radfarm@ncbj.gov.pl, www.ncbj.gov.pl/radfram

Eksperymentalne stanowisko do produkcji ^{99}Mo wiązką elektronów z akceleratora liniowego

Tobiasz Zawistowski

Promotor: dr hab. inż. Sławomir Wronka (prof. NCBJ)
Koopromor: dr hab. inż. Renata Mikołajczak (prof. NCBJ)
Promotor pomocniczy: dr Izabela Cieszykowska (NCBJ)

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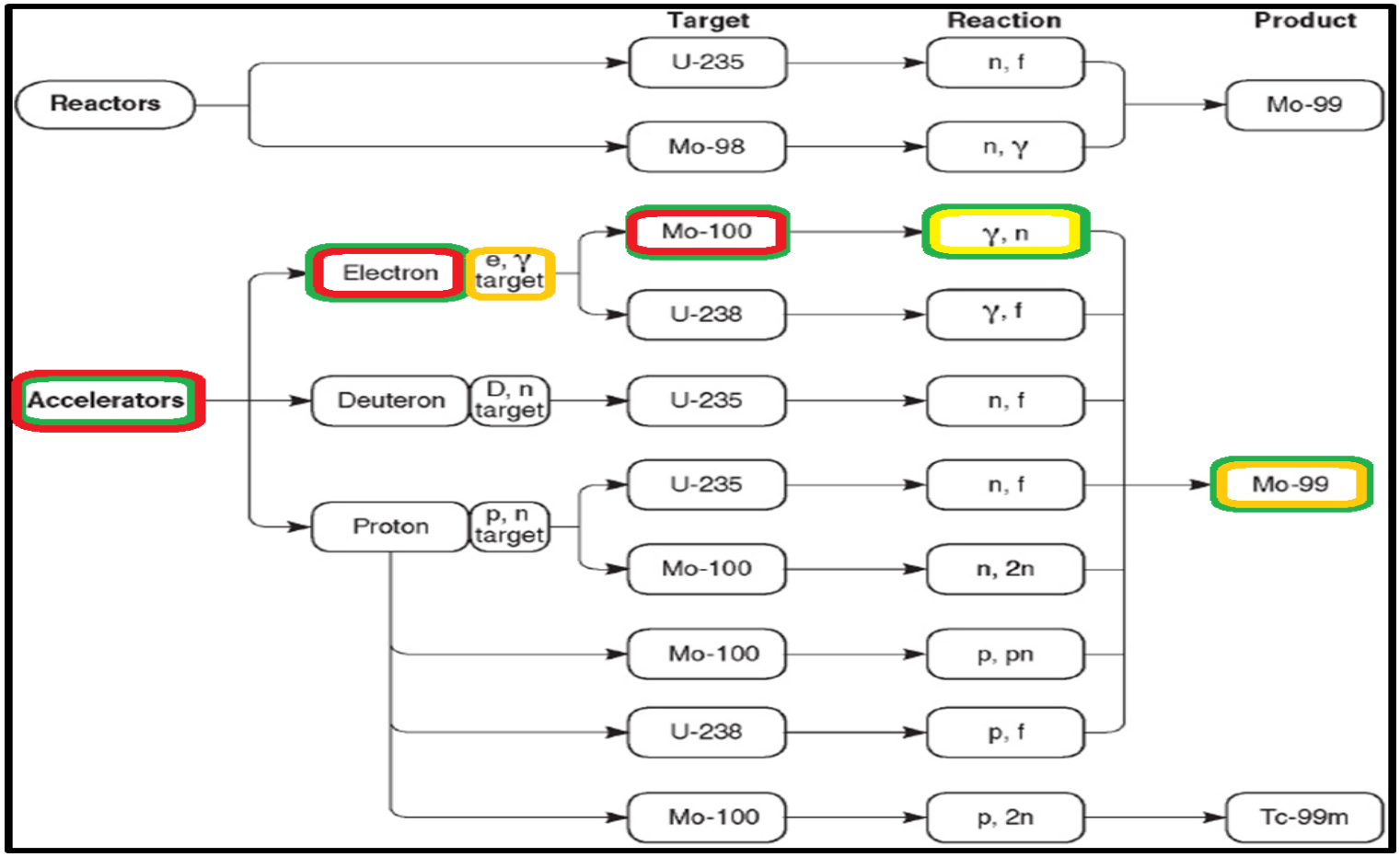

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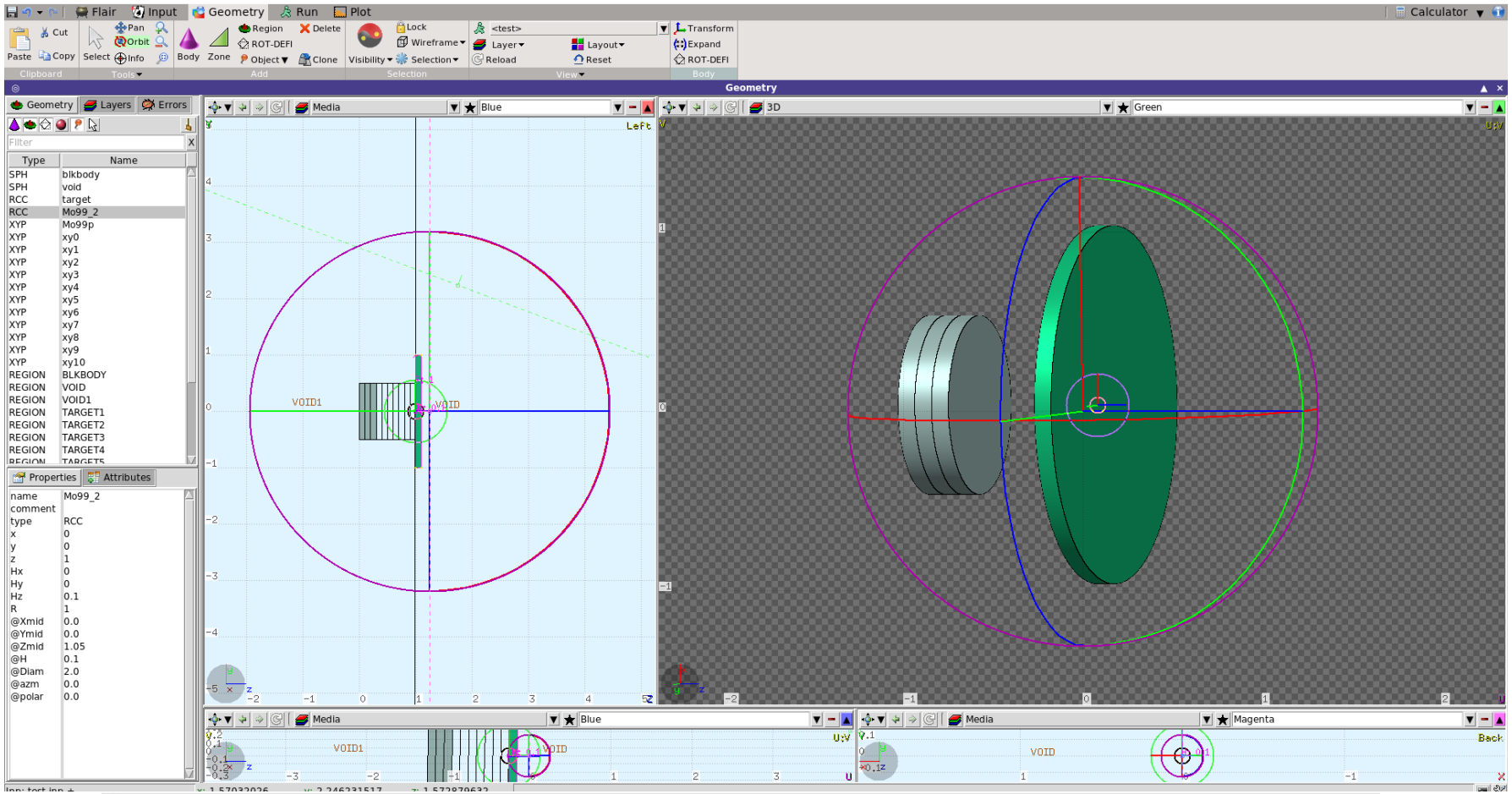

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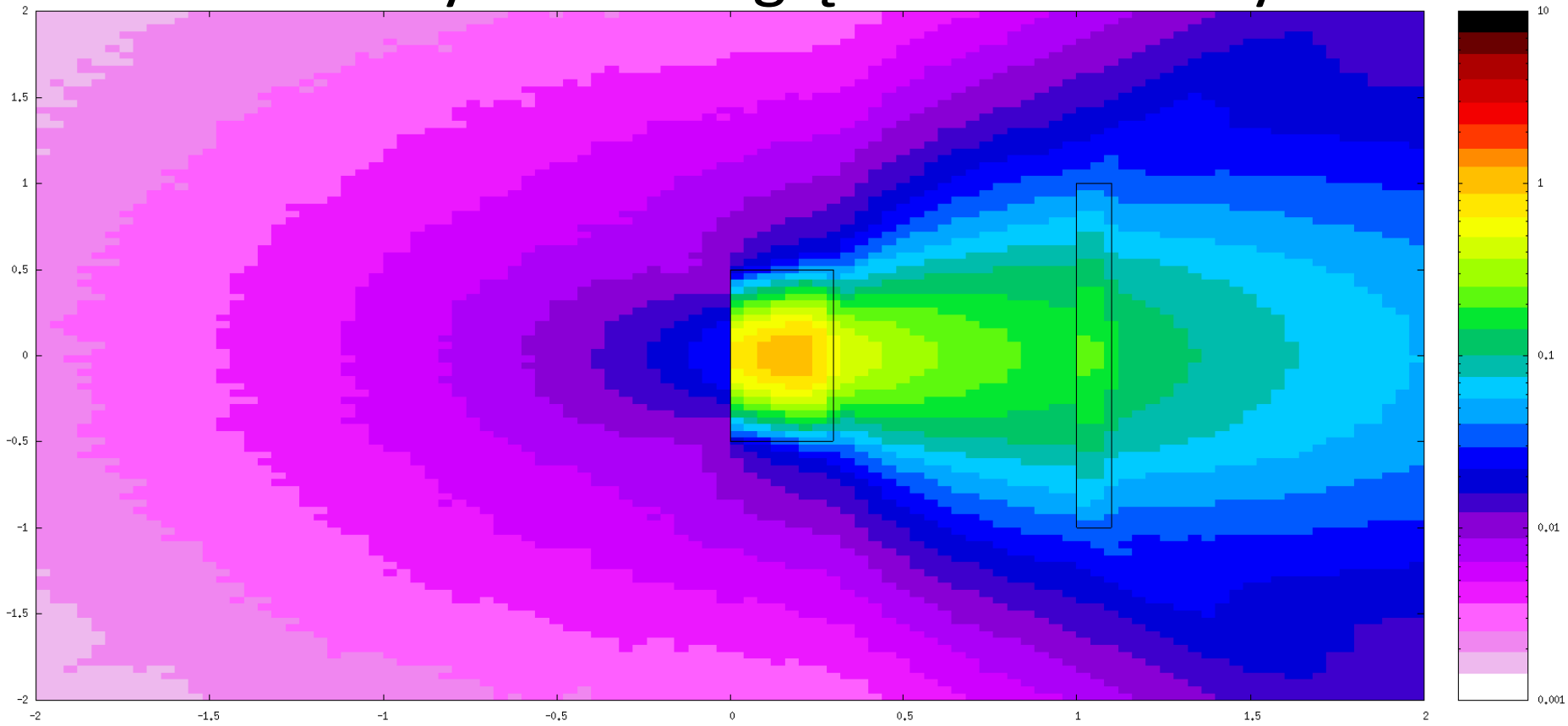
Intensywność względna - Elektrony



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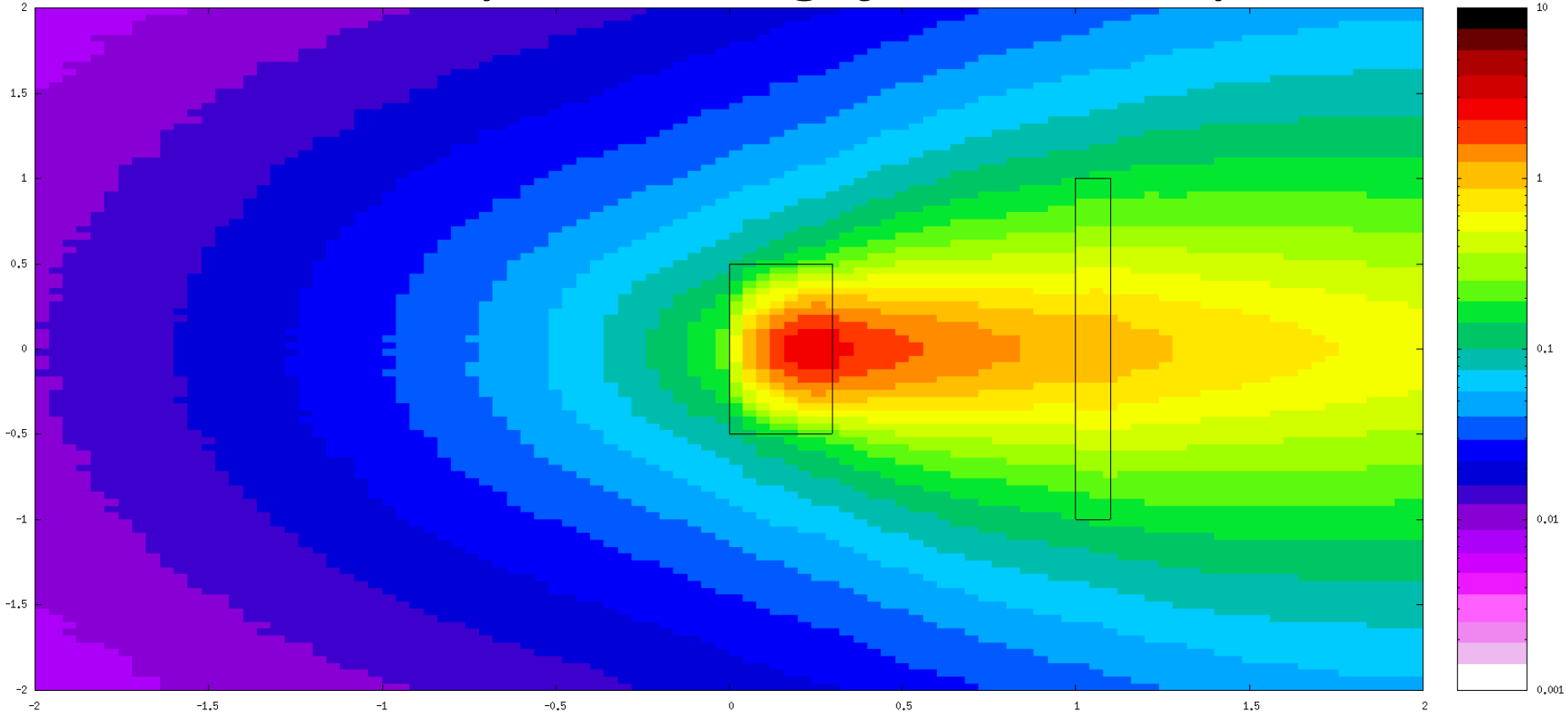
Intensywność względna - Fotony



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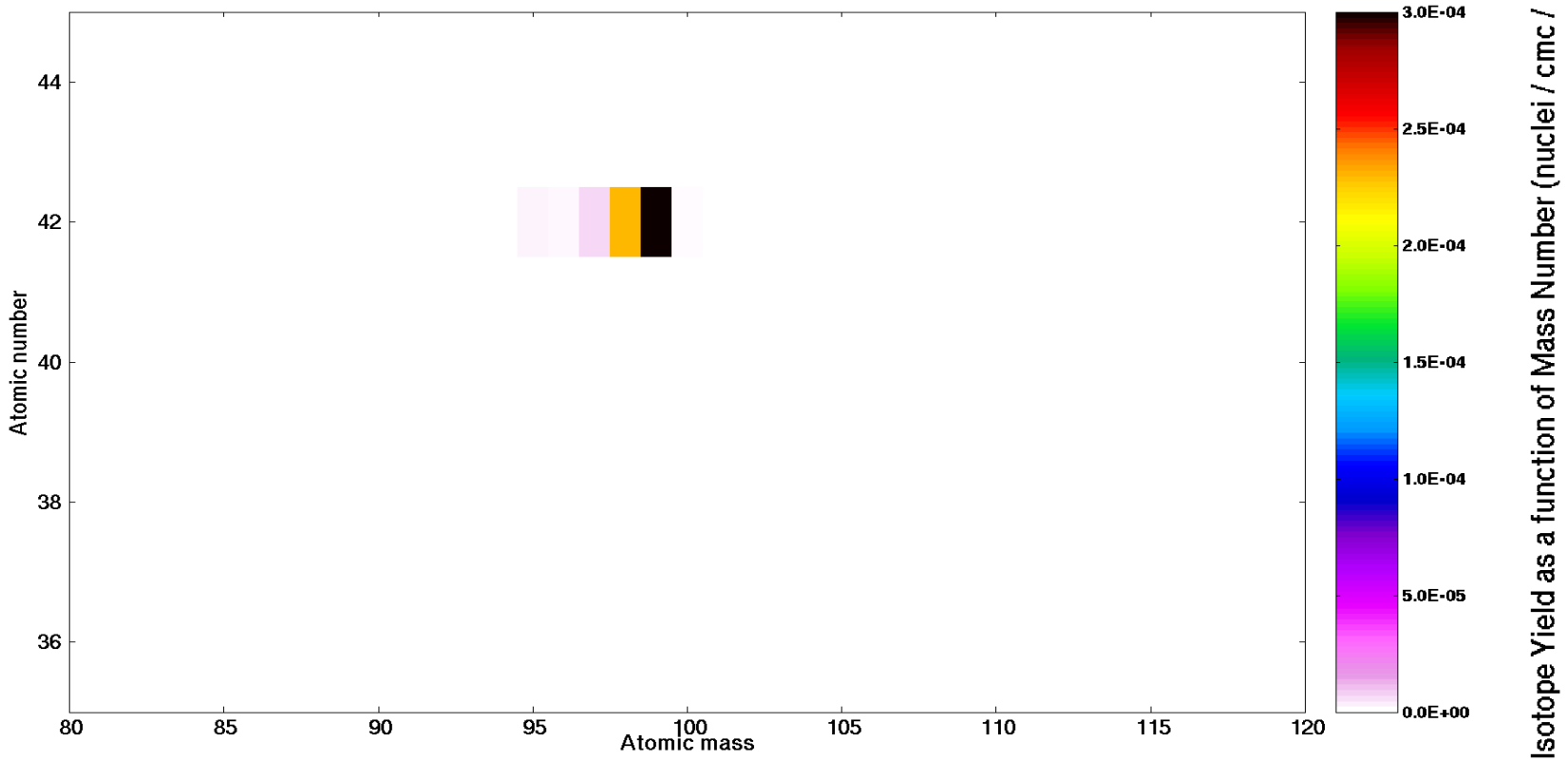
Izotopy



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Tarcza konwersji z odzysku:

- XRF sygnały od cynku
- Listek kolimatora?
- Wolfram ~ 95 %

Isotope	Abundance	Half-life ($t_{1/2}$)	Decay mode	Product
^{64}Zn	49.2%	stable		
^{65}Zn	syn	244 d	ε	^{65}Cu
			γ	–
^{66}Zn	27.7%	stable		
^{67}Zn	4.0%	stable		
^{68}Zn	18.5%	stable		
^{69}Zn	syn	56 min	β^-	^{69}Ga
^{69m}Zn	syn	13.8 h	β^-	^{69}Ga
^{70}Zn	0.6%	stable		
^{71}Zn	syn	2.4 min	β^-	^{71}Ga
^{71m}Zn	syn	4 d	β^-	^{71}Ga
^{72}Zn	syn	46.5 h	β^-	^{72}Ga

EIA	Z	Iso	J^π	Maxw.(n,g) [b]	(n,g) [b]	If [b]	Iab [b]	Macs30 [mb]
Zn	64	30	0+	0.731 ± 0.01				5.90e+1 ± 5.00e+0

EIA	Z	Iso	J^π	Maxw.(n,g) [b]	(n,g) [b]	If [b]	Iab [b]	Macs30 [mb]
U	235	92	7/2-		98.8 ±		419 ± 8	

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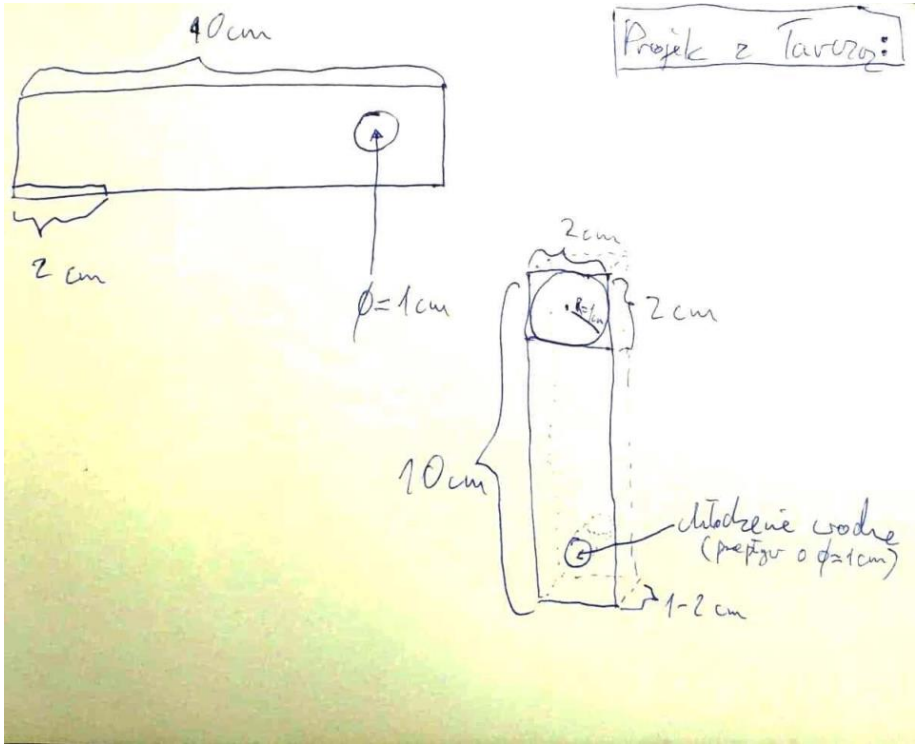
Pierwszy szkic „holderu” tarczy Mo^{100}



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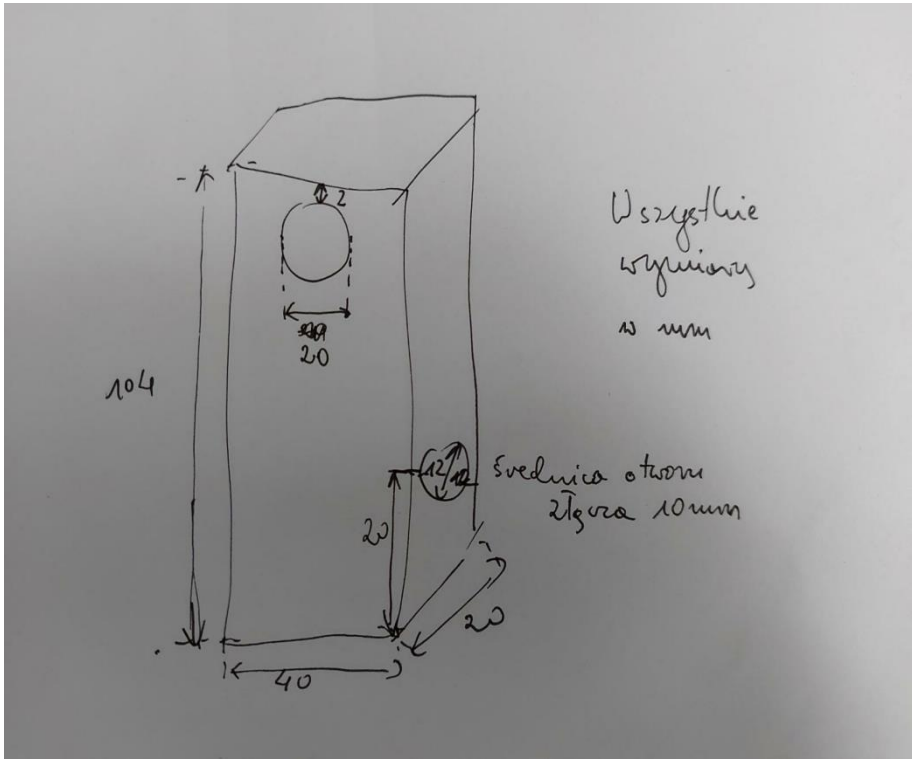
Drugi szkic holderu…



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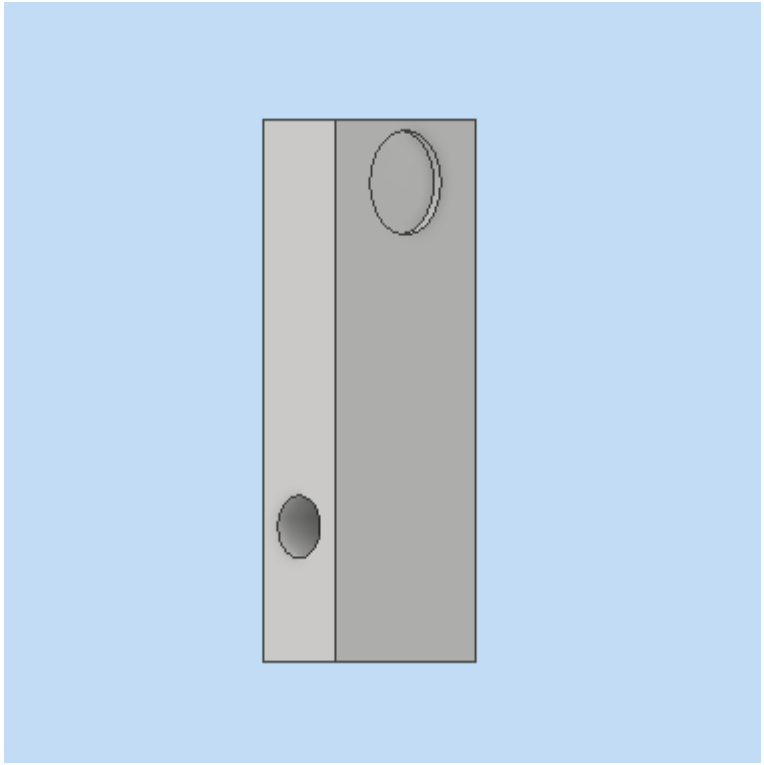
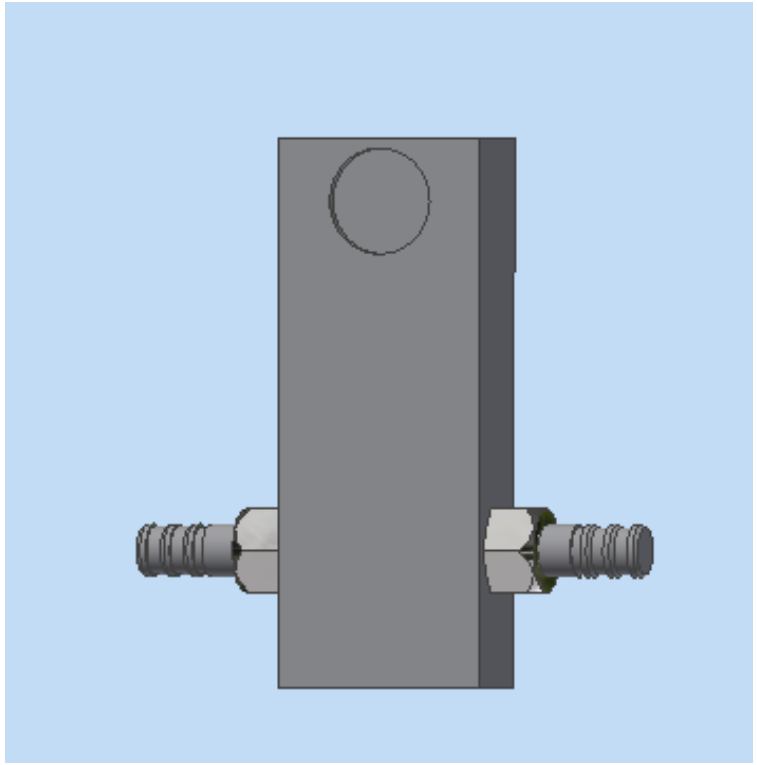
Oczekiwania…





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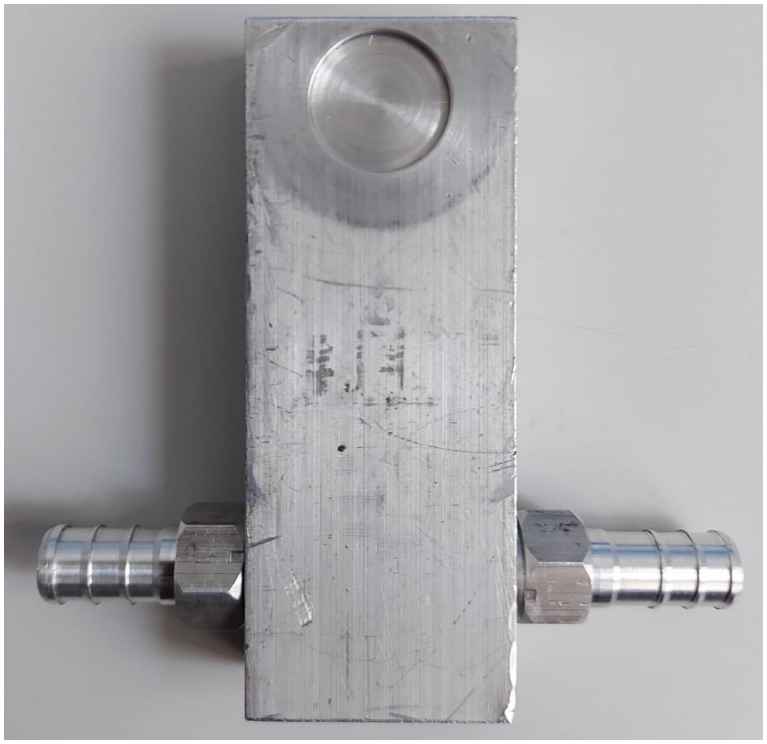
Rzeczywistość =]



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Mocowanie tarczy molibdenowej c.d.

Przewodność cieplna	W/m/K
Mo	130
Cu	380
Al	200

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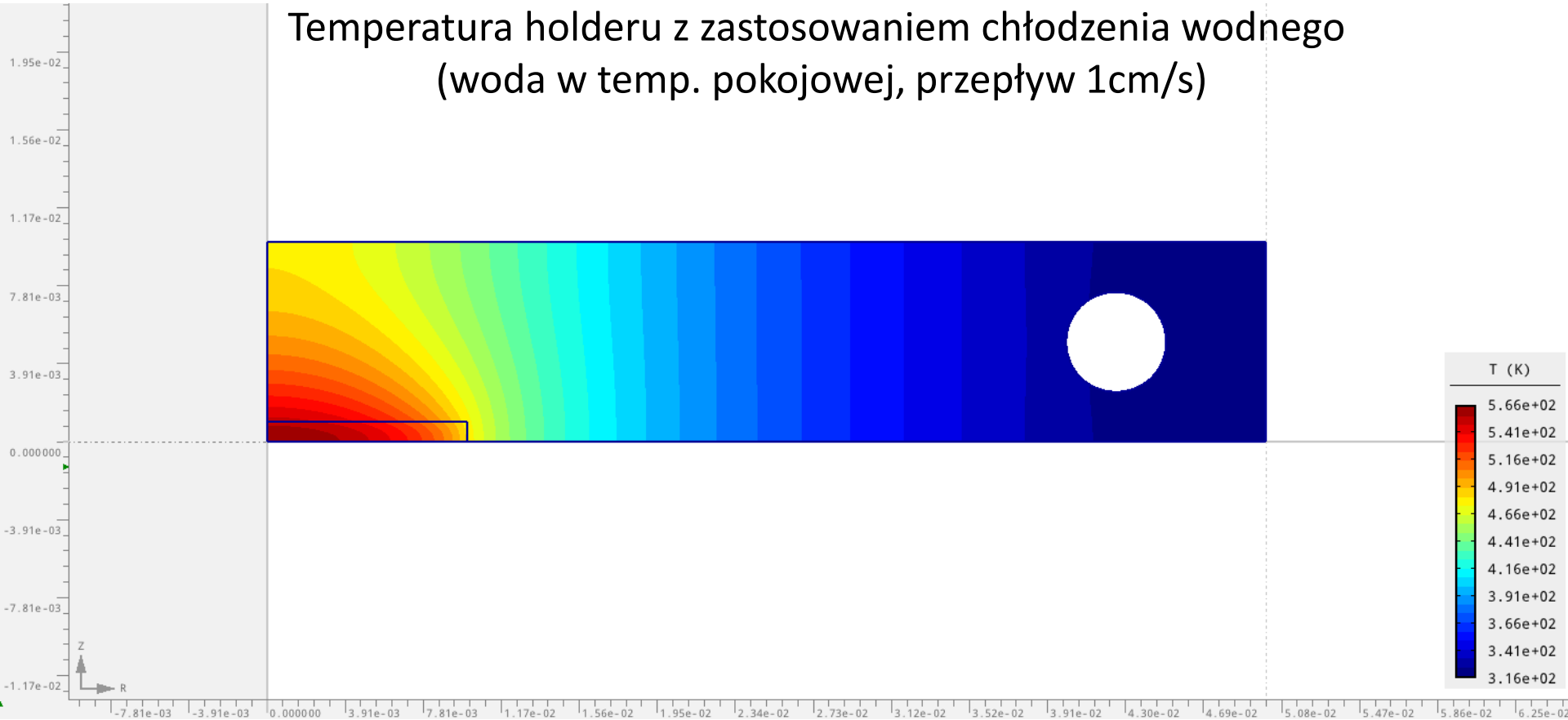
Temperatura holderu z zastosowaniem chłodzenia wodnego (woda w temp. pokojowej, przepływ 1cm/s)



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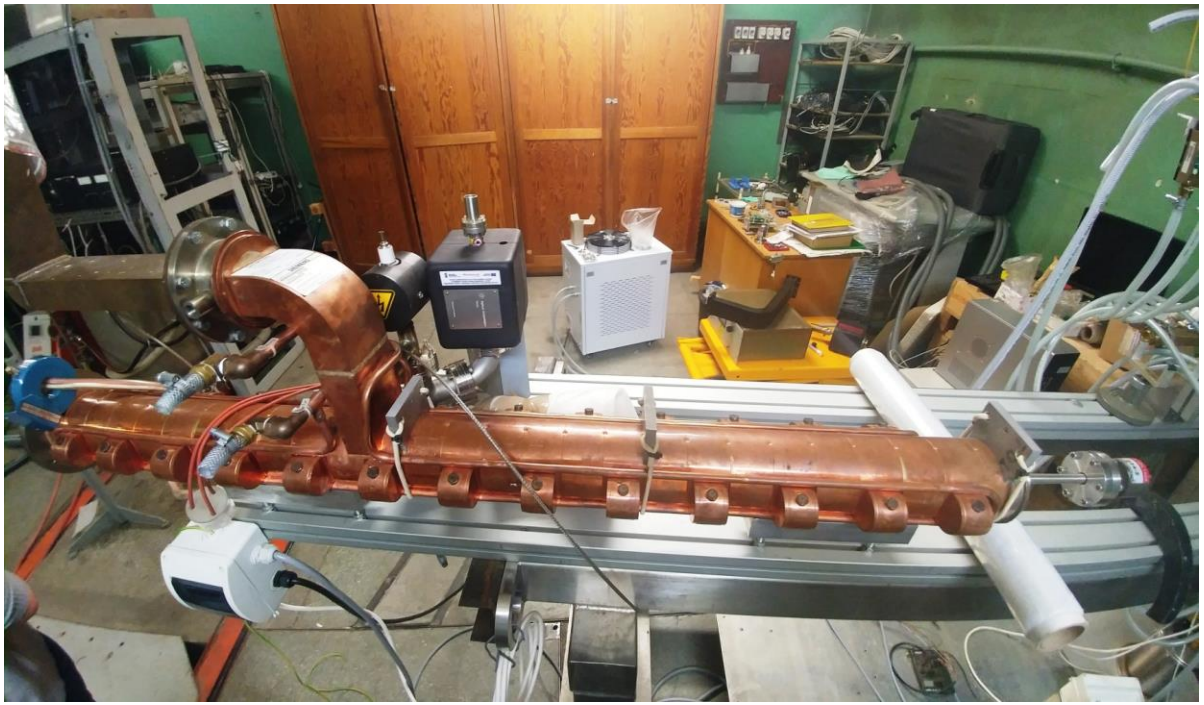
Powrót do „pierwotnego” konceptu

- 120 mA
- Sekcja:
- 20-25 MeV



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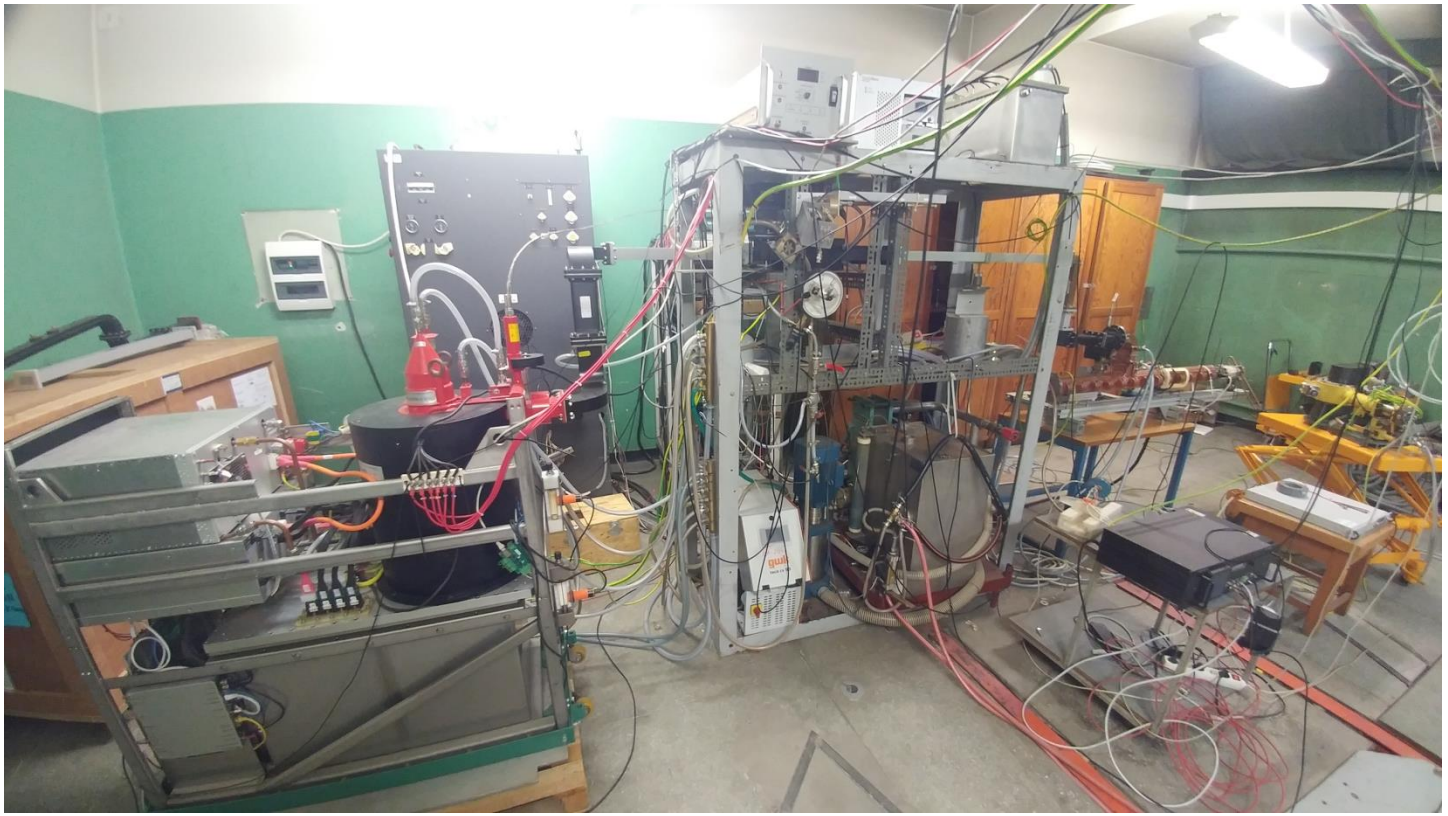

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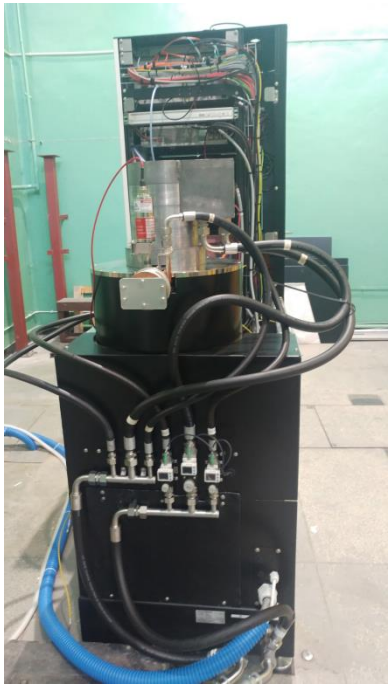
Nowy klistron 7 MW na hali (zamiast 5 MW)



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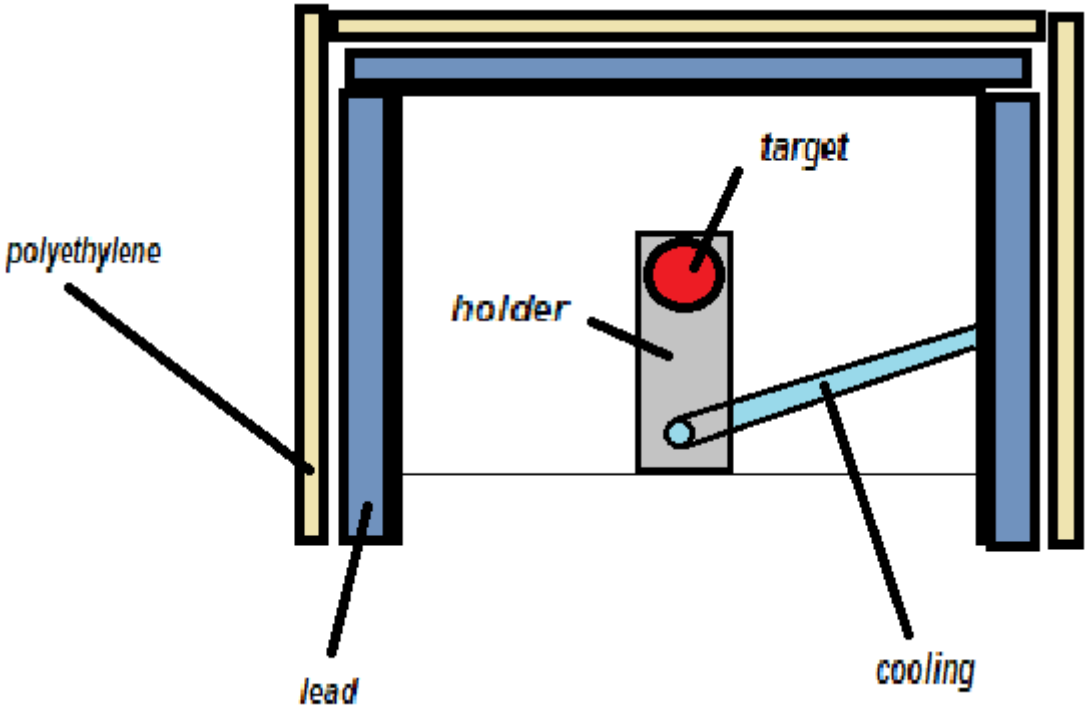
Osłony

- Ołów
  („jaskółki” ołowiane)
- Polietylen
  (płyty polietylenowe)



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Zasilacze cewek skupiających

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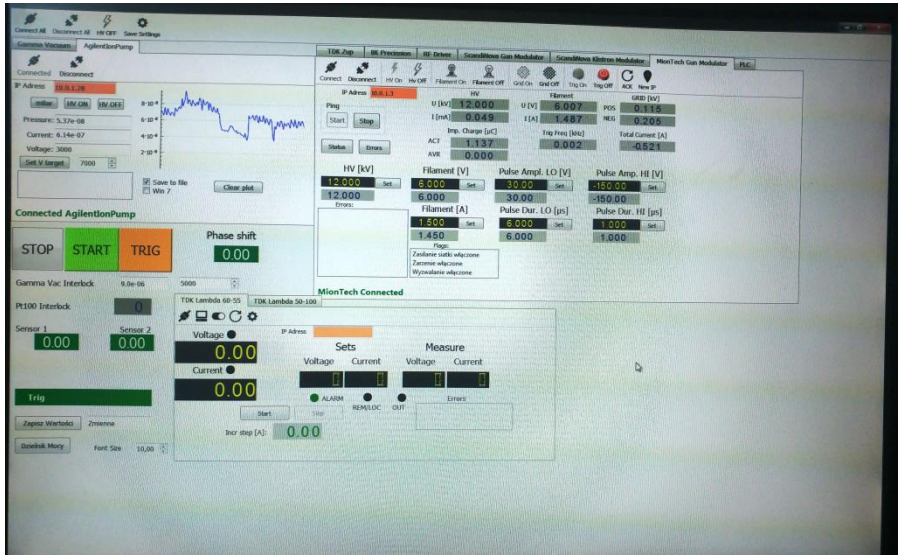
Pomiary optymalizacyjne sekcji

- Software napisany przez dra Tomasza Zakrzewskiego



	zasilacz [V]			
No. Zasilacza	1	2	3	4
podłączenie	1 i 2	3 i 4	5 i 6	7 i 8
HvPS U\ cewki	kwadrupo	Siemens	kwadrupo	kwadrupol
1050	8,2	0	1,8	0,9
1070	8,2	8	2,2	1,2
1090	8,2	8	2,4	1,2
1110	8,2	8	2,4	1,5
1130	8,2	8	2,9	1,5
1150	8,2	8	3,1	1,5
1170	8,2	8	3,5	1,5
1190	8,2	8	4	1,5

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Cewki „sterujące”

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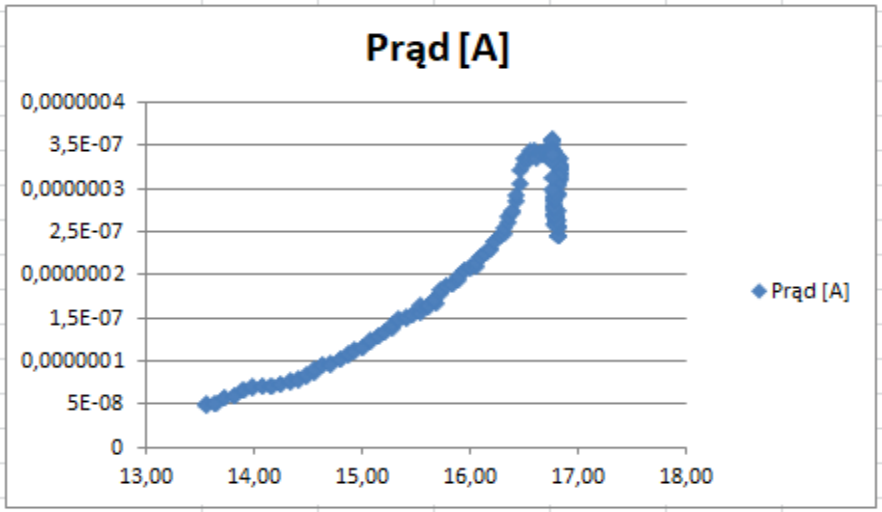
Natężenie prądu (zmierzonego na spektrometrze) w zależności od energi elektronów

	A	B	C	D	E	F	G	H	I	J	K	L	M	N	O
1								Energia z jednego wzoru [MeV]							
2	Max dla ok 4,5 V		2,95024	59,0047		-5,02E-08	5,0213E-08	13,55	3,5685375494E-07						
3	to odpowiada ok 90 A		2,95004	59,0008		-4,76E-08	4,7644E-08	13,55							
4			2,97605	59,5209		-5,02E-08	5,0174E-08	13,64							
5	sprawdzic energie		2,97419	59,4838		-5,11E-08	5,1123E-08	13,63							
6			2,99968	59,9937		-5,65E-08	5,6458E-08	13,72							
7			2,99933	59,9866		-5,78E-08	5,7763E-08	13,72							
8			3,02538	60,5075		-6,03E-08	6,0292E-08	13,81							
9			3,02411	60,4822		-6,05E-08	6,0451E-08	13,81							
10			3,04972	60,9945		-6,64E-08	6,6419E-08	13,89							
11			3,05119	61,0237		-6,75E-08	6,7486E-08	13,90							
12			3,0753	61,5059		-6,91E-08	6,9146E-08	13,98							
13			3,07605	61,5209		-7,05E-08	7,049E-08	13,98							
14			3,10217	62,0435		-7,17E-08	7,1715E-08	14,07							
15			3,10249	62,0498		-7,13E-08	7,132E-08	14,07							
16			3,12775	62,5549		-7,09E-08	7,0885E-08	14,15							
17			3,1281	62,5621		-7,19E-08	7,1913E-08	14,15							
18			3,1534	63,068		-7,27E-08	7,2743E-08	14,24							
19			3,15383	63,0767		-7,23E-08	7,2348E-08	14,24							
20			3,18249	63,6498		-7,55E-08	7,551E-08	14,33							
21			3,18237	63,6474		-7,67E-08	7,6696E-08	14,33							
22			3,20427	64,0854		-7,93E-08	7,9265E-08	14,40				0	0		
23			3,20498	64,0996		-7,85E-08	7,8514E-08	14,40				4,5	90		



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Przeprowadzka - Nowy budynek bunkra

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Nowy budynek i bunkier – sterownia stan na wrzesień 2021

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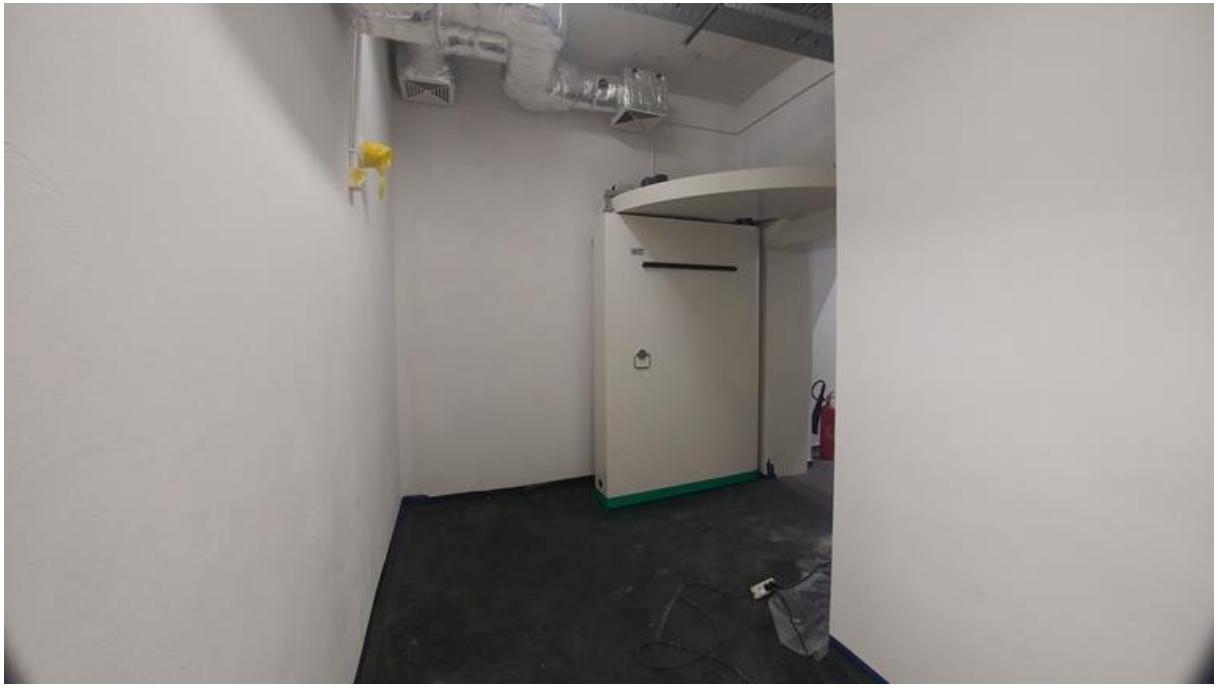
Wejście do bunkra



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Bunkier

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Wnętrze bunkra

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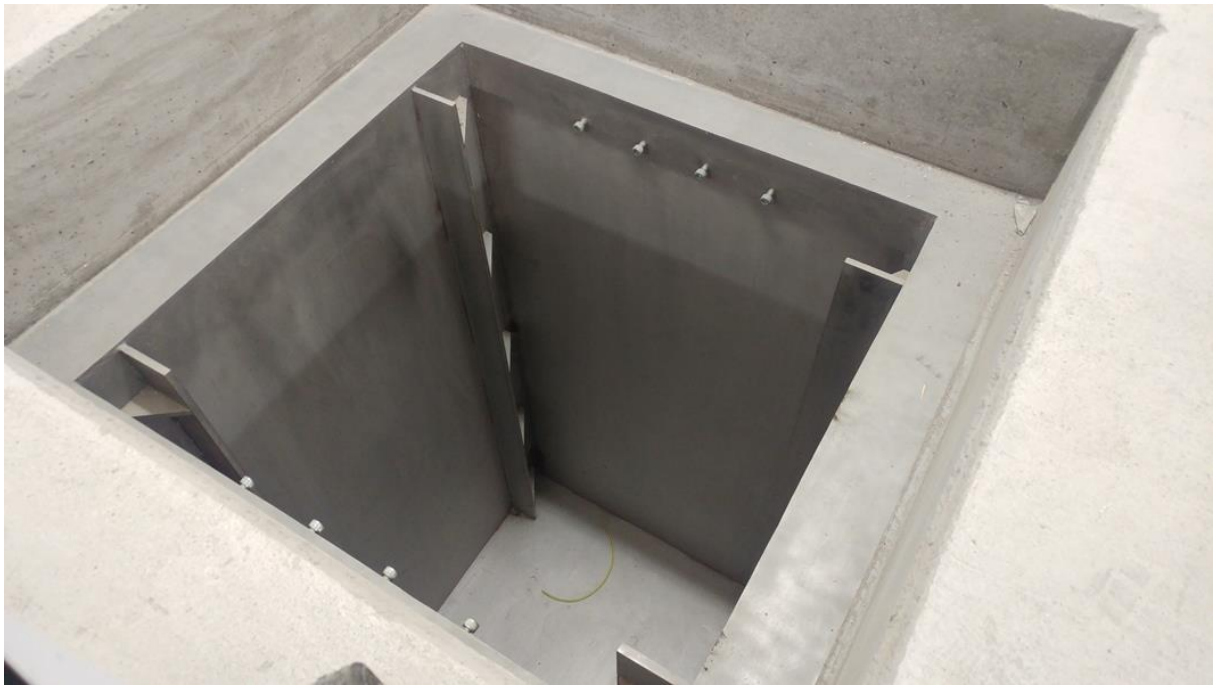
„zapadnia” na napromieniowane materiały



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Spektrometr (pomiar energii elektronów o E_{max} ok. 18 MeV)

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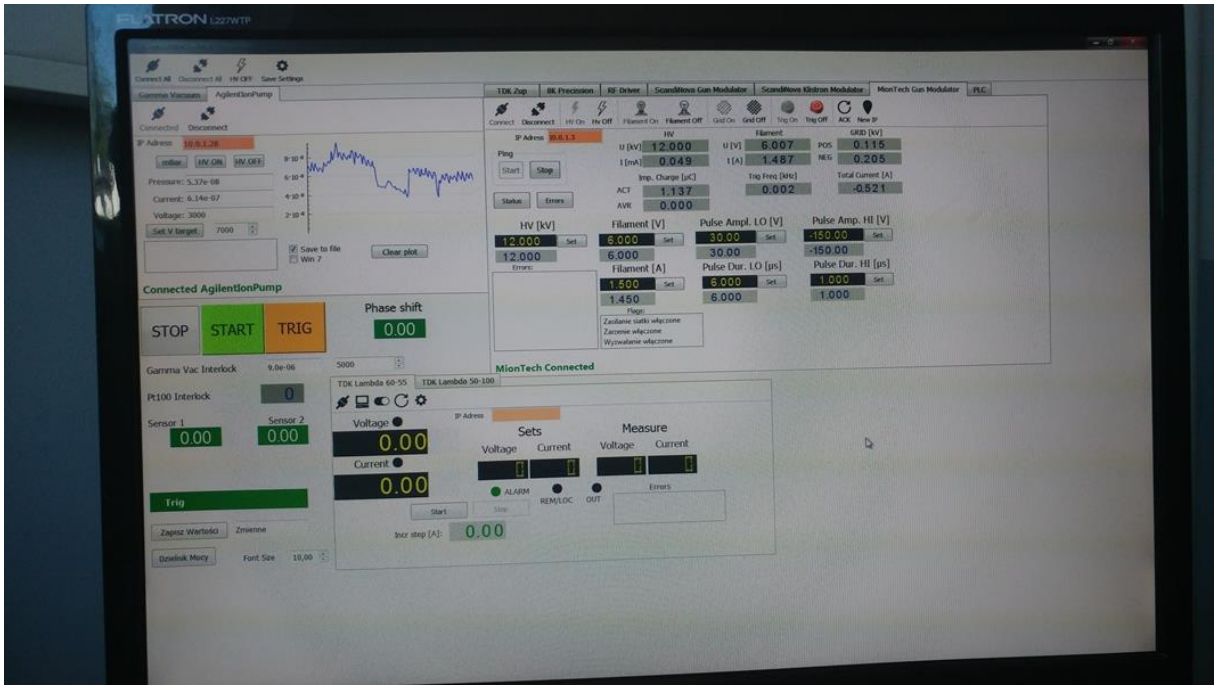
Program do sterowania akceleratorem



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Europejskiego Funduszu Społecznego

Tarcze

- 1600 °C – 2,9299g
- 1300 °C – 2,9988g

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Dwie tarcze do naświetlenia

Irradiation time (E=15 MeV, I≈100uA)	Target activity [MBq]	
	(fi=19.6 mm, d=1.2 mm) Volume=0.3619cm^3	(fi=18.8 mm, d=1.2 mm) Volume=0.3329cm^3
5 min	0,54	0,49
1h	6,34	5,83
24 h	129,65	119,29

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Detekcja Mo^{99}

- Detektor germanowy
- Aktywności do 1 MBq
  aby nie „zatkać” detektora
- Wymagana kalibracja
  dużo łatwiejsza niż w przypadku „kryształów”

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Detekcja Mo^{99} c.d.

- Rozpuszczanie tarczy
- ważenie tarczy
- 30% H_2O_2 do rozpuszczenia (3g tarcza – ok 33 mL): prędkość ok. 0.27 g/min (ok 0.091 g/mL)
- „zobojętnianie” 5M NaOH

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Detekcja Mo^{99} c.d.

- Pobrany 1 mL roztworu
- Aktywność całkowita roztworu powinna nie przekraczać 1 MBq

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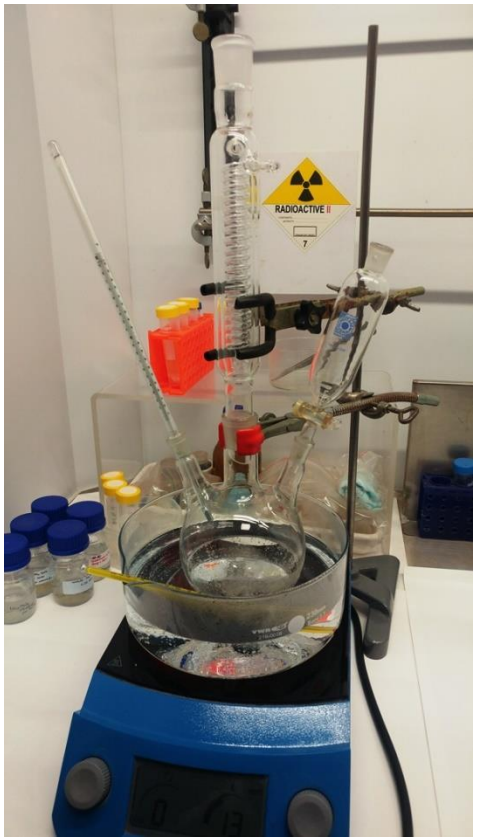
Rozpuszczanie tarczy

- Mieszadlo magnetyczne
- Plyta grzejna
- Łaźnia z wody dejonizowanej
- Chłodnica zwrotna
- Termometr z kolbą



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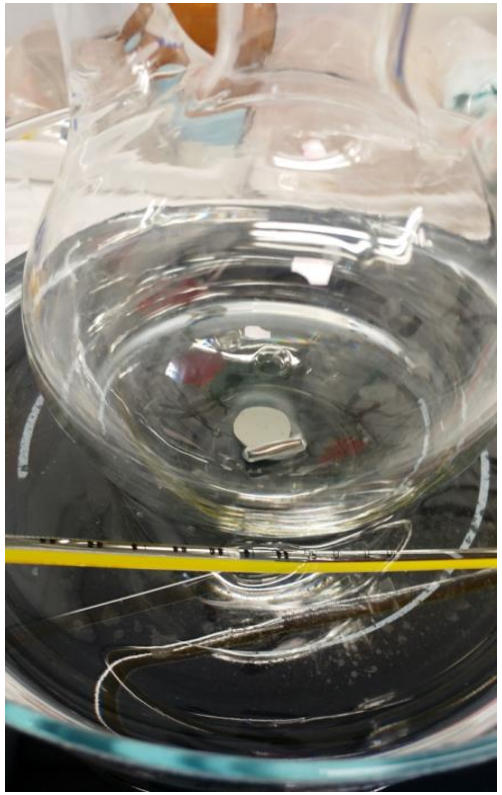
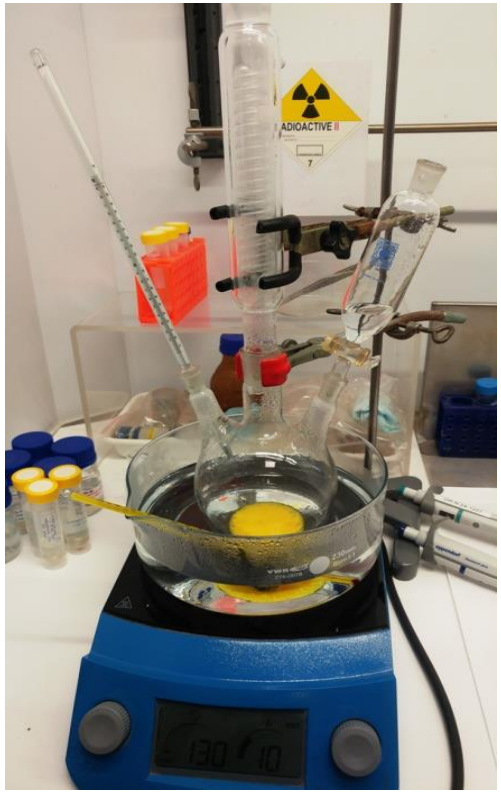
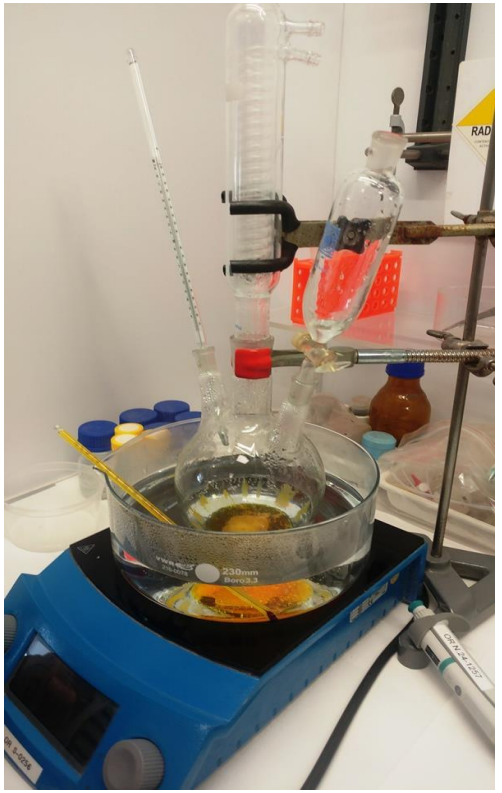
Proces rozpuszczania tarczy







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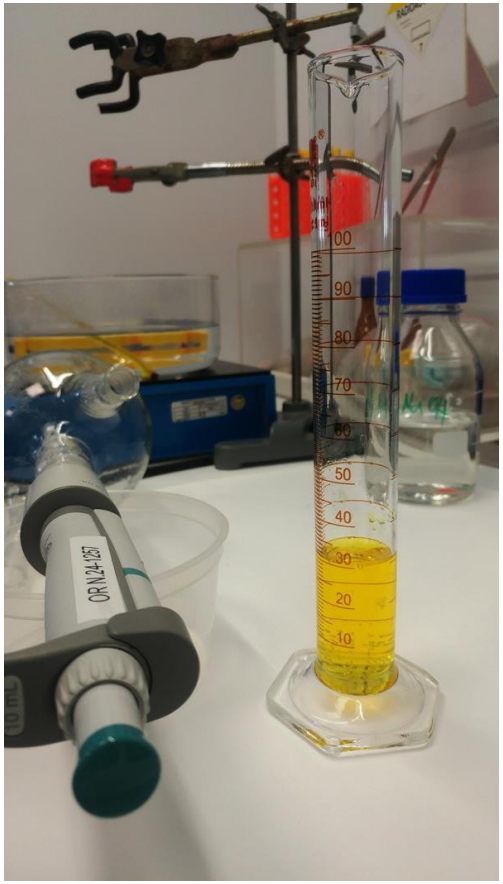

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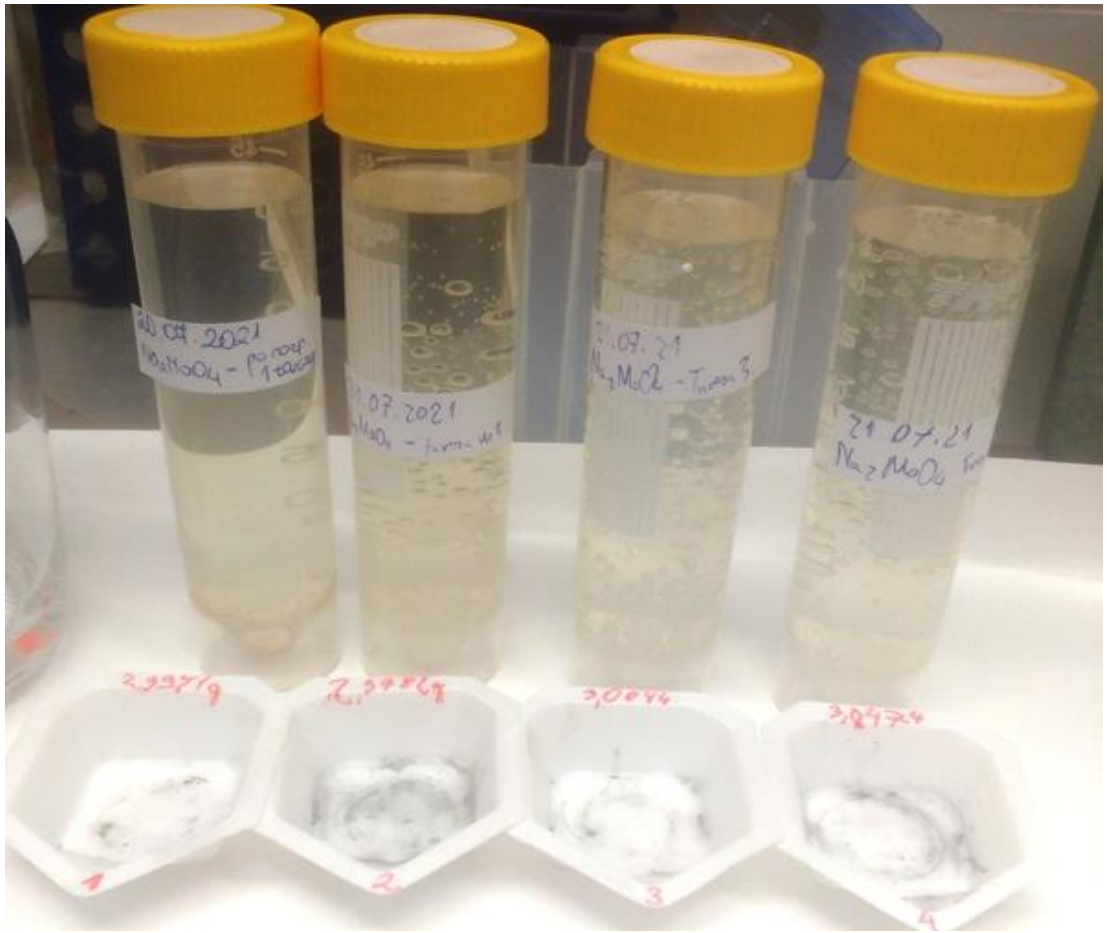
Europejskiego Funduszu Społecznego



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South African Production of High Specific Activity Mo-99 (Precursor for Tc-99m) via the (g,n) Reaction Making Use of the Szilard-Chalmers Effect for CRPF22068

(Necsa, South Africa Document Number: RL-NTPMO99-REP-20002)

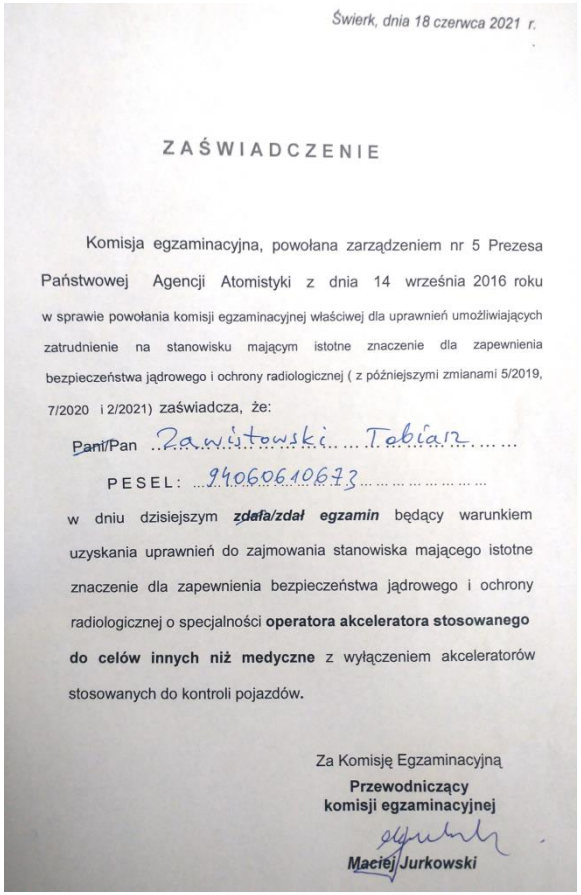
CONTRIBUTORS TO DRAFTING AND REVIEW

Dr Jan-Rijn Zeevart	South African Nuclear Energy Corporation (Necsa)
Mr Thato Sello	South African Nuclear Energy Corporation (Necsa)
Mr Phillip Pare	South African Nuclear Energy Corporation (Necsa)
Dr Carl Wagener	South African Nuclear Energy Corporation (Necsa
Dr Mark de Jong	Canadian Isotope Innovations Corporation (CII)
Mr Zawistowski Tobiasz	National Centre for Nuclear Research (NCBJ) in Poland

Dear Author,

Thank you for the source files of your manuscript.
It is my pleasure to inform you that the article entitled
"Simulation of 99Mo production from 30 MeV electron linear accelerator" will be published and will appear in a special issue of Acta Physica Polonica A Vol 139, No. 4 (2021).

Acta Physica Polonica A
Institute of Physics
Polish Academy of Sciences



Świerk, dnia 18 czerwca 2021 r.

ZAŚWIADCZENIE

Komisja egzaminacyjna, powołana zarządzeniem nr 5 Prezesa Państwowej Agencji Atomistyki z dnia 14 września 2016 roku w sprawie powołania komisji egzaminacyjnej właściwej dla uprawnień umożliwiających zatrudnienie na stanowisku mającym istotne znaczenie dla zapewnienia bezpieczeństwa jądrowego i ochrony radiologicznej (z późniejszymi zmianami 5/2019, 7/2020 i 2/2021) zaświadcza, że:

Pani/Pan Zawistowski Tobiasz

PESEL: 94060610673

w dniu dzisiejszym ***zdała/zdał egzamin*** będący warunkiem uzyskania uprawnień do zajmowania stanowiska mającego istotne znaczenie dla zapewnienia bezpieczeństwa jądrowego i ochrony radiologicznej o specjalności **operatora akceleratora stosowanego do celów innych niż medyczne** z wyłączeniem akceleratorów stosowanych do kontroli pojazdów.

Za Komisję Egzaminacyjną
Przewodniczący komisji egzaminacyjnej
Maciej Jurkowski

Fundusze Europejskie Wiedza Edukacja Rozwój
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Plan kontynuacji pracy

W celu porównania przewidywanych aktywności, wyliczonych na podstawie symulacji MC z programu FLUKA, potrzebuję przeprowadzić naświetlania i pomiary aktywności. Do dyspozycji na ten moment są 2 tarcze spiekane w atmosferze azotu (od Pana Tomasza Janiaka):

- 1) spiekana w 1600 oC – 2.9299g, 2) spiekana w 1300 oC – 2.9988g

Naświetlania ze względu na zwiększone promieniowanie w okolicy budynku nr 5 odbywałyby się w soboty. Przetransportowanie materiału napromieniowanego do wejścia na rampie (tyły ośrodka POLATOM) miałoby miejsce w poniedziałek, a następnie rozpuszczenie tarczy w laboratorium R&D POLATOM oraz pomiar 1 mL roztworu tarczy w detektorze germanowym skalibrowanym na pomiary izotop molibdenu 99. Transport tarczy po naświetleniu w pojemniku ołowianym (w związku z transportem na terenie NCBJ jest to do uzgodnienia z Panem Jerzym Wojnarowiczem - inspektorem OR).

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radfarm@ncbj.gov.pl, www.ncbj.gov.pl/radfram

Czynności		Opis		Miejsce i czas	Uwagi
1	Przygotowanie stanowiska przed naświetlaniami	Przygotowanie (wolframowej) tarczy konwersji	Podłączenie chłodzenia	NCBJ (bud. 5) np. piątek (po 16:00) lub sobota	Potrzebny detektor ze ZDAJ, aby zmierzyć poziom promieniowania po podłączeniu tarczy konwersji
		Podłączenie holderu tarczy molibdenowej	Uszczelki teflonowe i podłączenie chłodzenia (pompa od spektrometru)		
		Ustawienie osłon z jaskółek ołowianych oraz płyt polietylenowych			
2	Przeprowadzenie naświetlań tarcz molibdenowych	spiekana w 1600 °C – 2.9299g			Dozymetr indywidualny?
		spiekana w 1300 °C – 2.9988g			
	Transport tarcz do POLATOM	Transport zorganizowany w pojemniku ołowianym lub transportowym koszyku		Bunkier -> Polatom (poniedziałek)	Ustalenie miejsc z IOR
3	Rozpuszczanie tarczy	Przygotowanie stanowiska w dygestorium radiochemicznym	Osłony ołowiane + szkło ołowiane	Polatom - Laboratorium Badawczo Rozwojowe (poniedziałek)	
		Ustawienie szkła laboratoryjnego na statywie	• Chłodnica zwrotna • Kolba trójszyjna (z termometrem rtęciowym) • Wykraplacz • Cylinder miarowy		Dodatkowo potrzebne: mieszadło magnetyczne do kolby, łaźnia z wodą dejonizowaną oraz płyta grzejna
		Proces rozpuszczania	do rozpuszczenia: 30% H_2O_2 (tarcza ok. 3g wymaga użycia ok 33 mL perhydrolu), roztwór 5M NaOH		Dodatkowo potrzebne dozymetry pierścionkowe
4	Pomiar Mo-99	Detektor germanowy skalibrowany na pomiary (3 razy po 3 pomiary 1m L)	Pobrany 1 mL roztworu	Polatom - Laboratorium Wzorca Radioaktywnego	(Aktywność całkowita 1 mL nie może przekraczać 1 MBq)
5	Zebranie i analiza wyników	Wyznaczenie wydajności	Przedstawienie możliwości		Prośba o konkretne tarcze molibdenowe
		Porównanie wyników z symulacjami	Kolejnych pomiarów		

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Simulation of ^{99}Mo production from 30 MeV electron linear accelerator - Monte Carlo calculations in FLUKA

Tobiasz Zawistowski, Sławomir Wronka
National Centre for Nuclear Research (NCBJ), Sołtana 7, 05-400 Otwock, Poland

Introduction

The most widely used isotope for medical imaging is ^{99m}Tc. This radionuclide is produced via ^{99}Mo beta minus decay. A relatively new option is linac-based production of ^{99}Mo through the photonuclear reaction ^{100}Mo(γ,n)^{99}Mo with bremsstrahlung irradiation. The purpose of this study was to simulate the process of ^{99}Mo production using a 30 MeV electron linear accelerator with 100 µA average beam current. The Monte Carlo calculations were used for this purpose. The main goal of presented work was to optimize the process of irradiation the ^{100}Mo target with electron beam in order to produce ^{99}Mo with the highest efficiency.

Methodology

The simulation of irradiation of photon converter and photo-neutron targets depending on their geometry and thickness were performed. Tungsten and tantalum were investigated as photon converter targets. The Full Width Half Maximum (FWHM) of the beam dependance on bremsstrahlung photons energy was studied. The simulations were carried out using Monte Carlo method with FLUKA 2011.2x.4 program. The program code interface was: Flair_2.3-0. As the MC method requires high efficiency processors, the Świerk Computing Centre (CIŚ) software was applied for this purpose.

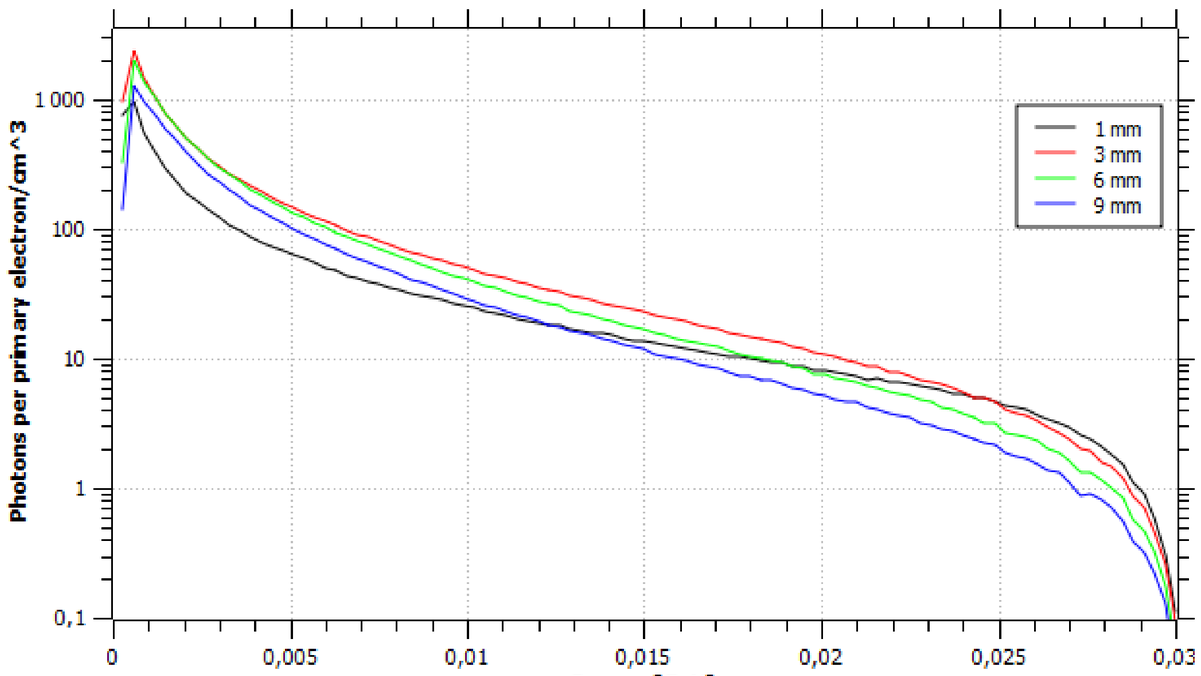

Fig. 2 – Bremsstrahlung photons rate depending on tungsten target thickness

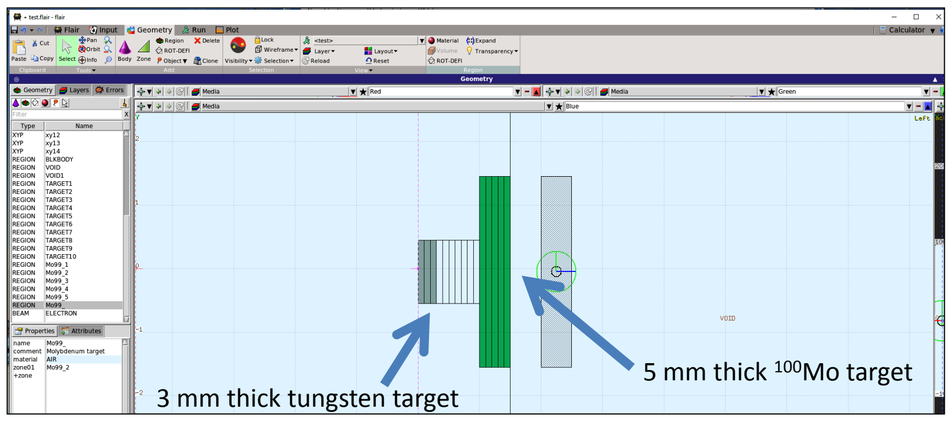

Fig. 1 – Geometry tab in Flair program and scheme of the targets system

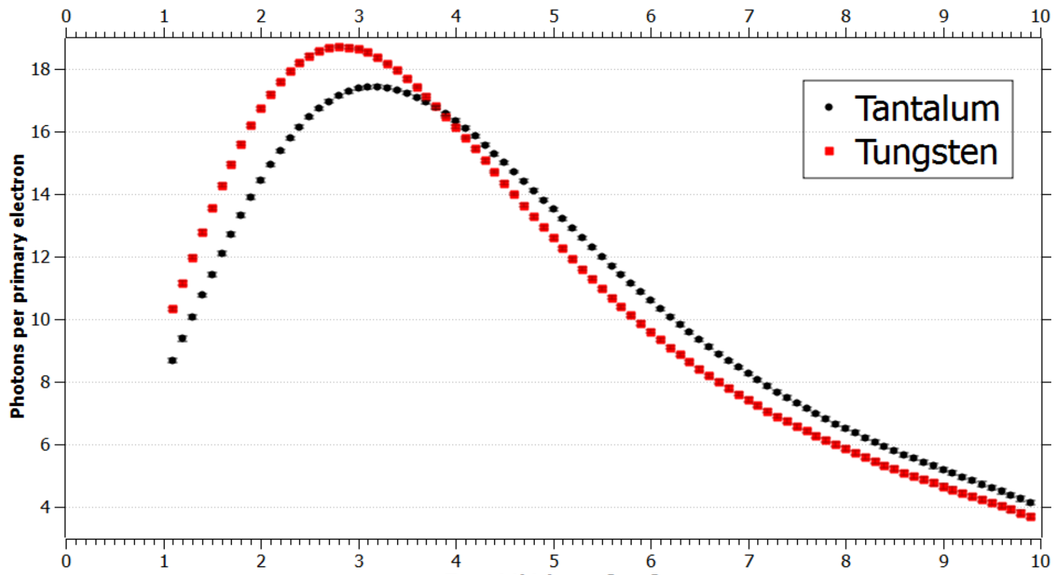

Fig. 3 – Number of photons in tantalum and tungsten depending on target thicknesses

Results

It was found that the FWHM of the beam parameter did not affect photons energy spectrum, therefore 4 mm FWHM beam parameter was chosen. The assumed geometry and scheme of targets system is presented in Fig. 1. An important issue was selection and optimization of the thickness of conversion target. Comparison of two converters is shown in Fig. 3. Tungsten is slightly more effective in generating bremsstrahlung photons when compare with tantalum. The highest number of photons was obtained for 3 mm tungsten target. This was confirmed by the measurement of photons number with energy above 10 MeV – Fig. 2 (treshhold energy for ^{100}Mo(γ,n)^{99}Mo nuclear reaction is 9.8 MeV). Performed simulation included studies on energy deposition in molybdenium target. It was found that during irradiation with photons the most exposed is first 2 mm of the molybdenum target (Fig. 4). Energy deposition is isocentric (Fig.5) and amounts about 500 J/s in the middle of the target. This indicates that in the real experiment a target cooling system will be needed. Activity of ^{99}Mo produced with 30 MeV electron beam depending on ^{100}Mo target irradiation time is shown in Fig. 6. It can be seen that as results of 72 hours irradiation of 30 mm in diameter and 5 mm thick ^{100}Mo target about 300 GBq (8.14 Ci) of ^{99}Mo will be produced.

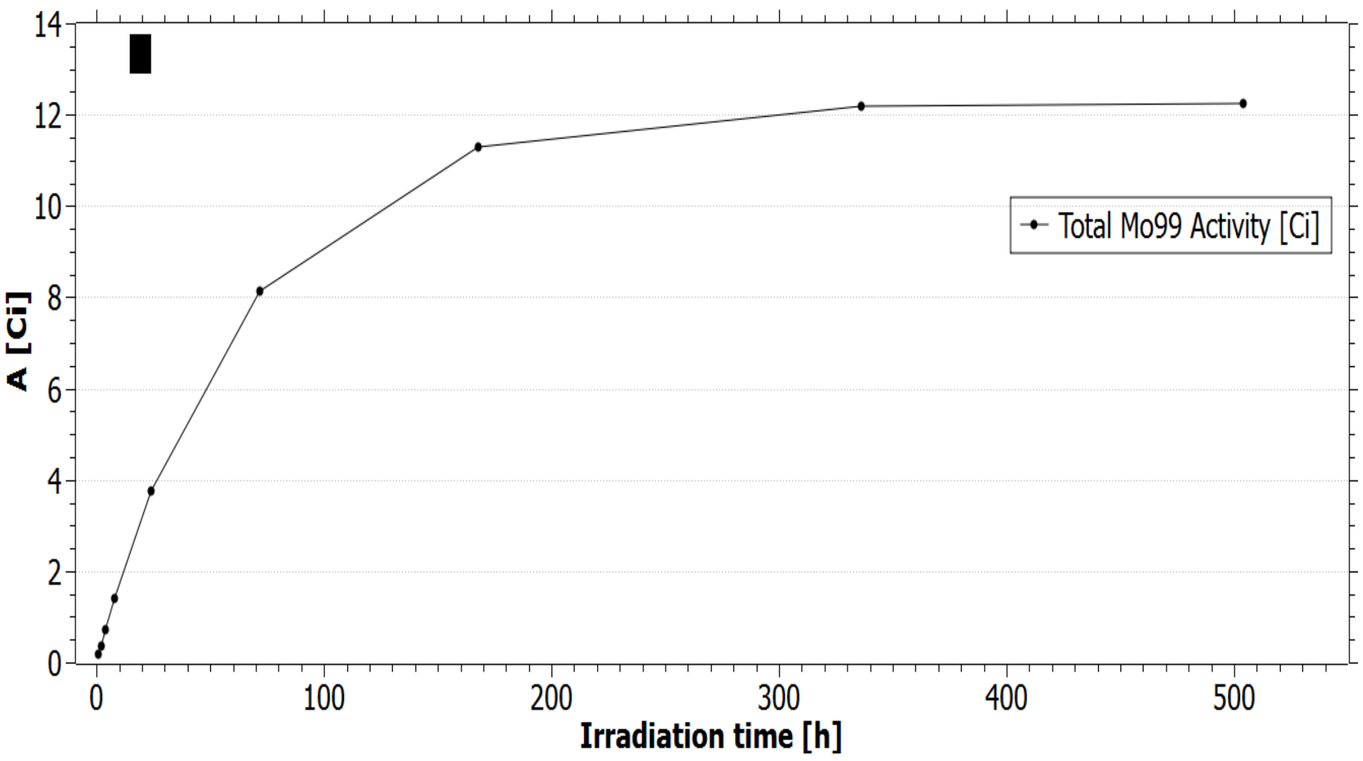

Fig. 6 – Molybdenum-99 activity (Ci) depending on irratiation time

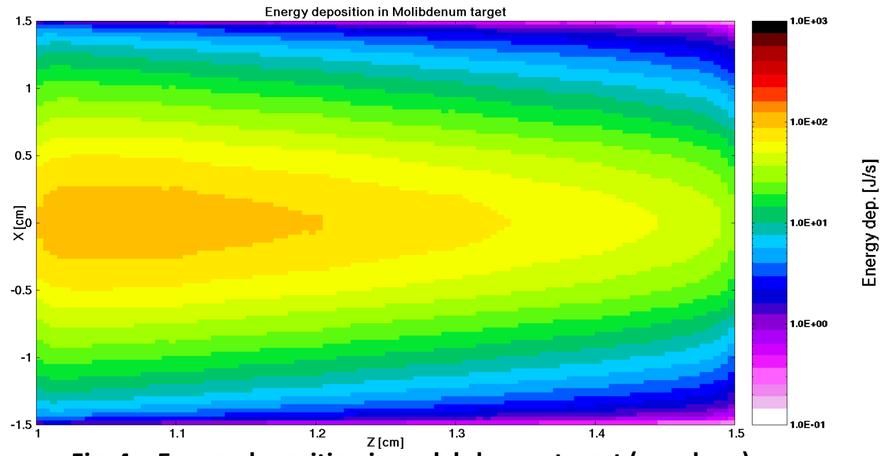

Fig. 4 – Energy deposition in molybdenum target (x-y plane)

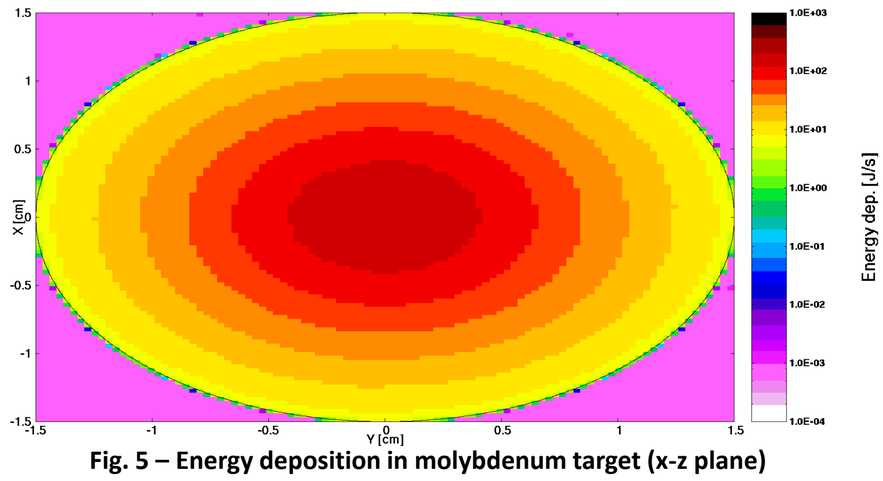

Fig. 5 – Energy deposition in molybdenum target (x-z plane)

Conclusions

The performed calculation showed that 3 mm thick tungsten disc is the most optimal convertion target used in production of ^{99}Mo according to nuclear reaction ^{100}Mo(γ,n)^{99}Mo. By the use of this convertion target it is possible to obtain 8.14 Ci of ^{99}Mo after 3 days of irradiation of ^{100}Mo target (ϕ=30 mm and 5mm thickness) with 30 MeV electron beam of 100 µA average beam current. The obtained results will be verified by experimental method in the near future.

Projekt realizowany w ramach Programu Operacyjnego Wiedza Edukacja Rozwój 2014-2020 współfinansowanego ze środków Europejskiego Funduszu Społecznego

Simulation of ^{99}Mo production from 30 MeV electron linear accelerator*

List of Authors:
T. Zawistowski, S. Wronka
National Centre for Nuclear Research, A. Soltana 7, Otwock-Świerk 05-400, Poland
corresponding email: tobiasz.zawistowski@ncbj.gov.pl

1. Abstract

The purpose of the research was to simulate the process of ^{99}Mo production. The most widely used isotope for medical imaging is ^{99m}Tc. This radionuclide is produced via ^{99}Mo beta minus decay. A relatively new option is a linac-based way of obtaining ^{99}Mo, through the photonuclear reaction ^{100}Mo(γ,n)^{99}Mo, with bremsstrahlung irradiation. The purpose of this study was to simulate the process of ^{99}Mo production using a 30 MeV electron linear accelerator with a 100 μA average beam current. The Monte Carlo calculations were used for this purpose. The main goal of the presented work was to optimize the process of irradiation of the ^{100}Mo target with an electron beam in order to produce ^{99}Mo with the highest efficiency. Parameters like FWHM of the beam and thickness of conversion target were investigated. The simulation was carried out using the Monte Carlo method with FLUKA 2011.2x.4 and the Flair_2.3-0_all. The study showed that the highest efficiency (photons production) among widely used conversion targets is at about 3 mm thick round tungsten plate. Results of the executed simulations showed that there is a possibility of obtaining 300 GBq of ^{99}Mo within 3 days of irradiation of 99% enriched ^{100}Mo target (ϕ=3 cm, 5 mm thickness).

Keywords: molybdenum-99, technetium-99m, irradiation, radioisotopes

2. Introduction

The most widely used isotope for medical imaging is ^{99m}Tc. This radionuclide is produced via ^{99}Mo beta minus decay. A relatively new option is linear accelerator-based production of ^{99}Mo through the photonuclear reaction ^{100}Mo(γ,n)^{99}Mo with bremsstrahlung irradiation. The purpose of this study was to simulate the process of ^{99}Mo production using a 30 MeV electron linear accelerator with 100 μA average beam current. The Monte Carlo calculations were used for this purpose. The main goal of the presented work was to optimize the process of irradiation of the 99,9% enriched ^{100}Mo target with an electron beam in order to produce ^{99}Mo with the highest efficiency for the proposed target system.

3. Methodology

The simulations of photon converter and photo-neutron targets irradiation depending on their geometry and thickness were performed. Tungsten and tantalum were investigated as photon conversion targets. Dependence of the FWHM of the beam on bremsstrahlung photons energy was studied. The simulations were carried out using the Monte Carlo method with the FLUKA 2011.2x.4 program [1]. The program code interface was: Flair_2.3-0. As the MC method requires high-efficiency processors, the Świerk Computing Centre (CIŚ – Centrum Informatyczne Świerk) hardware was applied for this purpose.

4. Results

It was found that the FWHM as the electron beam parameter did not affect photons energy spectrum, therefore (wider than optional 1 mm) default - 4 mm FWHM beam parameter was chosen. The assumed geometry and scheme of the target system are presented in Fig. 1. The gray rectangle (blue indicator on the left) means 3 mm of tungsten target and the green rectangle (blue indicator on the right) represents 5 mm molybdenum target. Space “material” used in the simulation was pure air.

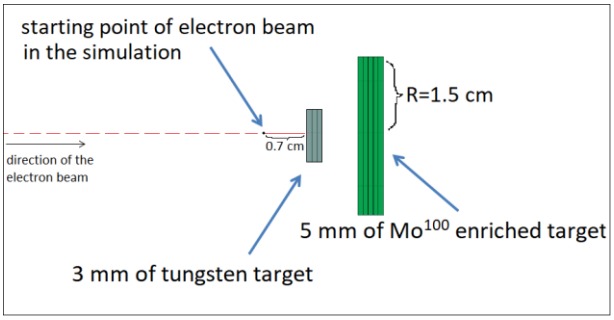

Fig. 1. – Geometry and scheme of the targets system

An important issue was the selection and optimization of the thickness of the conversion target. Two converters comparison is shown in Fig. 2. The highest number of photons was obtained for about 3 mm tungsten target. Maximum photon fluence can be obtained for a 3.2 mm tantalum target or 2.8 mm tungsten target.

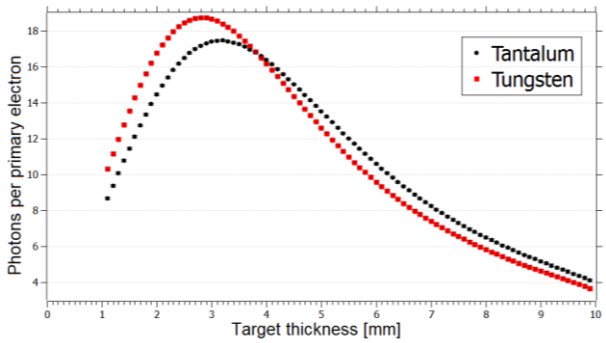

Fig. 2 – Comparison of the conversional targets

Tungsten is slightly more effective in generating bremsstrahlung photons when compared with tantalum. The photonuclear reaction (^{100}Mo(γ,n)^{99}Mo) needs photons with adequate energies. According IAEA data, the threshold energy for ^{100}Mo(γ,n)^{99}Mo photonuclear reaction is equal to 8.2942(+/-0.0004) MeV [2, 3]. However, in some papers authors give a value of 9.8 MeV [4, 5] or 9 MeV [6]. The measurement of the relative amount of high energy photons for different thicknesses of tungsten confirmed, that the highest number of photons (with energies above threshold energy) could be gained for 3 mm thickness.

The percentage share of the photons amount is different depending on converter thickness, but the highest amount of photons with desired energy is for the 3 mm tungsten conversion target. For clearer display results the photons corresponding to the highest amount were marked by a red line (Fig. 3.).

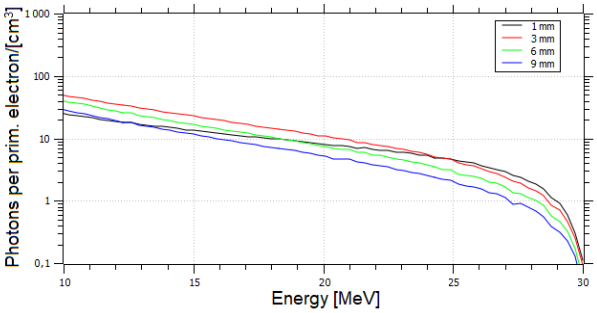

Fig. 3 – Bremsstrahlung photons depending on tungsten thickness

The performed simulation included studies on energy deposition in the molybdenum target. It was found that during irradiation with photons the most exposed is the first 2 mm of the molybdenum target (Fig. 4). Energy deposition is concentric (Fig.5) and the power equals about 500 J/s in the middle of the target. This indicates that in the real experiment a target cooling system seems to be necessary.

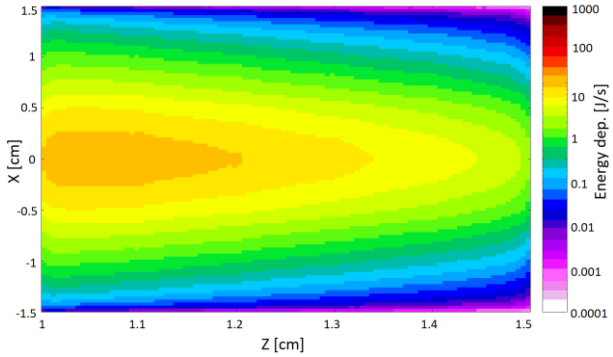

Fig. 4 – Energy deposition in molybdenum target (x-z plane)

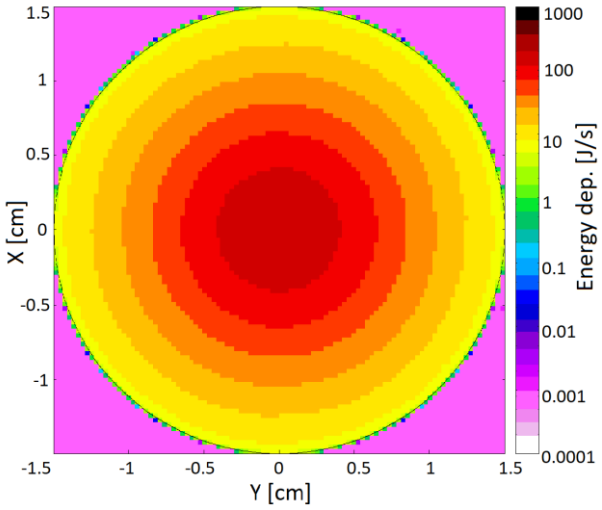

Fig. 5 – Energy deposition in molybdenum target (x-y plane)

The activity dependence of ^{99}Mo produced with 30 MeV electron beam on ^{100}Mo target irradiation time is shown on Fig. 6.

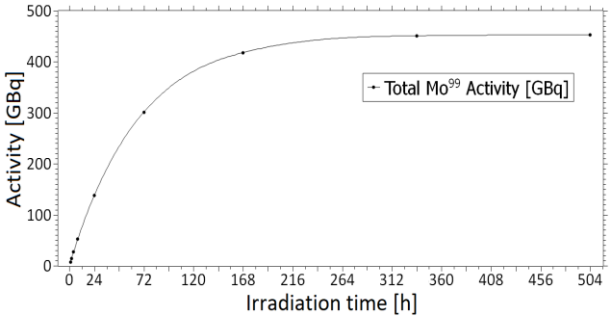

Fig. 6 – Molybdenum-99 activity [GBq] depending on irradiation time

It can be seen that as results of 72 hours irradiation of 30 mm in diameter and 5 mm thick ^{100}Mo target activity about 300 GBq of ^{99}Mo will be produced. Such a great amount of ^{99}Mo nuclei could provide material for fifteen 20 GBq molybdenum-technetium generators (for nuclear medicine). It could be an alternative way of obtaining ^{99}Mo to the method of production using nuclear reactors.

5. Conclusions

The performed calculations showed that a 3 mm thick tungsten disc is the most optimal conversion target used for the production of ^{99}Mo via the nuclear reaction: ^{100}Mo(γ,n)^{99}Mo. Using a conversion target, it is possible to obtain about 300 GBq of ^{99}Mo after 3 days of irradiation of ^{100}Mo target (ϕ=30 mm and 5mm thickness) with 30 MeV electron beam of 100 μA average beam current. The obtained results will be verified by the experimental method in the near future.

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South African Production of High Specific Activity Mo-99 (Precursor for Tc-99m) via the (g,n) Reaction Making Use of the Szilard-Chalmers Effect for CRPF22068

(Necsa, South Africa Document Number: RL-NTPMO99-REP-20002)

ABSTRACT

Experiments were conducted on a 35 MeV electron Linac where it was shown that the irradiation of Mo in the form of $^{nat}Mo(CO)_6$ by spallation gammas does indeed result in the formation of activated ^{99}Mo atoms. The $^{99}Mo(O)_3$ was separated from the $^{nat}Mo(CO)_6$ using sublimation. Enrichment factors between 1.3 and 307 were obtained and it was shown that these enrichment factors were heavily dependent on the efficiency of the sublimation.

INTRODUCTION

Background

South Africa is part of the CRP F22068 which involves "New Ways of Producing 99mTc and 99mTc Generators" In the 2017 RCMP [1] it was said that South Africa intends to use CRP F22068 to

> investigate the feasibility of Mo-99 production (for Tc-99m production) via the (γ, n) reaction. The main problem is the low specific activity of Mo-99 that will result. This can be overcome using the Szilard Chalmers / hot atom chemistry effect where the produced radionuclide can be separated from the non-activated isotopes of the same element. Classically, this has been demonstrated in the form of recoil where the activated nuclide is trapped in a capture material. This however is not the only case. If the oxidation states of the activated nuclide (in this case Mo-99) and the target material (Mo-100) after the activation are different, then the separation of the two is theoretically possible. Such separation has been reported in the literature [2-7]for the (γ,n) reaction , and will form the basis of the research. Therefore, the main aim is to show that the irradiation of Mo in the form of $Mo(CO)_6$ will result in activated ^{99}Mo atoms in a different chemical form and to find what enrichment factor can be obtained. If this is successful the following parameters will need to be investigated. 1. Recovery yield, 2. Purification from other radionuclides, 3. Target design.

Aim of Project

It was intended to demonstrate the production of Mo-99 using the Szilard and Chalmers effect [8] when irradiating $Mo(CO)_6$ with gamma rays. It was to be shown that those molybdenum atoms that undergo the nuclear reaction would have their oxidation state changed from 0 to VI and would react with the free oxygen present to form MO_3. $Mo(CO)_6$ was to be sublimated at between 90°C and 110°C, so as to remove the molecules where no reaction had occurred while leaving radioactive MO_3 in the target chamber. The results would be compared with those obtained in a Russian experiment as reported in [5, 7]. A collaboration was to be established between the South African Nuclear Energy Corporation (Necsa) in South Africa, the National Centre for Nuclear Research (NCBJ) in Poland and the Canadian Isotope Innovations Corporation (CII) in Saskatoon, Canada.

The aims of the CII program and their Linac, which is installed at the Canadian Light Source (CLS), was described in an article "Producing medical isotopes using X-rays" in [9] and is patented under the title "Production of Molybdenum-99 using electron beams" in [10]

MATERIALS AND METHODS

The collaboration was established with CII in Canada and with NCBJ in Poland and the experiments were conducted in Saskatoon in the last two weeks of February 2020.

In preparation for these experiments a sublimation procedure was designed and tested in South Africa. The necessary equipment was then transported to Canada where the experiments were conducted at the Canadian light source which is within walking distance of the CII offices.

The experiments were conducted on the CII Linac which was manufactured by a Canadian company, Mevex. It is a standing wave machine which uses microwaves to accelerate electrons to 35 MeV in a beam which has a maximum power of 40 kW. The peak current is 215 mA and the diameter of the beam is 5 mm. The power is adjusted by changing the duty cycle of the pulses while the 215 mA peak current is kept constant. We had the LINAC operated at its minimum power of 1 kW. The amplifiers would have become unstable should we have operated at a lower power.

The target for the electrons was a tantalum spallation target which generated the photons necessary for the ^{100}Mo(ɣ,n) reaction. It was water cooled. 50% of the power of the beam was absorbed by the tantalum and the remaining 50% was radiated out in the photon beam. Usually the photons would then impinge on specially fabricated enriched molybdenum-100 discs, but for this experiment the target was a few hundred milligrams of $Mo(CO)_6$ in the South African designed aluminium alloy canister.

After irradiation the activity of the target was measured in a dosimeter. Full gamma ray spectroscopy was done on the first two the targets using an HPGe detector calibrated with multi gamma and mono-gamma emitting NIST traceable sources.

After the radiation measurements, the $Mo(CO)_6$ from five of the six targets were sublimed under continuous vacuum at a pressure of 10 millibars and at a temperature of between 95-100 ^{o}C. A total of six targets where irradiated and the $Mo(CO)_6$ from five of these targets was sublimed.

After sublimation the residue was dissolved in 3M NaOH. Both this solution and the sublimate were then analysed using the HPGe detector mentioned earlier.

The results from the Gamma spectroscopy were first analysed using the manufacturer's software and subsequently the results were further analysed using an Excel spreadsheet. The Excel spreadsheet enabled all activities to be calculated as of **end of bombardment**.

RESULTS

First Target

All the activities that are mentioned have been calculated to represent the activity as at the time of **the end of bombardment.** The mass of the first $Mo(CO)_6$. sample was 235 mg. After it was irradiated for 1 hour, the activity of the Mo-99 739.5 keV gamma ray was 114 kBq (3.08 μCi) The spectroscopy was performed 180×10^3 s (49.9 hours) after irradiation. Unfortunately after this irradiation the sample was too burnt to be sublimated. This radiation level means that 65.0×10^{-15} moles of Mo-99 had been produced by the irradiation at the end of bombardment (EoB). This was found from the standard formula $n\ (moles) = \frac{A}{N_A \lambda}$ where the decay constant, λ, of Mo-99 is 2.918×10^{-6} s^{-1}. This is found from the Mo-99 half life of 237.51×10^3 seconds (or 65.98 hours) where $\lambda = \frac{\ln(2)}{T\,½}$. N_A is Avogadro's constant = 6.022141×10^{23} atoms per mole.

Second Target

For the second sample it was decided to keep the mass almost the same but to reduce the irradiation time by a factor of ten. This meant that for the second sample the mass of the $Mo(CO)_6$. was 206 mg but the irradiation time was only 6 minutes. In this case the activity of the Mo-99 739.5 x keV gamma ray in the sample was 10.151 kBq (0.274 μCi) at the end of bombardment. This figure was obtained in the same way as described above for the first sample. The gamma spectroscopy was performed 121 hours after irradiation. It was sublimated 4.24 days later. The activity of the Mo-99 739.5 k eV gamma ray in the sublimate at EoB, which we assume to be $Mo(CO)_6$, was only 81 Bq. This is in contrast to the residue that remained in the sample holder which had a Mo-99 activity of 1073 Bq and a mass of 3.9 mg.

In further experiments the total activity of the target was unfortunately not measured using gamma spectroscopy prior to the commencing of the sublimation. It was only measured using a dose calibrator which did not give usable results because of the number of different isotopes that were present in the target.

Composition of Residue and Sublimate

For initial calculations, the simplification was made that all the molecules in the sublimate were $Mo(CO)_6$ molecules where the isotopic composition of the Mo was equal to that of natural Mo.

Using the same formula, $n\,(moles) = \frac{A}{N_A \lambda}$ mentioned above, meant that there were 5776×10^{-18} moles of Mo-99 atoms in the target; 610.3×10^{-18} moles of Mo-99 in the residue and 46.1×10^{-18} moles of Mo-99 in the sublimate. 610×10^{-18} moles of $^{99}MoO_3$ has a mass of 89.7×10^{-18} kg which shows that most of the 3.9 mg of residue did not contain MoO_3 but can be assumed to contain un-sublimated $Mo(CO)_6$. This means that to convert the 3.9 mg of residue to moles of atoms, the molar mass of unenriched $Mo(CO)_6$ should be used. This also means that the efficiency of the sublimation has a large role to play in the enrichment factors. The mass of the residue was measured on a balance with a resolution of 0.1 mg. The mass of the sublimate was found by subtracting the mass of the residue from the mass of the full target. This had to be done as the sublimate absorbed water during the sublimation.

This also means that there was a deficit of $(5776 - 610 - 46.1) \times 10^{-18}$ moles of Mo-99. It has not yet been possible to account for this deficit of 5100×10^{-18} moles of Mo-99.

Using these simplifications, it is found that the 3.9 mg mass of residue contains approximately 14.77×10^{-6} moles of Mo atoms.

The concept of "fractional abundance" as described by HAYES [11] will be used to calculate the enrichment factor.

The fractional abundance of Mo-99, in the residue was

$$^{99}F_{residue} = \frac{\text{moles of Mo}-99}{\text{moles of all Mo isotopes}} = \frac{610.3 \times 10^{-18}}{14.77 \times 10^{-6}} = 41320 \times 10^{-15}.$$

Likewise the 202.1 mg mass of sublimate contains approximately 765.5×10^{-6} moles of Mo atoms. This means that the fractional abundance of Mo-99 in the sublimate was

$$^{99}F_{sublimate} = \frac{\text{moles of Mo}-99}{\text{moles of all Mo isotopes}} = \frac{46.1 \times 10^{-18}}{765.5 \times 10^{-6}} = 60.2 \times 10^{-15}.$$

From the mass of the un-irradiated $Mo(CO)_6$,the total number of Mo atoms, including all the Mo isotopes in the full target was 780.3×10^{-6} moles. To resolve the problem of the missing Mo-99 mentioned earlier, the number of moles of Mo-99 in the complete target can be taken to be equal to the sum of the moles of Mo-99 in the residue and the number of moles of Mo-99 in the sublimate. As the total activity of the target at the end of bombardment was not measured in subsequent experiments this approach had to be used to be consistent as opposed to using the activity in the target vial prior to sublimation. Hence $N_{Mo\text{-}99}$ in the target before sublimation was taken to be $610.3 \times 10^{-18} + 46.1 \times 10^{-18} = 656.4 \times 10^{-18}$ moles. This means that the fractional abundance in the full target was

$$^{99}F_{\text{full target}} = \frac{\text{moles of Mo}-99}{\text{moles of all Mo isotopes}} \frac{656.4 \times 10^{-18}}{780.3 \times 10^{-6}} = 841.16 \times 10^{-15}$$

If we define the enrichment factor ε to be $\frac{^{99}F_{residue}}{^{99}F_{target}}$ then

$\varepsilon = \frac{41320 \times 10^{-15}}{841.16 \times 10^{-15}} =$ 49.1 for target 2.

It could be argued that the number of moles of Mo-99 should be compared with the number of moles of Mo-100 (9.63% natural abundance) as the Mo-99 is procured following the Mo-100 (γ,n) Mo-99 reaction. However because the short bombardment does not cause any significant burn up, the abundance of the Mo-100 does not change in this experiment and so the number of moles of natural Mo can be used to describe the fractional abundance of Mo in the full target.

The remaining targets

Using similar methods for the remaining targets the following table of enrichment factors is obtained:

Table 1 Fractional Abundance of Mo-99 (^{99}F) and Enrichment Factors

	Full Target	Residue	Sublimate	Enrichment Factor	Sublimation **efficiency**
	$^{99}F_{\text{Full Target}}$	$^{99}F_{\text{Residue}}$	$^{99}F_{\text{ Sublimate}}$	ratio	%
Target 1	73010×10^{-15}				
Target 2	841.16×10^{-15}	41320×10^{-15}	60.2×10^{-15}	49.1	98.11%
Target 3	553.65×10^{-15}	27000×10^{-15}	169.2×10^{-15}	48.8	98.57%
Target 4	1830×10^{-15}	1126000×10^{-15}	1500×10^{-15}	614.1	99.97%
Target 5	1140×10^{-15}	1435×10^{-15}	624.6×10^{-15}	1.3	36.50%
Target 6	1220×10^{-15}	255537×10^{-15}	960.2×10^{-15}	209.9	99.90%

DISCUSSION

In Figure 1 below, the Enrichment Factor is plotted against the sublimation efficiency which is the percentage of the target that sublimed.

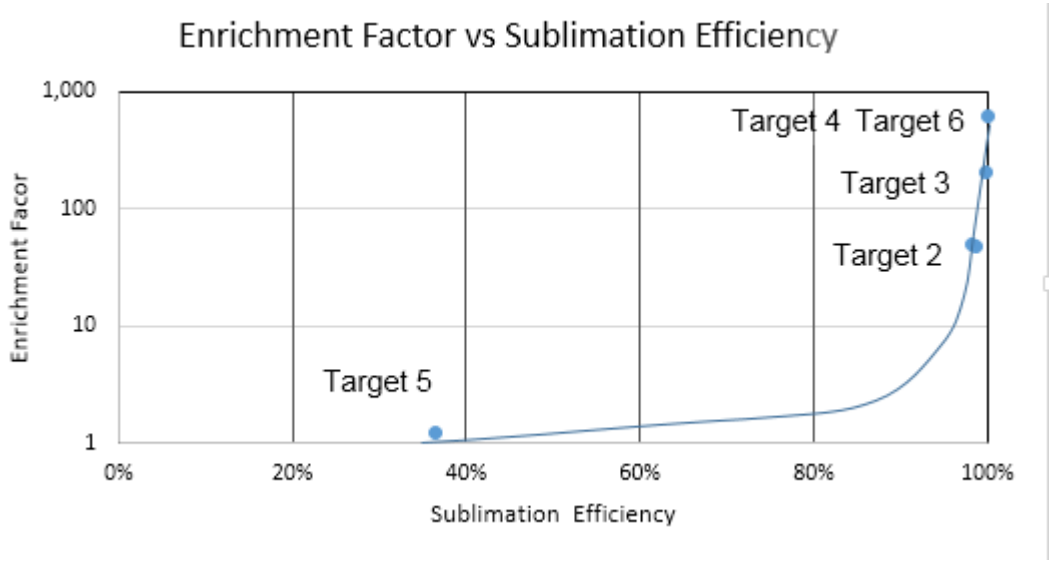

Figure 1 Graph of Enrichment Factor against the percentage of the target that sublimed which we call "sublimation efficiency".

It can be seen from this plot and from the data in table 1 above that the enrichment factor is heavily dependent on how much of the target sublimed. When there was almost complete sublimation then the mass of the residue was negligible as the mass of the $^{99}MoO_3$ is very small and it is only the Mo-99 that changes its oxidation number to form the molybdenum oxide. It was also noted that not all the Mo-99 formed $^{99}MoO_3$. Some of the Mo-99 remained as $^{99}Mo(CO)_6$ and sublimed thereby showing up in the gamma spectroscopy as 739.5 keV gamma activity.

Apart from the fact that no data was obtained to determine if an activity balance was obtained for targets 3 to 6, no measurements were made towards determining if a mass balance was obtained for all targets. The reason for this is that moisture went into the receiving vial and traps making it impossible to determine the mass of the $Mo(CO)_6$

Error Analysis of the Enrichment factor.

All masses where measured with a balance that had an accuracy of $\pm$ 0.1 mg = $\pm 1 \times 10^{-7}$ kg . This uncertainty only has a large effect on the mass of the residue for target 4 . The residue had been measured as 0.1 mg or 0.1×10^{-6} kg. This means that target 4 residue mass could be in the range of 0.00 mg < residue mass < 0.20 mg. The minimum could be defined by the mass of the $^{99}MoO_3$ if it was assumed that all the $Mo(CO)_6$ sublimed. This would result in the minimum number of moles of MoO_3 being the figure obtained from the activity of the Mo-99 in the residue which is 426×10^{-18} as shown in table 5 above. This means that ^{99}F fractional abundance of Mo-99 in the residue could be between unity and 426×10^{-12}. This means that the enrichment factor could be between 307 and 5.5×10^{11}. This minimum value of 307 for the enrichment factor is still larger than the enrichment factors obtained from the other targets and so it can be conservatively stated that the experiment yielded enrichment factors for all the targets between 1.3 and 307. The biggest

effect on the enrichment factor was the sublimation efficiency which is the amount of sublimation that had taken place. Table 6 below shows the error margins for all the enrichment factors and the graph in figure 1 above shows the effect of the degree of sublimation on the enrichment factor.

Table 2 Enrichment Factor error ranges based on mass uncertainties

	Enrichment Factor		
	Minimum	Nominal	Maximum
Target 2	47.9	49.1	50.41
Target 3	47.3	48.8	50.43
Target 4	307.0	614.1	5.5×10^{11}
Target 5	1.2593	1.2596	1.2599
Target 6	174.9	209.9	262.3

Target 2: Unaccounted for Activity

There is still a concern about the unaccounted for activity in target 2. An attempt was made to use the Nb-96 activity to investigate this problem further. While 1105 Bq of Nb-96 was detected in the full Target 2 before sublimation using the 658.871 keV gamma, Nb activity was not detected in the residue nor in the sublimate of Target 2. Besides the 739.5 keV gamma from Mo-99 there were no other gammas that were common to the full target, the residue and the sublimate.

CONCLUSION,

Unfortunately the stability of the $Mo(CO)_6$ molecule is such that it is difficult to irradiate it with the intensity that would be normally be required to produce Mo-99 at the desired rate. This makes it appear that the CII irradiation of pure Mo disks will produce more useful Mo-99 but the problem of low specific activity remains. The challenge would be to design a target arrangement that would not burn the $Mo(CO)_6$ before sublimation.

In future experiments the design of the experiment should allow for the following measurements to be done for each target after the end of bombardment:

1. The activity of the target vial prior to sublimation should be measured using gamma spectroscopy and not just with a dose calibrator.
2. The activity of the connecting tubes, recipient vial, cold trap and any other components should also be measured using gamma spectroscopy.
3. The mass of the connecting tube, recipient vial, cold trap and any other components should be measured while making sure that water is not absorbed.

Future work could also include an investigation into the stability of ^{100}Mo-Pc in a (ɣ,n) reaction as an alternative to the $Mo(CO)_6$ which we have investigated, and as a complement to the . (n,ɣ) reaction that Febrian et al investigated.[12, 13]

Further future work could also include the use of irradiated MoO_3 nano-particles where the recoil nuclei of ^{99}Mo after irradiation of the solution by bremsstrahlung are separated by means of diantipyrylmethane in sulphuric acid solution. This would be an extension of Dikiy et al's work. [14]

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ABBREVIATIONS

NECSA	South African Nuclear Energy Corporation
CII	Canadian Isotope Innovations Corporation
CLS	Canadian Light Source
NCBJ	National Centre for Nuclear Research (NCBJ) in Poland

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Raport ze stażu zagranicznego/~~krajowego~~

I. Informacje ogólne:

Imię i nazwisko doktoranta(tki):	Tobiasz Karol Zawistowski
Opiekun naukowy/Promotor (imię i nazwisko):	dr hab. Sławomir Wronka, prof. NCBJ
Jednostka:	Narodowe Centrum Badań Jądrowych
Data stażu:	16-29.02.2020 r.
Miejsce stażu:	CANADIAN ISOTOPE INNOVATIONS Corp.
Opiekun stażu:	Mark de Jong

II. Informacje o przebiegu stażu:

II.1. Cel stażu:

I was the intern at CANADIAN ISOTOPE INNOVATIONS Corp. (CII) and I took part in the experiment at Canadian Light Source (CLS). The purpose of the experiment was to explore the possibility of obtaining a high specific activity of molybdenum–99.

II.2. Opis przebiegu stażu:

The research at CLS was a part of the IAEA project: *"New Ways of Producing ^{99m}Tc and ^{99m}Tc Generators"*. Molybdenum–99, which undergoes nuclear decay to technetium–99m, is the main isotope used in nuclear medicine. The nuclear reaction leading to molybdenum–99 production using accelerators is **^{100}Mo(γ,n)^{99}Mo**. The Molybdenum–100 nucleus emits a neutron when a high–energy photon hits the target. In the experiment carried out at CLS, irradiation of the Molybdenum hexacarbonyl (Pic. 1) target took place in a linear electron accelerator with 1 kW beam power and 35 MeV beam energy. The Canadian Light Source linear accelerator is located in the underground bunker. It is surrounded by tons of concrete, lead and polyethylene plates. The photon beam necessary for the nuclear reaction was produced as a result of bremsstrahlung radiation of decelerated electron on the atomic nuclei of a 3 mm conversion target made of tantalum. Each target was weight in the holder (Pic. 2) made of aluminum.

Picture 1 – Container containing $Mo(CO)_6$

Picture 2 – Target holder

We have done 6 irradiations within 2 weeks of research. The material of the target was natural $Mo(CO)_6$ – white powder). The activity of each target was measured in dose calibrator just after irradiation (to check the level of radiation). Then the target was connected to a special tube (Pic. 3) which let to conduct the sublimation process (Pic. 4). Equipment necessary to sublimation was stored in the radiochemical fume hood in the industrial chemistry lab.

Picture 3 – Target holder and cooler tube

Picture 4 – Sublimation apparatus after sublimation

For each sublimation process I had to prepare a dewar with few liters of liquid nitrogen and the heating coat for the target. Molybdenum hexacarbonyl sublimes at about 105 Celsius degrees, after about 1 hour of sublimation we had sublimate ($Mo(CO)_6$) moved out of target holder. High purity germanium detector spectroscopy was used to identify isotopes produced during target irradiation. Even in natural molybdenum, there was possible to obtain high specific activity of ^{99}Mo. We had to compare the spectra of our sublimate and target after sublimation. Table 1 shows the results for target number 6 ($Mo(CO)_6$ – the sublimate and the MoO_3 – the content of the holder after sublimation).

Table 1 – Specific activity of Molybdenum–99 in target no.6

Target	Irradiation time [s]	Gamma Spectra ID	^{99}Mo Activity (Bq)	Ratio of MoO3 to $Mo(CO)_6$ ^{99}Mo Activity	Mass (grams)	Specific Activity (Bq/gram)	Ratio of MoO_3 to $Mo(CO)_6$ ^{99}Mo Specific Activity
$Mo(CO)_6$	180	MoO3_3600s	843	0,27	0,0005	1,69E+06	270,4
		Mo(CO)6_3600s	3080		0,4939	6,24E+03	
		MoO3_10800s	310	0,28	0,0005	6,20E+05	280,7
		Mo(CO)6_10800s	1091		0,4939	2,21E+03	

According to the Szilard–Chalmers effect, hot atoms of one element are more likely to react if their weight is greater (heavier isotopes). The ratio of ^{99}Mo specific activity in MoO_3 and $Mo(CO)_6$ is ≈280. The ratio of ^{99}Mo to other molybdenum isotopes ratio (in the product MoO_3) is also significantly greater than in natural molybdenum (natural abundance of ^{99}Mo is only about 10%). Picture 5 shows the energy spectrum of the dissolved target after sublimation. The technetium peak is higher because

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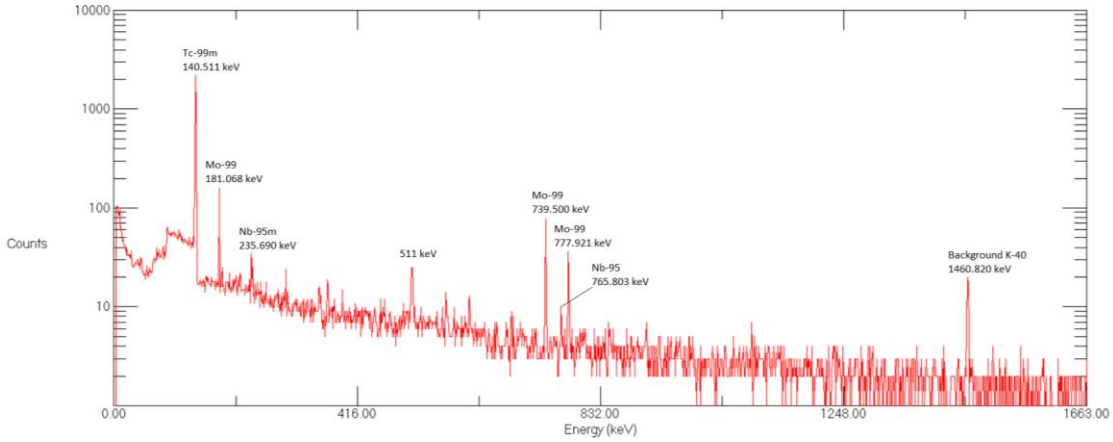
^{99m}Tc has ten times shorter half-life time ($T_{1/2}$≈6h) than ^{99}Mo. The MoO3 mass was right at the resolution of the scale used.



Picture 5– Spectrum of the dissolved target no. 6 after sublimation, measured in 10800s (live time)

II.3. Wnioski:

Together with scientists I still work on the data obtained and we will try to publish the results. The preliminary results definitely show that there is a significant enhancement of the specific activity under the experimental conditions used. However, the preliminary results do not indicate that it will be possible to use it in the event of a temporary reduction of production in reactors. This possibility requires much for work to show that it is feasible.

……	…….
(miejscowość i data)	(podpis doktoranta Interdyscyplinarnych Studiów Doktoranckich RadFarm)
…….	…….
(miejscowość i data)	(podpis Opiekuna Naukowego)
……	…….
(miejscowość i data)	(podpis Kierownika Projektu Interdyscyplinarnych Studiów Doktoranckich RadFarm)

Załącznik 1 – Porozumienie o organizacji stażu.

Projekt realizowany w ramach Programu Operacyjnego Wiedza Edukacja Rozwój 2014-2020 współfinansowanego ze środków Europejskiego Funduszu Społecznego

Accelerator-based method of ^{99}Mo production
The first foreign intership of a RadFarm PhD student

Tobiasz Zawistowski MSc, a student of the RadFarm interdisciplinary doctorate studies, was an intern at CANADIAN ISOTOPE INNOVATIONS Corp. (CII) (http://isotopeinnovations.com/) and carried out experiments at Canadian Light Source (CLS) (https://www.lightsource.ca/).

„Mr. Mark de Jong - Chief Technology Officer at Canadian Isotope Innovations Corp. was the supervisor of my internship" Tobiasz Zawistowski MSc says. „I met Mark in May 2019 during the 2nd Research Coordination Meeting on *"New Ways of Producing 99mTc and 99mTc Generators"* coordinated by POLATOM and organized in NCBJ. He mentioned that his team had been researching the accelerator-based method of ^{99}Mo production for past several years."

Molybdenum-99, which undergoes nuclear decay to technetium-99m, is the main isotope used in nuclear medicine. It is obtained primarily in nuclear reactors, such as the Polish research reactor MARIA at National Center for Nuclear Research (NCBJ) in Otwock-Świerk. However, the method of producing this isotope based on nuclear reactors has significant limitations, so scientists are looking for new ways to efficiently produce molybdenum-99, for example using accelerators. This issue is also being explored by a PhD student, Tobiasz Zawistowski MSc, who is building an experimental station at NCBJ for producing the ^{99}Mo with electron beam from a linear accelerator. "The nuclear reaction leading to molybdenum-99 production is crucial in my doctoral study. The Molybdenum-100 nucleus emits a neutron when a high-energy photon hits the target" says the PhD student. "The photon beam necessary for this nuclear reaction is produced as a result of inhibition of accelerated electrons, on the atomic nuclei of a conversion target made of e.g. tantalum or tungsten."

The internship, supported by RadFarm, gave the Polish PhD student the opportunity to participate in an experiment carried out at CLS as part of the IAEA project. NECSA researchers from South Africa (http://www.necsa.co.za/) were also involved in it. The PhD student will use the experience gained in conducting the actual molybdenum-99 production process while preparing his experimental stage of research at NCBJ. "In the experiment carried out at CLS, irradiation of the target containing molybdenum-100 took place in a linear electron accelerator with 35 MeV beam energy" explains Tobiasz Zawistowski MSc. "NCBJ is currently building an accelerator with 30 MeV beam Energy which is very similar to that available at CLS Laboratory. Mainly with the help of this Polish device I intend to perform an experimental part of my PhD thesis."

Mr. Tobiasz Zawistowski has very good memories of the time spent together with foreign colleagues: "Although the first week was marked by frost (the air temperature ranged from -20 to -35 degrees Celsius), it was really hot in the lab. Along with the Canadian scientists and colleagues from South Africa, I participated in an experiment exploring the possibility of obtaining high specific activity of molybdenum-99 in the containing material. The Canadian Light Source linear accelerator is located in the underground bunker. It is surrounded by tons of concrete, lead and polyethylene plates, protecting researchers from radiation."

The scientists are still working on the data obtained, so we have to wait a bit for the results to be published. However, the preliminary results of the research carried out by an international team indicate that the method used works and it will be possible to use it in the event of temporary reduction of production in reactors, which already took place in 2008. The accelerated molybdenum production method used on a larger scale may also be cheaper. It also generates less radioactive waste.

RadFarm project "Radiopharmaceuticals for molecularly targeted diagnostics and medical therapy", Agreement No. POWR.03.02.00-00-I009 / 17-00 under the Operational Program Knowledge Education Development 2014-2020 co-financed by the European Social Fund, is implemented in a consortium of four units, two research institutes, NCBJ and IChTJ, and two universities, UW and WUM.

The supervisors of Tobiasz Zawistowski's PhD thesis (being prepared at NCBJ) are Sławomir Wronka, PhD, DSc (prof. NCBJ), Renata Mikołajczak, BEng, PhD, DSc (prof. NCBJ) and dr Izabela Cieszykowska.