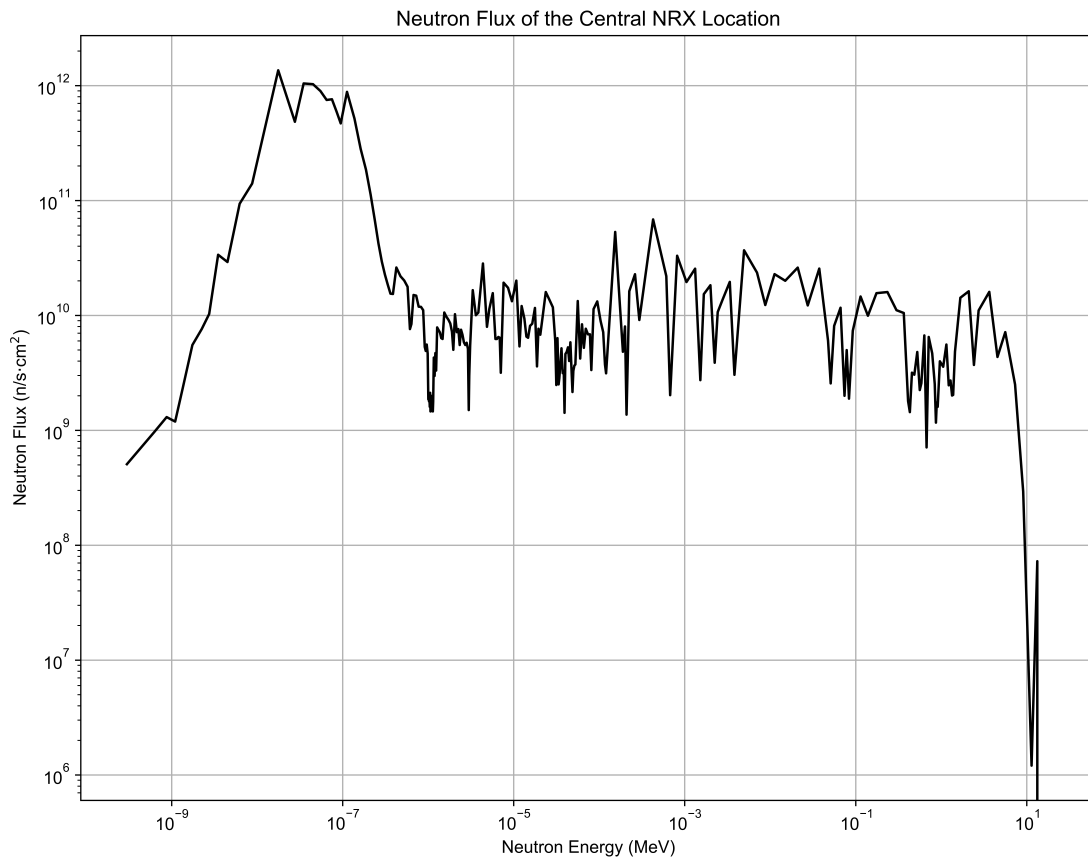


Figure 3.3: Neutron Flux in the central thimble location calculated using MCNP6.

One modification to an NRX-type reactor model that this analysis used was the assumption that the J-rod annulus could be modified to accommodate more material than how it is typically configured. Usually only 60 J-rod positions are used, however for this analysis it was assumed that

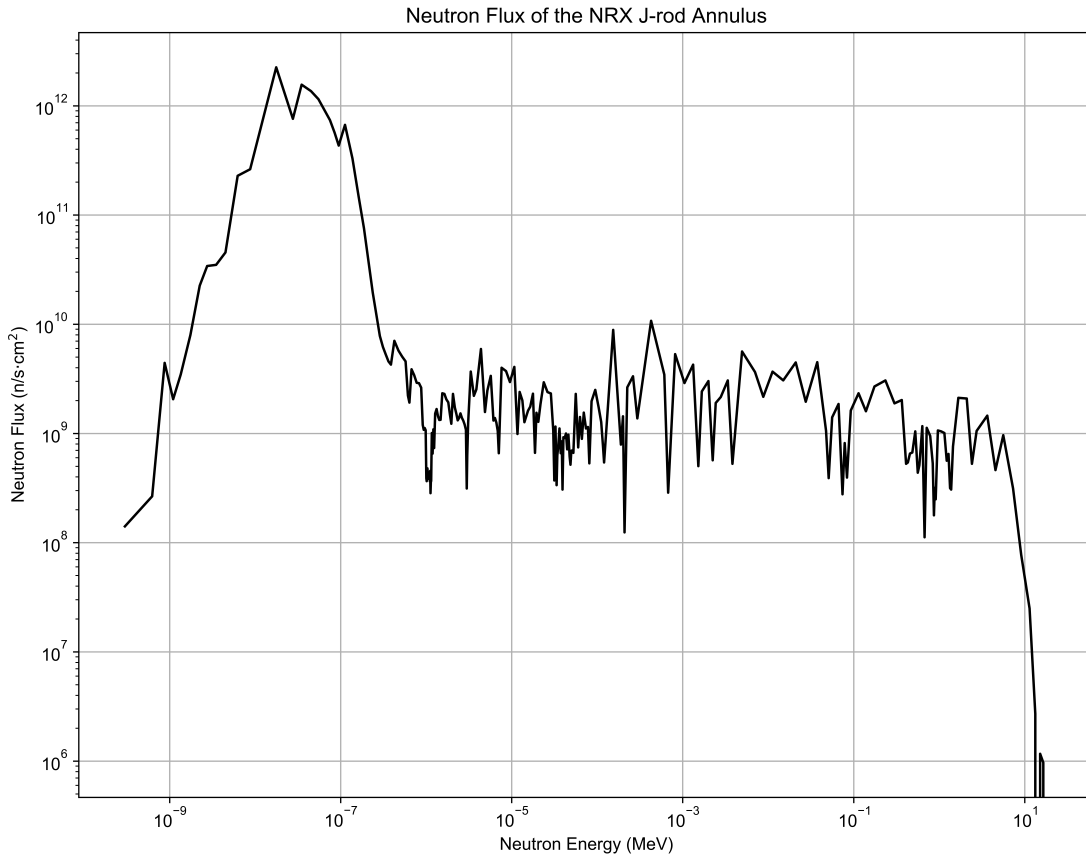


Figure 3.4: Neutron Flux in the J-rod annulus calculated using MCNP6.

the proliferator would modify the J-rod annulus to be as efficient as possible. The thorium was added in the form of metal rods with the same dimensions as the uranium fuel rods. The thorium target rods were similarly clad in aluminum with the same thickness as the normal fuel rods. 41 thorium target rods were placed in the J-rod annulus in the one sixth core MCNP6 model for a full core loading of 246 thorium targets. This was a total thorium loading of 2.7 tons, well under the IAEA SQ value for thorium. Renderings of the one sixth core MCNP6 model can be seen in Fig. 3.5 and 3.6.

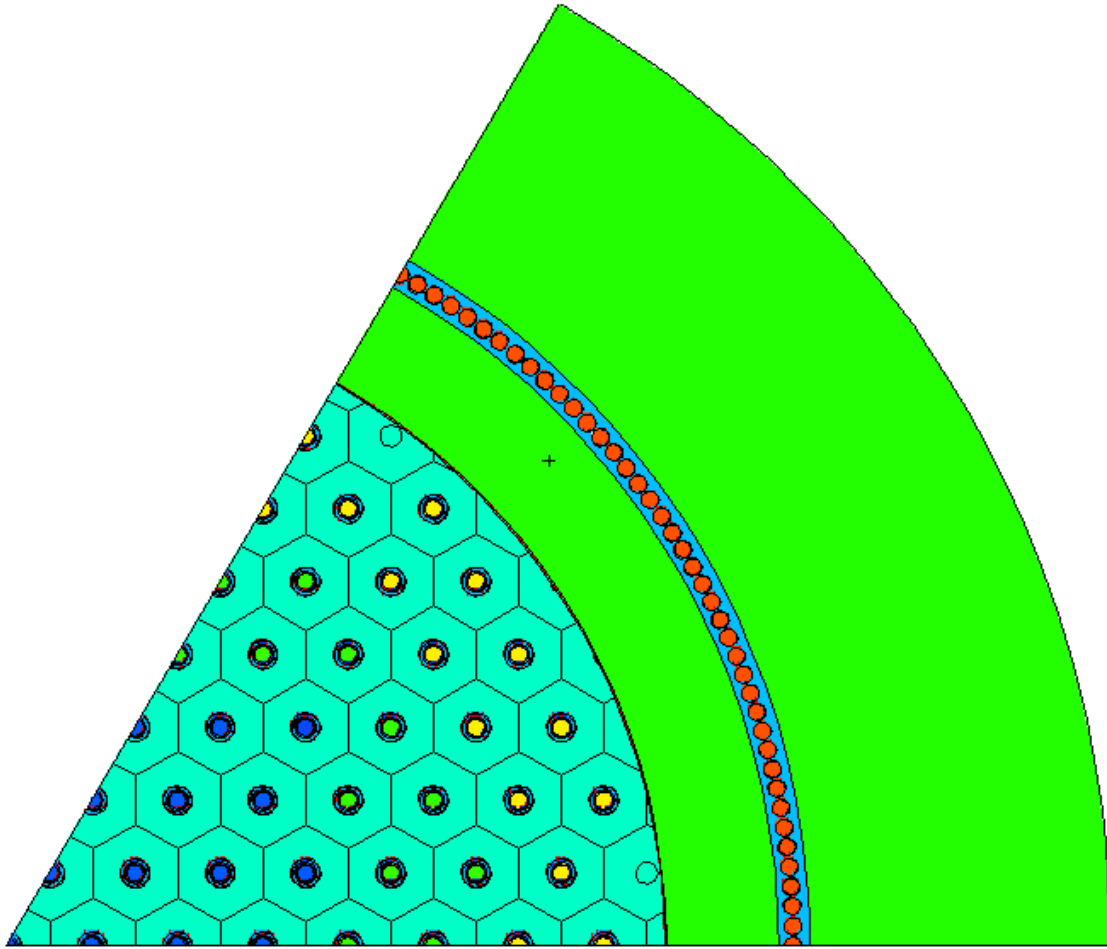


Figure 3.5: Rendering of MCNP6 model with J-rod annulus fueled (red color), in the XY view.

3.4 NRX Fuel Burnup Calculations

After the reactor core model had been loaded with thorium target rods, the final calculation that needed to be carried out was a measure of how much time was required to produce a pragmatic critical mass of ^{233}U . The burnup calculation utilized the CINDER-90 module of MCNP6, which uses reaction rates and cross sections found by performing Monte Carlo criticality calculations to calculate the isotopic concentration changes and burnup of fissile materials. The only material that was monitored was the thorium in the J-rod annulus, as it was assumed that the natural uranium fuel would be refueled often enough to maintain a constant reactor flux. This also presented significant computational savings as the natural uranium fuel would have significantly added to the

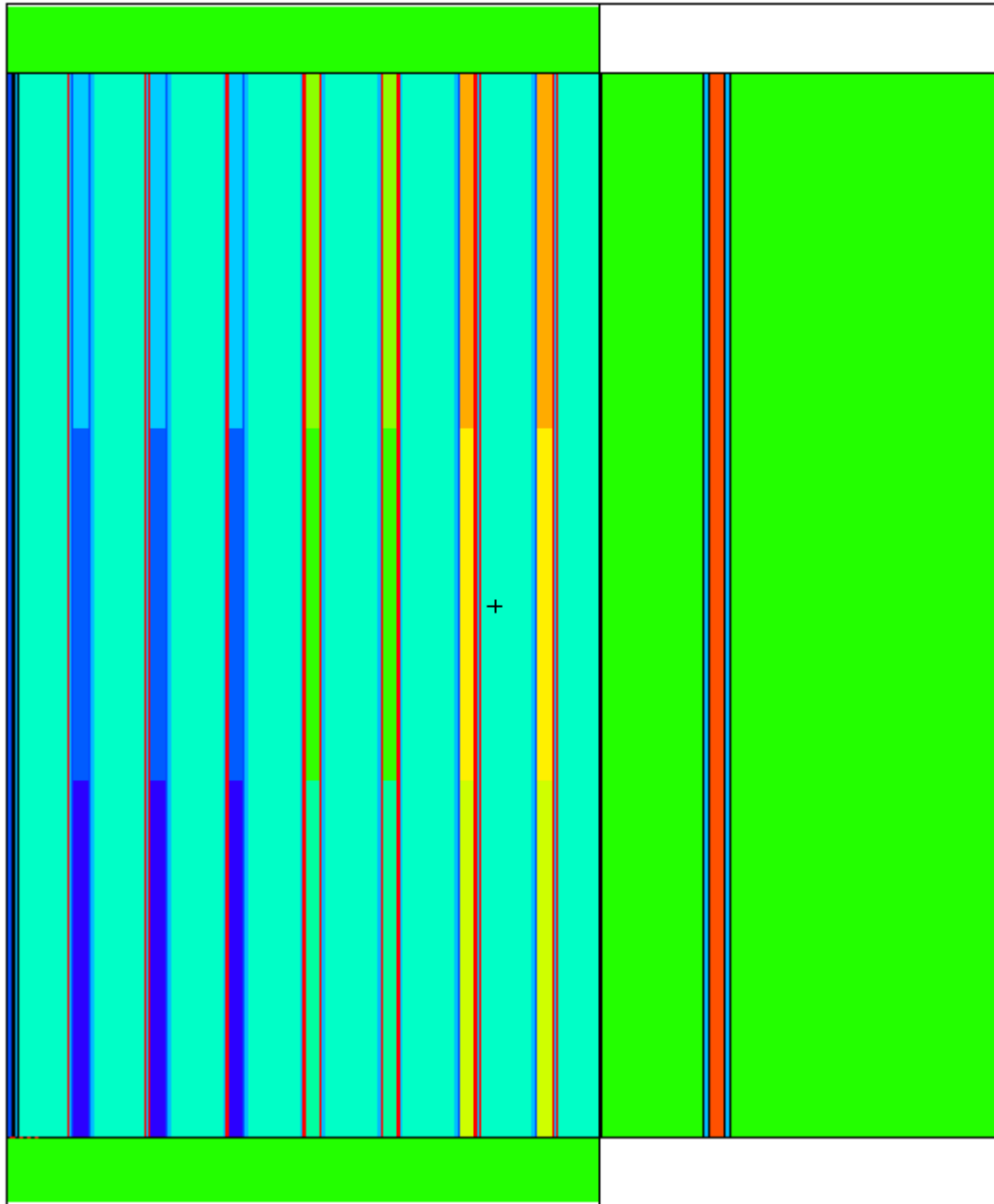


Figure 3.6: Rendering of MCNP6 model with J-rod annulus fueled (red color), in the XZ view.

workload of the multicore cluster this was performed on. The burnup calculation was conducted for a duration of 2 years, with 29 burn time steps (6 for the first month and 23 for the remaining 23 months). Each burn time step was performed with 4×10^4 particles and 205 active cycles. After one year, 1.32 kg of ^{233}U was produced with a contamination of 5.11 ppm of ^{232}U . This amount

of fissile material would satisfy the pragmatic critical mass of any of the compressed reflected geometries and would still be able to be used due to the low concentration of ^{232}U . For a 5 kg sphere of ^{233}U one year after separation with 5 ppm of ^{232}U , the material could be handled by one worker for 80 hours. The handling time for material produced in the model would be even longer, as this is significantly less material than the 5 kg sphere, and the model produced material can be handled much sooner than one year, which means work could be done before the in-growth of the main radiation hazard, ^{208}Tl . After 2 years, 2.79 kg of ^{233}U was produced with 10.1 ppm ^{232}U contamination. This mass would still be usable, and would satisfy the pragmatic critical mass for some of the unreflected geometric cases, depending on the volume compression. The production of ^{233}U can be seen in Fig. 3.7 and the ^{232}U contamination can be seen in Fig. 3.8.

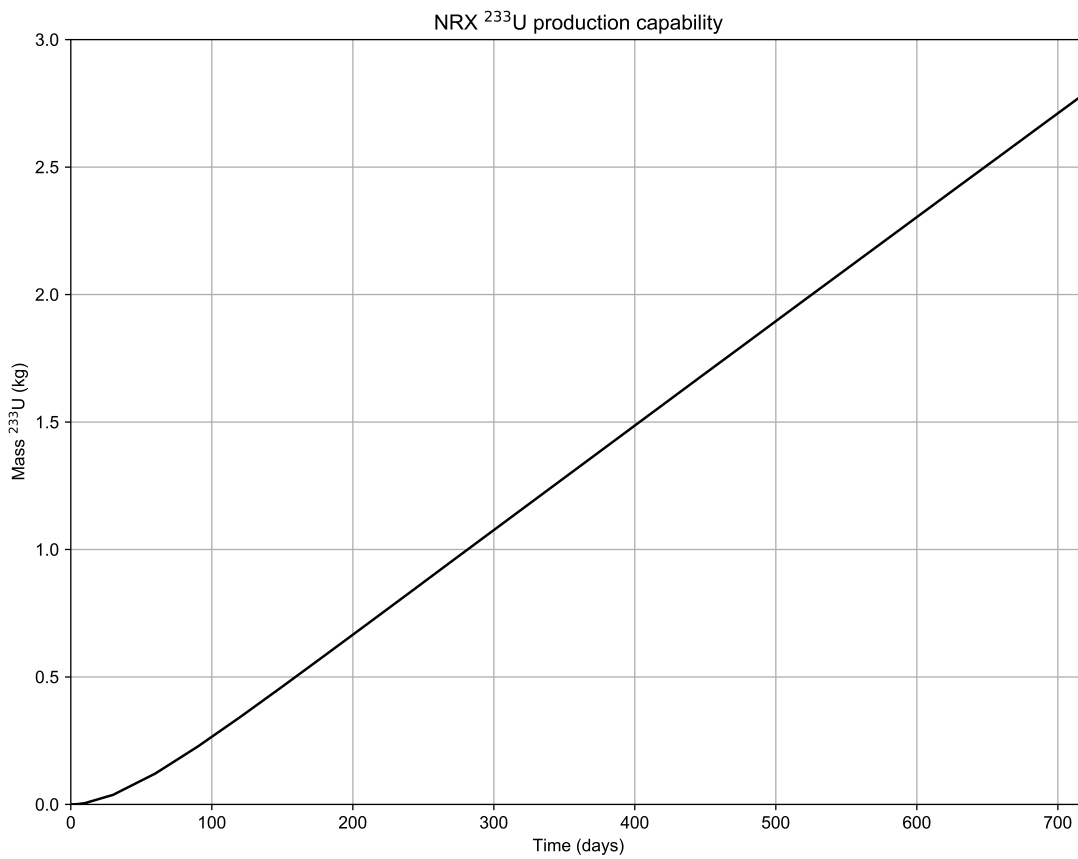


Figure 3.7: ^{233}U production within the NRX over the 2 year burn period calculated using MCNP6.

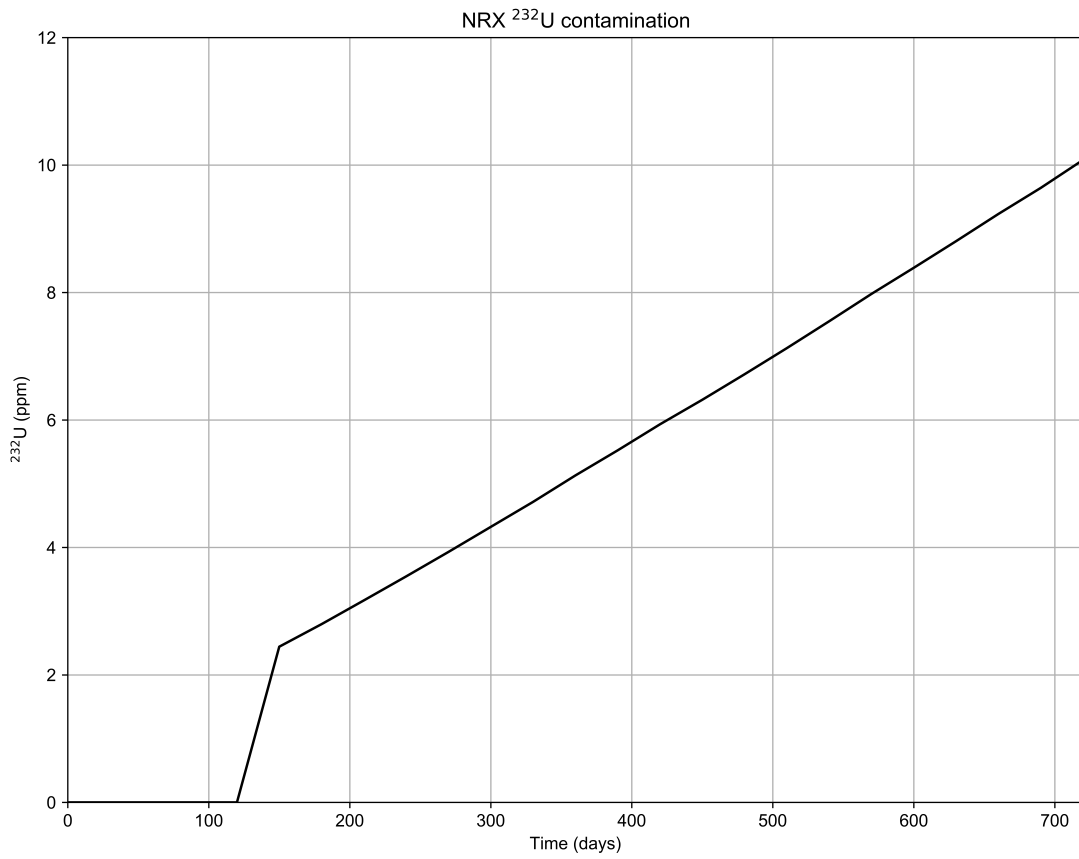


Figure 3.8: ^{233}U contamination ingrowth within the NRX over the 2 year burn period calculated using MCNP6.

4. ACCELERATOR-DRIVEN SUBCRITICAL REACTOR

4.1 Introduction to ADS

An exciting subject of research that has been developing more recently than the technology utilized in the NRX reactor are a group of systems classified as accelerator-driven systems (ADS). An ADS leverages a particle accelerator, such as a linac, synchrotron, or cyclotron, to produce high energy protons that can be used to bombard a heavy metal target and produce neutrons via spallation. Nuclear spallation occurs when a particle and nucleus collide with such force that many of the constituent nucleons are ejected from the nucleus. A heavy metal nucleus is capable of producing 20 to 30 neutrons per spallation event. An ADS would be able to use this spallation neutron source for a number of applications, like fissioning higher actinides, such as ^{237}Np , ^{241}Am , and ^{243}Am that exist in nuclear waste to produce power. Another use for an ADS would be to incorporate thorium into the nuclear fuel cycle, in which the spallation neutrons are used to breed ^{233}U from the thorium, which then continues the reaction. One advantage of an ADS is the inherent safety it possesses. As an ADS burns material that could not otherwise sustain a chain reaction, in an accident scenario all that needs to be done to stop the reaction is to turn off the particle accelerator. [29]

4.2 State of ADS Development

Currently there are no operating ADSs in the world. Several groups are working on producing an economically viable ADS, differentiating themselves with various accelerator technology and nuclear fuels. One of the most significant challenges in ADS development is building an affordable accelerator capable of producing protons with a high enough energy to produce an adequate flux of spallation neutrons. There are very few neutron spallation facilities globally, and many of them were constructed at extreme costs. It is estimated the Neutron Spallation Source (SNS) at ORNL cost up to \$1.4 billion dollars. Until the cost of accelerators begins to decline, the only way ADS can reach viability is at existing spallation facilities or at a great cost to a governmental

entity. One effort to lower the cost of accelerators that could make an ADS possible is taking place at Daresbury Laboratory in the UK. Construction is ongoing on the world's first non-scaling fixed field alternating gradient (NS-FFAG) particle accelerator. A NS-FFAG is much smaller and cheaper than typical particle accelerators and should be capable of accelerating protons to the requisite energy to produce spallation neutrons. While the facility at Daresbury will be accelerating electrons to a much smaller energy, should the effort prove viable the concept could be applied to produce an accelerator capable of driving an ADS.[30] If the NS-FFAG becomes an economically attractive method to accelerate the requisite protons of an ADS, many countries could be interested in ADS technology as a method of utilizing thorium resources for power production. Other ongoing efforts to produce an ADS are MYRRHA, which is being produced by a group of European Union (EU) countries led by Belgium, and the OMEGA project, which is a Japanese Atomic Energy Agency (JAEA) effort to leverage the existing Japan Proton Accelerator Research Complex (J-PARC) facility to power an ADS. MYRRHA seeks to produce an accelerator facility to power the system.[31] J-PARC already has a 400 MeV proton linac, 3 GeV rapid cycling synchrotron, and 30 GeV Main Ring synchrotron. These already functioning facilities form a very strong foundation for the JAEA to produce a viable ADS, and Japanese researchers are currently designing the systems that will become the ADS.[32] As a result, the spallation source MCNP model geometric and material properties as well as the proton beam qualities heavily drew upon literature concerning the OMEGA project.

4.3 ADS Modelling

The first step in producing a viable ADS model that could emulate what qualities a potentially real ADS could have is to accurately model the spallation source at the heart of the ADS. For this portion of the design, research conducted by JAEA scientists in support of the OMEGA project published in open literature was used. The proton beam used in the JAEA systems was found to be capable of producing 1 GeV protons with a current of 20 mA. This corresponds to a proton source strength of 1.25×10^{17} protons bombarding the heavy metal spallation target every second. The optimal beam shape was found to be a flat beam with a radius of 20.2 cm. This was modelled

Dimension	Value (cm)
Beam Radius	20.2
Beam Duct Inner Radius	22.5
Beam Duct Thickness	0.3
LBE Tank Inner Radius	32.5
LBE Tank Thickness	2
Active Core Height	100
Beam Window Thickness	0.2

Table 4.1: Key dimensions of the proposed JAEA ADS. [9] [10] [11]

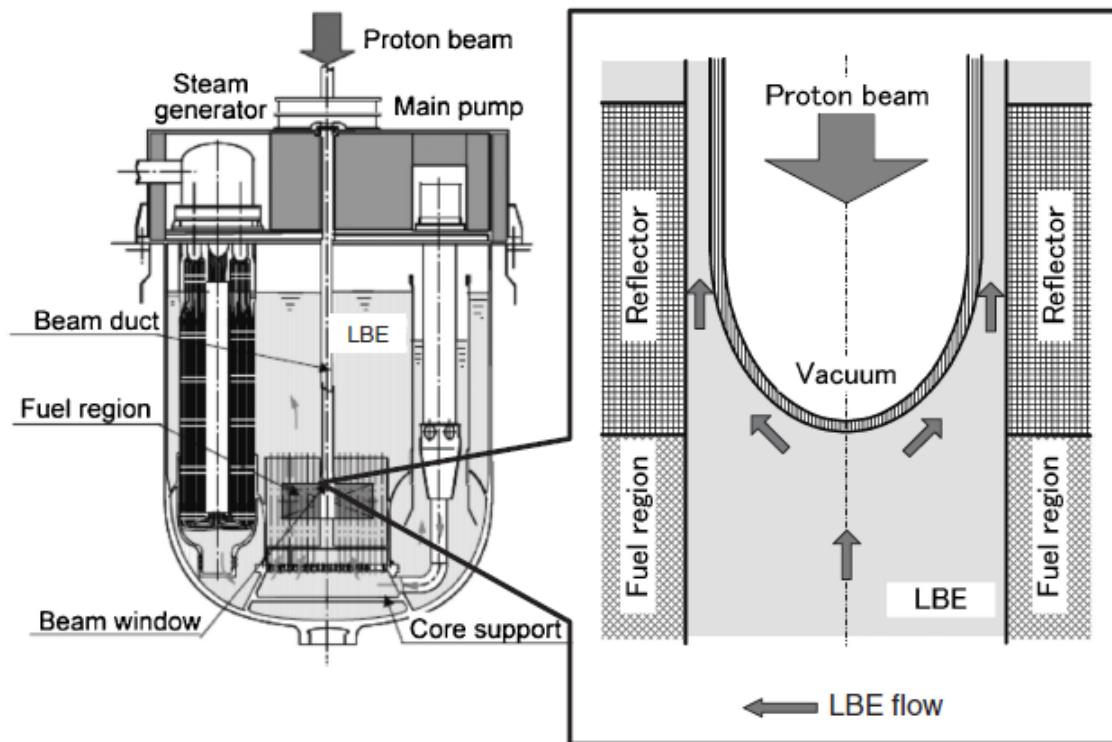


Figure 4.1: Detailed schematic of the JAEA ADS[5]

in MCNP6 with a pencil beam source definition card. The JAEA's proton beam targets a liquid lead-bismuth eutectic (LBE) to produce the spallation neutrons. LBE is used instead of pure lead because it lowers the target's melting point. LBE composed of 44.5% lead and 55.5% bismuth by weight melts at roughly 125 °C while pure lead melts at 330 °C. [33] The proton beam enters the

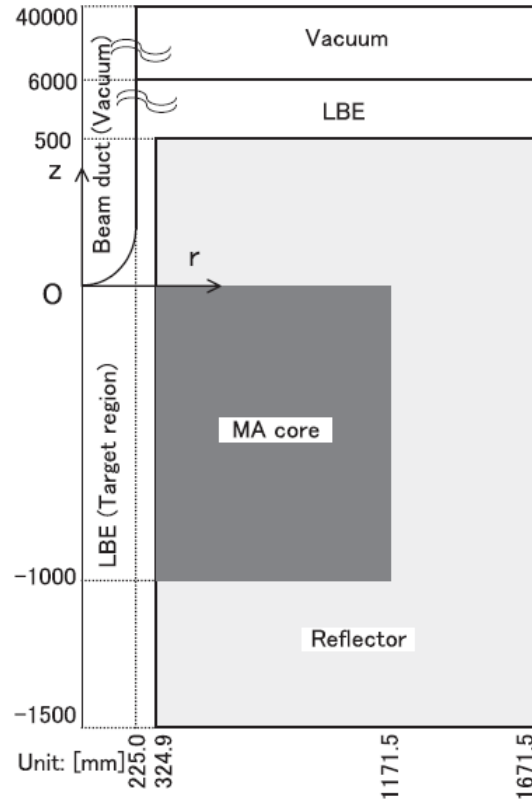


Figure 4.2: RZ view of the JAEA ADS with dimensions.[6]

Element	Concentration (wt-%)
Fe	65.27
Cr	14.14
Ni	15.87
Mo	2.34
Mn	1.54
Ti	0.22
Co	0.028
B	0.004
C	0.058
Si	0.5
P	0.026
S	0.004
N	0.003

Table 4.2: JPCA composition. [5].

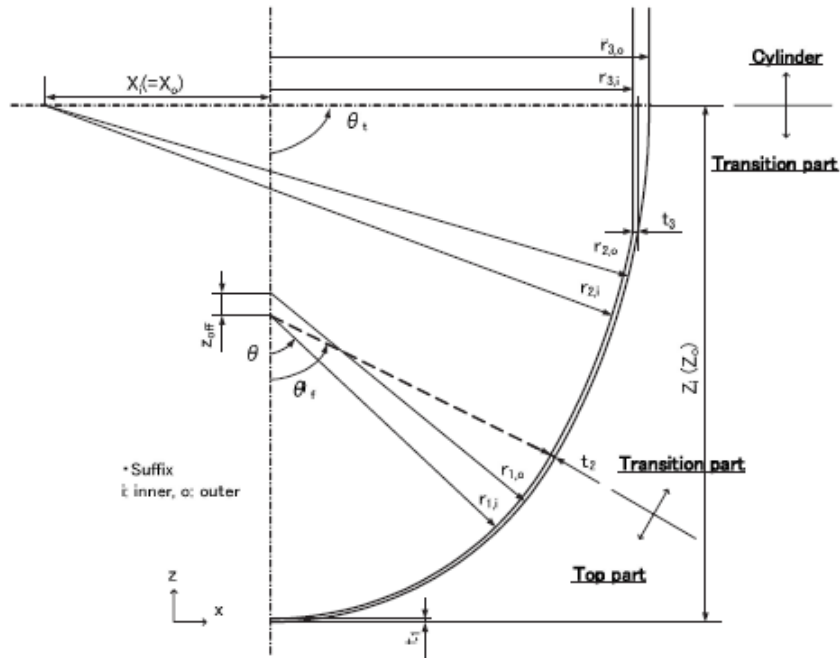


Figure 4.3: Detailed schematic of the beam window, in which the thickness transitions from 0.2 cm at the top to 0.3 cm in the cylindrical portion.[6]

top of the assembly and travels downwards through the beam duct, an evacuated chamber. Then the proton beam crosses through the ellipsoidal beam window to the cylindrical tank holding the LBE target. Pertinent dimensions of the beam duct, window, and LBE target can be found in Table 4.1 and the geometry can be seen in greater detail in Fig. 4.1, 4.2, and 4.3.

Another key detail gleaned from the JAEA research was what kind of metal alloy would be used to contain the LBE target. This alloy would have to be capable of holding molten metal for a long time, withstand high energy proton and neutron interactions, and not interfere with either particle type's flux. The preferred beam window material is an alloy called JPCA. It's chemical composition can be found in Table 4.2.

With the dimensions and compositions of the critical components scoped, the MCNP6 model was constructed. The first simplification that could be made was that only a very small portion of the beam duct needed to be modelled. Anything above the LBE target would not contain any reactor components and very few neutrons would travel directly upwards and then be reflected

down into the core surrounding the LBE. Both of these factors indicate that modelling more than a few cm above the beam window was unnecessary. Another simplification that could be made was the beam window geometry. Because the portion where the beam window thickness began to transition was outside of the radius of the beam itself, the entire beam window could be modeled as a sphere with a uniform thickness of 0.2 cm. After finalizing the neutron spallation source geometry, the irradiation positions needed to be selected. It was decided that the LBE target should be surrounded by graphite to moderate the high energy spallation neutrons. Surface flux tallies were taken at various radii surrounding the LBE target to find the most desirable moderation distance. Fig. 4.4 depicts the results of this investigation. It can be seen that the ideal distance from the LBE is roughly 15 cm from the boundary of the LBE tank at a radial value of 50 cm. Metallic thorium target rods clad in aluminium with the same radial dimensions as those utilized in the NRX analysis were placed into the core in a circle between 10 and 20 cm from the LBE target. In all, 336 rods were loaded surrounding the LBE target, for a total fuel loading of 3.7 metric tons. The MCNP6 model with full fuel loading can be seen in Fig. 4.5 and 4.6.

4.4 Target Burnup Calculations

After the ADS core was loaded with thorium target rods, it needed to be found what kind of transmutation capability this system had. After the total power, 0.32 MW, of the system was found by taking fission energy deposition tally for each thorium target rod, depletion calculations of the thorium rods were performed. Each burn step was performed with 5,000 particles and 230 active cycles. The burnup calculation was done for 2 full years. The burn was broken into 28 burn time steps (5 for the first month, 23 for the other 23 months). After 1 year of irradiation 3.06 kg of ^{233}U was produced, with negligible ^{232}U contamination. The production of ^{233}U can be seen in Fig. 4.7.

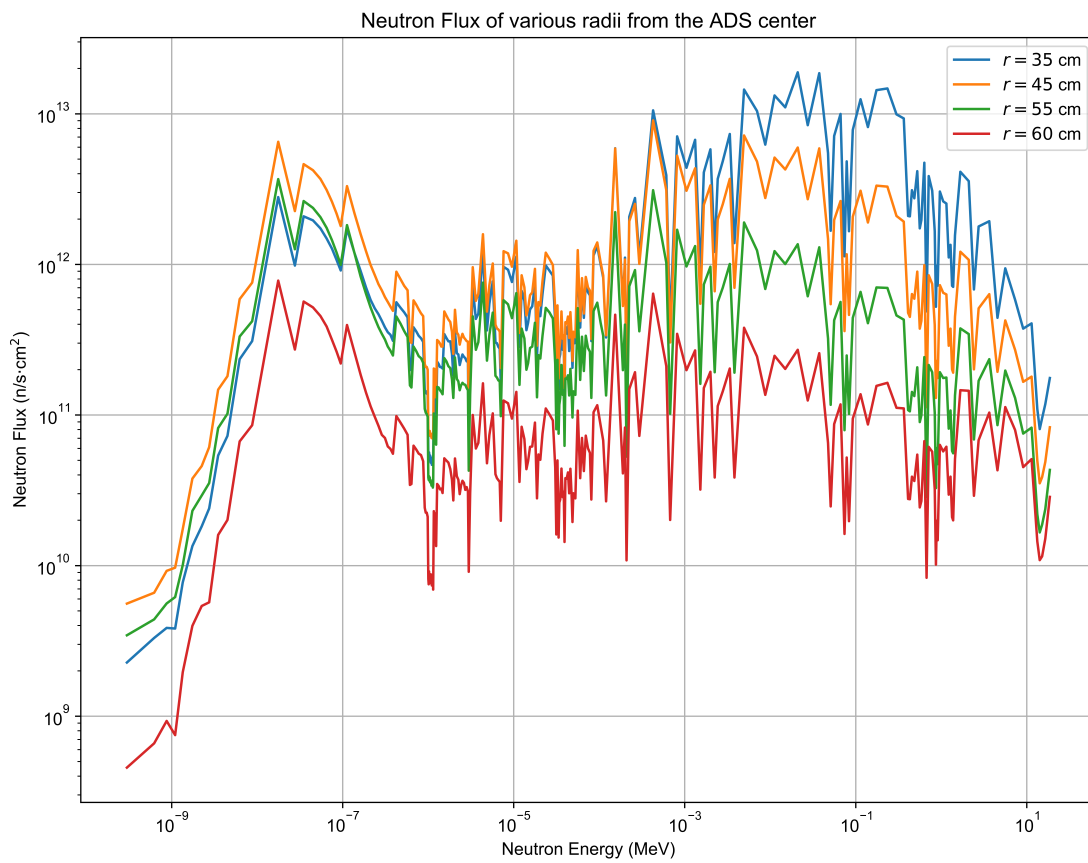


Figure 4.4: A survey of neutron flux spectra calculated using MCNP6 at different radial values away from the LBE target center.

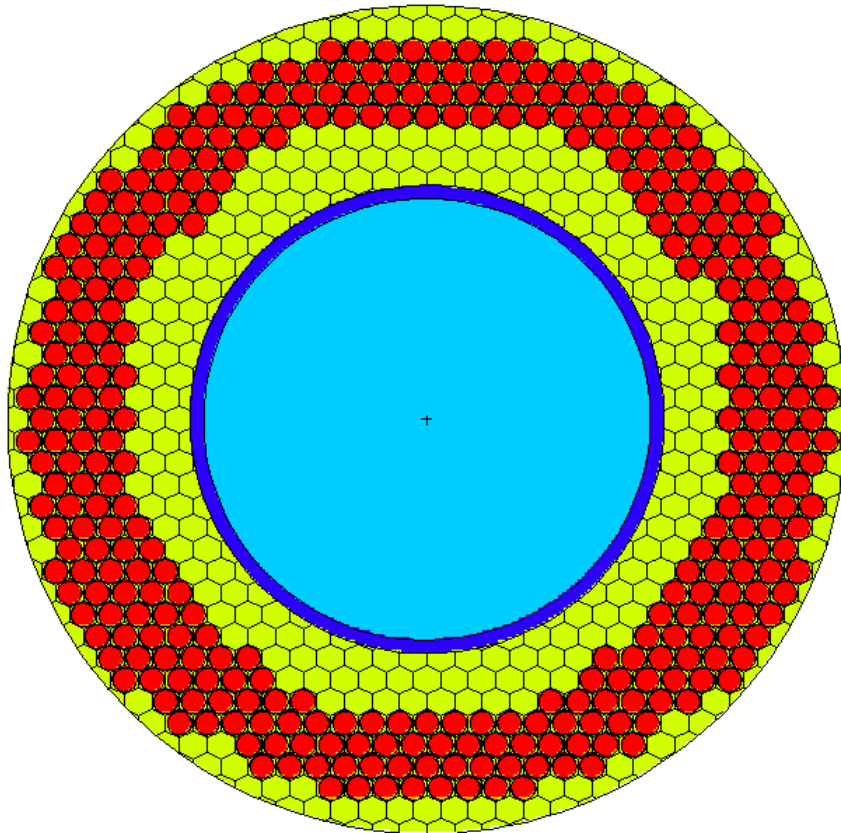


Figure 4.5: XY view of the MCNP6 model of the ADS, with light blue, dark blue, yellow, and red representing the LBE, JPCA vessel, graphite, and thorium target rods respectively.

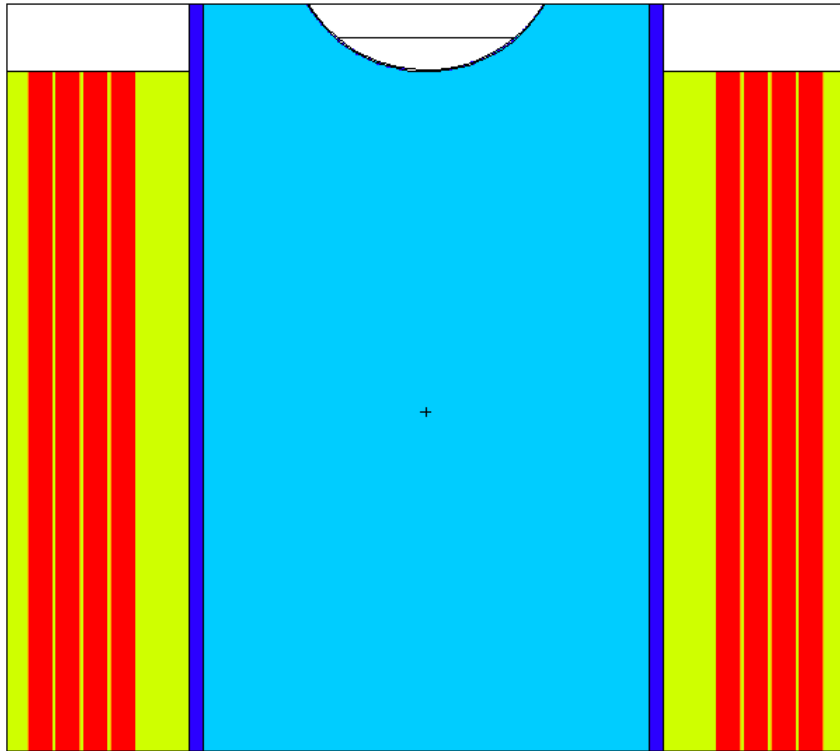


Figure 4.6: XZ view of the MCNP6 model of the ADS.

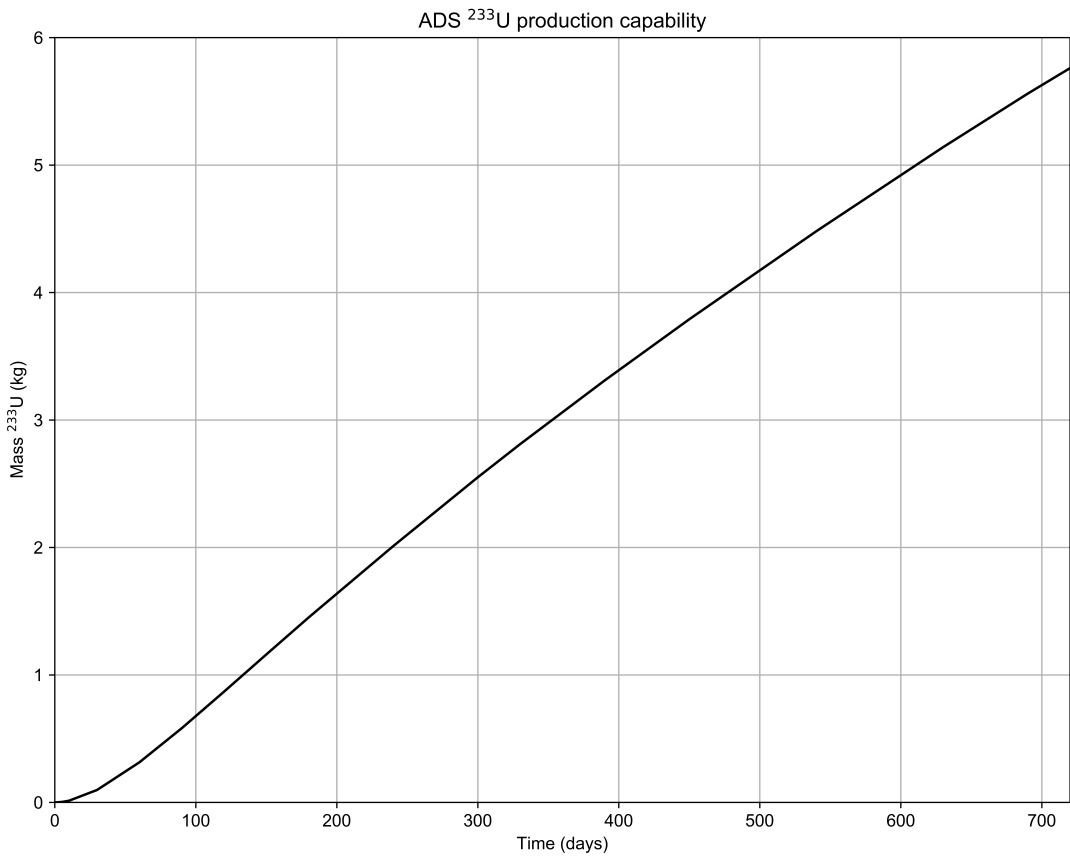


Figure 4.7: ^{233}U production within the ADS over the 2 year burn period calculated using MCNP6.

5. SUMMARY AND CONCLUSIONS

Monte Carlo simulations were carried out to find the pragmatic critical masses to judge fissile isotope productions. A model of the NRX reactor (40 MW_{th}) was used to determine the optimal location for thorium irradiation target rods, and fuel burnup calculations were done to find the quantity of ²³³U that could be produced in 2 years. Next, an ADS model was created from open literature. The optimal irradiation location was selected and another fuel burnup calculation was conducted to find the ADS 2 year ²³³U production capability. After the completion of the burnup calculations, the amount of ²³³U produced was compared to different material quantity benchmarks. This can be seen in Table 5.1. The NRX was able to produce 2.79 kg of ²³³U while the ADS produced 5.76 kg ²³³U for a two year burn period. Both of these quantities exceeded the reflected and unreflected critical mass benchmarks described in Chapter 2.

Material	Benchmark	Quantity
Th	Significant Quantity	20 t
	NRX Loading	2.7 t
	ADS Loading	3.7 t
²³³ U	Significant Quantity	8 kg
	Unreflected, 2× compressed	3.78 kg
	Reflected, 2× compressed	1.28 kg
	NRX, 2 years irradiated	2.79 kg
	ADS, 2 years irradiated	5.76 kg

Table 5.1: A comparison of Th loading and ²³³U production quantities with established benchmark quantities.

The ADS was not only able to produce a greater quantity of ²³³U than the NRX reactor, but the ADS produced ²³³U more efficiently as well. This can be seen in Table 5.2. This is despite the fact that the NRX reactor had a higher total system flux. One factor that contributed to the total quantity discrepancy is that a full ton more Th was able to be loaded into the ADS (3.7 tons compared to

2.7 for the NRX reactor). The disparity in the efficiency of the ADS and NRX can be explained by the fact that the ADS was dedicated solely to Th production, with the entire assembly surrounding the neutron spallation source catering to the needs of Th transmutation. Much of the neutron flux for the NRX was only in the actual reactor core, not the J-rod annulus that contained the irradiation targets. In the ADS case much more of the system flux was actually interacting with the Th target rods. The ADS-produced ^{233}U also had an advantage over the NRX-produced ^{233}U in that it was not affected by ^{232}U contamination. One point to note is that in reality, the NRX reactor would be much easier to produce than an ADS, which is massively complicated and hugely expensive, so much so that not a single economically viable ADS has been constructed yet. There exists some advantages to producing this material at a more economically viable and versatile facility.

Method	System Flux ($\frac{n}{s\cdot\text{cm}^2}$)	$\frac{\text{kg } ^{233}\text{U}}{\text{t Th}}$
NRX	3.86×10^{14}	1.03
ADS	8.5686×10^{13}	1.56

Table 5.2: A comparison of key system parameters as well as ^{233}U produced normalized for Th loading

In future consideration of this work, several factors that will need to be considered, namely the non-ideal nature of material manipulation and the non-ideal nature of how facilities operate. An extension of this analysis would need to account for the material losses that would be incurred in the creation of the thorium irradiation targets. Losses during milling, conversion to metal, and fabrication would all increase the amount of thorium needed to produce the pragmatic masses. Additionally there would be losses in the separation process after the fuel has been irradiated. These material considerations were outside the scope of this research effort, but would need to be addressed in some continuation of the work. Another potential factor in the transmutation process that was not accounted for in this work is that real world facilities have time when they are not operational. In this analysis, the amount of material produced during a specified period

of material depletion was found, but it would take more real time to reach that time period of depletion. For example, an NRX facility would have outage periods to perform maintenance that could not be done while the reactor operates. An NRX could also have shutdowns induced by environmental events, power outages, or even due to operational accidents. For a system like an ADS, irradiation time is reduced by the duty factor of the beam. The duty factor is found by comparing the portion of time during a cycle the beam is actually producing particle pulses compared to the total time of a cycle. If the duty factor is 100% then the beam can be considered to be continuously producing particles. This is the scenario that was modelled for this analysis. In reality, the type of system capable of producing high energy protons would have a lower duty factor. This would increase the real time needed to produce the same amount of ^{233}U . Additionally, particle accelerators experience what are called "beam trips", in which the beam is not operational or particles are not being delivered to the target. This can be caused by issues as simple as a slight misalignment of components due to vibrations or small fluctuations in power to the magnets which cause the particles to not deflect at the proper angle. Most beam trips last less than 10 s, but over the course of a long burn period these beam trips will start increasing the amount of real time required to produce the desired amount of ^{233}U . Lastly, ADS facilities will also need to undergo maintenance efforts that require the facility to not operate. All of these factors that cause a facility to be dormant were not factored into this research, and in a real scenario would mean more time would be required to produce the desired quantity of fissile material.

One question that must be asked is whether either of these production methods would be detectable by the international community. Because the amount of thorium used at these facilities is under an SQ, it is possible that this activity would not rouse the suspicion of the international community. Additionally, neither of these facilities would be subject to inspection, as they are not under IAEA safeguards. However, these facilities would be difficult to build without using many materials and technology that are considered dual-use, or that they can be used for civilian or military purposes. These items are monitored closely when traded between countries. Some materials that could be difficult to get if a country was under sanctions, or reveal the nations machinations if

merely monitored include: high purity graphite, the parts needed to build a 1 GeV proton accelerator (specifically the magnets), both the chemicals required as well as the glove boxes needed to separate the ^{233}U from the Th, and some of the metal alloys used, to name a few. While this line of research was purposely made to have the best chance of evading suspicion by the IAEA, the usage of dual-use materials would make hiding an endeavor like this very difficult, were someone monitoring a country's imports. A country would likely need to admit to at least having the facility. The international community would have very little chance to find what is happening at these facilities, as they wouldn't be subject to safeguards, but at least the global community would be aware that nuclear activities were occurring.

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