CHARACTERIZATION OF IRRADIATED FUEL AT TRIGA PUSPATI RESEARCH REACTOR

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This thesis is dedicated to my beloved family, for all the support and sacrifices throughout this wonderful journey of knowledge. Perseverance pays with meaningful step at a time. May this success spur wisdom and inspiration to my children.

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ABSTRACT

The TRIGA PUSPATI reactor is a custom model research reactor that allows storage of the irradiated fuel in the reactor tank, at the out-core. The fuel storage rack is positioned 1 m from the core, in the same vicinity of the reactor tank. In order to create a fuel-specific blueprint that validates the fuel type for safeguards and fuel characterization purposes, this study was undertaken to assess two fuel elements which were stored at the fuel storage rack. To achieve this objective, the study was divided into three main parts. The first part was focused on the assessment of neutron flux in the core using MCNP code. Profiles of fast neutrons and thermal neutrons were plotted and it was observed that thermal neutrons were fully thermalized at 158 cm with maximum flux of 1×10^{12} $n.cm^{-2}.s^{-1}$. As for the fast neutrons, the maximum flux occurred at the central thimble with the value of 5×10^{13} $n.cm^{-2}.s^{-1}$, and travelled 70 cm before became fully thermalized at the distance of 158 cm from the core. The second part, ORIGEN calculation code was used and data pertaining to radionuclide composition, energy of each radionuclide and burn up were obtained. The result of the calculation was series of photon energy emission rate in 18 energy groups as a function of irradiation or decay time and a summary table listing the principal nuclide contributors to each of the 18 energy groups. The data base produced was the fission products that were produced from spontaneous fission, and bremsstrahlung. The third part, the underwater autoradiography technique had been used in these experiments. The autoradiography analysis required a new and special-designed underwater gamma scanner rig to be developed. The underwater gamma scanner consisted of four main units; the arm block, the collimator, the stand and the main structure while the control system comprised electro-pneumatic system and control lifting system. The design of the scanner took into account conditions in the reactor pool and safety matters to avoid any incident or accident during the operation. The main results of this experiment were radiographic film images that had different brightness according to the type of wt% ^{235}U of irradiated fuel. The 3D pixel view showed the presence of AgBr in the control sample while the pixel of the film exposed to uranium 8.5 wt% fuel indicated that most of the AgBr reduced to silver build up in the film. Changes in 3D pixel view profiles analyzed for 12 wt% ^{235}U fuel exposure showed serious effects on photon impact on film. The effect of the blockade caused the pixel value to decrease by almost 29.3 % compared to the pixel value of the control sample and this made the resulting film darker than all analyzed samples. The film's gray value for exposure to 8.5 wt% ^{235}U was 128 compared to 78 for 12 wt% of fuel. Overall, when a specific fuel image was mapped with neutron-character data, then a blueprint that outlines the type and the characterization of irradiated fuel was established.

ABSTRAK

Reaktor TRIGA PUSPATI merupakan model khusus reaktor penyelidikan yang membenarkan penyimpanan bahan api terpakai di luar teras tangki reaktor. Kedudukan rak penyimpanan bahan api ini adalah 1 meter dari teras di dalam persekitaran tangki reaktor. Dalam mewujudkan cetakan biru bahan api khusus yang mengesahkan jenis dan pencirian bahan api bagi tujuan kawal selia, kajian ini dijalankan untuk menaksir dua unsur bahan api yang disimpan di rak penyimpanan bahan api. Untuk mencapai objektif ini, kajian telah dibahagikan kepada tiga bahagian utama. Bahagian pertama kajian ini memfokus kepada penaksiran fluks neutron di dalam teras menggunakan kod MCNP. Profil neutron laju dan neutron terma telah diplot dan diperhatikan, didapati bahawa neutron terma ternyahterma sepenuhnya pada jarak 158 cm dengan fluks maksimum sebanyak 1×10^{12} $n.cm^{-2}.s^{-1}$. Bagi neutron laju, fluks maksimum berlaku di pusat jidal reaktor dengan nilai 5×10^{13} $n.cm^{-2}$ s^{-1}, dan melalui jarak 70 cm sebelum ternyahterma sepenuhnya pada jarak 158 cm dari teras. Dalam bahagian kedua, kod pengiraan ORIGEN digunakan dalam pengiraan dan data berkaitan komposisi radionuklid, tenaga setiap radionuklid dan kadar bakar bahan api diperolehi. Keputusan pengiraan merupakan siri-siri kadar pelepasan tenaga foton dalam 18 kumpulan tenaga sebagai fungsi penyinaran atau masa reputan, dan ringkasan jadual senarai penyumbang radionuklid prinsipal kepada setiap 18 kumpulan tenaga. Pangkalan data yang dihasilkan adalah produk belahan yang terhasil daripada pembelahan spontan dan bremsstrahlung. Bahagian ketiga, teknik autoradiografi dalam air digunakan dalam eksperimen. Analisis autoradiografi memerlukan pelantar pengimbas gamma dalam air yang baharu dan direkabentuk khusus. Pengimbas gamma dalam air ini terdiri daripada empat unit utama; blok lengan, pengkolimat, tapak dan struktur utama sementara sistem kawalan terdiri daripada sistem elektro-pneumatik dan sistem angkat. Rekabentuk pengimbas ini mengambilkira keadaan di dalam kolam reaktor serta aspek keselamatan bagi mengelak sebarang kejadian atau kemalangan semasa operasi. Hasil utama eksperimen ialah imej filem radiografi yang mempunyai kecerahan berbeza mengikut jenis wt% ^{235}U bahan api terpakai. Pandangan piksel 3D menunjukkan kehadiran AgBr dalam sampel kawalan manakala piksel filem yang terdedah kepada bahan api 8.5 wt% ^{235}U menunjukkan kebanyakan AgBr terturun menjadi unsur perak dan menumpuk pada filem. Perubahan profil piksel 3D yang dianalisis untuk dedahan kepada bahan api 12 wt% ^{235}U menunjukkan keseriusan kesan hentaman foton terhadap filem. Kesan kepungan menyebabkan nilai piksel berkurangan hampir 29.3 % berbanding nilai pixel dari sampel kawalan dan ini menjadikan filem lebih gelap berbanding sampel-sampel lain yang dianalisis. Nilai kelabu filem bagi dedahan kepada 8.5 wt% ^{235}U ialah 128 berbanding 78 untuk bahan api 12 wt% ^{235}U. Secara keseluruhannya, apabila imej bahan api tertentu dipetakan dengan data ciri-ciri neutron, cetakan biru yang menggariskan jenis dan pencirian bahan api terpakai dapat dibangunkan.

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LIST OF ABBREVIATIONS

BU		Burn up
CT		central thimble
DNA-bare		delayed neutron activation-bare
DNA-Cd		delayed neutron activation-cadmium
DEC		cooling period where decay of fission products happens
FE		Fuel element
FOL		Full Operating Length
FPD		Full Power Days
FRR SNF RP		Foreign Research Reactor Spent Nuclear Fuel Repatriation Program
HLW		High level waste
IAEA		International Atomic Energy Agency
ID		Identification number
IRP		Irradiation power
ILW		Intermediate level waste
LLW		Low level waste
MCNP		Monte Carlo N-Particle
MCNPX		Monte Carlo N-particle
MWD		Megawatt-day
MWH		Megawatt Hour
ORIGEN		Oak Ridge Isotope Generation Calculation Code
Ppm		parts per million
PTS		pneumatic transfer tube
RTP	-	Reactor TRIGA PUSPATI
UZrH	-	Uranium Zirconium Hydride
VLLW		Very low level waste
Zr-nat		Natural Zirconium
ZZAAA		Atomic number (ZZ) and Atomic weight (AAA)

LIST OF SYMBOLS

Σ_a	total macroscopic absorption cross section for thermal neutrons.
σ_k	Spectrum averaged neutron absorption cross section of nuclides k
σ_i	Spectrum averaged neutron absorption cross section of nuclides i
Agi^0	Silver metal atom
Agi^+	Interstitial positively charged silver ion
Ag_v^-.	Interstitial negatively charged silver ion
Br^-	Bromide ion
E	neutron energy
E_f	energy release per fission
ΔE/E	average relative energy loss per collision
E	fast fission factor
F	body force per unit mass
f	thermal utilization factor or single particle probability distribution function
f_{ik}	Fraction of neutron absorption by other nuclides, which leads to formation of species i
H	height of the core
h	Planck's constant
H^+	Proton
h•	Photohole
HBr	Hydrogen Bromide
J_0	Bessel function
k_{eff}	effective multiplication factor
k_∞.	infinite multiplication factor
Φ	neutron flux
$\bar{\phi}$	Position and energy averaged neutron flux, which is also assumed to be constant over short intervals of time

η	equal of reproduction factor
λ	deBroglie wave-length
λ_a	thermal lifetime
ℓ	absorption mean free path
lij	Fraction of radioactive disintegration by other nuclides which lead to the formation of species i
m	neutron mass
m_U	metallic uranium mass in stack fuel rod
N_f	total number of fissions
n	number of neutrons per cubic centimeter
p	resonance escape probability
$p(k)_i$	irradiation power for a single fuel of ID k
v	distance the neutrons travel in 1 s
v	Velocity
t_i	period of irradiation for core configuration i
T	Time
X_i	Atom density of nuclide$i$
x	spatial position vector and location of particle
Z	Atomic number

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CHAPTER 1

INTRODUCTION

1.1 Background

The PUSPATI TRIGA Mark II reactor at the Malaysian Nuclear Agency or typically referred to by the acronym RTP that stands for Reactor TRIGA PUSPATI, is the one and only research reactor in Malaysia. TRIGA stands for Training Research Isotopes, General Atomics while PUSPATI stands for Pusat Penyelidikan Tenaga Atom Tun Dr. Ismail (the Tun Dr. Ismail Center for Atomic Energy Research). The reactor achieved its first criticality on June 28, 1982, and since then it has been operated for an average of 500 hours per year. In terms of the design, the RTP is a custom model that allow storage of the irradiated fuel in the reactor tank specifically at the out-core. The fuel storage rack is located 1 m from the core (Masood, 2016).

The RTP is loaded with 3 types of standard TRIGA Uraninum Zirconium Hydride (UZrH) fuels of differing ^{235}U composition: 8.5 wt%, 12 wt%, and 20 wt%. (Malaysian Nuklear Agency, 2015). According to the International Atomic Energy Agency (IAEA), Member States operating or having previously operated a research reactor are responsible for the safe and sustainable management of associated radioactive waste, including the spent nuclear fuel from the research reactor. Malaysian Nuclear Agency at the moment possesses six irradiated fuels in the storage rack located in the reactor pool. However, no irradiated fuel is declared by Malaysian Nuclear Agency as spent fuel yet. To declare an irradiated fuel as a spent fuel, it shall fulfil either one of the three conditions;

i. Burn up limit has been reached,

ii. Manufacturing defects such as small size, and
iii. Incident or accident involving fuel such as drop and dented.

For managing future spent fuel generated from the TRIGA research reactor, Malaysia has the option of participating in the take back program or defer the spent fuel management if the RTP will continue its operation using the current nuclear fuel. In 2014, the fuel supplier, General Atomics, through the government of United States of America offered Malaysia with the option to repatriate the spent fuel back to USA under the Foreign Research Reactor Spent Nuclear Fuel Repatriation Program or in short FRR SNF RP Program (IAEA 2008), with the condition that the reactor need to be shut down by 2016. However, Malaysia has decided to continue with the operation of the RTP for strategic reasons since the reactor is the only reactor in Malaysia, providing service and teaching tool in nuclear technology. The reactor needs to meet the demand of service especially in the Neutron Activation Analysis. At the same time, substantial investment has been expended for modernizing the reactor involving change of reactor console and upgrade of cooling system. Thus continuing the reactor in an operating state will maximize the benefit gained from the investment.

Whichever the case, it is important to fully understand the characteristics of the fuels that will be handled as exemplified in Figure 1.1. If the fuels will continue to be used, fuel characterization data is needed to prepare all documents as per regulator's requirement such as the Decommissioning Plan, Safety Analysis Report, Spent Fuel Management Plan or to prepare for safeguards inspection. In the spent fuel management plan, options are available between the closed loop and opened loop, but both options may involve probable disposal in Malaysia. If the fuels are meant to be repatriated, then fuel characterization data is imperative to design or identify suitable transfer cask and transport cask. In the case where the reactor has no more fuel for its operation, it is expected that the reactor will eventually undergo a decommissioning program. Depending on the level of decommissioning activities that will be performed, various types of waste will be generated such as very low level waste (VLLW), low level waste (LLW) or intermediate level waste (ILW).

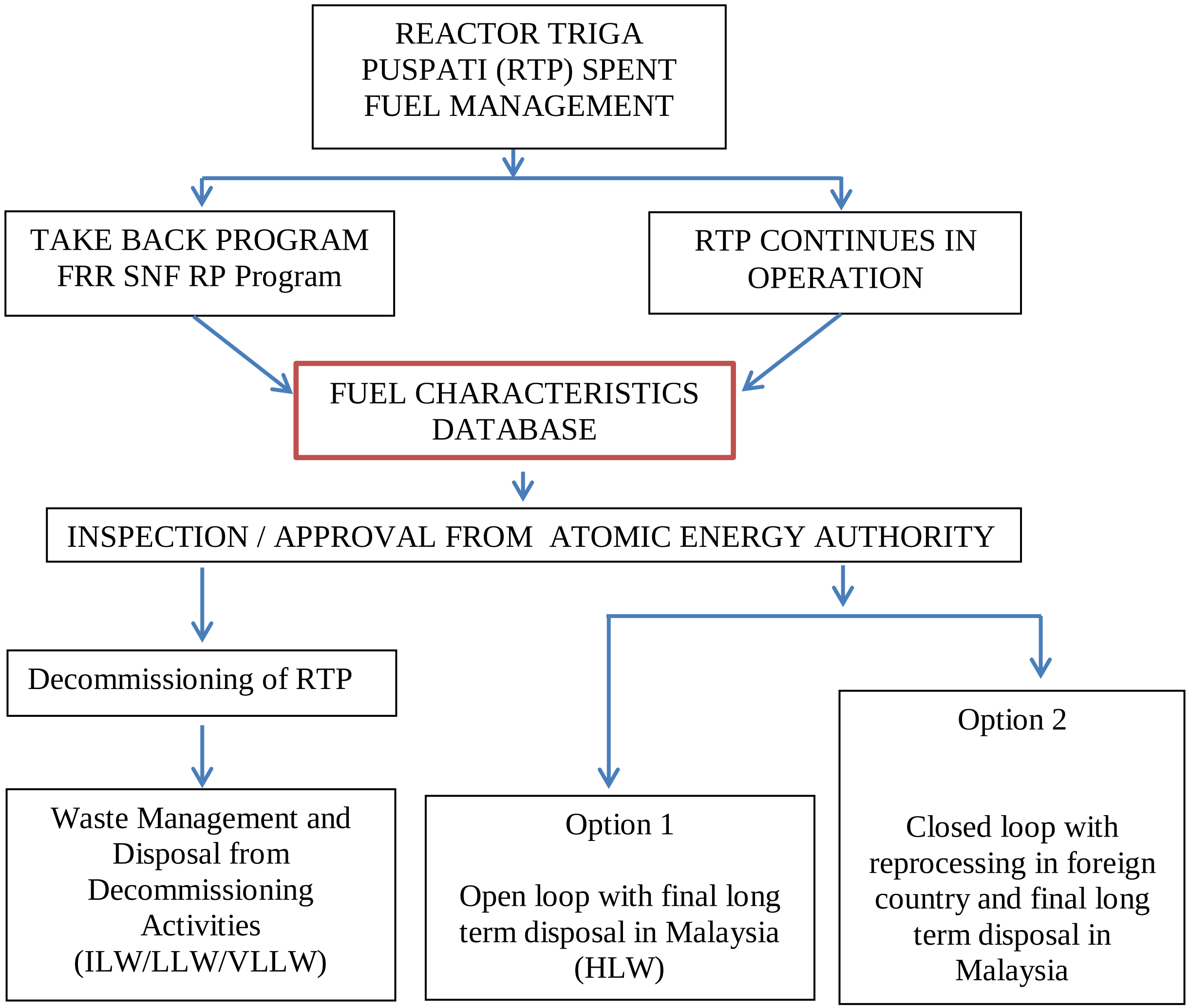

Figure 1.1 Role of Fuel Characteristics Database in Spent Fuel Management Plan

1.2 Problem Statements

It is recognized that at the moment, there is lack of facilities and appropriate instruments in Malaysian Nuclear Agency to assess and validate inventory of the fuel. Without the suitable facility, and appropriate instruments, it is difficult to perform safeguards inspection or to draw up spent fuel management plan based on fuel characteristic data. Therefore, this study is driven by two conditions which are seen as the gap in the current fuel management. The first condition is the lack of characterization data of the fuel and the second condition is the need to fulfil the safeguards requirement.

1.2.1 Lack of characterization data

In a fuel characterization program, information attributes to the characteristics, and behaviour of the spent nuclear fuel is determined. The main attribute for spent fuel is burnup, for which the parametric burn up results can lead to estimation of the actinide, activation, fission product, radionuclide inventory, decay heat, and dose rate (Sterbentz, 1997). With the availability of the characteristic data, it will support the decision for the transport, interim storage, safety analyses and repository post closure performance assessment.

There is a limited study on RTP characteristics. Ismail et.al. (2012) attempted preliminary calculation on radionuclides inventory in the fuel using assumed burn up data. Other researchers, Rabir and Usang (2015), determined characteristics data covering burn up and isotopic compositions of fuel elements of the RTP TRIGA MARK II reactor for a few selected irradiated fuels, generated from computer code simulation such as MCNP, TRIGLAV and ORIGEN. However, validation and comparison of the calculation results were not presented. There is also lack of quantitative analysis to support the calculational data, which may be derived from the lack of suitable facility capable to perform post irradiation analysis since the RTP is not equipped with attached experimental pool unlike the TRIGA MARK III model.

Characterization requires dedicated facility such as neutron activation measurement and proper simulation tools for modelling the neutronics, the dynamics and the thermal-hydraulics of the system. Although validating of calculation result is recognized to be imperative in any spent fuel characterization program, however no attempt earlier was made to perform validation test as the only suitable area to carry out such experiment involving irradiated fuel is in the confined reactor tank. Thus, it requires a specific and specially design test rig to be developed in order to conduct the validation experiments such as the underwater gamma scanner.

Furthermore, there are two irradiated fuel elements that have been stored in the fuel storage rack in the reactor pool for a very long time. The fuel elements are not being used due to defect from specification and faulty. As the fuel elements are kept in the reactor pool environment over a long period of time, hence the activation effect due to neutron exposure has now become a concern on these fuel elements. These two irradiated fuels have never been characterized so far.

1.2.2 Safeguards requirement

Management of nuclear fuel, be it spent fuel or irradiated fuel, is technically and administratively challenging as it also involves aspects of safeguards and security, besides the safety and technical aspect. Malaysia has signed the Non Proliferation Treaty on the 7th January 1968, and 2 years later on 3rd May 1970 the treaty was rectified. In line with this agreement, Malaysia is obliged to ensure any activities carried out in this country do not pose any proliferation risks (INFCIRC/182, 1973). In this decade, there have been more activities pertaining to fuel taken place at the RTP, for instance additional fresh fuel stock up, burning of fuel in the core, storage of irradiated fuel, and the recent is the construction of a spent fuel pool. Thus, there is a timely need for a safeguard analysis to be carried out to ensure that all activities are performed in accord to the requirements set forth in the Treaty.

Previous works using ORIGEN 2.2 by Usang et. al. (2015) only reported the dose rate of the fuel while Ismail et.al. determined radionuclide from a 20 wt% fuel. Clearly, there is inadequate data to fulfil the safeguards requirements in terms of nuclear material accountancy. Nuclear material accounting refers to activities carried out to establish the quantities of nuclear material present within defined areas and the changes in those quantities within defined periods (IAEA-SVS-15, 2008). This requires verification works to verify quantitatively the amount of nuclear material reported (IAEA, 2003). For TRIGA reactors, the safeguards concerns mostly revolve around the plutonium and ^{233}U production, open pool reactors with ease of refuelling and loading targets (Pan. P., et. al., 2012).

1.3 Research Goal

The goal of this research is to create a fuel-specific blueprint that would validate the fuel type for safeguards purpose and relates to other characteristics data of the fuel such as the radionuclides composition, energy, radioactivity and burn up. This blueprint will be a primary data towards establishing the spent fuel characterization database.

1.4 Research Objectives

In order to create the blueprint, the research will comprise of several works. Therefore, the specific objectives of this study are outlined below.

(a) To investigate neutron flux effect onto fuel burnup using MCNPX code for the irradiated fuels that has been stored in the RTP pool.

(b) To determine fuel element materials composition and density for the irradiated fuel ID No: 9009 and ID No: 9316 by using MCNPX code.

(c) To determine the irradiated fuel characterization such as the energy of gamma photon group, radionuclides mass and activity using ORIGEN code.

(d) To produce radiography image of the fuel element in order to map the spectrum of the energy captured by the radiography film with the neutronic calculation data.

1.5 Scopes of the research

In order to achieve the objectives of this research, the following scopes of work are outlined as follows;

(a) The neutronic calculation requires the use of several modelling simulation and computer codes. In this study, established and proven codes in this field will be used, for example ORIGEN and Monte Carlo N-particle (MCNPX) simulation code.

(b) Design, build and commission a reactor pool-in test rig. An innovative underwater radiography scanner was developed in this study which was designed and fabricated to allow experiment to be conducted within the tight space in the reactor tank, close to the reactor core. This rig will be used to create the fuel blueprint. Being the first of its kind, the rig need to pass, approved and commissioned before it can be used in the reactor. It is also subject to rigorous safety protocol that needs to be met as specified by the Nuclear Malaysia's Safety Committee in the letter dated 15 Mac 2017, reference no. NM/JKKR/Memo Klt.1 (2). Among the requirements are the finite-element-mechanical strength analysis and the estimated dose exposure to operator. The communication letter and the approval from the Safety Committee are attached in **Appendix A.**

(c) Selection of fuel for the research. Only two fuel elements will be selected for further non-destructive analysis using the radiography technique. These are the fuels that are kept at the storage rack in the pool. Fuels that have been irradiated in the core will remain active therefore, the fuel need to be taken out of the reactor core and left to ‘cool’ for certain period of time, in order to cool it from decay heat and radioactivity before characterization works can be carried out. Therefore, the fuels in the core or have been irradiated recently do not qualify for immediate characterization.

(d) Security aspect is out of scope of this study. The results from this study will be covering the safety and safeguards aspects only.

(e) At the moment, Malaysian Nuclear Agency did not register any spent fuel declaration to the Atomic Energy Licensing Board. With respect to the current standing, the term ‘irradiated fuel’ is more appropriate to be used to refer to the fuels that are already placed at the fuel storage rack. Hence, the term ‘irradiated fuel’ will be used instead of ‘spent fuel’ throughout the thesis.

1.6 Significance of the research

1.6.1 Verification of neutron effect onto burn up of the irradiated fuel at the storage rack

The RTP is a custom model that allow storage of the irradiated fuel in the reactor tank, at the out-core. The fuel storage rack is positioned only 1 m from the core, in the same vicinity of the reactor tank. In the reactor core, the neutrons are thermalized and diffused through elastic and inelastic neutron-nucleus scattering collisions. The thermalization and diffusion parameters of the neutron flux and the neutron density spectra differ according to the average energy. It is assumed that possible leaked neutrons i.e neutrons that escape from the vicinity of the fissionable material in the reactor core; travel relatively far distance from the core, reaching to the fuel left at the storage rack. Hence there is a possibility of fast neutron being thermalized and absorbed by the fuels stored at the storage rack and induce fission. According to Ueda et.al (1993), the burn up parameters can be obtained from the measurement of the neutron flux produced by burn up dependent neutron source in a spent fuel. Hence present study would verify as whether the thermal or fast neutron would influence the burn up of the irradiated fuel stored in the same vicinity of the reactor core.

1.6.2 Characterization of irradiated fuel

The fuel elements used in RTP have never been characterized in detail due to technical limitation and unavailability of suitable post-irradiation facility. Characterization data is needed to determine what is the best approach and good practices for the entire back end management of the spent fuel that include handling, packaging, transportation, treatment and perhaps disposal. The evaluation of neutron and gamma ray source distribution in spent fuel is important for the shielding design and criticality safety analysis of spent fuel storage facilities (Sasahara et al, 2004). Moreover, understanding the neutronic physics is pertinent to evaluate the burn up of the nuclear fuel, to profile nuclear isotopes for safeguard purpose, and also to determine the residual reactivity of the irradiated fuel (Bolind, 2014). The characteristics of the fuel are also needed to be described in the RTP decommissioning plan. With the characterization details collected in the study would support a more pragmatic plan can be formulated for the management of spent fuel in Malaysia.

1.6.3 Conformance to regulatory requirements on safety and safeguards

From the blueprint output, this study is able to demonstrate the conformance to regulatory requirements from the aspect of safety and safeguards. While only two fuel elements will be used for detail analysis, the developed test rig can be used by other researchers to continue working with other fuel elements of interest, leading towards a much more substantial fuel element database developed for the one hundred over fuels in the RTP.

1.6.4 A new facility designed and developed

The underwater radiography scanner developed in the research is possible to be used in other facility, not only limited to research reactor. In addition, the scanner will find its applicability in other research reactor all around the world particularly those that have limited or no post-irradiation facility. This scanner will find its usefulness for inspection, monitoring, dose measurement and validating simulation calculation.

1.7 Thesis outline

This thesis is structured and written in 5 Chapters. Chapter 1 introduces the objectives of the study, scope, problem statements and define the significance of the study conducted. Chapter 2 begins by reviewing the history of the TRIGA reactor and describes the fuel used in this study. This chapter also provide theories related to reactor physics, neutron physics, neutronic calculation and outline the radiography principle and processes applied in the subsequent works. This chapter critically review the status and development taking place with respect to fuel characterization for RTP. Chapter 3 highlights the methodology adopted in the study which encompasses the process flow, procedures, calculation code simulation as well as analytical laboratory work undertaken for the radiography analysis. The development of a new rig used in this work is also described in Chapter 3. Chapter 4 is the Results and Discussions, depicting the calculational output and radiography analysis results, followed by corresponding discussions of the results to establish arguments and comparative view with other works of similar field. Chapter 5 draws up conclusions of the study and offer recommendations for future works in order to improve or complete the fuel blueprint database initiated in this study using the radiography technique. Such work can be continued for TRIGA PUSPATI or can be emulated in other research reactor in the world using the scanner developed in this study.

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LIST OF PUBLICATIONS

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2. M.A.S Husain, S.Hashim, S.K.Goshal,M.H.Rabir,N.Zakaria,M.R.M.Zin, D.A.Bradley. Determination of out-core fuel burnup in TRIGA PUSPATI. Malaysian Journal of Fundamental and Applied Science.
   Status: accepted for publication on 25 February 2019.

3. Mohamad Annuar Assadat Husain, Suhairul Hashim, Mohamad Hairie Rabir, Norasalwa Zakaria, Muhamad Rawi Mohamed Zin, David Bradley. Investigation on Neutron Flux Effect Onto Irradiated Fuel Burn Up Stored in the Rector TRIGA Puspati. Atom Indonesia. 2019. vol 45 (1) pg 59-65.

4. Mohamad Annuar Assadat Husain and Norasalwa Zakaria. Radiography Analysis Using Newly Developed Underwater Gamma Scanner For Establishing Fuel Blueprint. Paper presented as oral presentation at the European Research Reactor Conference, RRFM/IGORR, 24-28 March 2019.

5. Suhairul Hashim, Mohamad Annuar Assadat Husain, M. Hairie Rabir, M.Rawi M.Zin. Determination of Out-core Fuel Burn-up on TRIGA PUSPATI Reactor. Paper presented as poster presentation at the IAEA Conference on Spent Fuel Management, SFM#19, 24-28 June 2019

6. Mohamad Annuar Assadat Husain and Suhairul Hashim. An overview of Burnup Calculation For Spent Fuel Characterization Using ORIGEN 2.2 at TRIGA PUSPATI. 4th International Science Postgraduate Conference 2016, UTM. 22-24 February 2016.