Mass Spectrometry in Nuclear Forensics

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Overview

- Illicit trafficking/smuggling of nuclear materials is of great concern
- Measurements on interdicted materials to trace their origin and to detect undeclared nuclear activities
- Isotopic composition of U and Pu (depends on isotopic enrichments, reactor irradiation history, cooling)
- Variety of Nuclear Analytical Techniques being developed for Nuclear Forensics (mass spectrometry, radiometry, LIBS, portable XRF)
- Inorganic mass spectrometry occupies a unique place for determining isotopic composition, amount and trace constituents present
- Natural variations in the isotopic composition of O, S, Sr and Pb important for geolocation of the source material

Different Grades of Uranium and Plutonium

U grade	% of ^{235}U
DEPLETED U	< 0.71%
NATURAL U	About 0.71%
LEU (Low Enriched U)	> 0.71% to < 20%
HEU (High Enriched U)	> 20% to < 90%
Oralloy (Weapons Grade U)	90% or more

Pu grade	% of ^{240}Pu
REACTOR GRADE Pu	> 18%
FUEL GRADE Pu	> 7% to < 18%
WEAPONS GRADE Pu	< 7%

M.S. Technique Used	Application
Thermal Ionisation Mass Spectrometry (TIMS)	Isotopic composition and amount
Inductively Coupled Plasma source Mass Spectrometry (ICPMS)	Trace Impurities
Stable Isotope Ratio Mass Spectrometry (SIRMS)	Isotopic composition of Oxygen, Sulphur
Secondary Ion Mass Spectrometry (SIMS)	Particle Analysis
Gas Chromatography Mass Spectrometry (GCMS)	Residual Solvents/Chemicals

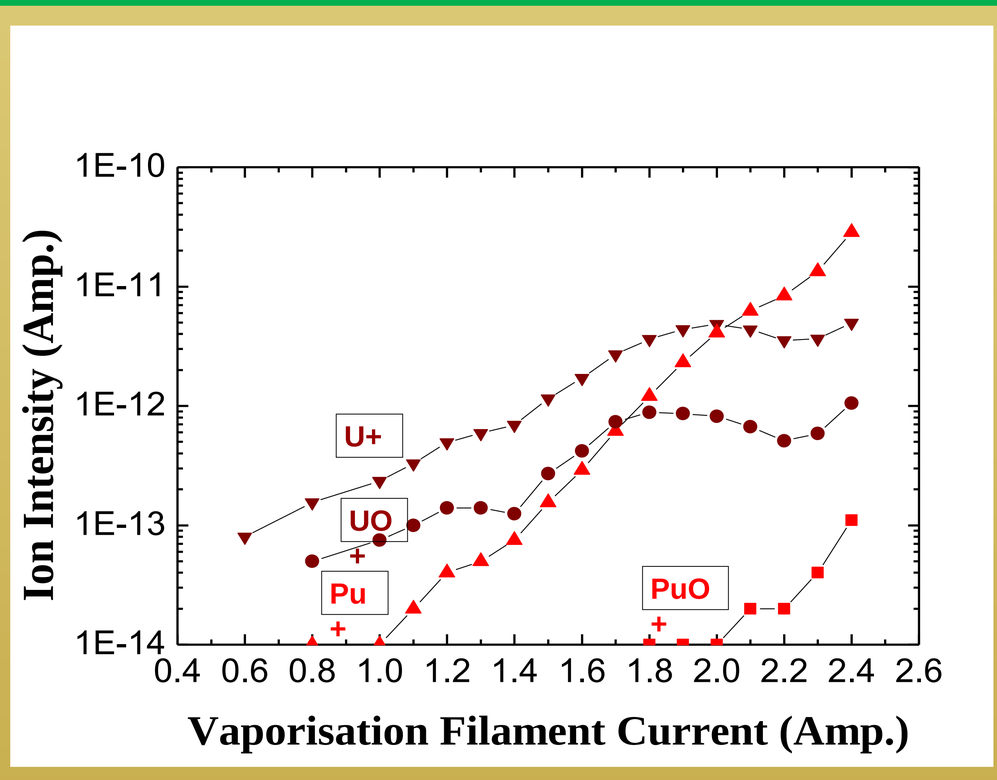
$$I(^{238}T) = I(^{238}U) + I(^{238}Pu)$$

$$I(^{238}Pu) = I(^{238}T) - I(^{238}U)$$

$$= I(^{238}T) - I(^{235}U) \times R(^{238}U/^{235}U)$$

$$= I(^{238}T) - I(^{235}U) \times R(^{238}UO/^{235}UO)$$

Evaporation and Ionization Behaviour of U and Pu in TIMS



Different Chronometers for Pu Age Determination

Parent (Half-life)	Daughter	Spikes Needed	Remarks
Pu-238 (87.7 yrs)	U-234	Pu-239, U-235	Low Abundance of Pu-238, Isobaric interference from U-238
Pu-239 (24110 yrs)	U-235	Pu-244, U-233	Pu-244 Spike availability restricted/limited
Pu-240 (6553 yrs)	U-236	Pu-244, U-233	Pu-244 Spike availability restricted/limited
Pu-241 (14.4 yrs)	Am-241, Np-237	Pu-244, Am-243	Spikes not available, Am-241 and Np-237 by ICPMS, γ spectrometry for ^{241}Am
Pu-242 (3.76×10^5 yrs)	U-238	----------	Long half-life of parent , not of interest

Mixture (SM-89)	$^{238}Pu/(^{239}Pu+^{240}Pu)$ Alpha Activity Ratio	$^{238}Pu/^{239}Pu$ Amount Ratio by	
		AS	TIMS
1	0.7956(0.15%)	0.003014	0.003014 (0.21%)
2	1.5468 (0.32%)	0.005860	0.005853 (0.23%)
3	3.0130 (0.69%)	0.011417	0.011524 (0.09%)
4	5.0848 (0.30%)	0.019267	0.019354 (0.07%)

CONCLUSION:

Pu-238 can be a useful chronometer for age determination of high burn-up Pu