Final Report DE-SC0020140

DOE Office of Nuclear Physics (NP)

Proposal Name:
Novel Methods for Th-229 Production through Fast Neutron Irradiation of Th-230 and Charged Particle Irradiation of Th-230 and Th-232.

Project Dates August 1, 2019 – July 31, 2022

Principal Investigator: Lawrence Heilbronn

Project Overview:

The overall goal of the research was to investigate accelerator-based methods for producing ^{229}Th, which in turn is used as a generator for ^{225}Ac, a promising alpha emitter used for the treatment of cancer via targeted alpha therapy (TAT). The research was conducted in collaboration with groups at Lawrence Berkeley National Laboratory, led by Dr. Lee Bernstein, and Oak Ridge National Laboratory, led by Dr. Justin Griswold. Irradiations of ^{232}Th and ^{230}Th targets with proton, deuteron and neutron beams were conducted at LBNL. The irradiated targets were then shipped to ORNL for radiochemical separation and analysis. University of Tennessee graduate student Naser Burahmah conducted the separations and analysis at ORNL, and also provided transport model calculations of the yields and cross sections of ^{229}Th and other actinides to compare with the experimental data on those isotopes. The comparisons between the experimental data and TALYS, PHITS, and MCNP code calculations provide insight into the codes' predictive capabilities and their usefulness in developing further novel methods for producing isotopes of interest to the nation.
The research was initially scheduled according to Table 1 below, with indicated milestones based on a quarterly (3-month) schedule.

Due to COVID pandemic restrictions starting in March of 2020, irradiation schedules at LBNL were altered to accommodate staffing availability. This also meant that travel by UTK and ORNL personnel to LBNL for the irradiations was no longer possible, but the LBNL team did an excellent job, and all proposed irradiations were performed.

Table 1. General timetable of subtasks required to complete each specific aim.

Specific Aim #	Sub-Task #	**Description**	**FY19**				**FY20**			
			Q1	Q2	Q3	Q4	Q1	Q2	Q3	Q4
1	1.1	Prepare ^{230}Th target for neutron irradiation	x	x						
	1.2	Develop ^{230}Th target for charged particle irradiation	x	x	x	x				
	1.3	Prepare ^{232}Th target for charged particle irradiation				x				
2	2.1	Perform neutron irradiation of ^{230}Th @ LBNL			x					
	2.2	Chemical processing and data analysis @ ORNL				x	x			
	2.3	Interpretation via comparison w/reaction models			x	x				
	2.4	Prepare manuscript/submit for publication						x		
3	3.1	Perform charged particle irradiations of ^{230}Th and ^{232}Th @ LBNL					x			
	3.2	Chemical processing and data analysis @ ORNL						x	x	
	3.3	Interpretation via comparison w/reaction models					x	x		
	3.4	Prepare manuscript/submit for publication								x

Project Highlights:

Neutron irradiation of a ^{230}Th target was performed at LBNL, and radiochemical separations of the target were conducted at ORNL. Gamma spectroscopy analysis of that experiment continues. Deuteron bombardment on a stacked ^{232}Th foil were conducted at LBNL, yielding results for deuteron interactions at 49.6 MeV, 41.4 MeV, 35.2 MeV and 31 MeV. A ^{230}Th target was fabricated for charged particle irradiations, and a deuteron irradiation was performed at LBNL ate the end of this research grant. Table 2 shows the timeline of the milestones achieved during the research. Mr. Burahmah successfully defended his Ph.D. in July of 2021 based on the work he did with the deuteron irradiations. Results on several protactinium isotopes were reported in the thesis and are shown in figures 1-4, along with comparisons to PHITS[1] model calculations run under three different physics models ("NASA", "MWO", "Kurotama"). Other measurements from deuteron bombardment on ^{232}Th targets are shown, where they exist. Agreement between the measurements conducted under this research grant and previous measurements is good. Model calculations are shown for both direct cross sections (dashed lines), and cumulative cross sections that take into account feeding into the isotope from decay of other isotopes (solid lines). The measurements correspond to the cumulative cross sections as there is no way to determine if the measured isotope came from direct interactions or feeding. The ^{229}Pa cross sections are the first reported cross sections from d + ^{232}Th interactions. The production of ^{229}Pa is important for the production of ^{229}Th as it decays via electron capture directly to ^{229}Th.

Differences between model calculations and experimental data reported in the Physical Review C article show that, for the most part, PHITS[1] and TALYS[2] generally fit the shape of the excitation functions, although they typically overestimate or underestimate the cross sections. Both PHITS and TALYS overestimate the (d,2n) cross sections by a factor of 2-3, but in other cases show better agreement. TALYS overestimates the (d,4n) and (d,5n) reactions, with PHITS showing better agreement there. TALYS underestimates the (d,n) reaction, with PHITS again showing better agreement there.

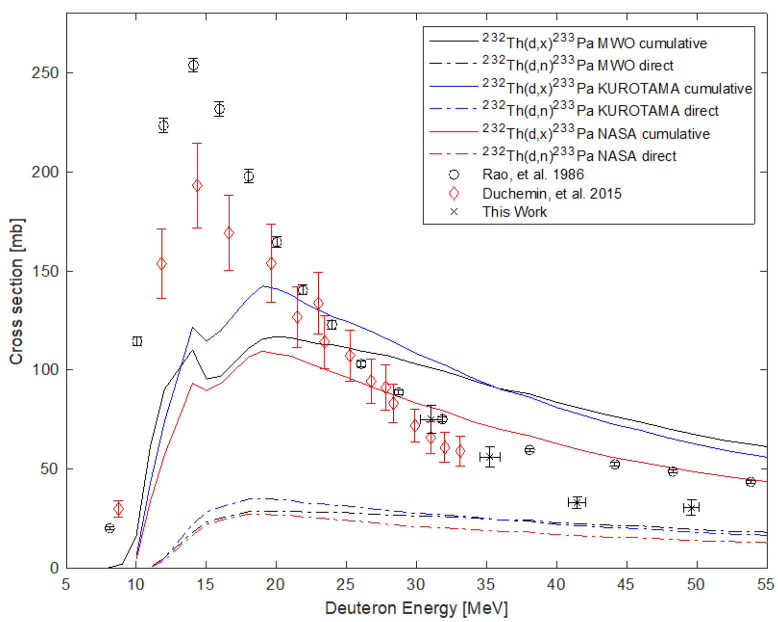

Figure 1. Effective cross sections and cumulative cross sections from PHITS for ^{232}Th(d,x)^{233}Pa reaction. Cross sections from experiments by Rao and Duchemin are shown along with data from this project.

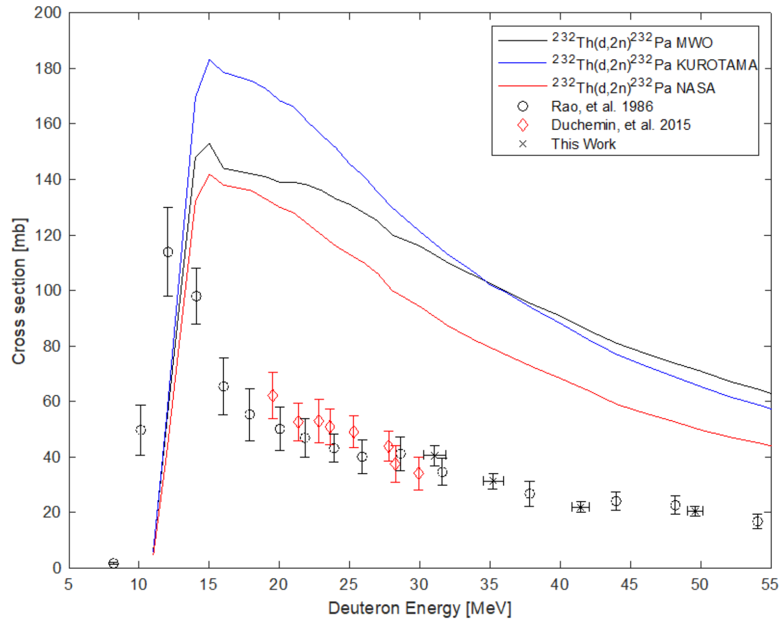

Figure 2. Effective cross sections and cumulative cross sections from PHITS for $^{232}Th(d,x)^{232}Pa$ reaction. Cross sections from experiments by Rao and Duchemin are shown along with data from this project.

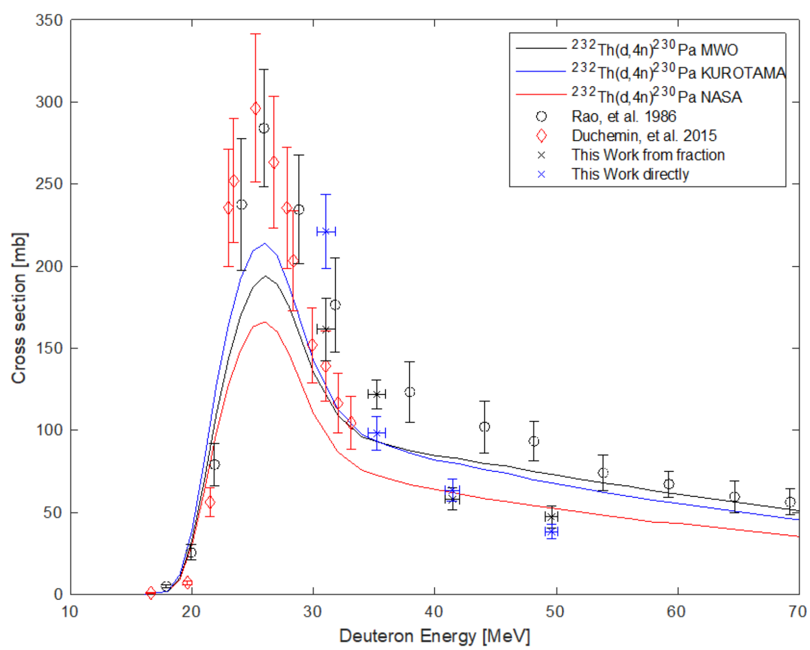

Figure 3. Effective cross sections and cumulative cross sections from PHITS for $^{232}Th(d,x)^{230}Pa$ reaction. Cross sections from experiments by Rao and Duchemin are shown along with data from this project.

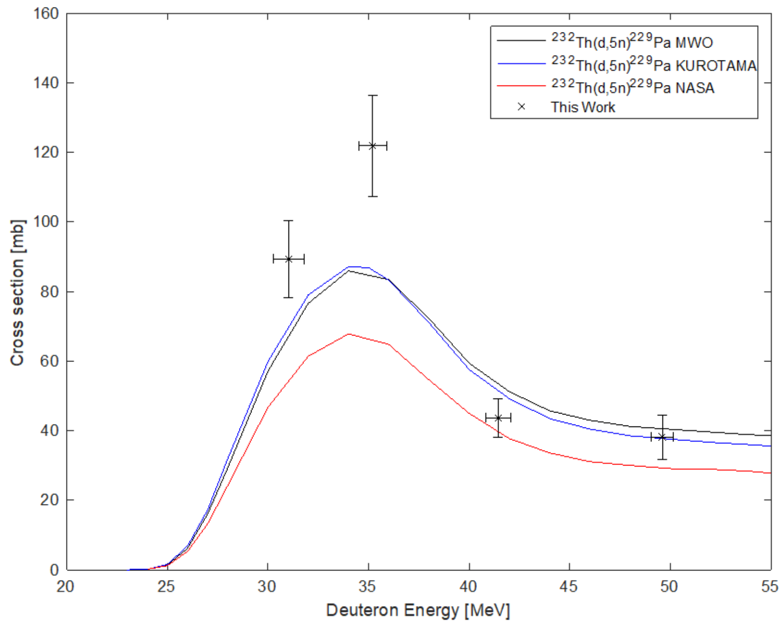

Figure 4. Effective cross sections and cumulative cross sections from PHITS for $^{232}Th(d,x)^{229}Pa$ reaction. These are the first reported cross sections for this reaction.

Table 3 shows the measured effective cross sections for the five Pa isotopes measured in this work. These results were also published in Physical Review C, as well as Mr. Burahmah's work on transport model calculations that were published in Applied Radiation and Isotopes. Details of the publications resulting from this research are shown in Table 4.

Table 2. Actual timeline of milestones after adjusting for COVID restrictions.

Sub-Task #	Milestone	Projected Start Date	Projected Completion Date	Milestone Result	Description of Milestone	Critical Path (Y/N)?
1.1	Prepare ^{230}Th target for neutron irradiation	FY19 Q4	FY20 Q1	Achieved (FY19 Q4)	Characterize Th-230 Stock via gamma spectroscopy and mass spectrometry. Develop target and viable substrate for depositing Th-230 nitrate stock that is suitable for irradiation, shipping, and post irradiation processing.	Y
1.2	Perform neutron irradiation of ^{230}Th @ LBNL	FY20 Q1	FY20 Q2	Achieved (FY20 Q1)	Ship material to LBNL. Participate in any discussion of irradiation parameters that will affect total activity produced (i.e. beam current,	Y

Sub-Task #	Milestone	Projected Start Date	Projected Completion Date	Milestone Result	Description of Milestone	Critical Path (Y/N)?
					irradiation time, incident particle energy etc.) Visit LBNL to observe irradiations and make future target improvements as necessary.	
1.4	Interpretation via comparison w/reaction models	FY20 Q1	FY20 Q2	Ongoing (FY21 Q4)	Perform transport model calculations of isotope yields from Th-230 neutron irradiations	N
2.1	Prepare 232Th target for charged particle irradiation	FY20 Q3	FY20 Q3	Achieved (FY21Q2).	Obtain thin Th-232 foils from commercial vendors. Characterize foil dimensions. Ship to LBNL for irradiation.	Y
2.2	Perform charged particle irradiations of ^{230}Th and ^{232}Th @ LBNL	FY20Q4	FY21Q1	Achieved (FY21Q2)	Proton and deuteron irradiations of thorium targets	Y
2.3	Chemical processing and data analysis @ ORNL	FY21Q1	FY21Q2	Achieved (FY21Q4)	Radiochemical separations and gamma analysis of irradiated targets	Y
2.4	Interpretation via comparison w/reaction models	FY20Q4	FY21Q1	Achieved (FY21Q2)	Transport model calculations of experimental irradiations and subsequent isotope yields	Y
3.1	Develop ^{230}Th target for charged particle irradiation	FY19 Q4	FY21 Q4	Achieved (FY21Q2)	Using Th-230 stock characterized in Task 1.1, deposit thorium as thin film through electrophoretic deposition.	Y

Table 3. Effective measured production cross section (in mb) of selected radionuclides with 31-50 MeV deuteron energies.

Radionuclide	Half-life	31 MeV	35.2 MeV	41.4 MeV	49.6 MeV
^{228}Pa	22 h	0	2.3 ± 0.2	34.9 ± 3.4	31.6 ± 3.5
^{229}Pa	1.55 d	89.3 ± 9.6	121.9 ± 12.7	43.7 ± 5.0	38.2 ± 5.1
^{230}Pa	17.4 d	161.4 ± 16.8	71.9 ± 7.6	58.0 ± 6.1	47.3 ± 5.4
^{232}Pa	1.32 d	40.6 ± 3.3	31.3 ± 2.6	21.9 ± 1.8	20.4 ± 1.7
^{233}Pa	26.98 d	75.0 ± 6.4	56.1 ± 8.4	33.1 ± 2.9	30.5 ± 3.0

Table 4. Naser Burahmah's publications from this research.

Title	Journal	Link
Transport model predictions of ^{225}Ac production cross sections via energetic p, d and α irradiation of ^{232}Th targets	Applied Radiation and Isotopes	Applied Radiation and Isotopes 172 (2021) 109676 https://doi.org/10.1016/j.apradiso.2021.109676
^{229}Pa cross section measurements via deuteron irradiation of ^{232}Th	Physical Review C	PHYSICAL REVIEW C 108, 024609 (2023) DOI: 10.1103/PhysRevC.108.024609
Production of Protactinium-229 via Deuteron Irradiation of Thorium-232	UTK Ph.D. Thesis	https://trace.tennessee.edu/utk_graddiss/7028/

After graduation in July 2021, Dr. Burahmah continued his work at ORNL through a separate contract between UTK and ORNL. This included the analysis of the n + ^{230}Th data taken with Mitch Allmond's Compton-suppressed HPGe detector array, as well as the analysis of the FY21Q2-3 irradiations of a stacked ^{232}Th target (four foils sandwiched between beam energy degraders) with deuterons (incident energy 50 MeV, estimated beam energies on four targets 49.6, 41.3, 35.0 and 30.8 MeV). Additional gamma spec measurements are continuing to measure long-lived isotopes and the buildup of daughter products.

Fabrication of the ^{230}Th target for proton irradiations was completed at the end of this project. Uniformity of the target was an issue throughout the entire fabrication, but in the end a target was fabricated that was sufficiently large and uniform to allow for deuteron irradiation conducted at the end of this research grant.

Further details of the results from this research may be found in the publications listed in Table 4.

Summary:

Success: The measured Pa cross sections from d + ^{232}Th interactions at four deuteron energies are reported, with the ^{229}Pa cross sections measured for the first time.

Success: Gamma-ray spectroscopy measurements continue to determine the production of longer-lived isotopes produced in the deuteron bombardments.

Success: Gamma-ray spectroscopy measurements continue to determine ^{229}Th and other longer-lived isotopes from the n + ^{230}Th bombardments.

Success: Ph.D. degree for the UTK student.

Success: One thesis and two journal publications at the time of this report.

This report was written by Lawrence Heilbronn, UTK PI.