Experience and Recent Developments in Nuclear Forensics at the Institute of Isotopes, Budapest

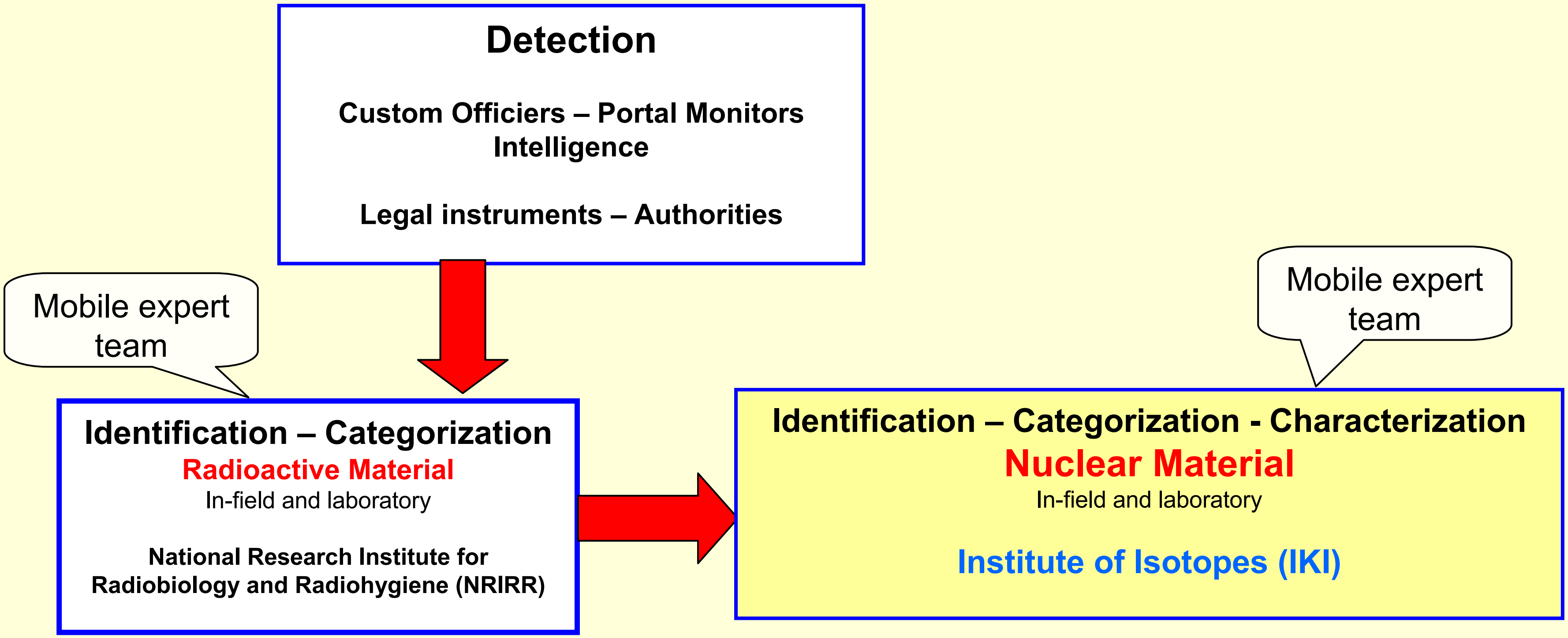
**T. Biró, J. Bagi, J. Huszti, L. Lakosi, C.T. Nguyen, Z. Révay,
Z. Stefánka, E. Széles, L. Szentmiklósi, Z. Varga, J. Zsigrai**

Institute of Isotopes, Hung. Acad. Sci., Budapest, Hungary

Outline

- Preparedness — national infrastructure
- Capabilities — forensic laboratory
- **Recent developments** — **detection**, **characterization**

Status of Hungarian preparedness



Simplified scheme of forensic investigation following the seizure of nuclear or other radioactive material

Status of Hungarian preparedness

milestones

- History of illicit trafficking (NM) 1991-

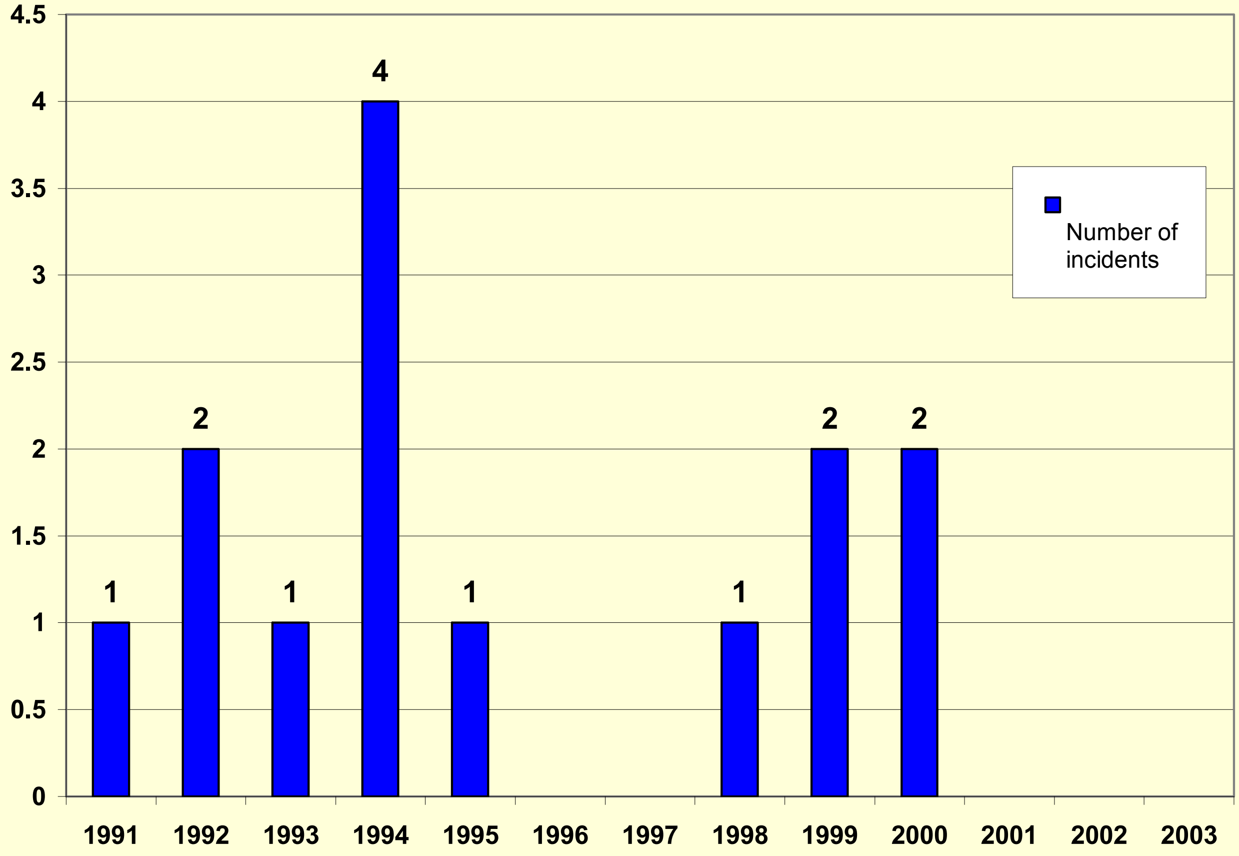
  tasks: detection-categorization-characterization
- Governmental decree: procedures, roles 1996
- Traditional technique: γ -spectrometry
- ITWG Model Action Plan: 1st simulation exercise, 2002
- Intercomparison exercise: Round Robin HEU, 2002
- Implementation of HR mass spectrometry 2005

Status of Hungarian preparedness

Techniques available

- Detection: border monitors, etc
- Response: identification (categorization)
  mobile expert teams – ensuring safety – securing source
- **Characterization**: laboratory investigation
  - γ-spec (HPGe) — U, Pu
  - Mass spec (ICP-SFMS) — U
  - Other (SEM, α-spec, etc)
  - Special techniques: neutron counting — Pu-Be
  - **Not possible**: processing Pu bearing material

Status of Hungarian preparedness



Illicit trafficking incidents in Hungary involving nuclear material

Status of Hungarian preparedness

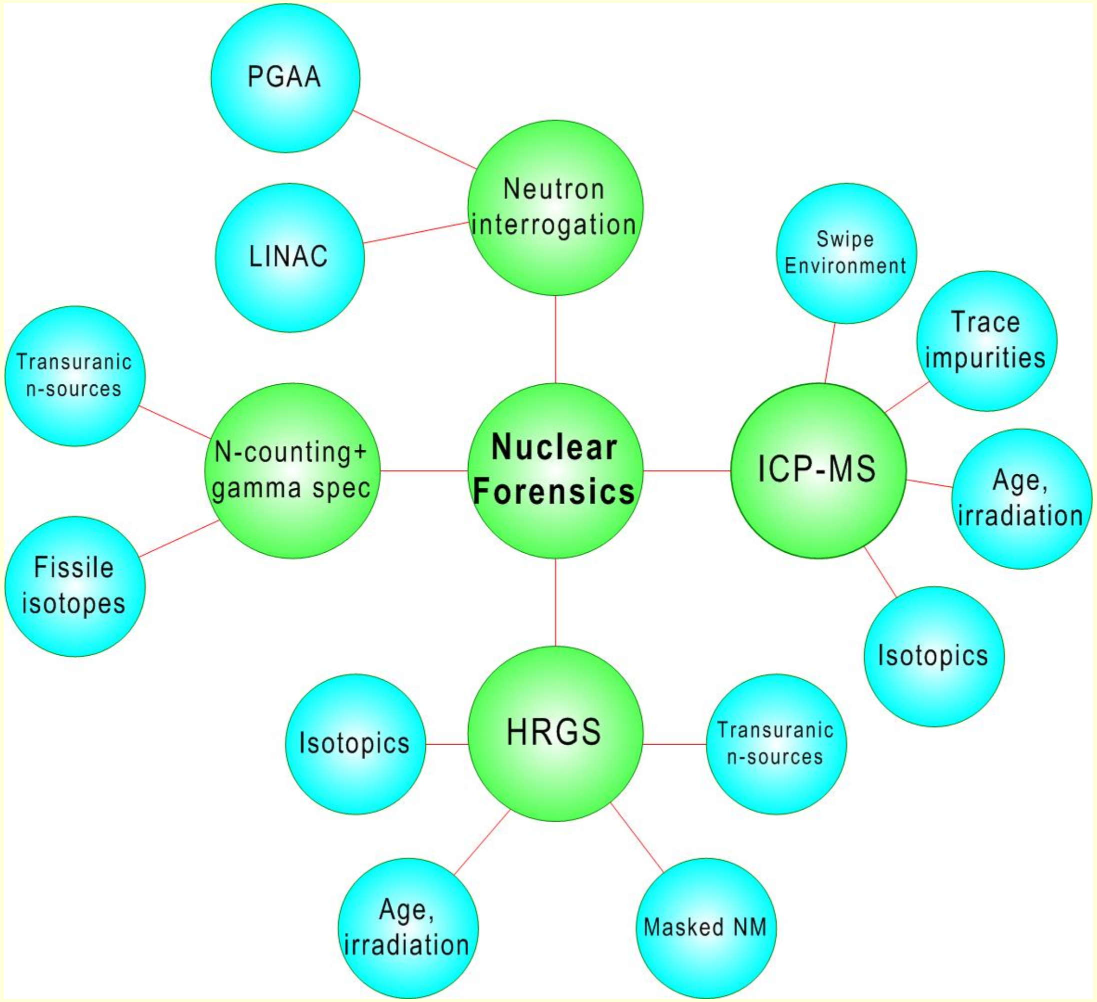
Portal monitors implemented at border crossings

Status of Hungarian preparedness

Parameter (indicator)	Information	Method (Inst. of Isotopes)
Dimension	Type of reactor, intended use	**micrometer,** measuring gauge
U content	Chemical composition	Titration, HKED, **ICP-MS**
Pu content	Chemical composition	AS, **ICP-MS**
Uranium isotopic composition	Reactor type, intended use	TIMS, MC-ICP-MS , SIMS **ICP-MS, LA-ICP-MS, HRGS**
Production date	Date of fabrication	AS, TIMS, MC-ICP-MS **ICP-MS, LA-ICP-MS**
Impurities	Production process, production facility	**ICP-MS,** ICP-OS. AAS, XRF
Microstructure	Fabrication method	**SEM,** TEM
Oxygen isotopics	Geographical location	GC-MS

Methods applied for characterization of nuclear materials

Status of Hungarian preparedness



Nuclear forensics at Institute of Isotopes

Capabilities for routine identification, categorization and characterization of NM

U bearing materials

- In-field categorization by **HRGS** (HPGe)
- Characterization
  - **γ-spectrometry:** isotopics, matrix, amount, age ($^{214}Bi/^{234}U$), presence of reprocessed U (^{232}U) using **HPGe** and low-background shielding
  - **mass-spectrometry:** isotopics, age ($^{230}Th/^{234}U$), presence of reprocessed U (^{236}U) and chemical impurities using **ICP-SFMS** and **LA-ICP-SFMS**
  - Scanning electron microscopy (with EDX probe)

Routine capabilities - facilities

Low background iron chamber

ICP-SFMS with LA unit (UP213)

Pu bearing materials

- In-field categorization by HRGS
- Characterization of sealed sources (Pu-Be) by neutron (coincidence) counting and HRGS

Note: non-sealed material cannot be handled in the laboratory because lack of safe facilities

Validation

- **Comparisons** of results obtained by gamma- and mass-spectrometry
- Participating in **inter-laboratory comparisons** (IRMM, ITWG, IAEA)
- **Joint analysis** of selected uranium-oxide pellets with JRC-ITU

Example 1

HRGS (IKI)	ICP-SFMS (IKI)	RR
1978 ± 3 y	August, 1979 (± 6 months)	Feb-July 1979

Production date (HEU sample from ITWG, Round Robin exercise)

Validation

Example 2

Joint analysis with Institute for Transuranium Elements (JRC-ITU)

	MC-ICP-MS	TIMS	IDMS	HRGS	HRGS	LA-ICP-SFMS
	ITU	ITU	ITU	ITU	IKI	IKI
^{232}U					3.2(9) E-8	
^{234}U	0.0346 (5)	0.0347 (21)	0.0345 (33)	**0.025(20)**	0.0362 (24)	0.0358 (9)
^{235}U	2.5136(14)	2.5121 (14)	2.5119 (30)	2.51 (12)	2.562 (34)	2.529 (19)
^{236}U	0.451 (22)	0.470 (44)	0.470 (86)	-	**0.380 (24)**	0.474 (24)
^{238}U	97.000(21)	96.9823(20)	96.9829(12)	97.47(12)	97.021 (34)	96.961(20)

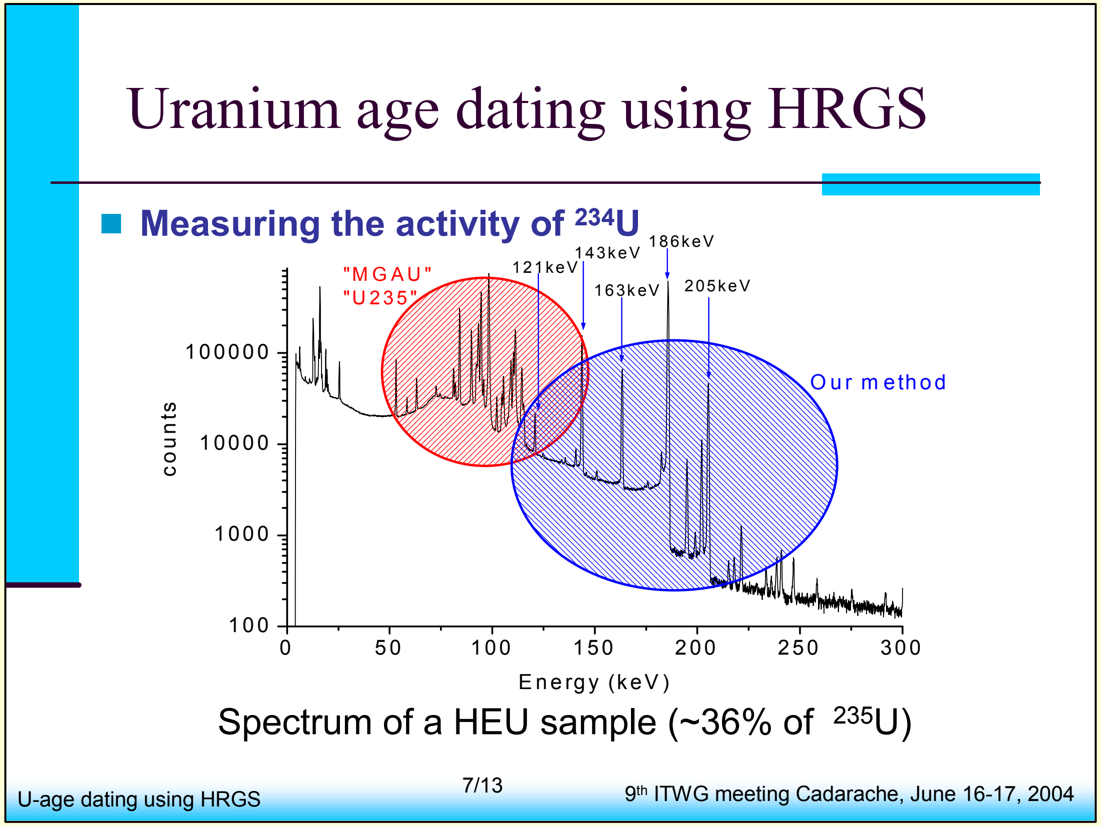
Isotopic composition of an LEU pellet analyzed (w% ± 2s)

Recent and current developments (selected)

- **Age (production date) determination by gamma- and mass spectrometry**
  - **gamma-spectrometry**: measurement of ^{234}U and ^{214}Bi
  - **mass-spectrometry**: measurement of ^{234}U and ^{230}Th
- **Mass spectrometry: environmental (swipe) samples, particle analysis (laser ablation)**
- **Pulsed neutron sources (detection of shielded (hidden) nuclear material)**
  - **4 MeV linear accelerator** (LINAC)
  - **Prompt gamma activation analysis** (PGAA)

Recent and current developments

Age: gamma-spectrometry: measurement of ^{234}U and ^{214}Bi



^{214}Bi / ^{234}U

^{234}U 121 keV planar LEGe

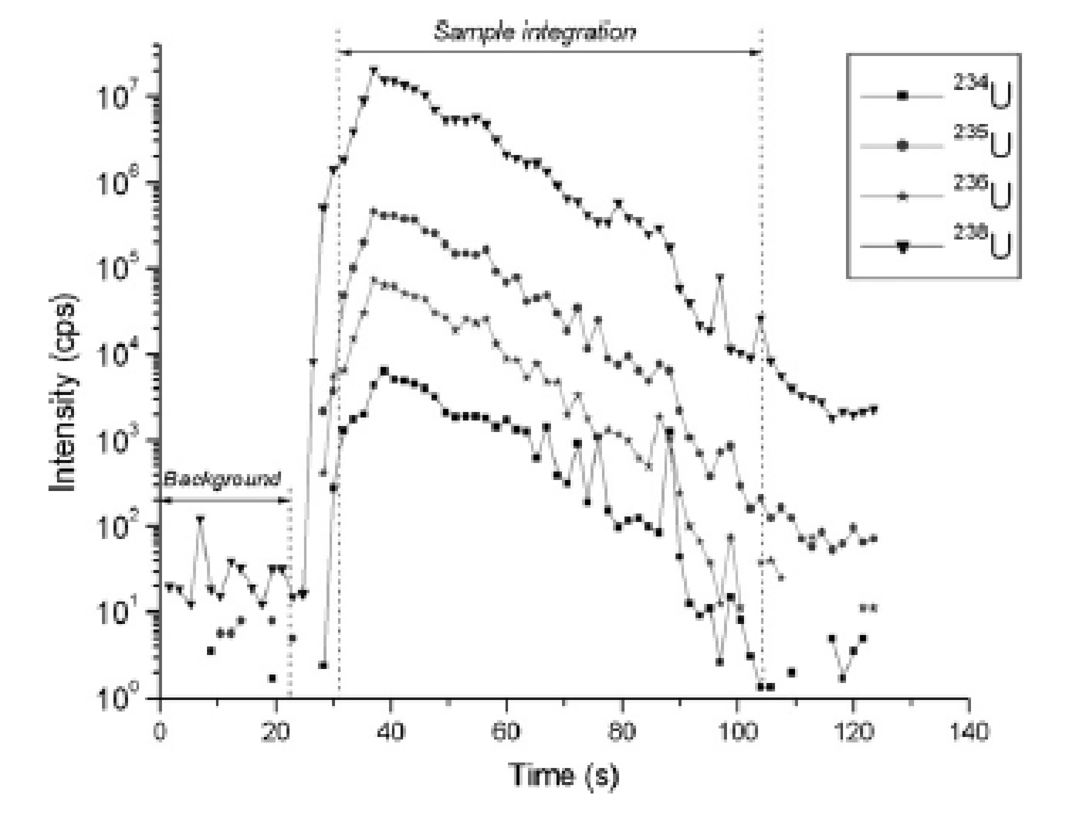
^{214}Bi 609 keV coax HPGe

via $^{234}U/^{235}U$ and $^{234}U/^{238}U$

intrinsic calibration

Group of gamma emissions used in U age dating vs. the group used by MGAU code (status in 2004, new version 4.0 MGAU)

Recent and current developments
particle analysis (laser ablation)



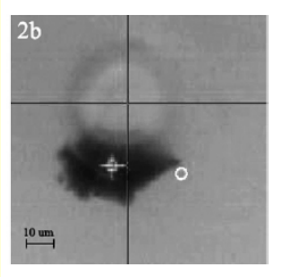
UP-213 (Nd:YAG 213 nm)

Beam diam: 20 µm

Energy 50%

Abs. detection limit: pg-fg

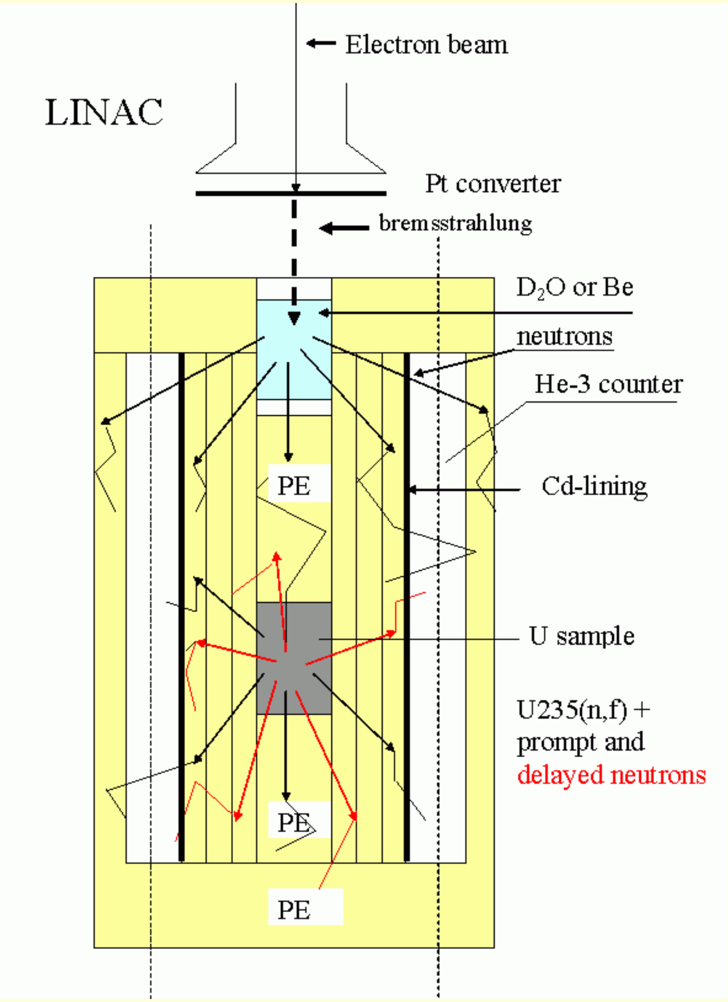
Down to 10 µm (1 µm?)



Laser ablation signals of a LEU particle (21 x 25 µm) using soft and partial ablation

Recent and current developments

Pulsed neutrons: 4 MeV linear accelerator (LINAC)



Experimental setup of LINAC neutron source and detector system

LEU

^{235}U: 0.5 g **^{238}U: 30 g**

20 sec (500 cycles)

100 g D_2O 2 μA (mean)

HEU

^{235}U: 10 mg

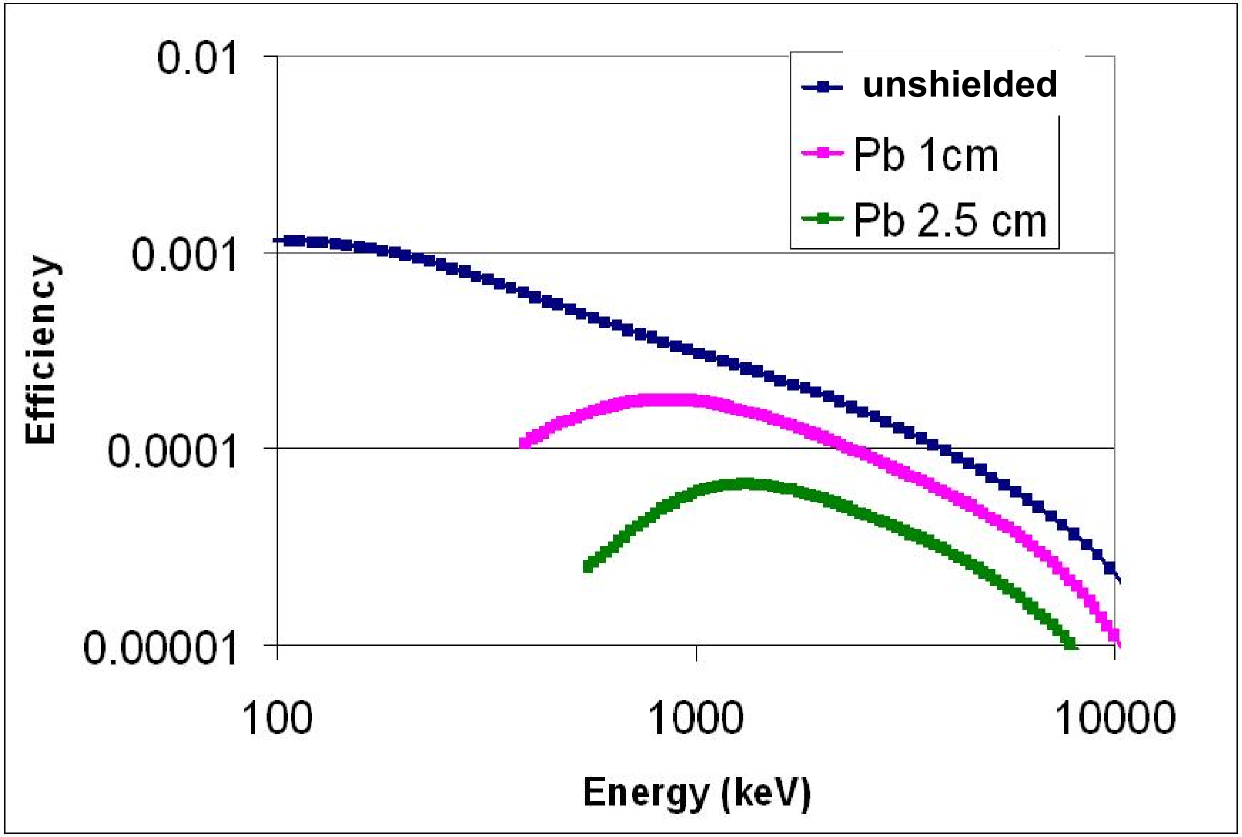
20 sec (500 cycles)

100 g D_2O 10 μA (mean)

Prompt gamma activation analysis (PGAA)

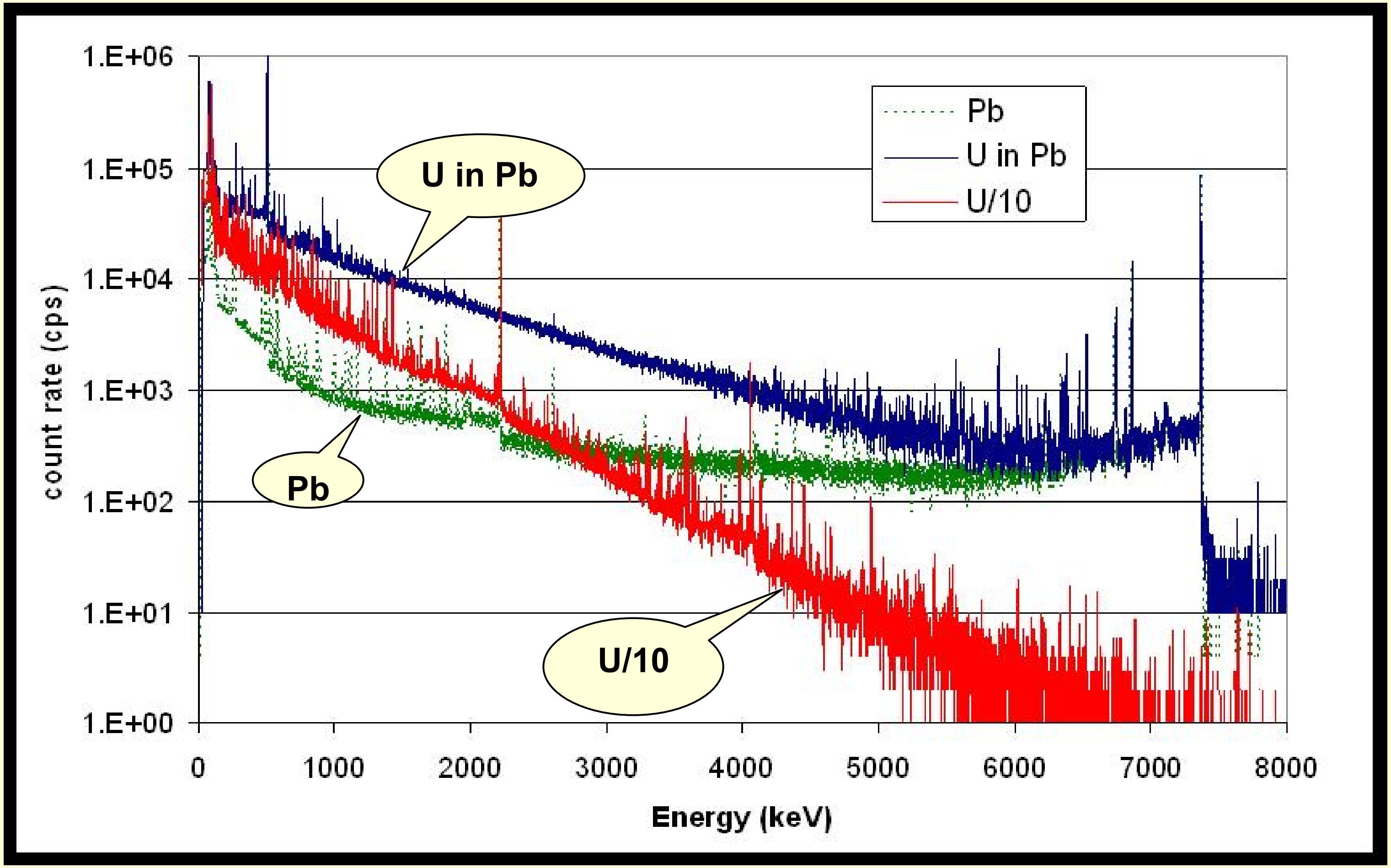
- Budapest Neutron Center (WWR-S research reactor) thermal neutrons are cooled to about 5 meV energy
- The neutrons induce prompt and delayed gamma emission in the target
- Due to the penetrating ability of the neutrons and the transmission of high energy (typically 4-6 MeV) prompt gamma rays uranium can be detected and identified even inside a thick lead container
- PGAA technique has been combined with neutron tomography: **PGAI-NT**

Recent and current developments: PGAA



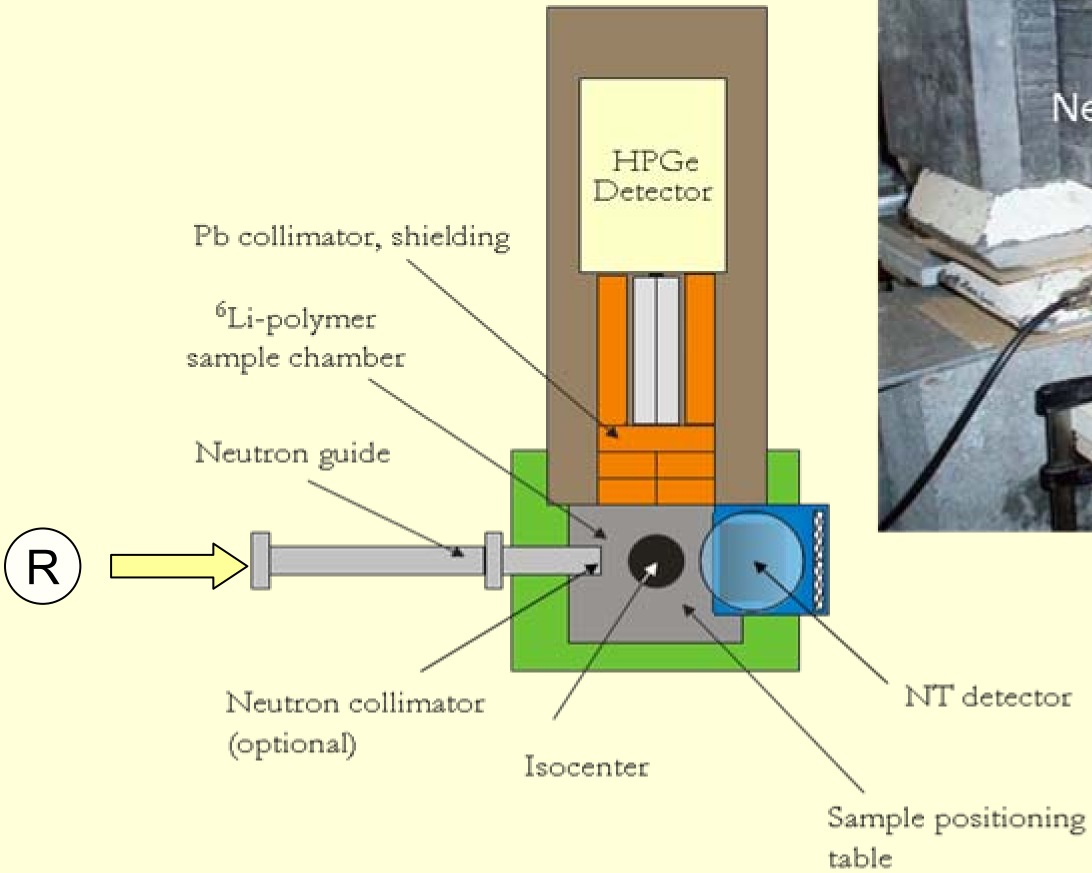
Detector efficiencies at different lead thicknesses depending on γ-energy

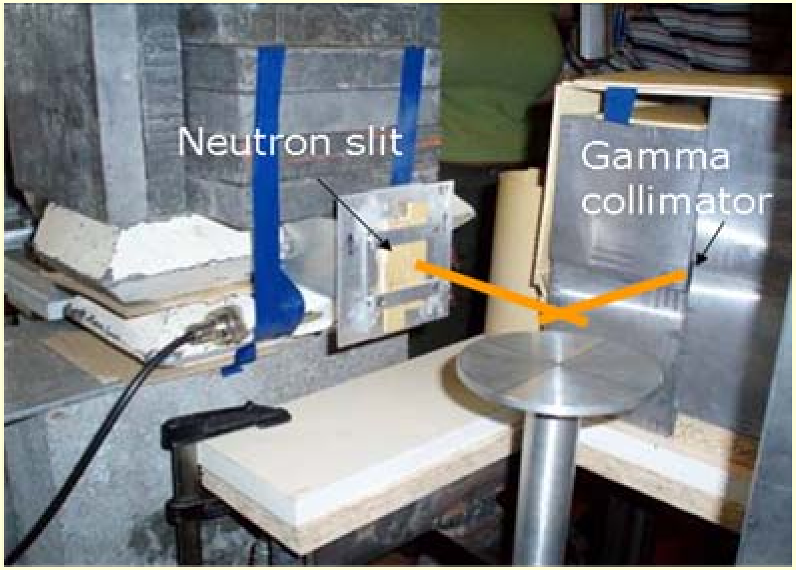
Recent and current developments: PGAA



Prompt gamma spectra of lead and uranium in lead shielding

Recent and current developments: PGAI-NT



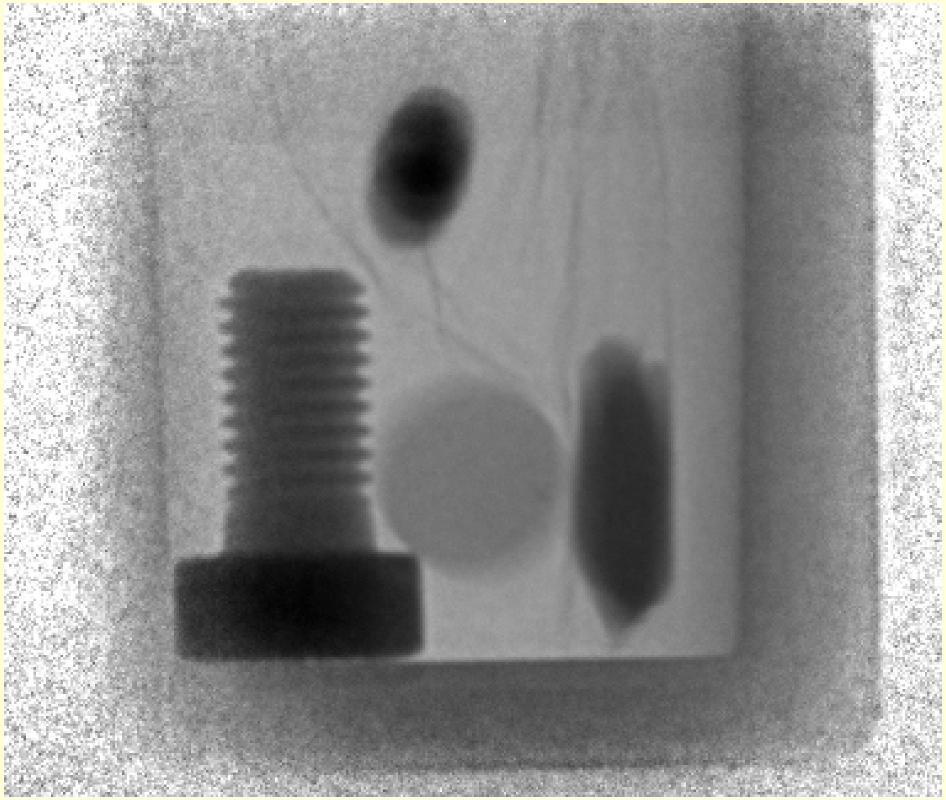


Beam size: 25 x 25 mm

Flux density: 3 x 10^7 cm^{-2} s^{-1}

Set up of PGAI-NT

Recent and current developments



Steel screw,

3 copper balls,

Aluminium cylinder and

Uranyl acetate (or U_3O_8) powder

(in a 25 μm thick teflon bag)

Inside a lead container

Materials identified by γ-spec

PGAA-NT 3D image of uranium and other items (Fe, Cu and Al)

Conclusions

- Routine capabilities are satisfactory for the
  - in-field categorization and
  - laboratory characterization of confiscated nuclear material (full analysis should be performed at JRC-ITU if needed)
- R&D have been carried out to
  - Improve accuracy and reliability of routine methods
  - Extend applicability and find new indicators by novel methods
- R&D results
  - Neutron interrogation studies provide scientific basis for the development of new detection methods (shielded and hidden NM)
  - Gamma-spectrometric methods for uranium age determination and to detect reprocessed uranium
  - Mass-spectrometric methods for uranium age determination, to detect reprocessed uranium, assay of transuranic and trace element fingerpints
  - Laser ablation assisted mass-spectrometry proved to be reliable and sufficiently precise, promising for particle analysis

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Thank you for your attention !