Neutron Activation Analysis in the environmental monitoring, homeland security and medicine

Michał Silarski

UJ Particle Physics Phenomenology and Experiments Seminar, Kraków, 04.04.2022

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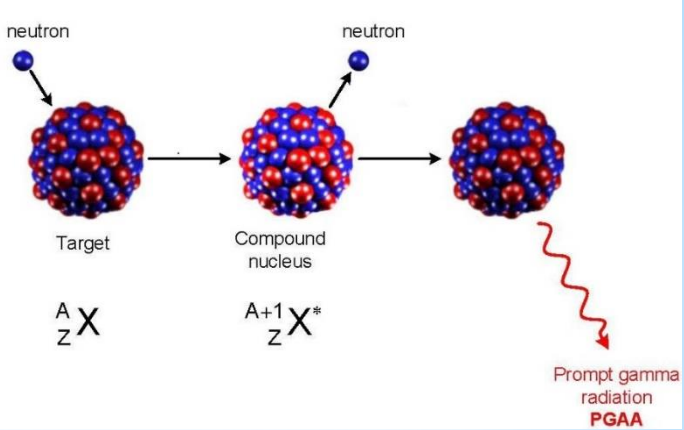
Introduction

❖ Neutrons: particles with great application potential

- ✓ highly penetrating (nondestructive probes)
- ✓ „sensitive” to light atoms



❖ Some applications:

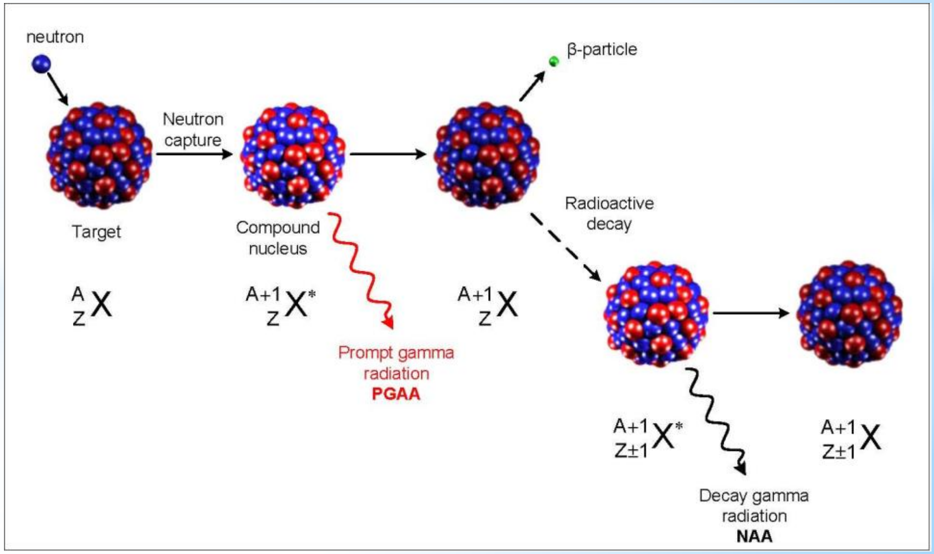
- ✓ Neutron radiography
- ✓ **Oncology (neutron therapy, breast cancer detector)**
- ✓ **Counter-terrorism/IED detection**

photograph

neutron

X-ray

http://mnrcdev2.ucdavis.edu/radiography.html

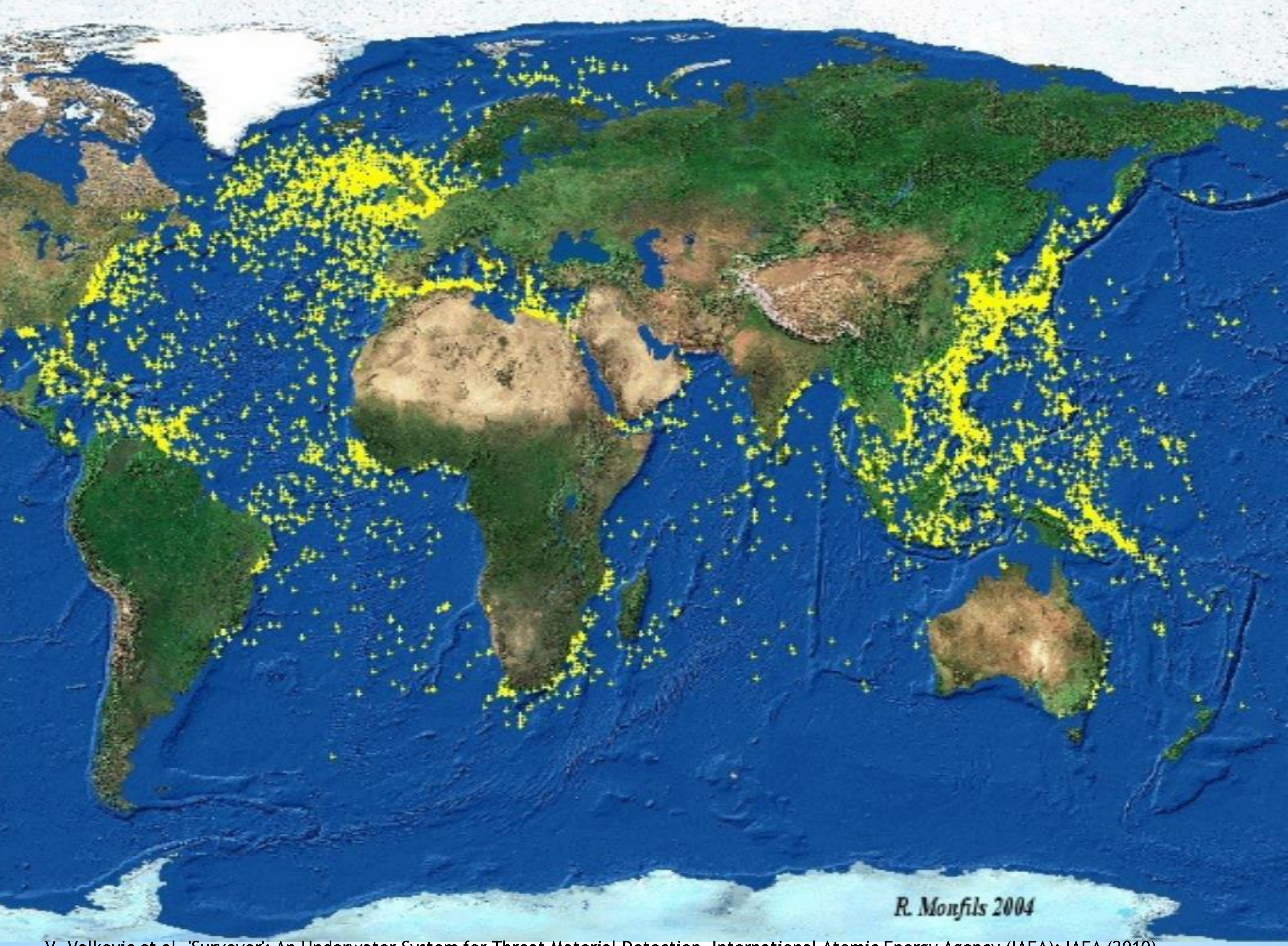


https://nmi3.eu/neutron-research/techniques-for-/chemical-analysis.html

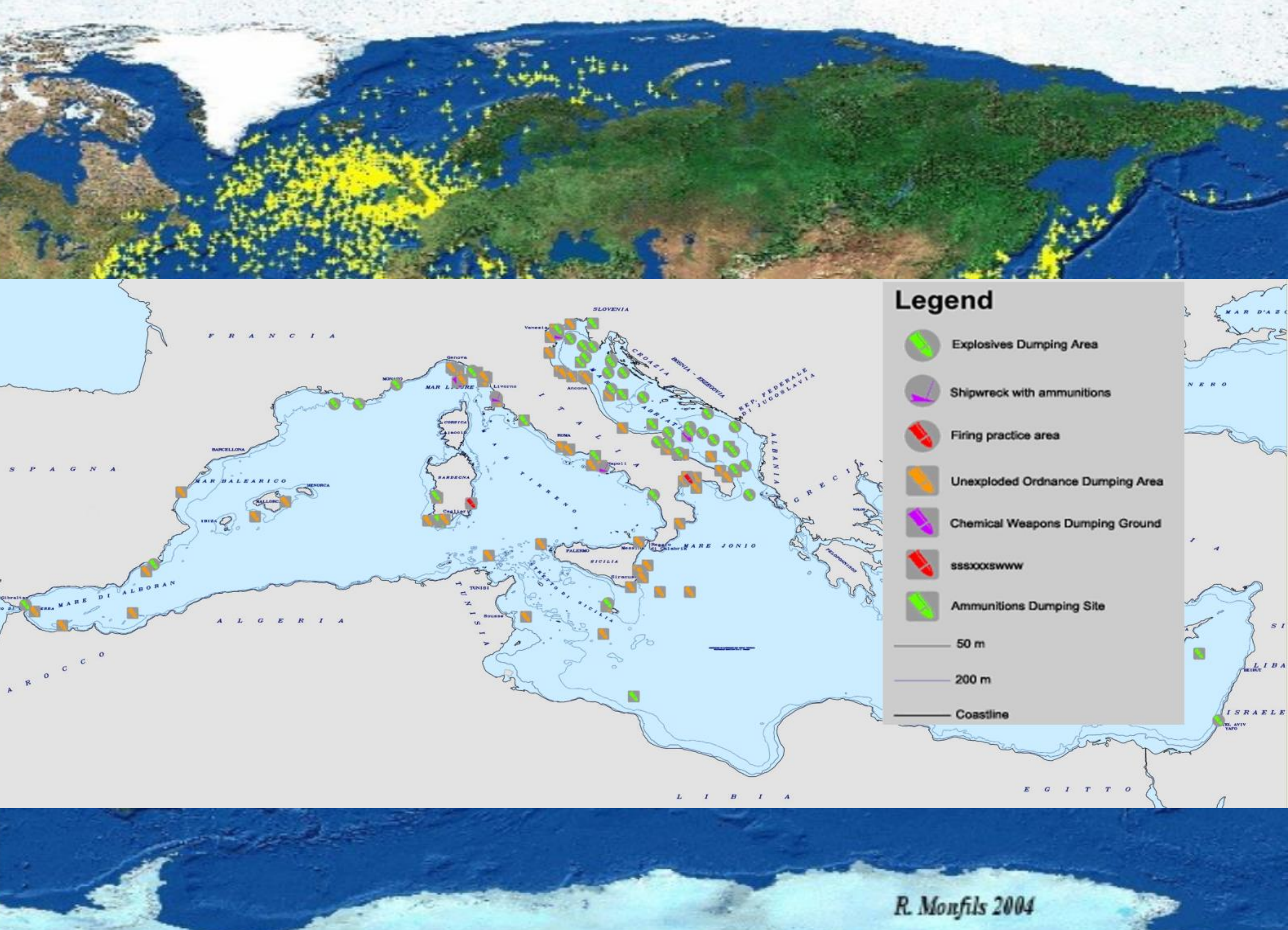
*PART I: Underwater non-invasive threads detection

* Motivation

Unknown amount of sunken chemical weapons is scattered across the Baltic Sea… and not only!



V. Valkovic et al. 'Surveyor': An Underwater System for Threat Material Detection. International Atomic Energy Agency (IAEA): IAEA (2010).



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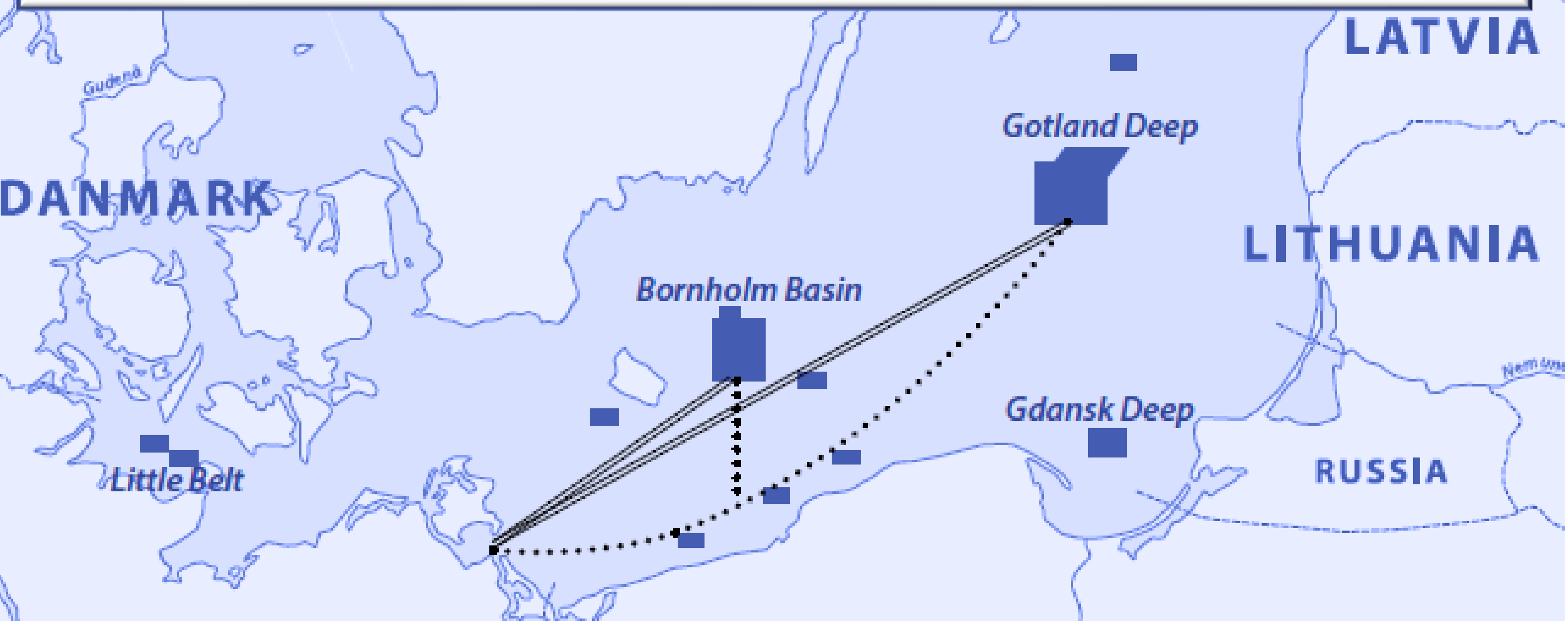
* Motivation

- ❖ **Menace to merchant navy**
- ❖ **Serious threat for people and environment**
  („Fake amber” on the coast, mustard gas „fished” out of the sea)

- ❖ **Clearing the sea bottom due to the construction of Nord Stream gas pipeline : 100 million euro**



Motivation

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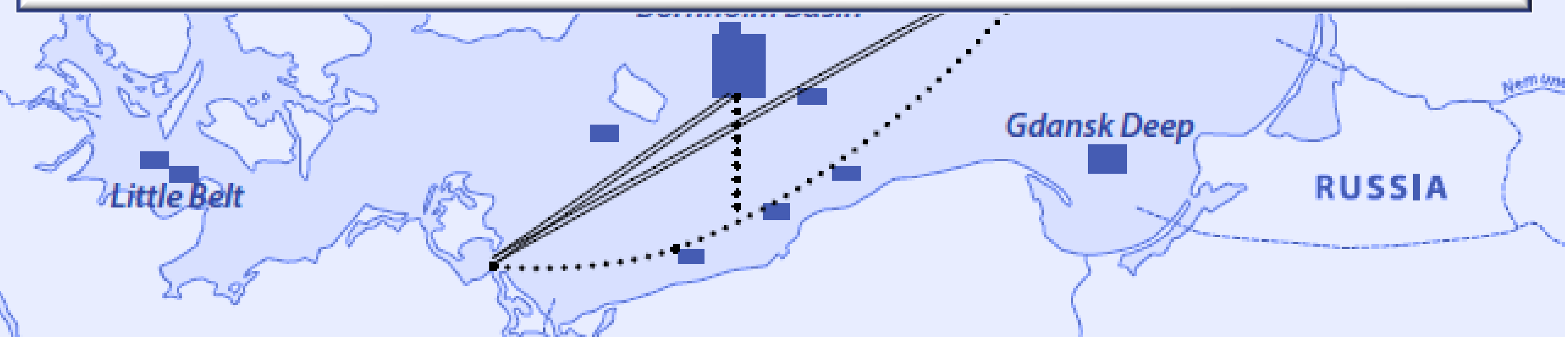
Detection methods for underwater hazardous materials: sonars / robots

- Recognition of shapes and density of objects ("chemically blind" methods)
- They usually require confirmation by a qualified sapper



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Detection methods for underwater hazardous materials: sonars / robots

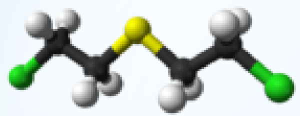
- ❖ Recognition of shapes and density of objects ("chemically blind" methods)
- ❖ They usually require confirmation by a qualified sapper
- ❖ Expensive, inefficient and slow

Possible alternative/improvment: Neutron Activation Techniques

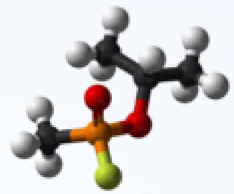
* Motivation

❖ Main agents to deal with:

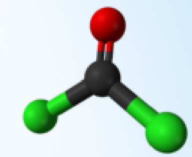
Mustard gas ($C_4H_8Cl_2S$)



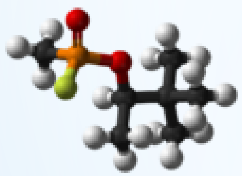
Sarin ($C_4H_{10}FO_2P$)



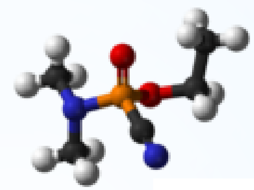
Fosgen ($COCl_2$)



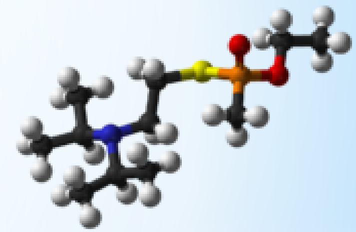
Soman ($C_7H_{16}FO_2P$)

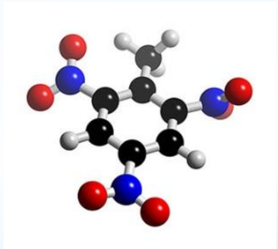


Tabun ($C_5H_{11}N_2O_2P$)



VX ($C_{11}H_{26}NO_2PS$)





TNT ($C_7H_5N_3O_6$)

❖ **High economic and environmental costs have been preventing so far any activities aiming at extraction of these hazardous substances.**

* Neutron Activation Techniques

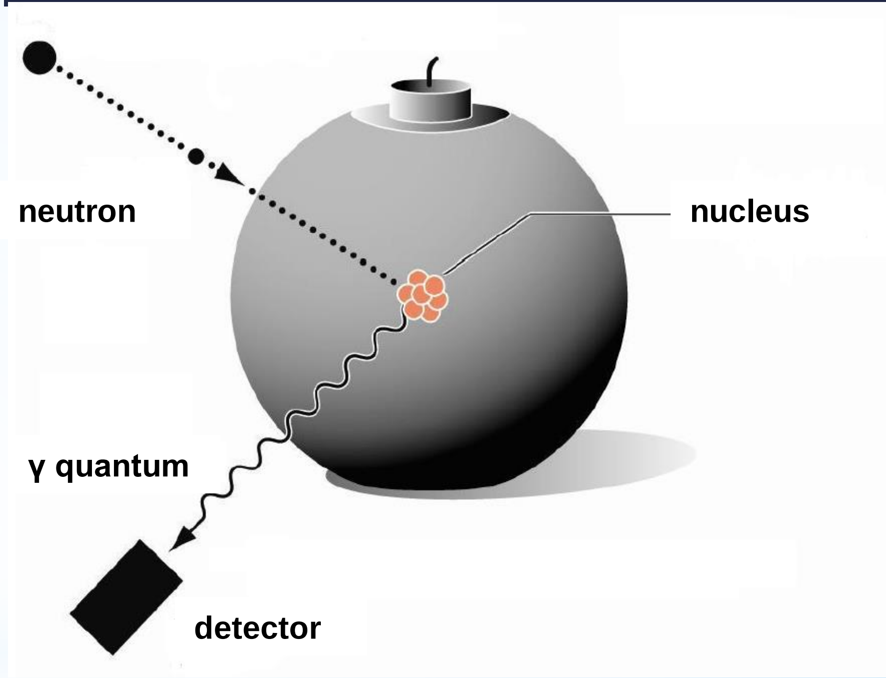
❖ Novel methods of **nondestructive** chemical threat detection based on neutron activation:

Thermal neutron capture (sources, D+Dgenerators)

Neutron inelastic scattering (D+D/**D+T** generator)

D+T → α + n

n + nucleus → nucleus + γ + n



Excited nuclei emit gamma quanta of energy characteristic of the element

Relative content of elements ⇔ Stoichiometry → **Identification**

Neutron Activation Techniques

- **Signature:**
  gamma quanta of the following nuclei: **C** (4.44 MeV), **O** (6.13 MeV), **N** (10.83 MeV), **Cl** (1.17 MeV, 7.79 MeV), **S** (2.32 MeV), **P** (1.27 MeV), **F** (0.11 MeV, 0.197 MeV)

- High penetration allows detection of explosives which are hidden in vehicles, buried, etc.

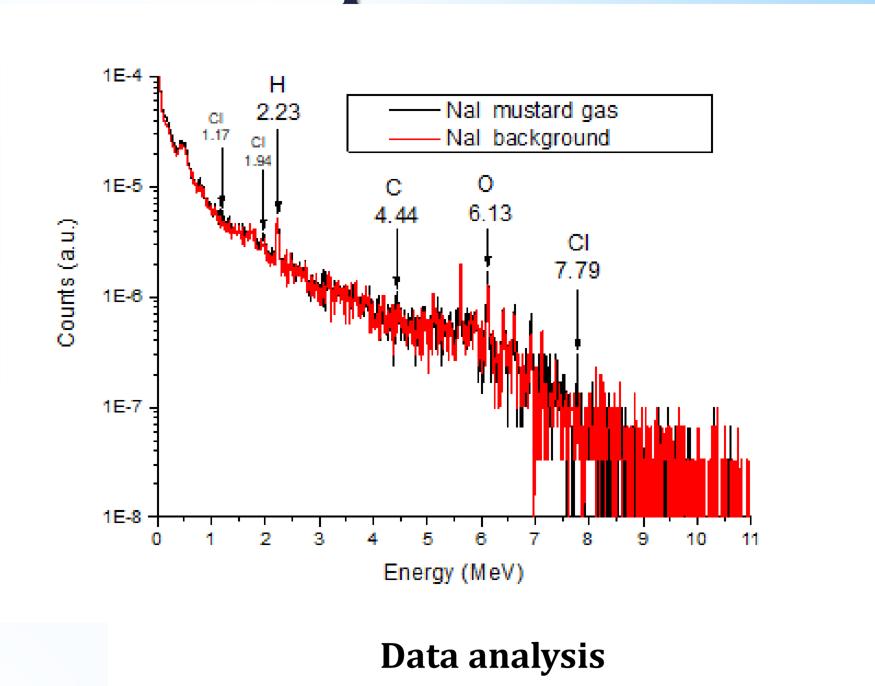
Drawbacks:

- Small cross sections for some of the elements
- Decreased mobility due to detector cooling
- High neutron flux needed
- Insensitivity to the structure of molecule
- **High neutron attenuation in water**
- **High background from oxygen and hydrogen**

P. Saska, Szybkobieżne Pojazdy Gąsienicowe (24) nr 1, 5 (2009)

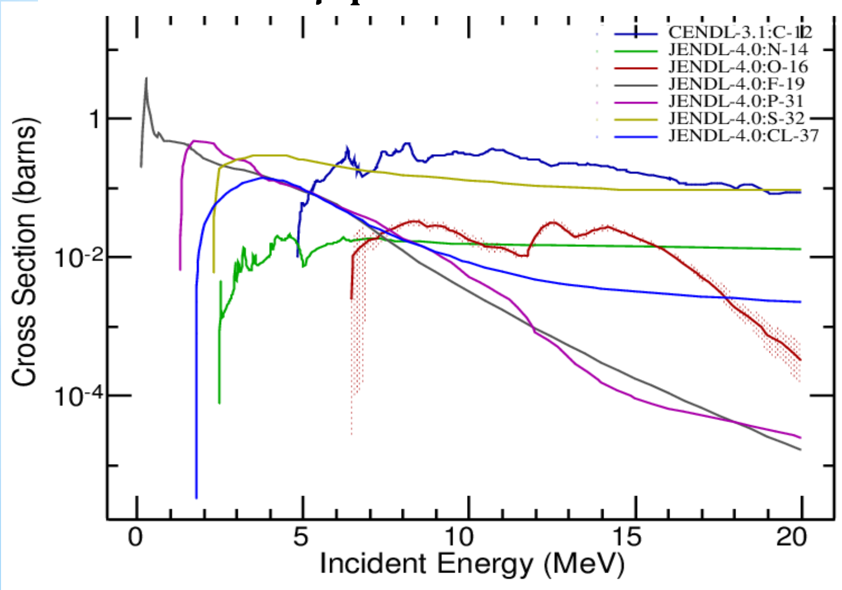
Neutron Activation Techniques

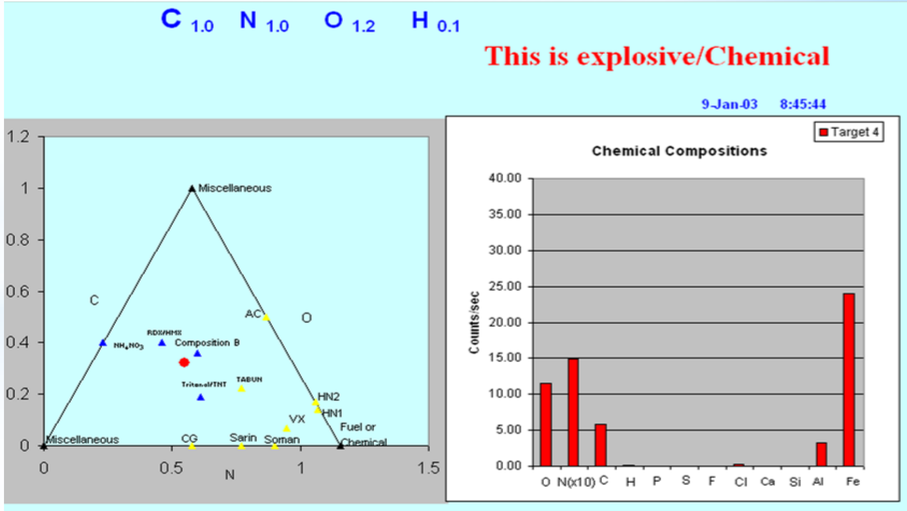
http://www.calseco.com/



Data analysis

γ quanta detection





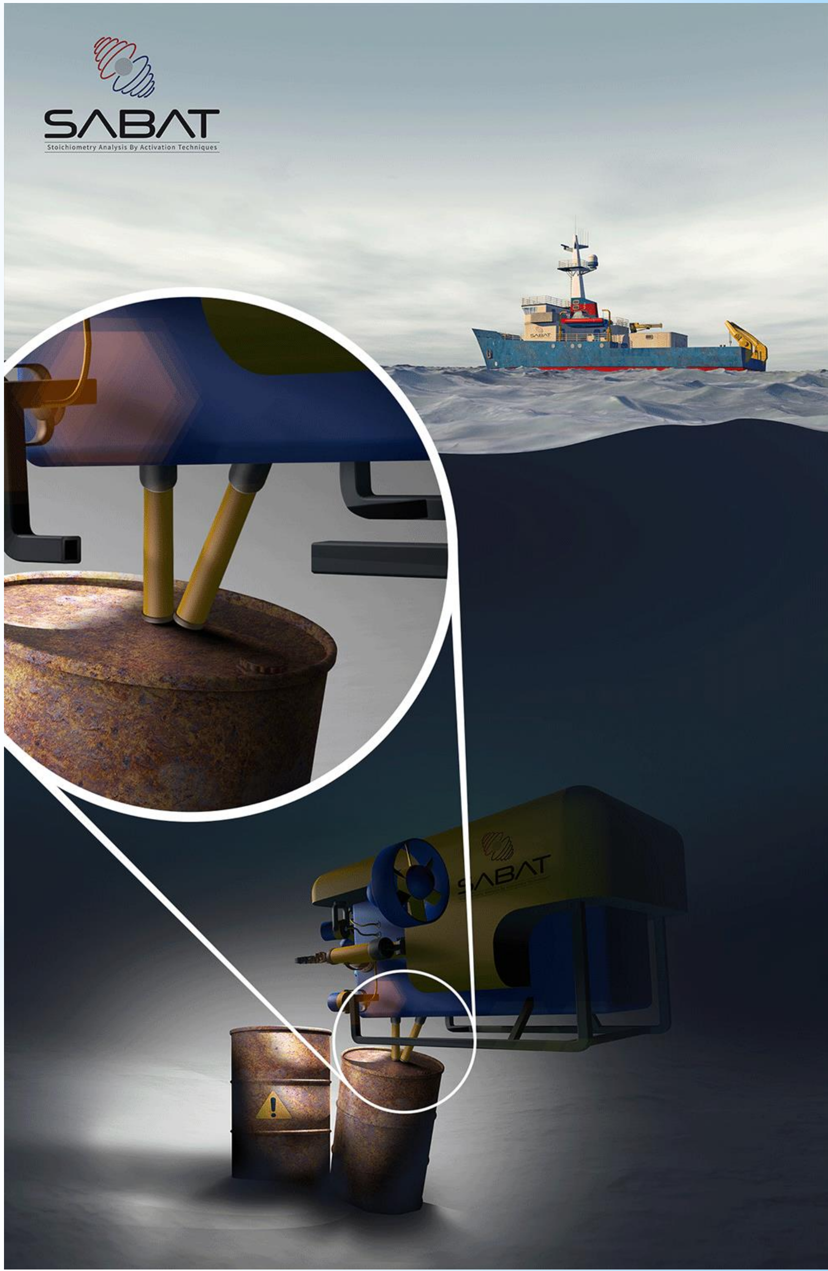
http://www.calseco.com/

Comparison with database of known substances & identification

The SABAT project (Stochiometry Analysis By Activation Techniques)

- The 14.1 MeV neutron generator with α particle detection
- Neutron and γ quanta attenuation in water minimized by guides filled with air or some other gas
- Pulsed generator & correlated α particles detection ⇔ tomographical picture of the chemical composition
- Changeable position, length and orientation of guides
- Position sensitive detector (**scintillator**)

(M. Silarski, P. Moskal, Patent PL 223751; EP 15738491.8;US 15/509,013)

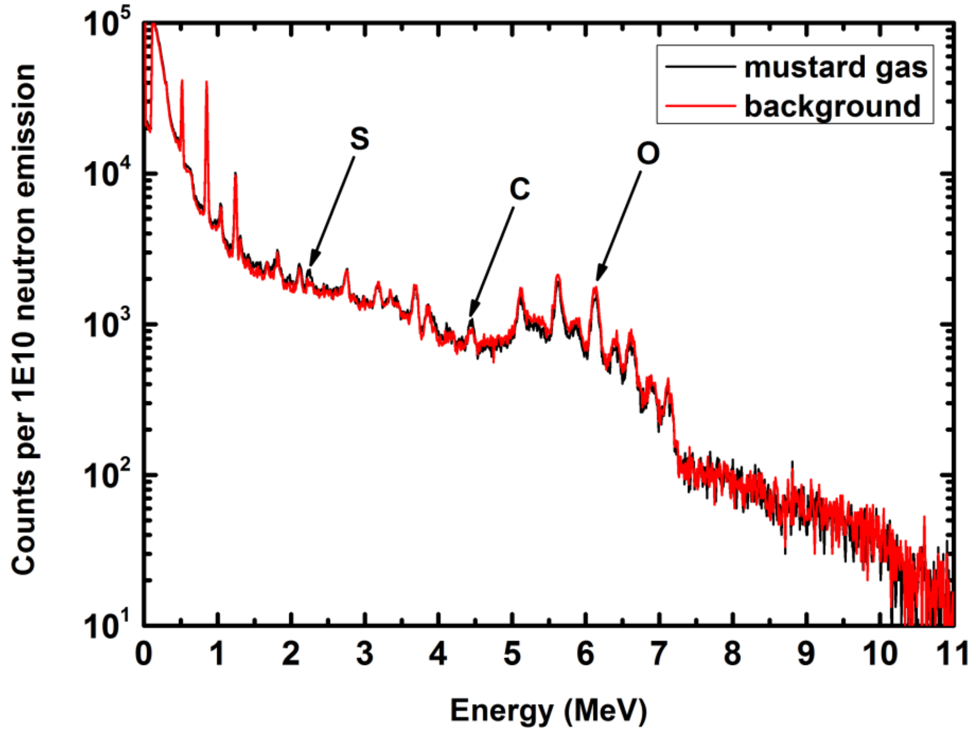


The SABAT project (Stochiometry Analysis By Activation Techniques)

- MCNP simulations:
  - pulsed D-T generator (Thermo Scientific P385)
  - 2” x 2” $LaBr_3$:Ce detector
  - mustard gas container: 100 x 100 x 40 cm^3
  - measrement time: 10 s

- Separation of the neutron capture γ quanta allows for identification

- Neutron scattering (γ quanata) may enable tomografic image reconstruction



Projekt SABAT (Stochiometry Analysis By Activation Techniques)

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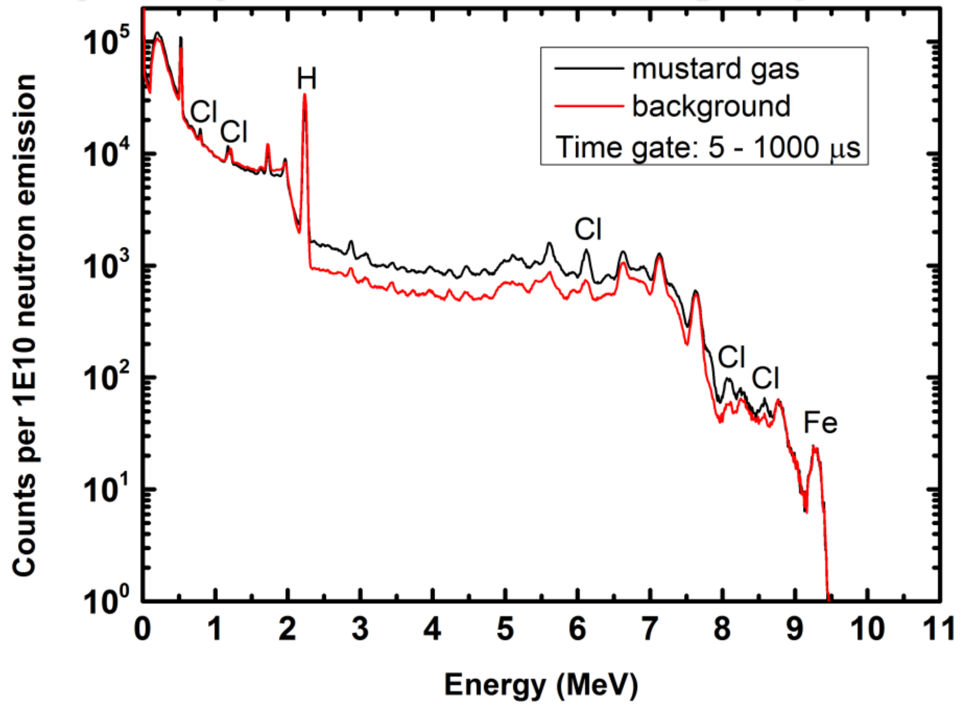


Elemental ratio	Signal	Background
Non-elastic scattering		
S(2.23)/O(6.13)	0.36 ± 0.04	0.22 ± 0.04
Cl(2.12)/O(6.13)	0.45 ± 0.04	0.29 ± 0.02
C(4.44)/O(6.13)	0.31 ± 0.03	0.19 ± 0.04
Neutron capture		
Cl(6.13)/H(2.23)	0.053 ± 0.003	0.014 ± 0.002

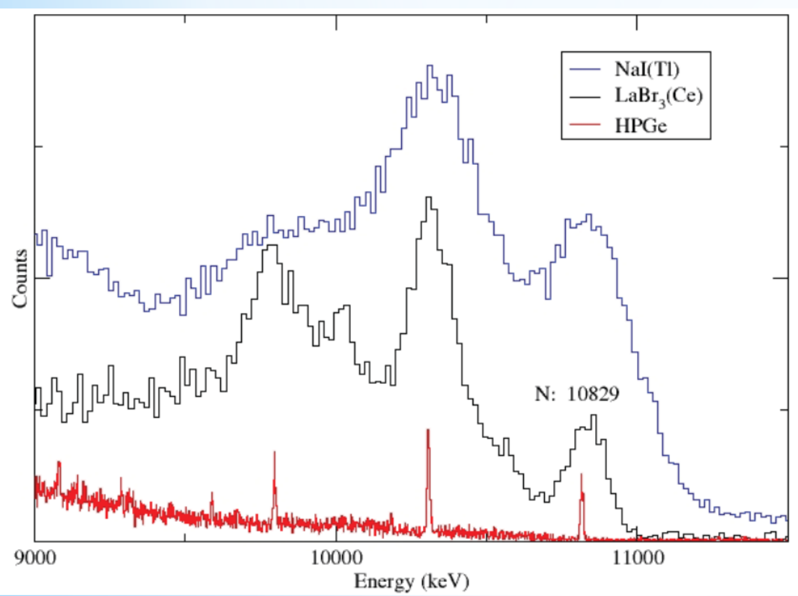
Elemental ratio	Signal	Background
TNT ($C_7H_5N_3O_6$)		
C(4.44)/O(6.13)	0.36 ± 0.04	0.133 ± 0.02
N(10.83)/Cl(2.12)	0.0112 ± 0.0028	0
CLARK I ($C_{12}H_{10}AsCl$)		
C(4.44)/O(6.13)	0.47 ± 0.05	031 ± 0.03
CLARK II ($C_{12}H_{10}AsN$)		
C(4.44)/O(6.13)	0.56 ± 0.05	031 ± 0.03

P. Sibczyński, M. Silarski et al., JINST 14 (2019) P09001--P09001

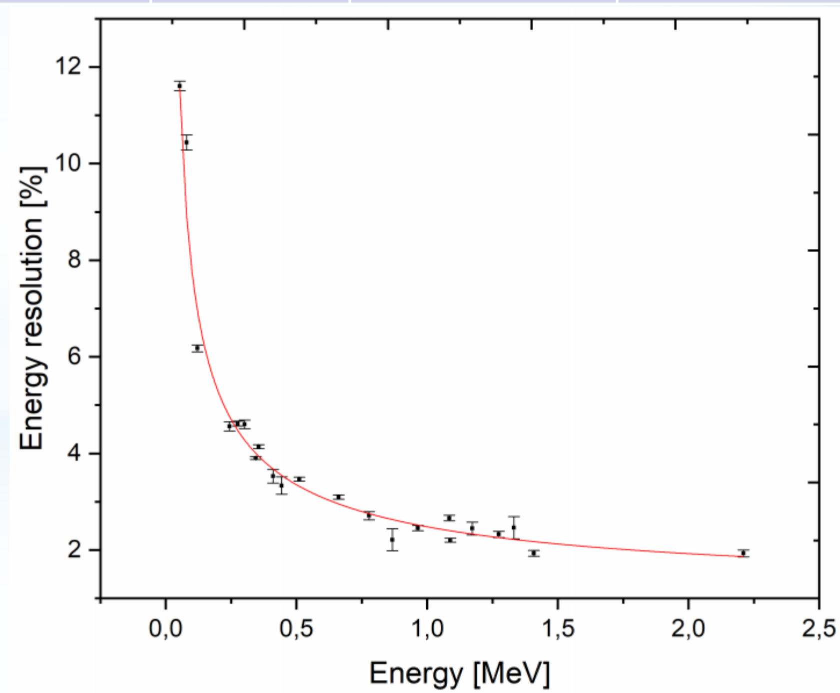
The SABAT project (Stochiometry Analysis By Activation Techniques)

- ❖ Mobility and compactness requires substitution of semiconductor detectors
- ❖ Natural candidates: inorganic scintillators

	BGO	NaI:Tl	$LaCl_3$:Ce	$LaBr_3$Ce:Sr
Average atomic number	28	**32**	28	**41**
Density [g/cm³]	7.1	**3.7**	3.9	**5.3**
Energy resolution (@662 keV) [%]	12	**7**	3.3	**2.8**



IEEE Nuclear Science Symposium,San Diego, CA,10/29/2006,11/04/2006



A. Miś, master thesis, Jagiellonian University (2020)

Status of the SABAT project

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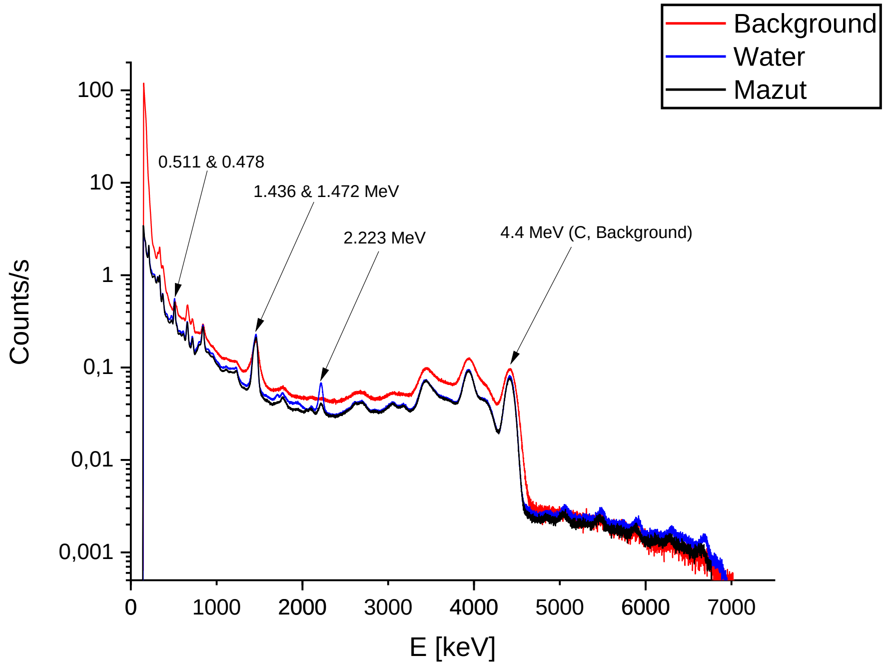
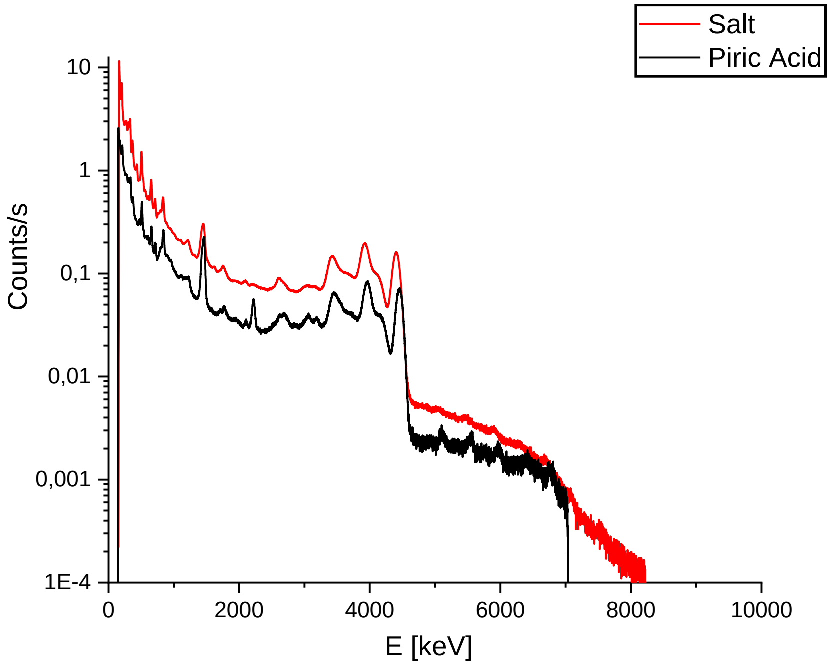
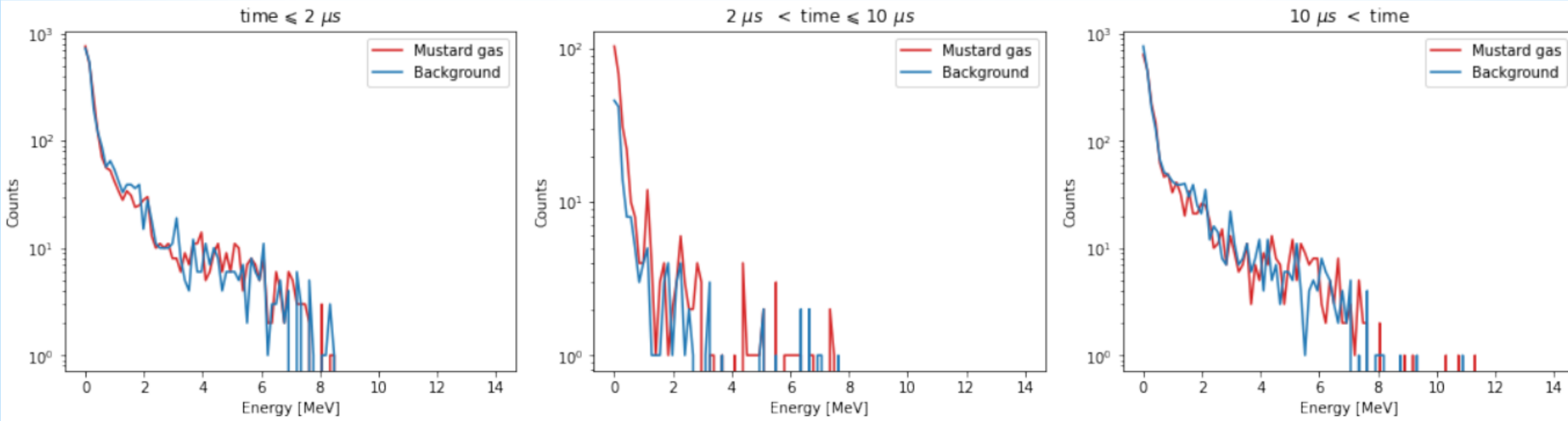




	Mazut		Water		Picric Acid ($C_6H_3N_3O_7$)	
	R	σ_R	R	σ_R	R	σ_R
H/O	15,79	0,58	68,81	0,85	61,7	1,8
C/O	305,0	8,4	416,8	5,5	452,15	9,64
H/C	5,05	0,24	14,84	0,44	18,72	0,58

Status of the SABAT project

- A first attempt to use neural networks to support the identification of an illicit material (based on Monte Carlo simulations)
- Input data: histograms of detector energy depositions for 3 time slots after the emission of a neutron:



- Feedforward Neural Network with two hidden layers with 16 and 8 neutrons, respectively (with L2 regularization)

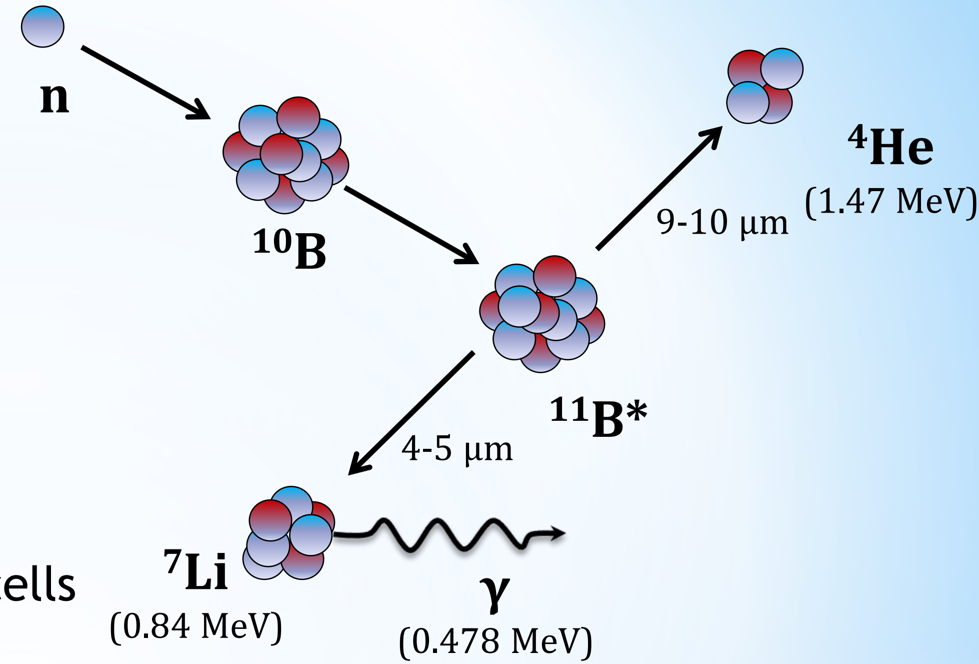
	Dataset type	Sample size	Batch size	Learning rate	Epochs	Mean accuracy	$\sigma_{Accuracy}$	Mean loss	σ_{Loss}
$LaBr_3 : Ce$	A	20000	750	0.0001	100	0.99781	0.00260	0.02456	0.01244
	B	5000	750	0.0001	100	1.00000	0.00000	0.00632	0.00322
$NaI : Tl$	A	2000	750	0.0001	100	0.99246	0.00708	0.03745	0.02450
	B	5000	750	0.0001	100	0.99993	0.00013	0.00936	0.00538
BGO	B	5000	750	0.0001	100	0.99999	0.00003	0.00929	0.00556
LSO	B	5000	1000	0.0001	100	0.99993	0.00017	0.00642	0.00383

D. Janik, Master thesis, Kraków 2020

*PART II: Boron Neutron Capture Therapy

The Boron Neutron Capture Terapy

- Therapy used against highly malignant and therapeutically resistant tumors:
  - gliobastoma multiforme
  - malignant melanoma
  - head and neck recurrent cancers
  - Malignant pleural mesothelioma
- Irradiation with (epi)thermal neutrons
- ^{10}B transfered selectively to the tumor cells
- High LET within a single cell



Nuclide	Reaction	σ [b]
^{6}Li	(n,α)	940
^{10}B	(n,α)	9835
^{155}Gd	(n,γ)	61100
^{157}Gd	(n,γ)	259000

(Wolfgang A.G. Sauerwein, A. Wittig, R. Moss, Y. Nakagawa „Neutron capture therapy”, Springer)

http://www.jsnct.jp/e/index.html; Kawasaki Medical School

The Boron Neutron Capture Terapy

❖ So far only ^{10}B and Gd were considered as „targets" for NCT

$$D_{tot} = CBE_B D_B + 3.2 D_n + 3.2 D_H + D_\gamma$$

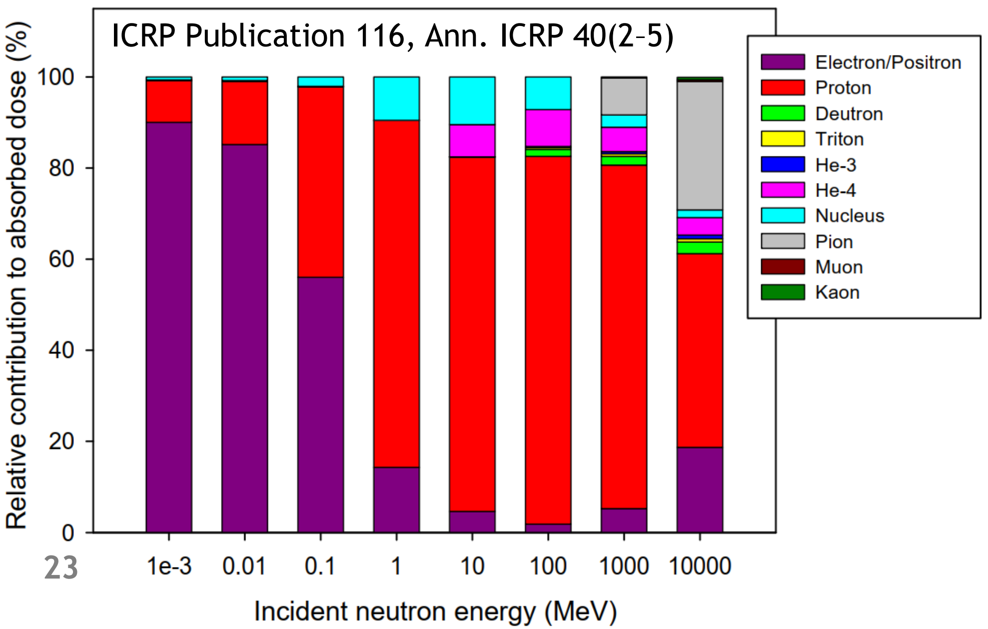
- $CBE_B D_B$ → $^{10}B(n,\alpha)^7Li$
- $3.2 D_n$ → $^{14}N(n,p)^{14}C$
- $3.2 D_H$ → p recoil
- D_γ → $^1H(n,\gamma)^2H$

CBE: Compound Biological Effectiveness

3.2: Relative Biological Effectiveness

❖ The in-air neutron beam recommendations by the International Atomic Energy Agency:

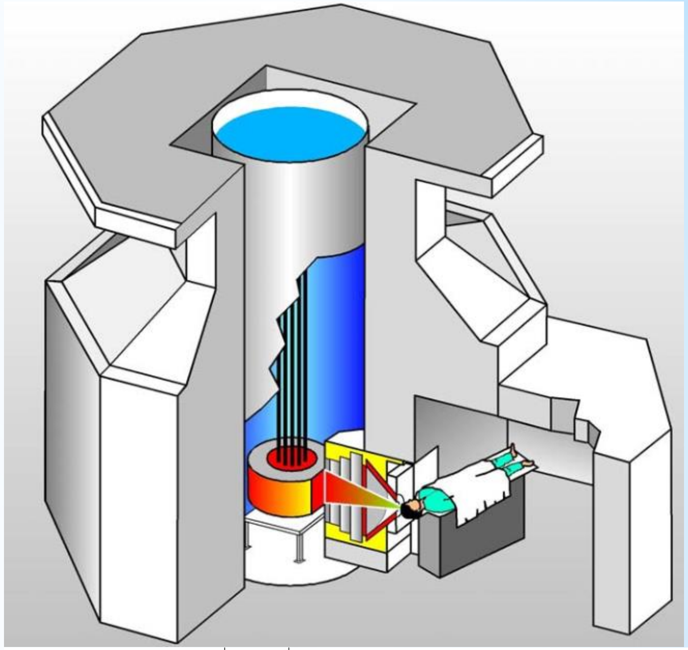
Parameter	Recommendations
Φ_{epi} [$cm^{-2}s^{-1}$]	$>10^9$
$\Phi_{epi} / \Phi_{thermal}$	> 20
Φ_{epi} / Φ_{fast}	>100
D_{fast}/Φ_{epi} [Gy cm^2]	$< 2*10^{-13}$
D_γ/Φ_{epi} [Gy cm^2]	$< 2*10^{-13}$



H. Naeem et al.,
Journal of the Korean Physical Society, 70 (2017) 816

Neutron sources for BNCT

- **Reactors**
  - High neutron flux (e.g. Maria: ~10^{14} $cm^{-2}s$)
  - Expensive and complex
  - low public acceptability
  - require complicated licensing procedures



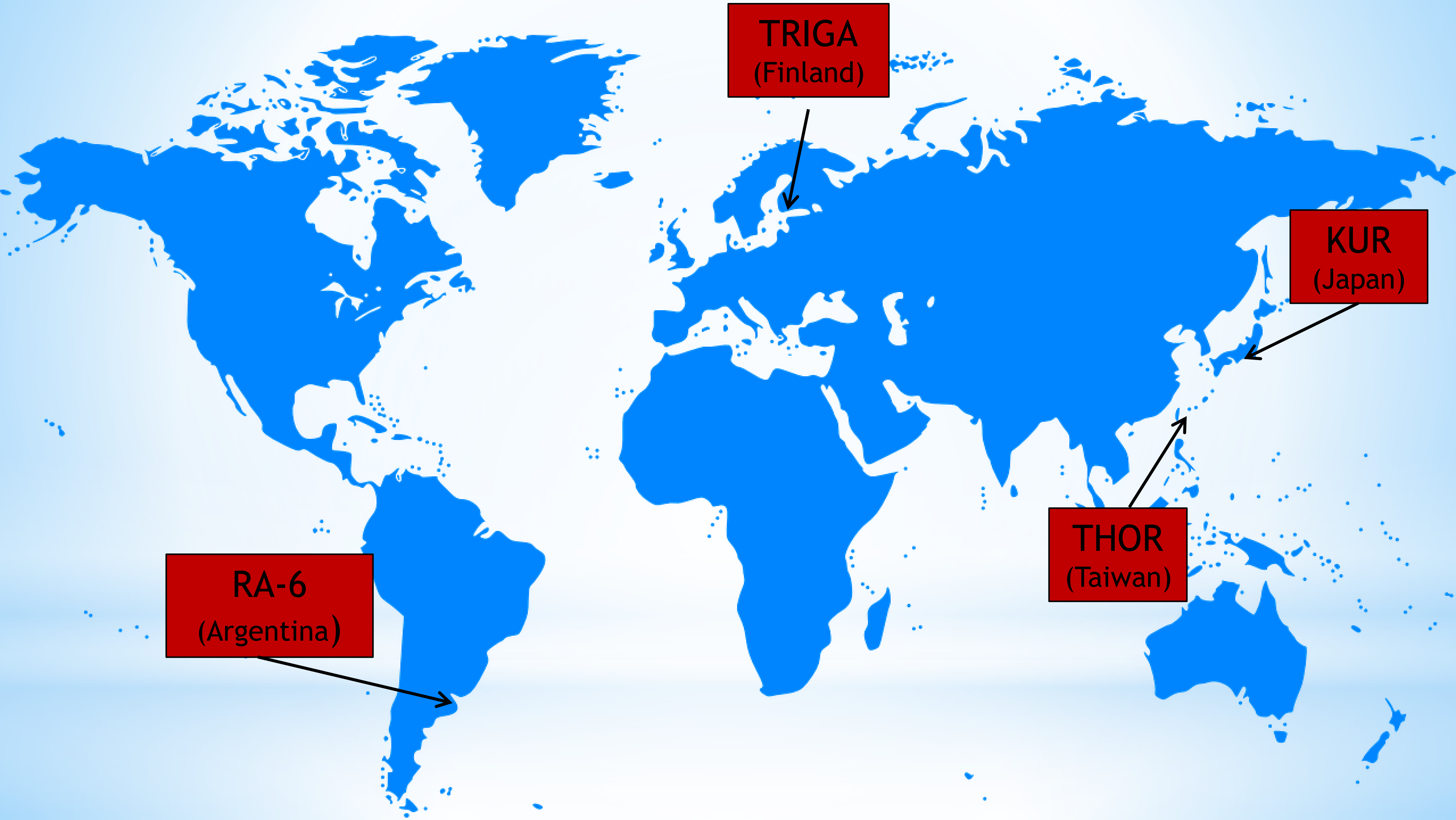
Reactor BNCT beams

core neutrons (fast→epithermal)

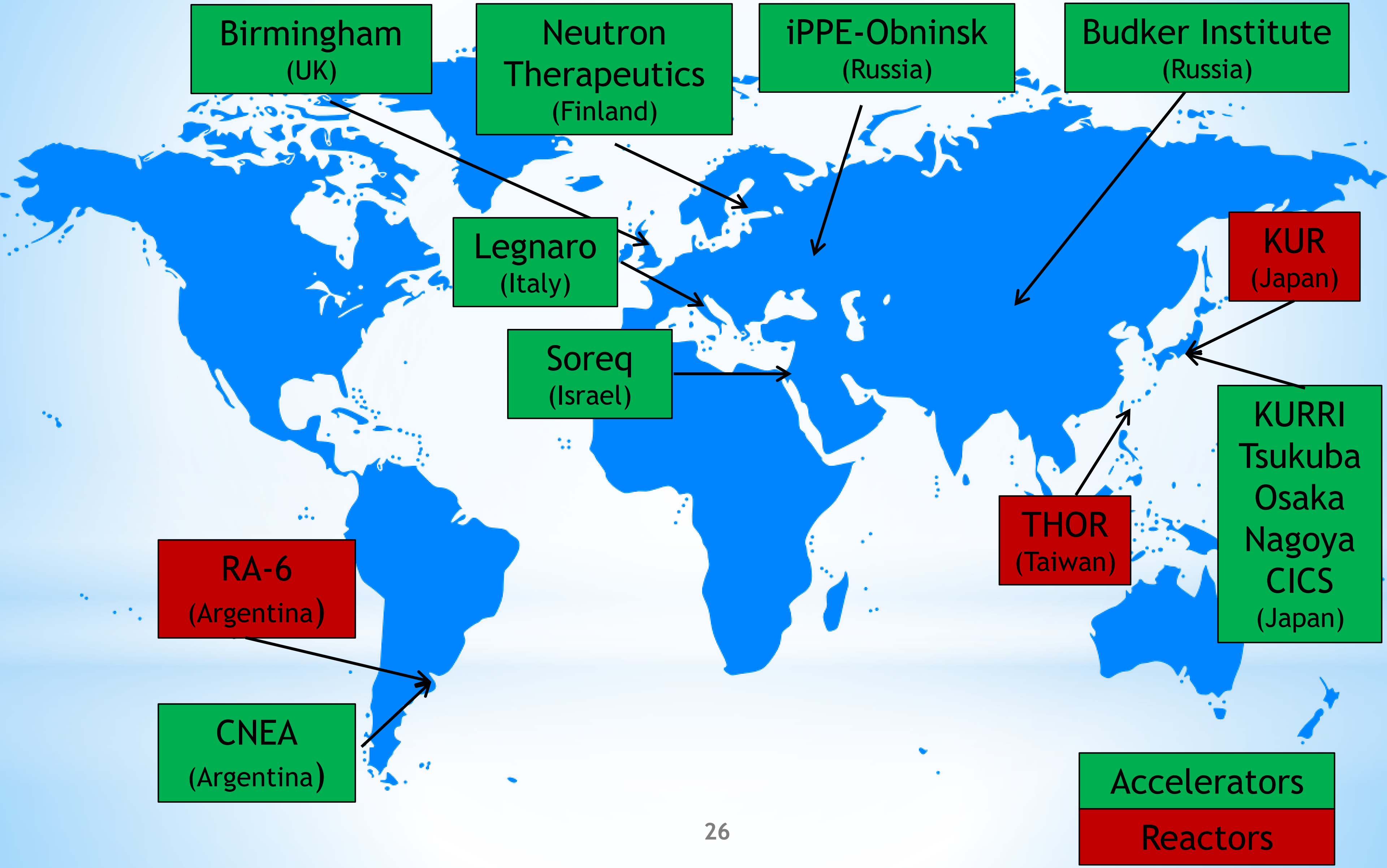
fission converter (thermal→ epithermal)

- **Accelerator sources**
  - Expensive
  - Require a lot of space
  - Most popular reactions: $^{7}Li(p,n)^{7}Be$ and $^{9}Be(p,n)^{9}B$
  - Target cooling problems for high proton intensity

BNCT facilities around the world (2016)

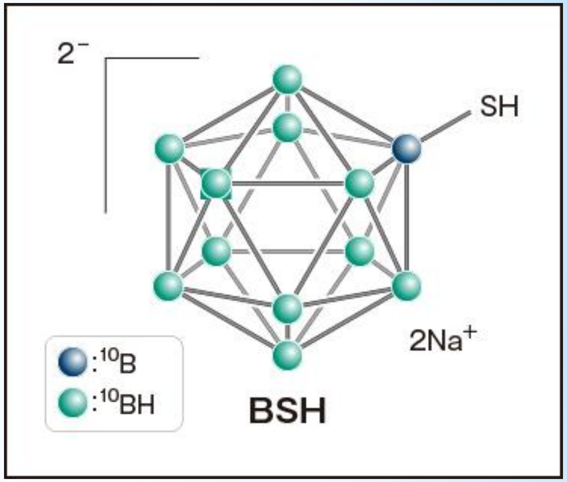


BNCT facilities around the world (2016-2020)



Boron carriers

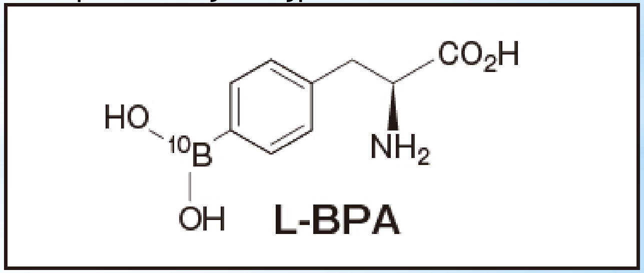
- Selective accumulation (Tumor/Normal >3) with ^{10}B concentration of 20 to 40 ppm
- Low toxicity
- Not metabolized in the tumor
- No pharmaceutical effects themselves (boron delivery molecule only)



http://www.jsnct.jp/e/about_nct/houso.html

- BSH (disodium mercaptoundecahydrododecaborate)
  - Low accumulation inside tumor cells (it stays in the intercellular spaces)

- L-BPA (L-p-Boronophenylalanine)
  - Administered combined with a water-soluble substance such as D-fructose
  - does not accumulate in slowly proliferating malignant cells

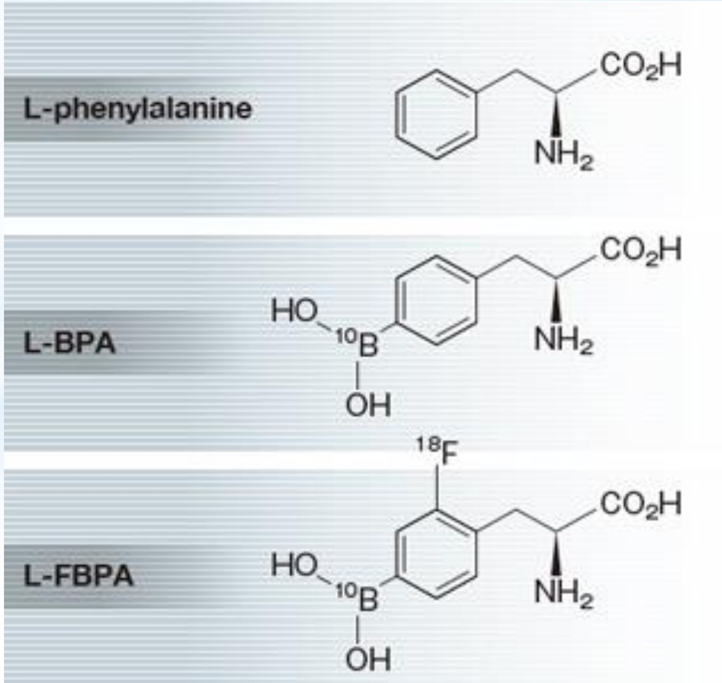


Dose and boron distribution monitoring

- **Magnetic Resonance Imaging (MRI)**
  - Non-invasive imaging of boron distribution
  - Sensitive to ^{10}B and ^{11}B isotopes

- **Activation Gamma Radiation Analysis**
  - Gamma quanta due to neutron capture on ^{10}B (E_γ = 0.478 MeV)
  - Radiation of tissue activation (H,C,N,…)
  - A fast method that allows in vivo imaging also during therapy

- **Positron Emission Tomography**
  - Boron carrier labeled with β^+ active element:
  - L-F-Boronophenylalanine [Imahori Y. et al. J Nucl Med. 39 (1998) 325]
  - ^{64}Cu-labeled BSH-3R-DOTA [Y. Iguchi et al., Biomaterials 56 (2015) 10]
  - Non-invasive imaging of boron distribution in the patient's body at each stage of therapy (resolution ~ 4-6 mm)



http://www.jsnct.jp/e/about_nct/pet.html

PGRA system developed within the Polish Consortium For the BNCT

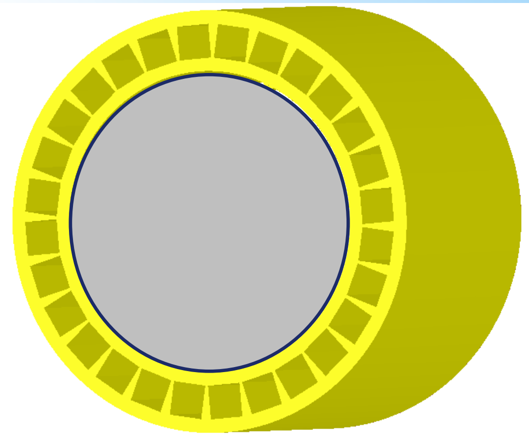
- No dose monitoring system based on PGRA exists yet

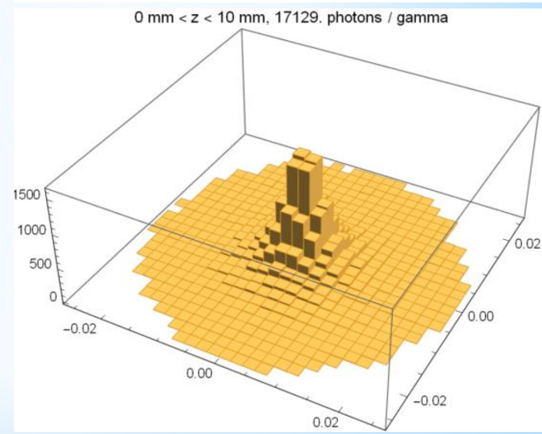
- Trials to date have included SPECT and Compton cameras optimized for the 478 keV boron capture line

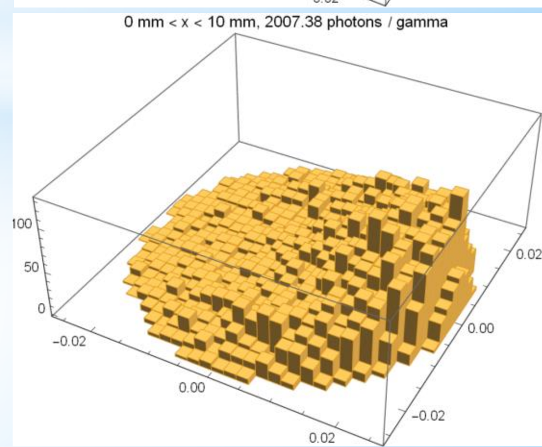
- The system based on:
  - ✓ Scintillation detectors with $LaBr_3$:Ce:Sr crystals
  - ✓ Readout by a silicon photomultiplier array (positional sensitive detector)
  - ✓ Reconstruction of the momentum direction of the registered gamma quantum
  - ✓ Active anti-Compton shields

    [M. Gierlik et al., Nuclear Instruments and Methods in Physics Research A 788 (2015) 54–58]
  - ✓ The use of detectors analogous to Compton cameras

    [M. Kim, et al., Nuclear Engineering and Technology, https://doi.org/10.1016/j.net.2020.07.010]

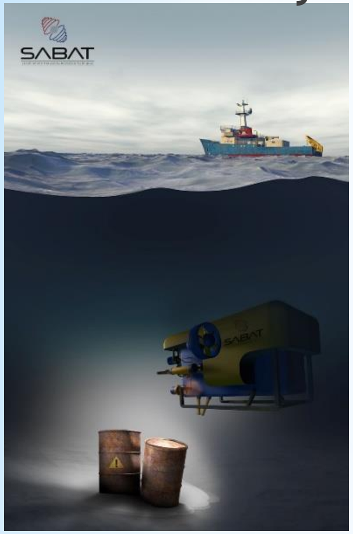




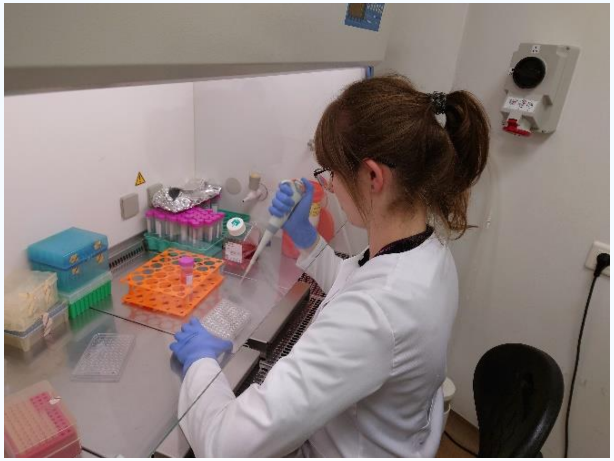


* Neutron Activation Analysis Laboratory

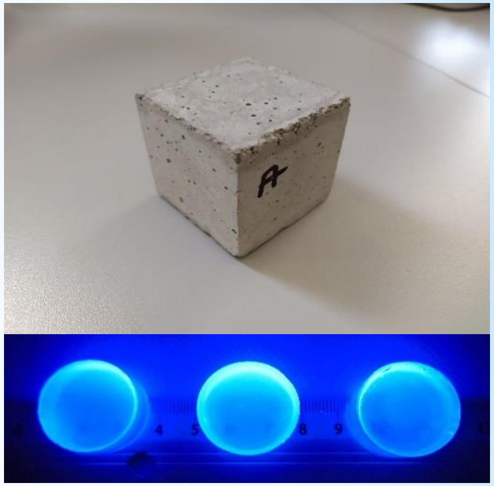
* Homeland security

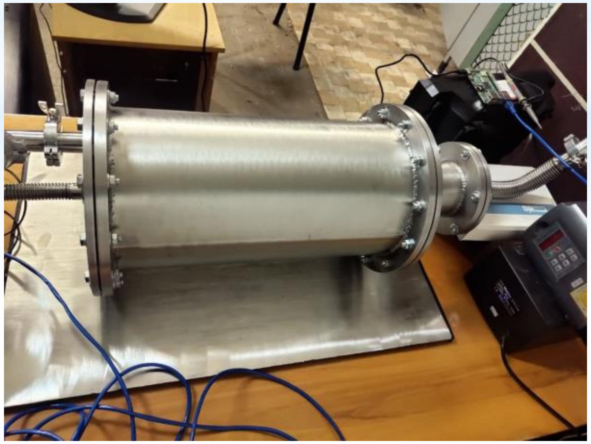


* Medicine and medical physics



* Material research





*Thank You for attention

Goya, *Witches sabbath*