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Modeling and Design of Micro Flux Monitors for Neutron Fluence Measurements

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OAK RIDGE National Laboratory

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Nuclear Energy and Fuel Cycle Division
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ABSTRACT

Measuring neutron flux is critical to understanding the characteristics of nuclear reactors and can provide information on the energy spectrum, total neutron fluence, and operational history of a facility. Such measurements are typically performed by irradiating high-purity materials in an operating reactor for a known length of time. By measuring and identifying activation products in the irradiated materials and referencing the energy-dependent cross section of each isotope, the neutron energy spectrum and total fluence can be determined. Historical techniques use a variety of small, individually pressed foils or wires for dosimeter devices, which can result in excessively high levels of radioactivity and complications in handling after irradiation. The present work describes a new neutron dosimetry concept, named the *micro flux monitor*, that uses semiconductor device fabrication processes to deposit thin films of flux monitoring materials in precise geometries onto a SiC wafer. This approach allows multiple dosimetry materials to be located on a single device, simplifying irradiation and subsequent analysis. The geometry, and therefore mass, of each material can be custom-tailored for a specific application—considering the expected neutron flux, duration of irradiation, and device retrieval time of a facility. Additionally, hundreds of identical dosimetry monitors can be produced simultaneously, reducing fabrication costs. Finally, the extremely thin metal dimensions of these devices could improve the fidelity of neutron fluence measurements by reducing neutron self-shielding and gamma self-attenuation effects.

1. INTRODUCTION

Understanding the neutron energy spectrum, power density, and operational history of nuclear reactors is critical for reactor safeguards monitoring to indicate fuel reloading, higher power operation, or extended reactor cycle lengths. Measuring these properties can be accomplished using the well-established technique of neutron dosimetry [1], which determines the neutron energy distribution, flux, and total fluence of specific locations in a reactor. Neutron dosimetry functions through the process of neutron capture and subsequent nuclear reactions of specific isotopes placed in a neutron field. Activation products of these reactions are measured following irradiation and are used—along with the dosimeter mass, neutron cross section, and irradiation time—to determine the neutron spectrum and fluence.

The dosimeter materials, generally small wires or foils, are selected for their sensitivity over a specific neutron energy range, and multiple materials are typically irradiated simultaneously for more accurate measurements. The activity of each material following irradiation can differ greatly depending on the neutron cross section of the target material and the half-life of the product isotope. If the dosimeter materials are encapsulated together, logistical issues with handling can arise because the high dose rates require remote handling or radiation shielding. Additionally, excessively high activity in some reaction products can complicate the measurement of low-activity isotopes, requiring the dosimeter materials to be physically separated and analyzed independently. Finally, the finite thickness of the dosimeter foil or wire can cause neutron self-shielding during irradiation and gamma self-attenuation during post-irradiation measurements, adding uncertainty to the analysis.

This work presents a novel iteration of neutron dosimeter devices that uses semiconductor device fabrication techniques to deposit very thin dosimeter materials—on the order of tens of nanometers—in specific geometries, micrometer lengths, onto a substrate for irradiation. As a result, the mass of deposited material can be controlled to tens of nanograms, which enables precise quantities of multiple dosimetry materials to be collocated on the same device. This dosimetry device, named the *micro flux monitor* (MFM), can be custom-tailored to a specific application, considering the expected neutron flux, duration of irradiation, and device retrieval time of a facility.

In addition to being a highly customizable dosimetry device, the MFM offers the following additional benefits:

- Compatibility with corrosive or high-temperature environments of advanced/small modular reactors (A/SMRs) by using an appropriate substrate, such a SiC.
- Lower dosimeter radioactivity which permits a single, concurrent gamma spectrometry measurement for all dosimeter materials – versus sequential measurements of each separate material – to streamline safeguards monitoring processes.
- Reduction in radioactivity for minimal occupational dose.
- Improved measurement accuracy through reduction in self-shielding and self-attenuation with ultra-thin metal foils.
- Minimal flux perturbation due to the small mass of the deposited materials.
- Potential for tamper-resistant design features such as layering dosimeter materials to conceal the device composition.

- Deposition of dosimeter materials in nearly any geometry, including unique quick-response (QR) codes for identification.

- Mass production using established semiconductor fabrication processes, which reduces manufacturing cost.

- Multiple identical devices that can be irradiated together and analyzed by separate, independent laboratories.

The following discussion presents the results of modeling and design considerations for a prototype MFM device that will be fabricated and tested in Oak Ridge National Laboratory's (ORNL) High Flux Isotope Reactor (HFIR) [2] later this year, 2024. The modeling process began with down-selection of dosimetry materials that are compatible with semiconductor fabrication instruments and with other dosimeter materials when collocated. Next, neutronics modeling of the pneumatic tubes (PTs) in HFIR was performed to determine neutron spectrum and flux intensity anticipated during testing. Irradiation of proposed MFM materials was simulated using the ORIGEN [3] isotopic depletion and decay code—part of the SCALE nuclear modeling and simulation suite—referencing the modeled flux values in the HFIR PTs. The results from these simulations were used to determine the mass of dosimeter materials required to achieve 10 nCi of activity in each product isotope under different irradiation scenarios, informing the geometry (thickness and diameter) required for fabrication. Finally, an overview of the geometry and materials that will be used for the prototype MFM devices is presented.

2. METHODS OF NEUTRON DOSIMETRY

Neutron dosimeters are a well-established analytical tool for measuring fluence rate, fluence, or the neutron spectrum of nuclear reactors. The principle of operation is to irradiate several dosimeter materials in high-flux locations of a reactor, then extract and subsequently measure neutron activation products in the irradiated materials. The dosimeter materials are selected for their sensitivity in a specific neutron energy range and generally, though not always, produce gamma-emitting activation products that can be measured using a high-purity germanium detector (HPGe). To measure the neutron energy spectra in a reactor that spans several orders of magnitude (from eV to MeV), multiple neutron dosimeters with sensitivities in different energy ranges are typically irradiated together. For example, ^{54}Fe and ^{47}Ti could be selected for high-energy threshold reactions, ^{59}Co and ^{93}Nb for intermediate neutron energies, and ^{197}Au and ^{115}In for thermal neutrons, as shown in Figure 1. Once irradiated, the dosimeters will have residual radioactivity predicted by

$$A = N_0 \bar{\sigma} \phi \alpha (1 - e^{-\lambda t_1})(e^{-\lambda t_2}) , \quad (1)$$

where:

A	=	activity (Bq) of the product nuclide at the time of counting
N_0	=	initial number of target atoms
$\bar{\sigma}$	=	average cross section (b) of target isotope
ϕ	=	neutron flux (n/cm^2/s)
α	=	branch ratio of activation products
$(1 - e^{-\lambda t_1})$	=	buildup of the activation product nuclide during t_1 irradiation time (s)
$(e^{-\lambda t_2})$	=	decay of activation product following irradiation until measurement at time t_2 (s)
λ	=	decay constant for product nuclide ($-\ln(2)/T_{1/2}$), where $T_{1/2}$ is the half-life (s)

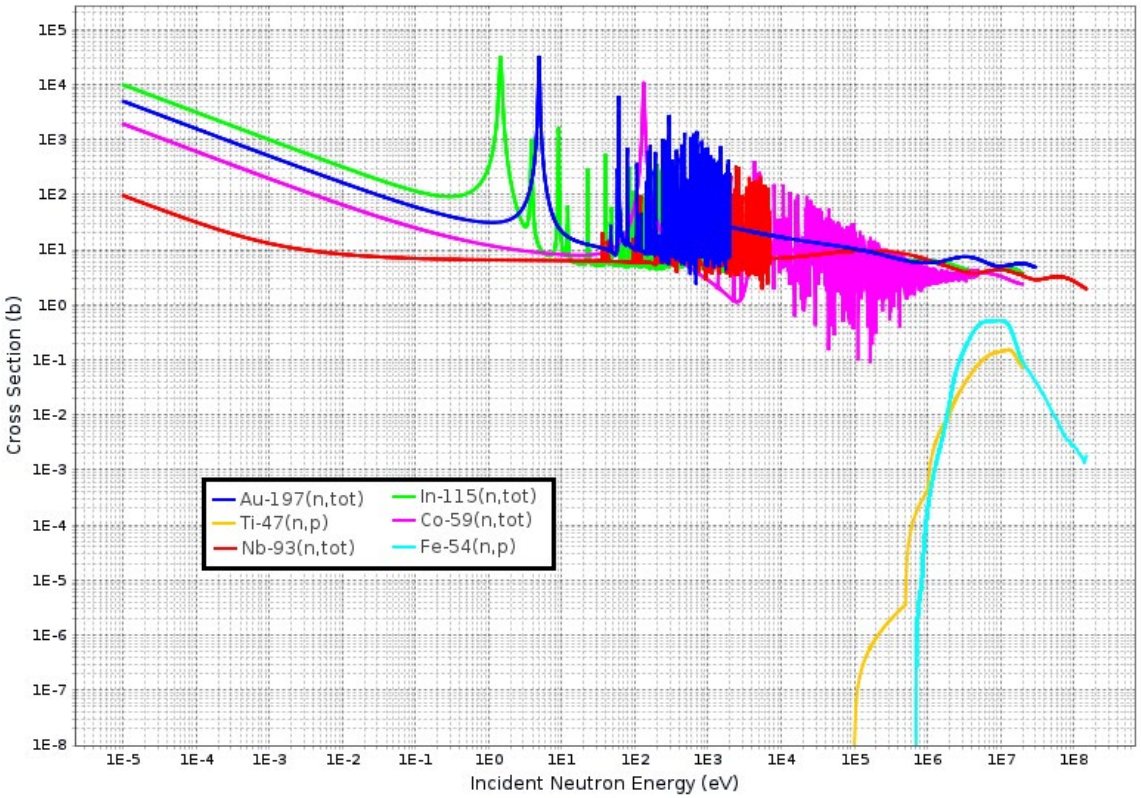

Figure 1. Neutron cross sections of several isotopes commonly used for neutron spectrum measurements.

After measuring the activities of each monitor, neutron spectrum adjustment and deconvolution codes such as STAYSL PNNL [4] and SAND-II [5], respectively, can be used to determine the neutron flux spectrum and neutron fluence.

Many ASTM standards have been published to guide dosimetry measurements, including detector calibration and analysis methods [6], procedures for performing neutron dosimetry [7, 8], and methods for measuring reactions in specific materials such as Ni [9], Al [10], and U [11]. Standards also exist to guide the selection of the appropriate dosimetry materials [12] and for performing neutron spectrum adjustment [13] calculations from dosimetry activity measurements. However, even with these standards and practices, complications can arise when performing fluence measurements in high-flux facilities.

Historically, fluence monitors are in the form of foils or wires, have masses of 1–1,000 mg, and are fabricated as small as possible to minimize neutron self-shielding and gamma self-attenuation. However, even using the smallest wires or foils requires correction factors to account for these effects and can increase uncertainty in the final results. Certain dosimeter materials can generate curies of activity in high-flux reactor environments, requiring remote handling or hot cell facilities for analysis, and complicated radiological disposal pathways. Some long-lived isotopes, such as ^{60}Co, are often diluted in a different metal to 1 wt% to reduce this excessive radioactivity. Due to this high residual activity, disassembly and extraction of individual monitors are typically required, with separate gamma counting to avoid excessive dead time in the HPGe detector. It is also necessary to protection from contamination and use high-purity dosimeter materials to avoid contribution from side reactions that produce the measured product but are not related to the reaction being studied. Finally, the mass and composition of each monitor material must be tracked from assembly, through irradiation, to gamma analysis, which can be challenging because of the monitors' compact size.

3. MICRO FLUX MONITORING DEVICE

The MFM device seeks to alleviate many of the previously mentioned issues of performing neutron dosimetry using conventional fluence monitors. The motivation for this device is to use well-established semiconductor clean room metallization processes, such as electron-beam (e-beam) evaporation, to fabricate thin film neutron dosimeter foils onto a substrate, thereby controlling the initial number of target atoms in a predictable and repeatable manner. This would enable deposition of a precise quantity of flux monitor material with sufficient mass to perform an accurate flux measurement—but with minimal residual radioactivity to allow for manual handling, concurrent HPGe analysis with other monitor materials, and a simplified disposal pathway. First, an assessment of suitable materials for MFM devices was performed.

3.1 MATERIALS

The STAYSL PNNL software suite provides neutron cross section data for 81 different nuclear reactions from 44 elements commonly used for neutron dosimetry measurements. Down-selection of proposed MFM materials began with these 44 materials, listed in Table 1. Figure 2 shows a representation of the nuclear reactions in these materials, with the left axis corresponding to the target isotope, the bottom axis showing the product isotope following neutron capture, and the right axis grouping isotopes from the same element. Reactions from actinides, the majority of which produce fission products, were not included. Green squares in Figure 2 represent a specific reaction pathway between target isotopes on the left axis and product isotopes on the bottom axis. Red highlighted regions depict nuclear reactions from different target elements that result in the same product isotope, which can be problematic in the MFM device. For example, ^{59}Co will produce ^{60}Co, as well as several other reaction products; however, ^{60}Ni and ^{63}Cu can also produce ^{60}Co via (n,p) and (n,a) reactions, respectively. If Co, Ni, and Cu are located on the same dosimetry device, determining the contribution of each reaction to ^{60}Co production would be challenging.

Table 1. 44 unique materials used for neutron fluence measurements in the STAYSL PNNL code.

Li	V	Nb	Ta
B	Cr	Mo	W
F	Mn	Rh	Au
Na	Fe	Ag	Hg
Mg	Co	Cd	Pb
Al	Ni	In	Bi
Si	Cu	I	Th
P	Zn	La	U
S	As	Pr	Np
Sc	Y	Cg	Pu
Ti	Zr	Tm	Am

For down-selection of MFM materials, those that do not produce a delayed gamma reaction—Li, B, and F—were excluded, since the measurement method utilizes delayed gamma spectrometry after irradiation. Additionally, “cover materials”—Cd and Gd—used for filtering thermal neutrons in dosimetry measurements were not considered. Next, actinide elements—Th, U, Np, Pu, and Am—were removed because depositing these materials using e-beam evaporation is not currently available in ORNL clean room facilities. Other elements that are not conducive to e-beam evaporation—Na, P, S, As, I, La, Pr, Tm, and Hg—were eliminated for similar reasons. Finally, because MFM devices are intended for use in high-temperature A/SMRs, materials with a melting point below 500°C—Zn, In, Pb, and Bi—were removed as neutron dosimeter materials.

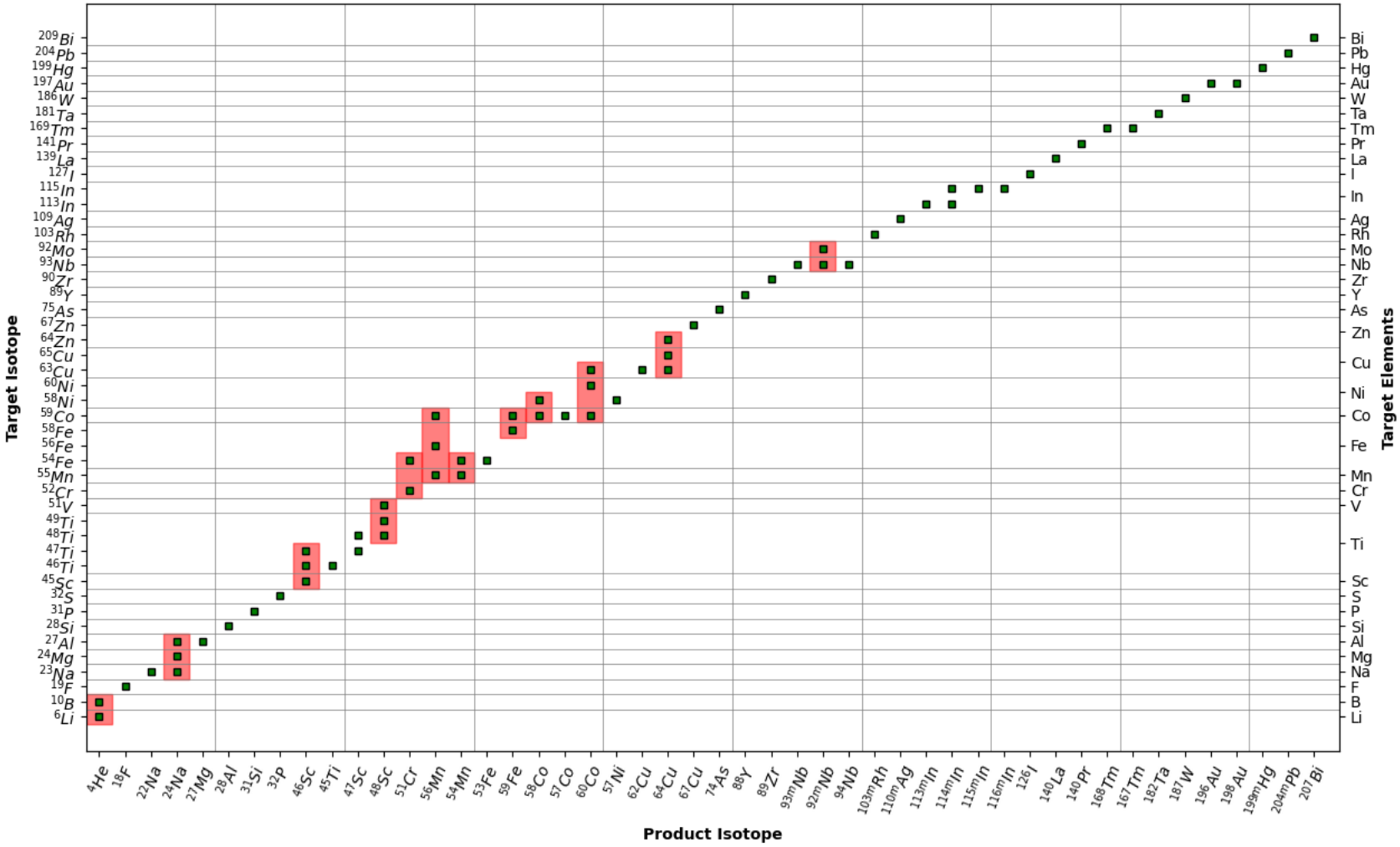

Figure 2. 68 of the 81 nuclear reactions documented in STAYSL PNNL code.
Note that cover reactions (Cd and Gd) and actinide reactions are not included.

The remaining 21 elements that are suitable for both neutron dosimetry measurements and e-beam deposition are listed in Table 2; however, certain caveats apply to several materials. Si is included in the substrate material (SiC) and will not be useful as a dosimeter in this application. However, Si could be used in future designs with a different substrate material. Y is highly reactive and relatively expensive, Zr is an export-controlled material, Nb can have deleterious reactions with the W crucible of the e-beam tool, and W can form volatile oxides. With the exception of these 5 materials, the nuclear reactions from the down-selected materials conducive to MFM devices are shown in Figure 3.

Table 2. 21 candidate materials for dosimeters in micro flux monitors.

Mg	V	Ni	Mo
Al	Cr	Cu	Rh
Si	Mn	Y	Ag
Sc	Fe	Zr	Ta
Ti	Co	Nb	W
			Au

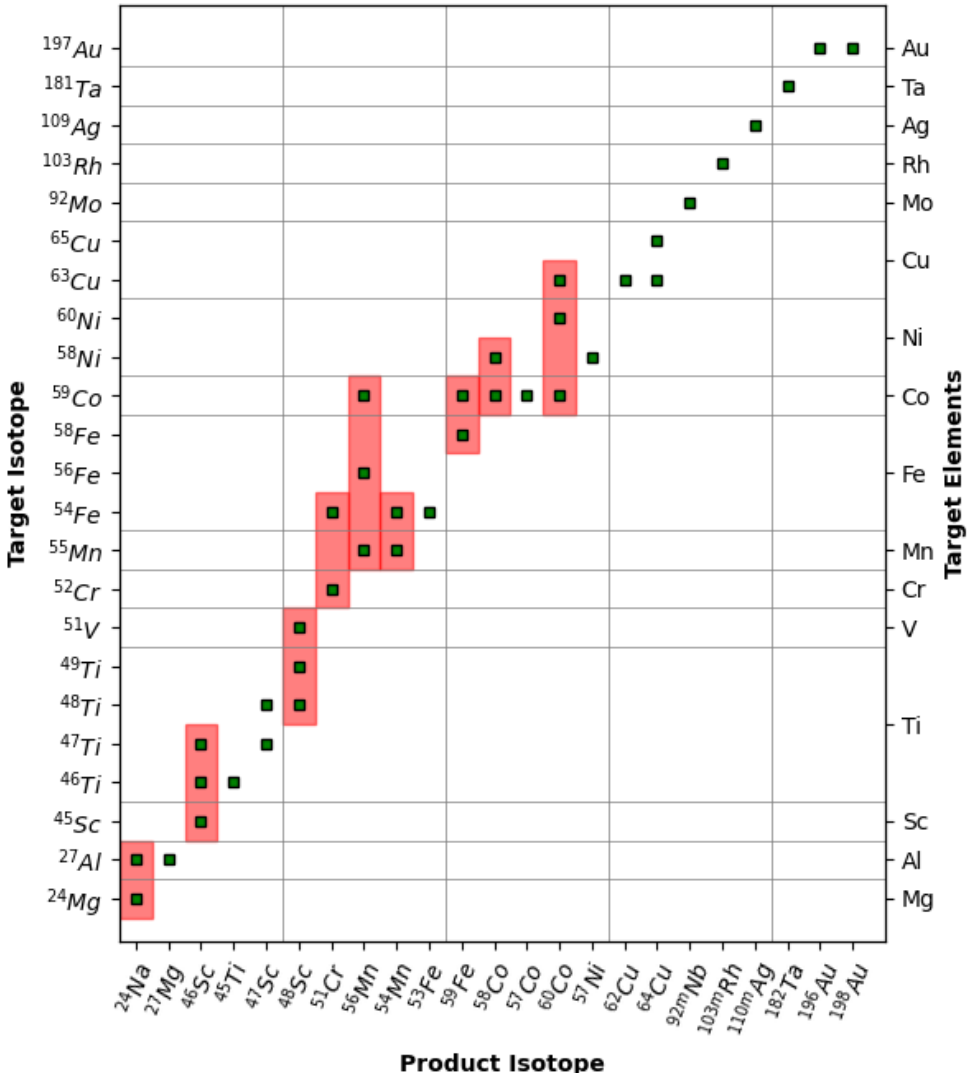

Figure 3. Nuclear reactions of 16 materials down-selected for use in MFMs.

In total, 16 elements are the most viable candidates for fabricating MFM devices. These 16 elements are composed of 23 naturally occurring target isotopes that produce 23 product isotopes through 38 neutron capture reactions. Several of these reactions yield the same product isotope from different target isotopes, which is problematic if the two materials are collocated in the same dosimetry device. These cross-competing reactions are highlighted in red boxes in Figure 3. A compatibility matrix of these materials is depicted by element in Table 3, with 'X' marks indicating materials that cannot be located on the same MFM device.

Table 3. Dosimetry materials which cannot be collocated with one another.

Al	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	
X										Mg
										Al
		X								Sc
			X							Ti
										V
						X				Cr
						X	X			Mn
							X*			Fe
								X*	X	Co
									X*	Ni

3.2 DEVICE FABRICATION

Prototype MFM devices will be fabricated in the Nanoscale Science and Technology Laboratory (NSTL) and the Center for Nanophase Materials Sciences (CNMS) cleanroom facilities. 4-inch wafers of high-purity SiC will be used as substrate materials to deposit dosimetry metals. The surface area of each metal will be controlled using standard lift-off deposition processes. First, the wafer will be coated with a thin layer of photoresist and/or lift-off resist. Next, a customized photomask will expose geometrical shapes onto the photoresist using an ultraviolet (UV) light source. The wafer will then be washed in developer to dissolve regions of the resist that were exposed to UV light, leaving the geometric shapes from the photomask transferred into the photoresist. The area of these shapes will determine the surface area of the metal dosimeters used in the MFM. The wafer will be placed in an e-beam evaporation tool, which uses an electron beam to evaporate various metals under high vacuum. Metal vapors are then deposited on the wafer, and the deposition rate of target material can be monitored using an in-situ quartz resonator in the tool. Once the deposition is complete, the wafer is placed in a solvent to dissolve all remaining photoresist on the wafer. Any metal that was deposited on top of the resist is removed, leaving metal deposits on the wafer in the geometric patterns from the photomask. This lift-off process thus allows for control of the thickness and surface area—and therefore the volume and mass—of each dosimeter material. Following liftoff, the metal may be sputtered with a protective coating, such as Si_3N_4, and cut using a dicing tool to separate the MFM devices from each other.

Although there are 16 candidate dosimeter materials for MFM devices, each material requires individualized process parameters in the e-beam tool and calibration of deposition rates, which can be time intensive. Therefore, consideration for which materials are currently available in the e-beam evaporator reduced the preliminary MFM design to 4 monitor materials: Al, Ni, Ti, and Au. Three additional materials—Mo, Cr, and ZrO_2—are also available but may require additional process characterization and are considered "stretch goal" monitor materials in the initial prototype.

3.3 MFM LAYOUT

The MFM design layout will include several geometrical patterns to resolve key questions concerning the feasibility of this technique. These questions include:

1. Is the area of circular monitors more accurately controlled than rectangular patterns?
2. Is the dosimeter mass more sensitive to area or thickness?
3. Is the deposited material fully dense?
4. Can the monitor materials be stacked over one another?
5. Can the monitor material be patterned accurately in a unique arrangement, such as a QR code?

To answer these questions, several patterns will be deposited into 5 × 5 mm cells on the SiC wafer. Several hundred MFM devices can be created on each wafer, which will then be separated using a dicing tool.

3.3.1 Coplanar Design

Circular and rectangular patterns of the 4 MFM materials will be deposited in a coplanar design, as shown in Figure 4. This coplanar patterning allows each flux monitoring material to be irradiated and analyzed via gamma spectrometry without self-shielding or self-attenuation from the other materials, although these effects are expected to be negligible. The metallization pattern will also be deposited in 4 different surface areas to determine how accurately the volume of material can be controlled through lithography. If the metallized area can be precisely controlled, then the activity of each dosimeter material should follow a linear trend. The circular and rectangular patterns will have the same surface area and should, in theory, result in the same residual activity. Comparing measurements from both will help determine whether linear or circular metallization patterns are more uniform. The metallized surface area of each coplanar design is reduced by a factor of 2 and ranges in size from 2.63 mm^2 to 0.38 mm^2. The MFMs will also have a unique serial number deposited on the bottom for traceability. The serial number will be made from Cr, which can also be used as a dosimeter material.

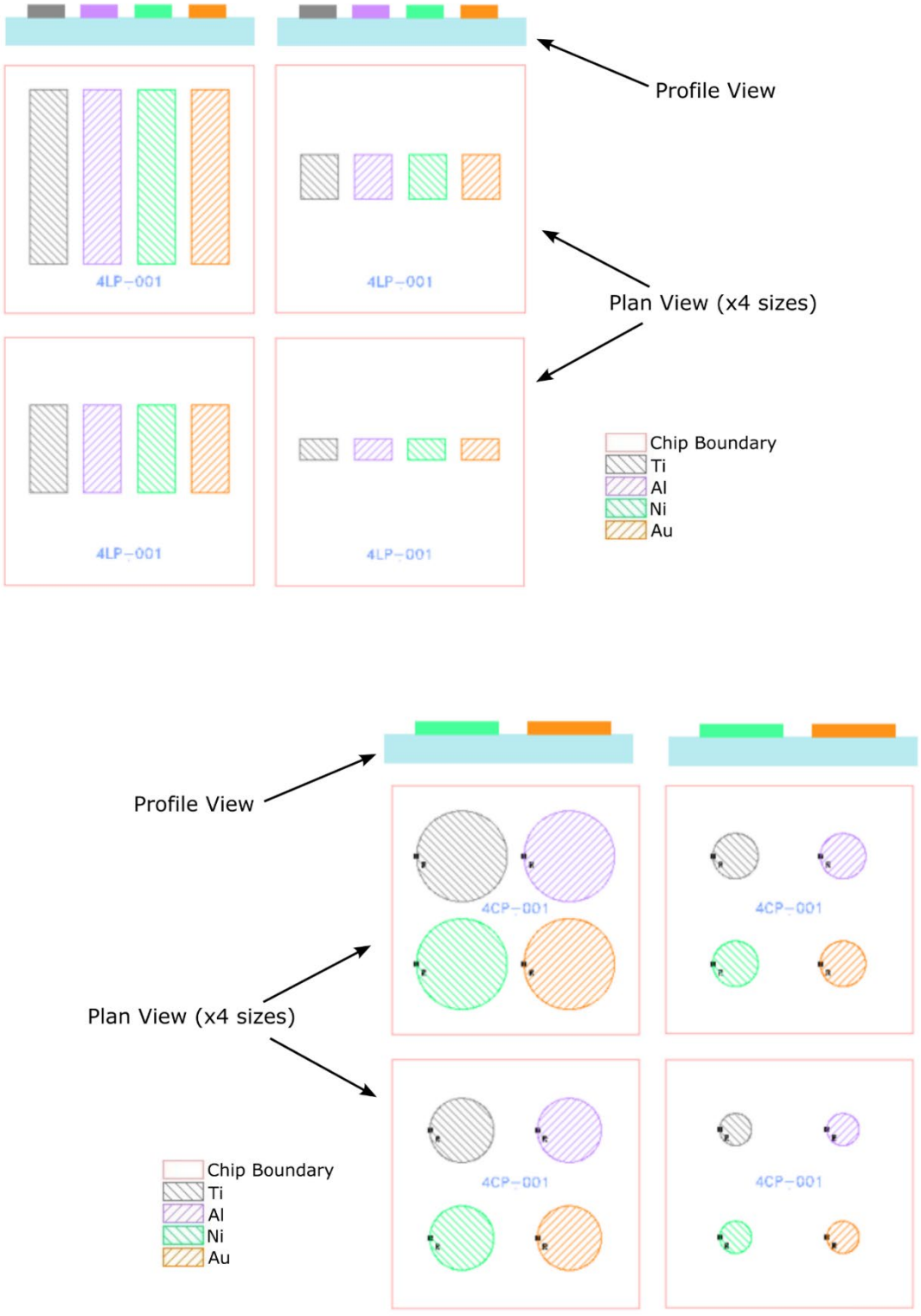

Figure 4. Metallization patterns for coplanar MFM prototype devices. Linear patterns (upper) with various surface areas, along with circular patterns (lower), will be deposited and compared.

3.3.2 Colinear Design

The colinear design MFMs, shown in Figure 5, have the same rectangular and circular pattern as the coplanar design, however each dosimeter material is deposited in a vertical stack. In this orientation, it is possible for neutron shielding or gamma attenuation to occur. However, the thickness of each layer is so small that these effects should be negligible. If experiments show that this effect is negligible, and the colinear design makes for an effective dosimeter, this will make fabrication significantly easier and cost effective. Depositing a colinear stack of metals requires only a single photolithography exposure, metal deposition, and lift-off. The coplanar fabrication method requires these steps to be repeated for each deposited material. Since the dosimetry metals are deposited sequentially without venting in the e-beam tool, the risk of contamination in the interface of each layer is mitigated. However, the precision and certainty in the thickness of each layer may be reduced.

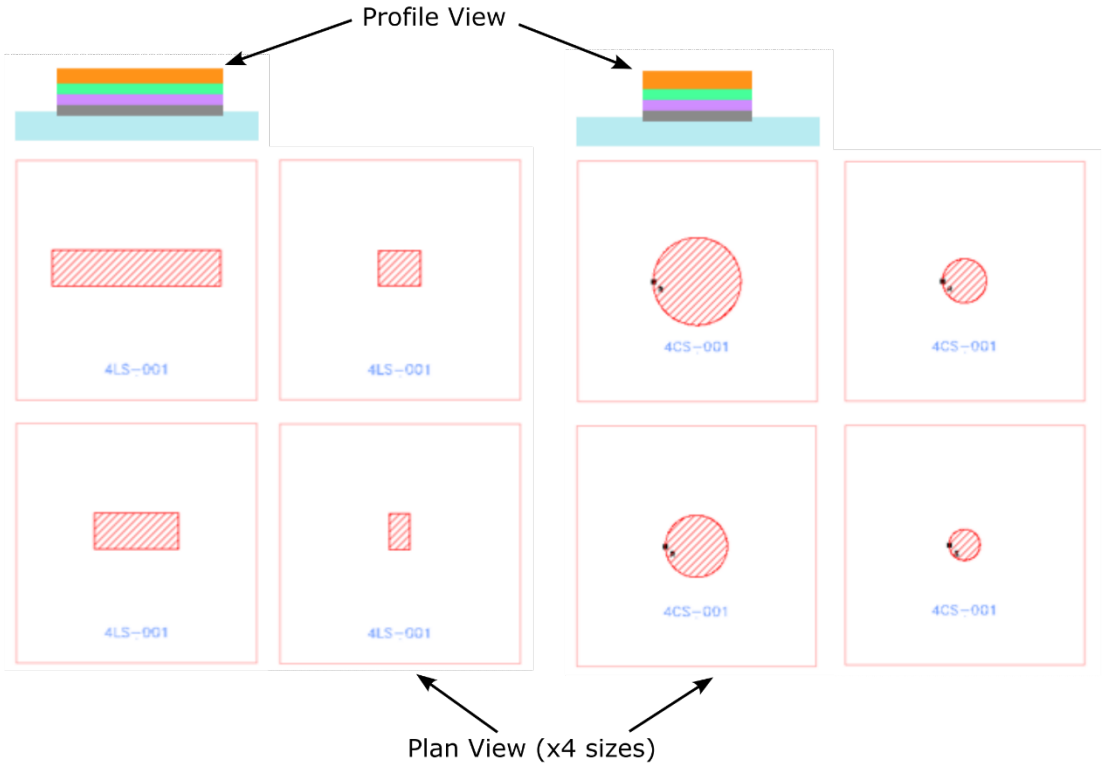

Figure 5. Linear and circular metallization patterns for stacked colinear MFM design.

3.3.3 QR Pattern Design

Simple QR codes will also be deposited on select MFMs, as shown in Figure 6. QR codes could be useful as both an identification mechanism and the dosimeter material in MFM devices. If the relatively complicated pattern of a QR code can be accurately and repeatedly deposited, then MFMs could be individually and uniquely fabricated, each with a slightly different amount of dosimeter material. The quantity of metal could then be recorded with the QR code for reference during gamma counting.

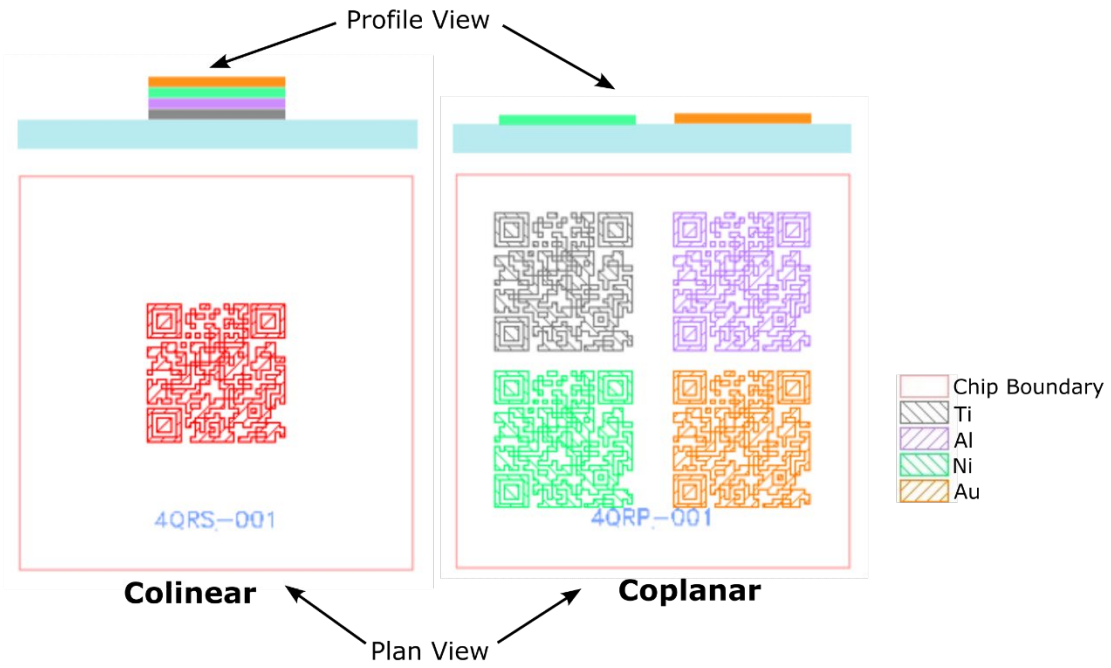

Figure 6. QR pattern in colinear (left) and coplanar (right) layout for prototype MFM devices.

3.3.4 Material Purity Reference Samples

In addition to the previously mentioned metalized patterns, metals will be deposited individually on SiC as reference samples for neutron activation analysis (NAA). This will allow for investigation of impurities in each metal that could cross-contaminate gamma measurements and spectrum adjustment calculations. Uncoated samples of SiC will also be analyzed using NAA to measure impurities that may be present in the substrate itself or in the residual material from photoresist, developer, cleaning solutions, or the wafer dicing tool.

3.4 IRRADIATION TESTING

Prototypes of the MFMs will be tested in the HFIR [2], located at ORNL. HFIR is a light-water–moderated and –cooled research reactor, capable of producing extremely high neutron flux. The reactor operates at 85 MW thermal power, with cycles typically lasting 24 days. MFMs will be inserted into HFIR using the NAA laboratory, located in the reactor building.

The NAA laboratory contains two PT irradiation facilities, which can transport small capsules, called *rabbits*, into and out of the reactor pressure vessel during a HFIR cycle. PT-1 transports rabbits to a vertical experiment facility (VXF) in the permanent Be reflector of the reactor, whereas PT-2 transports rabbits to the slanted experiment facility (EF) on the periphery of the reflector. The PTs can irradiate rabbits for short duration experiments (e.g., several seconds) as well as much longer multi-day irradiation experiments. Irradiations longer than 1 minute will be accomplished in graphite rabbits to ensure that the container does not melt in PT-1. The differences between polyethylene rabbits and graphite are expected to be small and will be examined by measurement through comparison of the production rates in each. Multiple irradiations for several different durations are planned for each of the MFM prototypes to assess feasibility. Following irradiation, the MFMs will be analyzed using one of the several lead-shielded HPGe detection instruments in the NAA laboratory.

4. MODELING

To inform the MFM design, an estimate of the activity in each dosimeter material was required. This was accomplished by first modeling the anticipated neutron spectrum and flux in the PT-1 and PT-2 irradiation positions. Calculated flux values were then used in the ORIGEN depletion code to determine activity levels and the required mass of each material under different irradiation scenarios.

4.1 NEUTRON FLUX MODELING OF PT-1 AND PT-2

Energy- and time-dependent neutron flux were calculated in the PT-1 and PT-2 locations during a typical 26-day HFIR cycle using the ORNL-developed HFIRCON [14] radiation and depletion code (version 1.0.5). The HFIRCON code is a collection of several nuclear modeling programs designed to perform time-dependent, coupled radiation and depletion calculations of experiments in HFIR. The code incorporates the LAVA Model INTerrogator (LAVAMINT) for calculation of cell volumes, the AutomateD VAriaNce reducTion Generator (ADVANTG) code for creating weight windows and source biasing [15], the ORNL-Transformative Neutronics/MCNP5 (ORNL-TN/MCNP5) code for higher efficiency radiation transport modeling [16], the ORIGEN code for isotopic depletion [3], and various C and Python wrappers for data transfer between programs. Use of this code provides neutron flux energy spectra specific to each day of a HFIR cycle, which can change over the course of a HFIR cycle due to movement of the HFIR control plates.

Using a previously developed MCNP model of the HFIR core [17], the PT-1 and PT-2 locations were modeled as a 1 cm diameter cylinder and sphere, respectively, as shown in Figure 7. Neutron flux tallies were calculated with a 238-group structure [18] for follow-on calculations in ORIGEN, as well as a 100-group structure used in STAYSL PNNL. Ten days over the course of a 26-day HFIR cycle were modeled to generate neutron flux spectra specific to each day of the HFIR cycle. Energy-dependent neutron flux spectra were averaged and normalized to 1 to determine the average spectrum for PT-1/PT-2, as shown in Figure 8. The total neutron flux in PT-1/PT-2 for each day modeled is depicted in Figure 9.

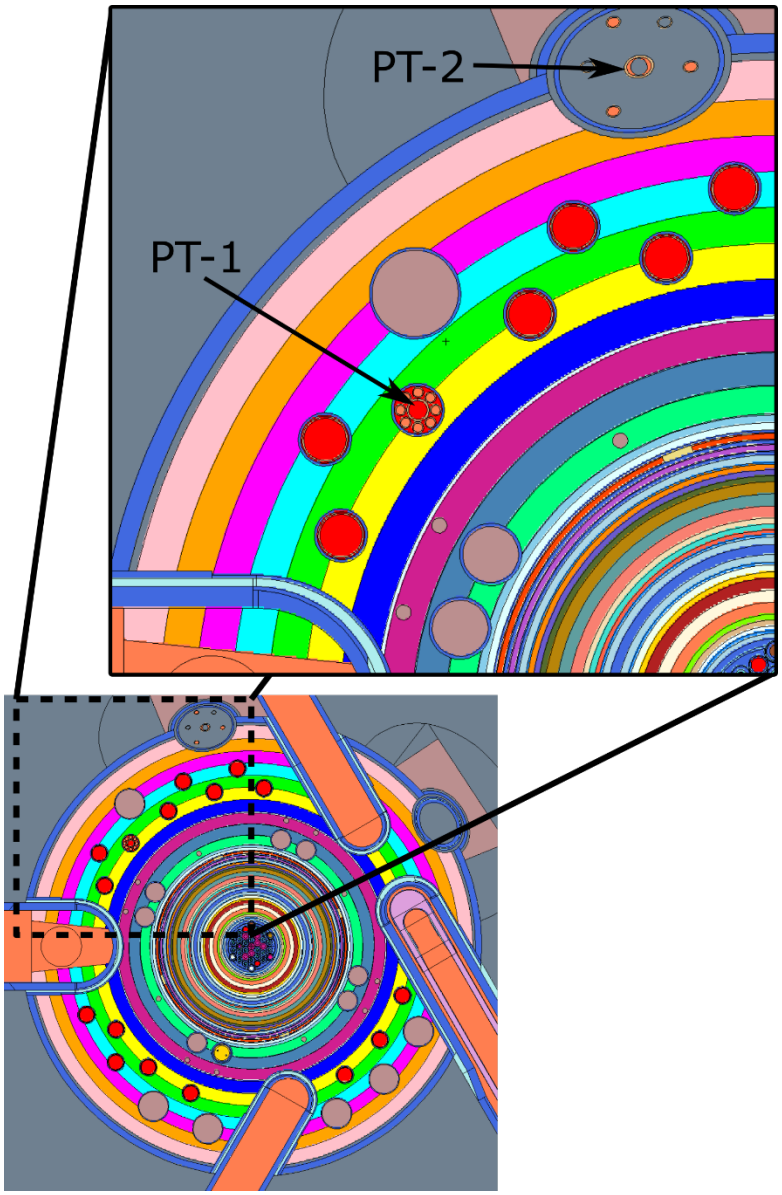

Figure 7. Cross-sectional view of HFIR midplane from MCNP model, showing locations of PT-1 and PT-2.

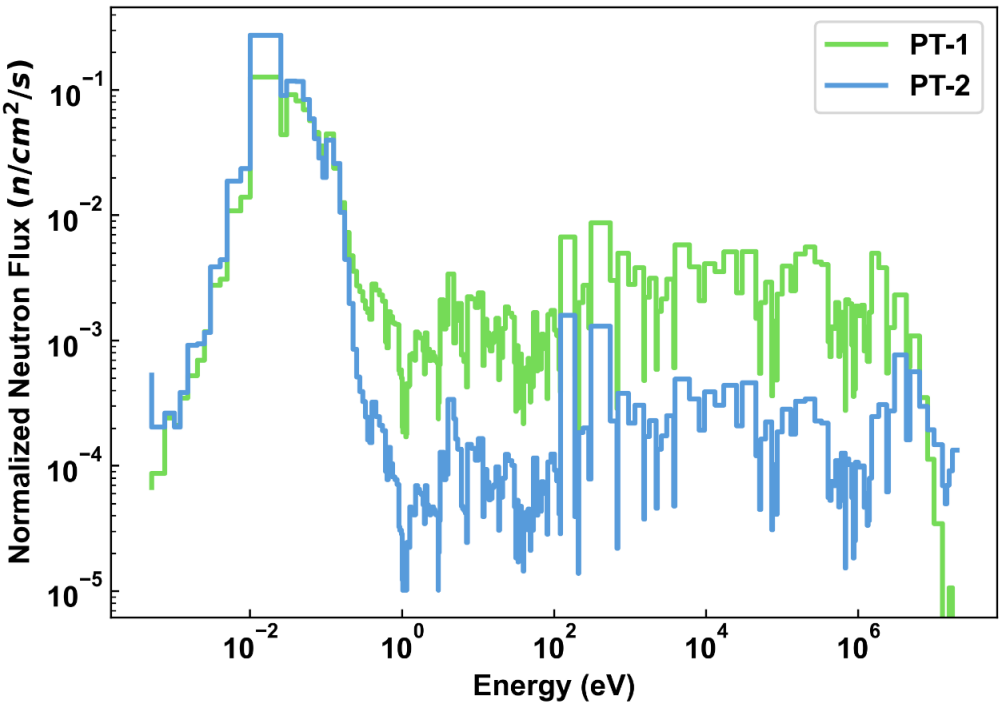

Figure 8. Normalized 238-group neutron flux spectra for PT-1 and PT-2.

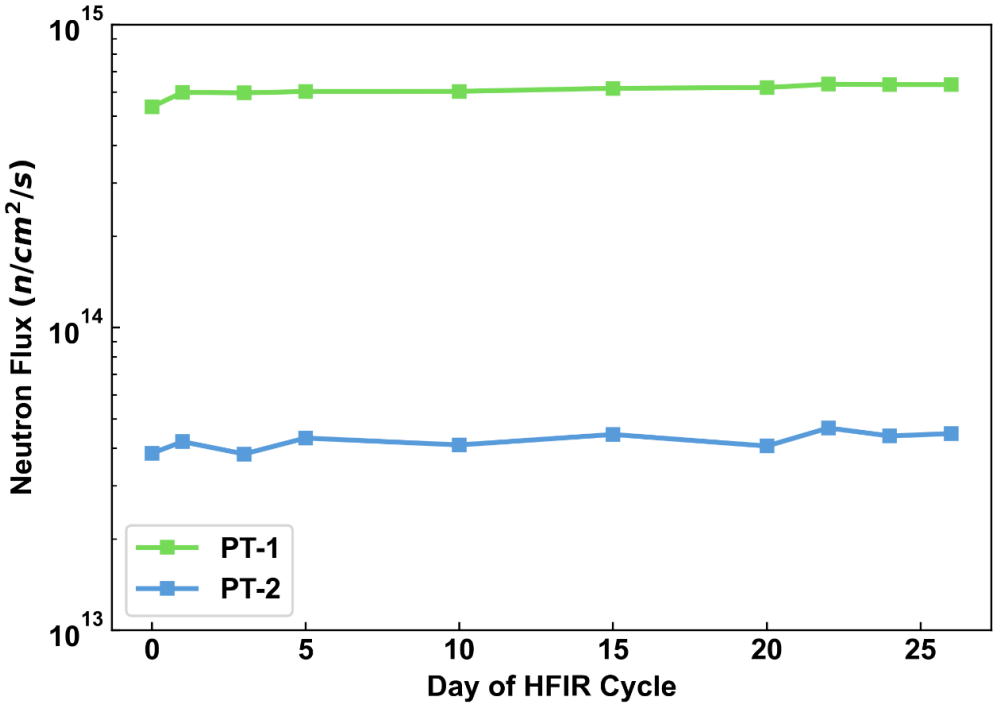

Figure 9. Total neutron flux in PT-1 and PT-2 for 10 days of a 26-day HFIR cycle.

4.2 ACTIVATION USING ORIGEN

Transmutation of neutron dosimeter materials was modeled using ORIGEN [19] (version SCALE 6.2.3) to determine the activity of product isotopes under different irradiation scenarios. Neutron flux spectra and total neutron flux, calculated using HFIRCON, were input into ORGIEN to irradiate 1 mg quantities of each down-selected material listed in Table 2. Each material was irradiated for 4 different lengths of time—1 hour, 1 day, 1 month, and 1 year—and isotopic activities were calculated after decay intervals of 10 minutes, 30 minutes, 1 hour, 1 day, and 1 week.

The short irradiation time intervals of 1 hour and 1 day are representative of planned experiments using the PT facilities in the NAA laboratory, whereas the longer time intervals of 1 month and 1 year are characteristic of end use applications of MFM devices. Even longer irradiation intervals (multiple years) could be modeled to replicate the operation of some planned A/SMRs designs, however, these time scales were not included in the current work. The decay time intervals represent the length of time anticipated between irradiating an MFM and performing gamma spectrum measurements. The short time intervals (< 1 day) are typical lengths of time between irradiation and counting in the NAA laboratory, whereas the 1 day and 1 week time intervals represent the amount of time necessary in an industrial facility to physically retrieve MFMs from an in-core location during an outage. The activity of all 2,239 isotopes tracked in ORIGEN as well as the associated gamma spectra in 1 keV energy bins were calculated for each material, irradiation, and decay combination. Results of these simulations were then stored for additional calculations to aid in the design of MFM devices.

Results from irradiating the dosimeter materials in the PT-1 position of HFIR using ORIGEN were used to determine mass and volume requirements. The activity of the primary product isotope was determined for several time intervals following the irradiation. The residual activity of the primary product isotope was then scaled to 10 nCi, representing the "goal" activity for each material. This scaling factor was then applied to the 1 mg of target material irradiated and divided by the material density, giving the volume of material necessary to achieve 10 nCi in these irradiation and decay scenarios. Determining the volume required is essential to specifying the fabrication recipe (metal contact area and thickness) for the MFM devices. Higher residual activities can be achieved by scaling these results using a desired scaling factor and the density of the target material. The following sections describe the diameter of a metal contact required to achieve 10 nCi in each material, assuming a 200 nm contact thickness. Complementary calculations that assume a metal contact diameter and determine a metal thickness were also performed but are not included.

4.2.1 Au

Au, using the $^{197}Au(n,g)^{198}Au$ reaction, is an excellent material for use in MFM devices. The relatively large thermal neutron capture cross section (98.7 b) and moderate half-life (2.696 d) means only a small quantity of target material is needed to produce sufficient quantities of the product isotope. As shown in Table 4, a 200 nm thick Au deposition requires a circular contact less than 20 μm in diameter to achieve 10 nCi of ^{198}Au. These dimensions are easily achievable using UV photolithography and e-beam metal deposition. General trends in the required quantity of Au indicate that more material is necessary if the time period between irradiation and gamma counting is long; very long irradiation periods also result in increased Au consumption due to burnup.

Table 4. Au contact diameter (µm) required to achieve 10 nCi in ^{198}Au (200 nm thickness).

		Time Until Counting				
		10 min	**30 min**	**1 hour**	**1 day**	**1 week**
Irradiation Time	**1 hour**	8.0	8.0	8.0	9.1	19.7
	1 day	2.2	2.2	2.2	2.5	5.5
	1 month	2.1	2.1	2.1	2.4	5.2
	1 year	5.0	5.0	5.0	5.8	12.2

4.2.2 Ni

Ni has 3 relevant threshold reactions for neutron dosimetry measurements: ^{58}Ni(n,2n)^{57}Ni, ^{58}Ni(n,p)^{58}Co, and ^{60}Ni(n,p)^{60}Co. Due to the small cross sections of these reactions (^{58}Ni = 4.6 b, ^{60}Ni = 2.9 b), large amounts of target material are required to produce detectable amounts of the product isotopes. As shown in Table 5 and Table 6, contacts as large as 10 and 437 mm in diameter would be required to produce 10 nCi of ^{58}Co and ^{60}Co, respectively, in a 1 hour irradiation, which is not practical for this application. However, for much longer irradiations, the required contact size drops to ~0.5 mm in diameter. Production of ^{57}Ni also requires large deposition contact areas; however, this isotope has a relatively short half-life ($T_{1/2}$ = 35.6 h), which negates any benefits from long irradiations, since the product isotope decays away. Therefore, Ni would be a suitable MFM dosimeter material for long duration irradiations but will be challenging to use in short irradiation periods in the NAA laboratory.

Table 5. Ni contact diameter (µm) required to achieve 10 nCi in ^{58}Co (200 nm thickness).

		Time Until Counting				
		10 min	**30 min**	**1 hour**	**1 day**	**1 week**
Irradiation Time	**1 hour**	10,659	10,539	10,371	8,046	7,952
	1 day	2,032	2,029	2,025	1,938	1,976
	1 month	592	592	592	592	609
	1 year	582	582	582	582	599

Table 6. Ni contact diameter (µm) required to achieve 10 nCi in ^{60}Co (200 nm thickness).

		Time Until Counting				
		10 min	**30 min**	**1 hour**	**1 day**	**1 week**
Irradiation Time	**1 hour**	437,741	423,551	419,393	418,823	419,281
	1 day	84,717	8,4603	84,568	84,577	84,668
	1 month	8,248	8,248	8,248	8,247	8,256
	1 year	327	327	327	327	327

4.2.3 Cr

Cr activates via the ^{52}Cr(n,2n)^{51}Cr threshold reaction. Due to the moderately long half-life ($T_{1/2}$ = 27.7 d) of this dosimeter material, the residual radioactivity remains relatively unchanged for short (minutes) or extended (days) periods between irradiation and gamma counting. This fact, combined with the small cross section of ^{51}Cr, means smaller contact diameters are required for long irradiations, as shown in

Table 7. However, the majority of ^{51}Cr comes from thermal neutron capture in ^{50}Cr (4.3% natural abundance), which makes Cr questionable as a threshold monitor unless shielded with Cd or Gd covers.

Table 7. Cr contact diameter (µm) required to achieve 10 nCi in ^{51}Cr (200 nm thickness).

		Time Until Counting				
		10 min	30 min	1 hour	1 day	1 week
Irradiation Time	1 hour	309.6	309.7	309.7	313.5	337.9
	1 day	63.6	63.6	63.6	64.4	69.4
	1 month	13.9	13.9	13.9	14.0	15.1
	1 year	11.0	11.0	11.0	11.2	12.0

4.2.4 Ti

Ti undergoes several threshold reactions when irradiated and has a similar efficacy to Ni as a MFM dosimeter material. Only the $^{46}Ti(n,p)^{46}Sc$ and $^{48}Ti(n,p)^{47}Sc$ reactions produce residual activities in reasonable quantities, as shown in Table 8 and Table 9. Moderately sized (< 2 mm) contacts can be used for long duration irradiations. However, the low cross section of Ti isotopes makes this material impractical for short irradiation MFMs.

Table 8. Ti contact diameter (µm) required to achieve 10 nCi in ^{46}Sc (200 nm thickness).

		Time Until Counting				
		10 min	30 min	1 hour	1 day	1 week
Irradiation Time	1 hour	89,334	89,339	89,347	89,701	91,958
	1 day	18,273	18,273	18,275	18,348	18,808
	1 month	3,546	3,546	3,546	3,561	3,651
	1 year	1,717	1,717	1,717	1,725	1,767

Table 9. Ti contact diameter (µm) required to achieve 10 nCi in ^{47}Sc (200 nm thickness).

		Time Until Counting				
		10 min	30 min	1 hour	1 day	1 week
Irradiation Time	1 hour	14,218	14,238	14,269	15,756	29,327
	1 day	3,047	3,051	3,058	3,377	6,279
	1 month	1,316	1,316	1,316	1,465	2,731
	1 year	1,312	1,312	1,312	1,479	2,691

4.2.5 Al

Al is a useful MFM dosimeter material for applications in which gamma spectrometry can be performed very quickly following irradiation. The short half-lives of ^{27}Mg ($T_{1/2}$ = 9.46 min) and ^{24}Na ($T_{1/2}$ = 14.6 h) require analysis within approximately 1 h for sufficient counts. This material will be used for MFM testing in the HFIR NAA facility but could be challenging to use in other facilities with long retrieval times.

Table 10. Al contact diameter (μm) required to achieve 10 nCi in ^{27}Mg (200 nm thickness).

		Time Until Counting				
		10 min	30 min	1 hour	1 day	1 week
Irradiation Time	1 hour	1,069	2,225	6,681	–	–
	1 day	1,063	2,212	6,639	–	–
	1 month	737	737	737	–	–
	1 year	738	738	738	–	–

Table 11. Al contact diameter (μm) required to achieve 10 nCi in ^{24}Na (200 nm thickness).

		Time Until Counting				
		10 min	30 min	1 hour	1 day	1 week
Irradiation Time	1 hour	8,033	8,094	8,188	13,933	389,892
	1 day	2,085	2,101	2,126	3,622	100,699
	1 month	1,701	1,701	1,701	3,031	85,353
	1 year	1,703	1,703	1,703	3,235	80,142

4.2.6 Additional Dosimetry Materials

Although Au, Ni, Cr, Ti, and Al will be used for prototype MFM devices, several other materials would be very suitable in future iterations. Sc, Mn, Co, Cu, Ag, Ta, and W all produce sufficient residual activities with very small initial quantities. These reactions also have significant cross sections in the intermediate and thermal energies, which could yield higher-fidelity neutron spectrum measurements compared to those of the prototype materials described above—most of which are high-energy-threshold reactions. However, depositing these materials requires additional fine-tuning of deposition parameters but could be proposed in future work.

5. CONCLUSIONS

Neutron dosimeters are a well-established analytical tool for reactor surveillance and can provide information regarding flux, fluence, and the neutron spectrum in nuclear reactors. Measuring these characteristics can be critical for safeguards monitoring to indicate fuel reloading, higher power operation, or extended reactor cycle lengths. This report describes a novel application of neutron dosimetry measurement devices that will use semiconductor fabrication techniques to deposit small, precise quantities of dosimeter materials onto a substrate. These MFM devices have been modeled using ORIGEN to determine the mass and physical dimensions required for sufficient radioactivity. Prototypes of these devices will be fabricated in several patterns and irradiated in HFIR later this year.

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