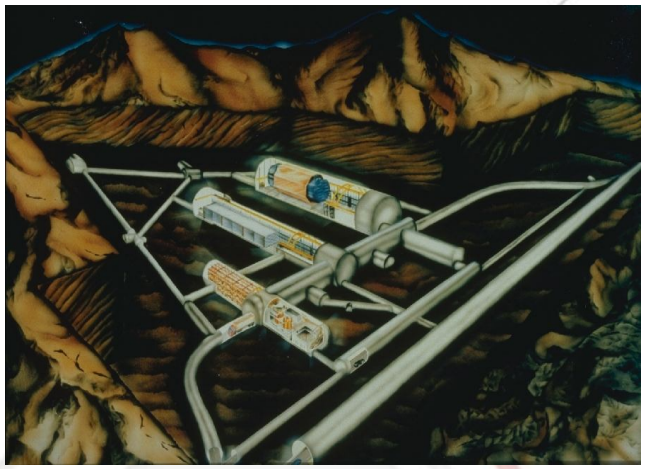
University of Milano Bicocca - Dep. of Physics
and
INFN of Milano Bicocca

Ultra sensitive Neutron Activation Measurements of ^{232}Th in Copper

M. Clemenza

Introduction: Low Level counting techniques for bulk measurements

Underground facilities may shields experiments from the background induced by cosmic radiation, but isolating the physics process from radioactive background due to the experimental apparatus itself, remains one of the crucial challenge.



What is considered Low Contamination?

For future 0νββ decay experiments we consider for some materials:

$^{232}Th < 10^{-12}$ g/g (< 3 uBq/kg)

$^{238}U < 10^{-12}$ g/g (< 12 uBq/kg)

Bulk measurements:

γ rays spectrometry with Ultra low bkg HPGe

HR-ICP-MS (High Resolution Inductive Coupled Plasma Mass Spectroscopy)

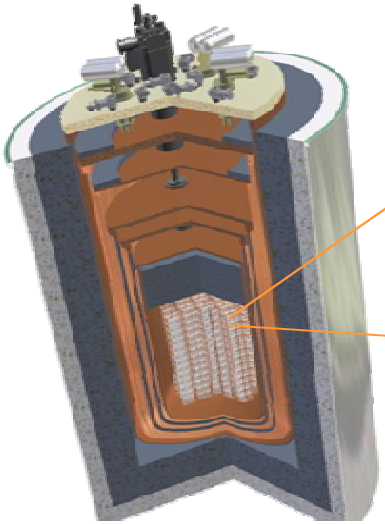
RNAA (Radiochemical Neutron Activation Analysis)

.....

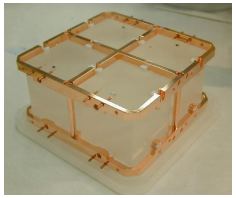
Introduction: Low Level counting techniques Copper Radio purity

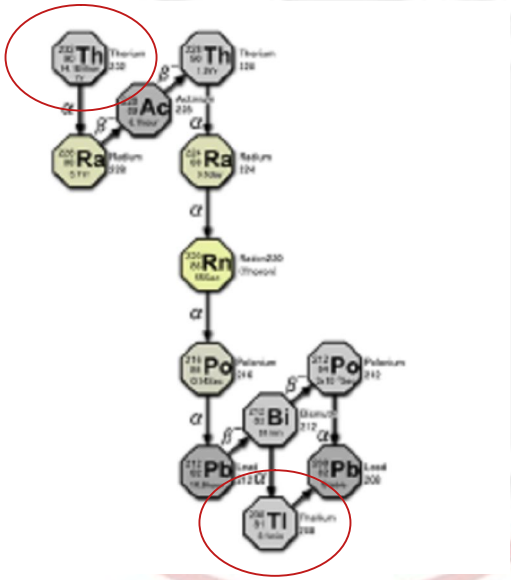
Copper, thanks to its low content in Primordial radionuclides like ^{232}Th, ^{238}U, is a material used frequently for shields, holders and others objects close to the detectors of many experiments of Rare Events Physics, so becomes crucial have a tools able to probing sensitivity of the order of $<10^{-12}$ g of contaminants / g of Copper.

CUORE Experiment: Shields and Frames are made by Copper



4 TeO_2 Crystals
Single Module





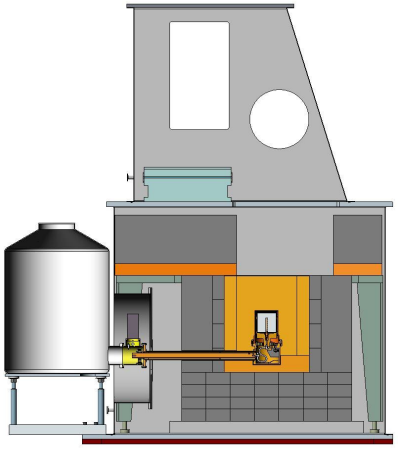
2614 keV ^{208}Tl Q_{value} $0\nu\beta\beta$ ^{130}Te: 2527 keV

Introduction: Bulk Contamination measurements techniques

γ rays spectrometry HPGe	Neutron Activation Analysis	HR-ICP-MS
Mainly on solids	Solids and liquids	Liquids /dissolved solids
Sensitivity on daughters	Sensitivity on primordial	Sensitivity on primordial
Huge amount of materials few tens of kg	Moderate amount of materials few tens of g	Small amount of dissolved solids
Long running time measurement few months	Medium running time measurement :few weeks	Short running time measurement: few minutes
Well established technique	Technique under development for low contaminations	Technique under development

complementary approach

Introduction: γ Spectrometry with Ultra low bkg HpGe



G. Heusser

GeMPI

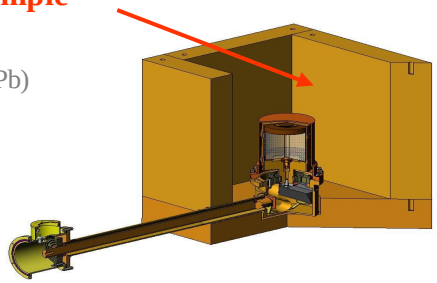
Operated at

LNGS

(3800 m w.e.)

effective volume of sample chamber ~ 15 l

(e.g. 125 kg Cu or 157 kg Pb)



High purity copper (NOSV) directly placed underground after electrolysis

D.L.: 16 μBq/kg ^{238}U (^{226}Ra)

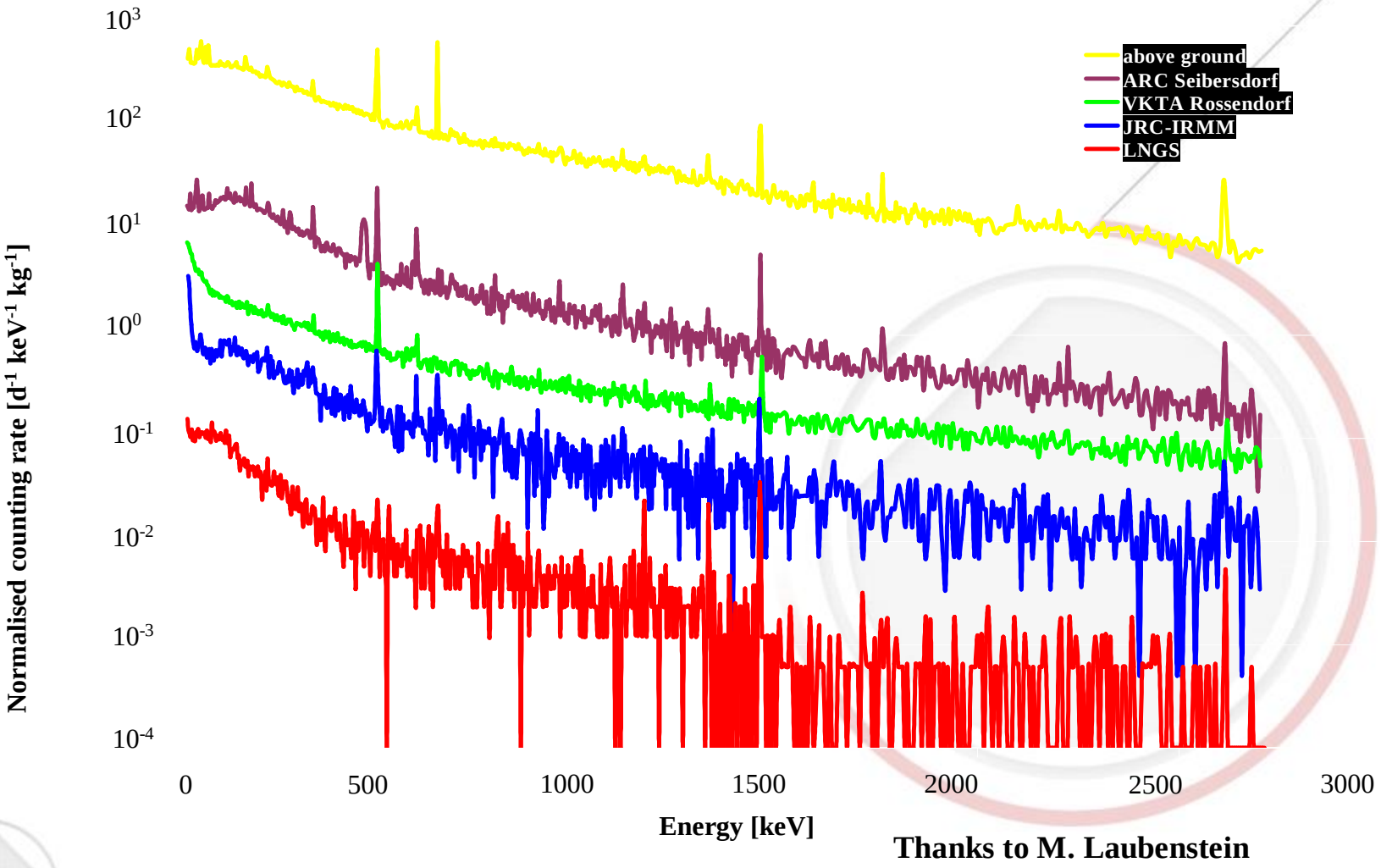
12 μBq/kg ^{232}Th (^{228}Th)

D.L.: 1.3 x 10^{-12} g/g ^{238}U

4 x 10^{-12} g/g ^{232}Th

Thanks to M. Laubenstein

Introduction: γ Spectrometry with Ultra low bkg HpGe



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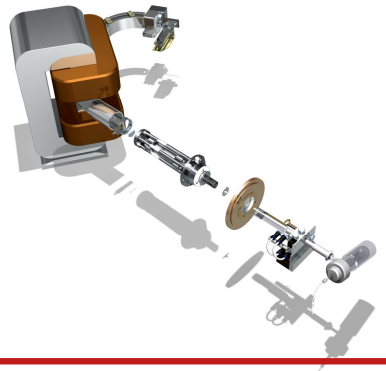
Introduction: HR-ICP-MS

HR-ICP-MS: High Resolution Inductively Coupled Plasma Mass Spectroscopy



High Sensitivity for ^{238}U ^{232}Th: 1- 0.1 ppq = $\mathbf{10^{-15}}$ - $\mathbf{10^{-16}}$ g/g in liquid matrix !!

For Copper:

Loss in Sensitivity due to the dilution factor (dissolve Cu in HNO_3 and dilute 10-100 times) and for the presence of Th and U in Ultrapure HNO_3 and Ultrapure water

Reduced Sensitivity for ^{238}U ^{232}Th to10 - 0.1 ppt = $\mathbf{10^{-11}}$ - $\mathbf{10^{-13}}$ g/g in solid matrix

Introduction: RNAA Strenghts and weakness

Borexino: very sensitive measurements on liquids (scintillators and water)

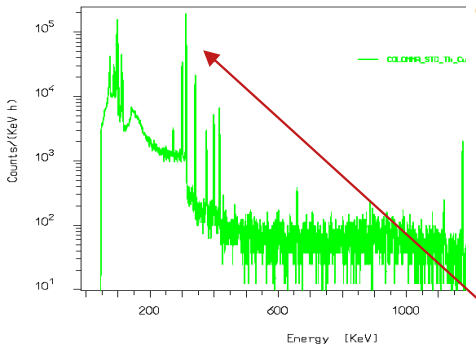
	^{238}U	^{232}Th
sensitivity for scintillator	$<9.4\ 10^{-17}$ g/g	$<2.4\ 10^{-15}$ g/g
sensitivity for nylon	$2.1\ 10^{-12}$ g/g	$5.1\ 10^{-12}$ g/g

matrix with nuclei with low cross section for neutron capture, respect to Th,U

No interfence due a contamination of solvents, acids, water…..

$$^{232}Th(n,\gamma)^{233}Th \xrightarrow{\beta^-} {}^{233}Pa \xrightarrow{\beta^-} {}^{233}U$$

22,6 m *27 d*



Gamma ray:

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17.26	0.004(AP)
28.375(5)	0.11(4)
40.35(1)	0.039(8)
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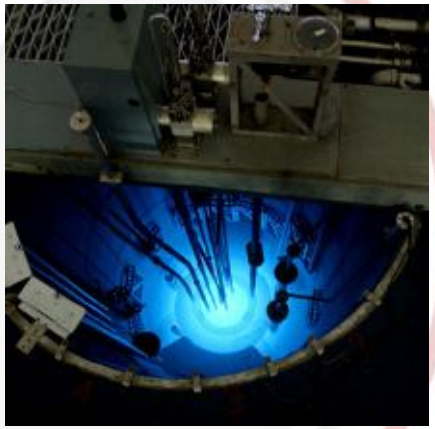


Copper: Activation of the sample

Introduction: NAA Strenghts and weakness

$$^{232}Th(n,\gamma)^{233}Th \xrightarrow[22,6\ m]{\beta^-} {}^{233}Pa \xrightarrow[27\ d]{\beta^-} {}^{233}U$$

Activation of the Copper

^{64}Cu $t_{1/2}$ 12.7 h

^{66}Cu $t_{1/2}$ 5.1 m

Waiting time 1 month after irradiation

Activation of the trace elements in the Copper

^{75}Se $t_{1/2}$ 120 d

^{192}Ir $t_{1/2}$ 74d

^{198}Au $t_{1/2}$ 2,7d

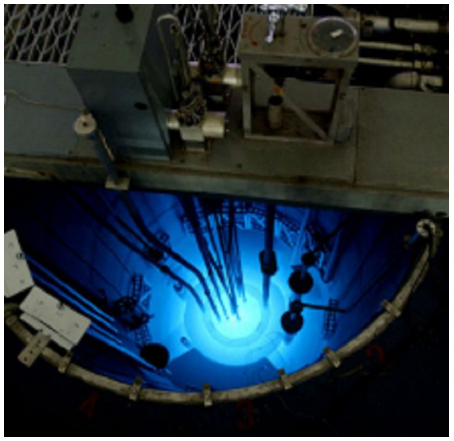
^{124}Sb $t_{1/2}$ 60d

^{110m}Ag $t_{1/2}$ 250d

^{60}Co $t_{1/2}$ 1925d

Separation of the activated element with Ion Exchange Resin

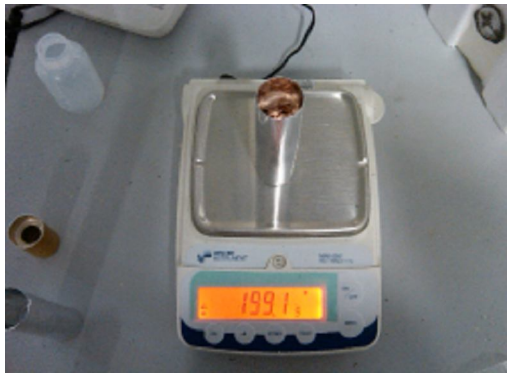
NAA ^{232}Th in Copper: Irradiation



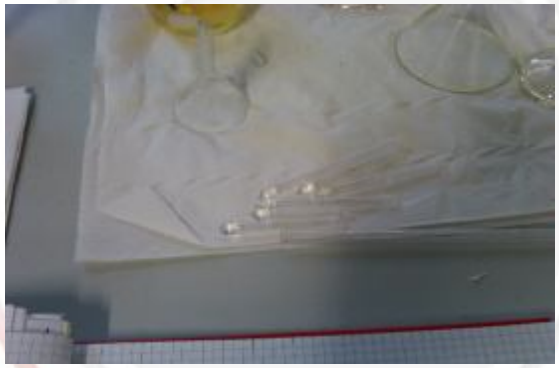
TRIGA MARK II University of Pavia 250 kW

Irradiation time: 30h Central Channel

Flux: 10^{13} n cm^{-2} sec^{-1}



Sample:199 g Cu



10ul Standard Solution

1000 µg/ml ^{232}Th

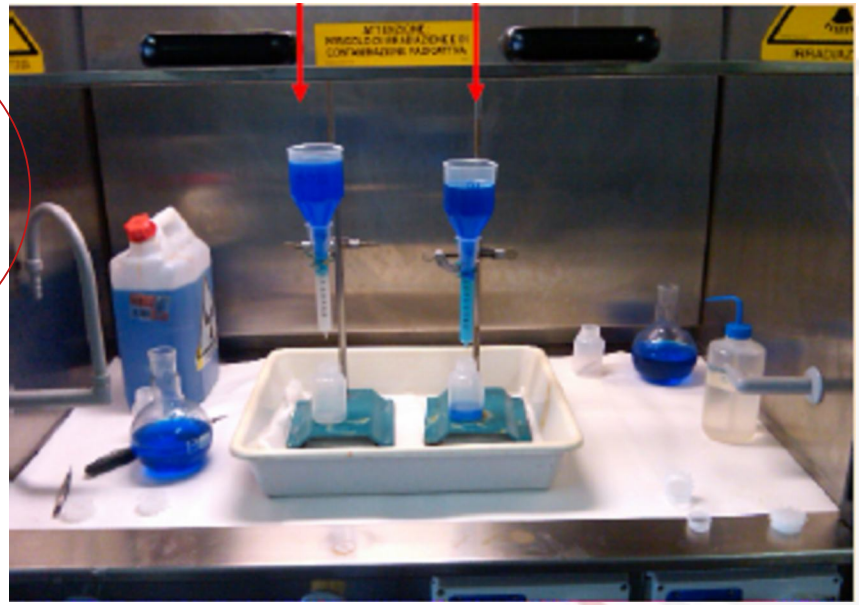
NAA ^{232}Th in Copper: Radiochemical procedure

Dissolution of the Sample:

$$3Cu + 8HNO_3 \longrightarrow 3Cu(NO_3)_2 + 4H_2O + 2NO$$

199g n.a. $Cu(NO_3)_2$

10ul STD solution ^{232}Th



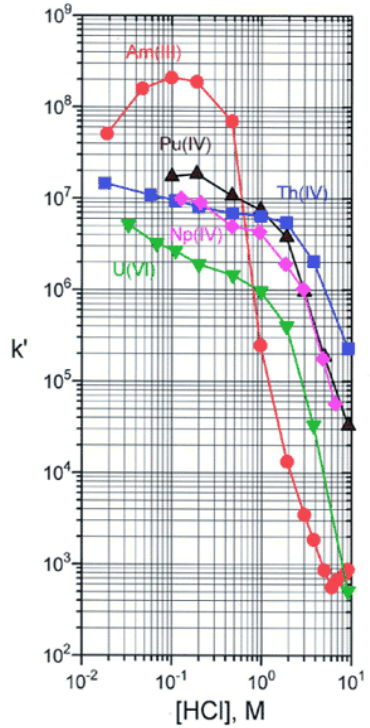
199g Irradiated $Cu(NO_3)_2$

Separation of ^{233}Pa from the other elements presents in the Copper with Ion Exchange Actine Resin Column (Eichrom)

^{232}Th in Copper: Actinide Resin Eichrom

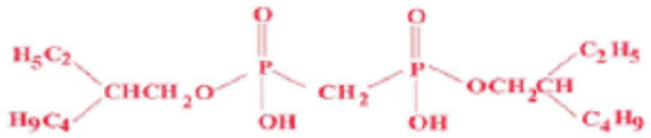
Polimeric Compound with High Affinity for all Actinides

The uptake of actinide elements by Actinide Resin



DIPEX® Extractant

P,P'- di(2-ethylhexyl)methanediphosphonic acid



$H_2DEH[MDP]$

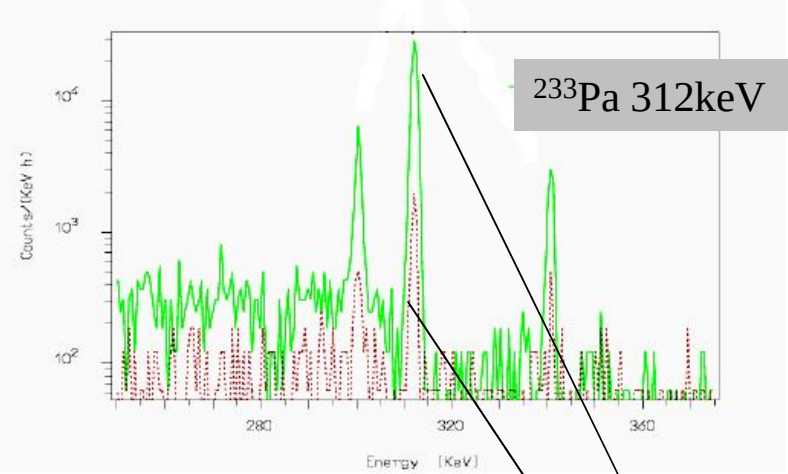
Horwitz, et al. (HP197)

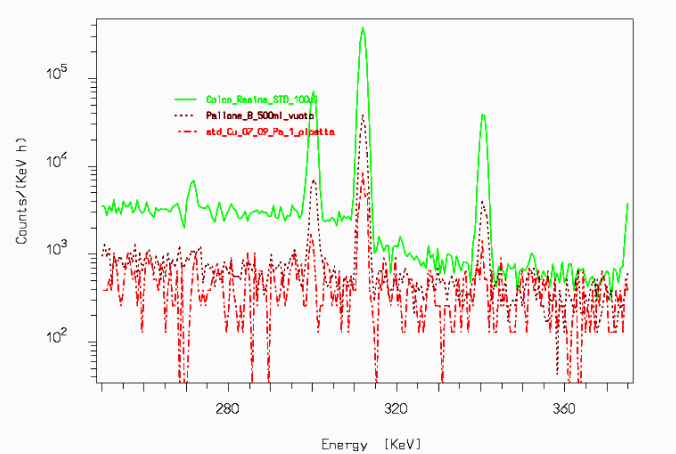
High Retention Factor

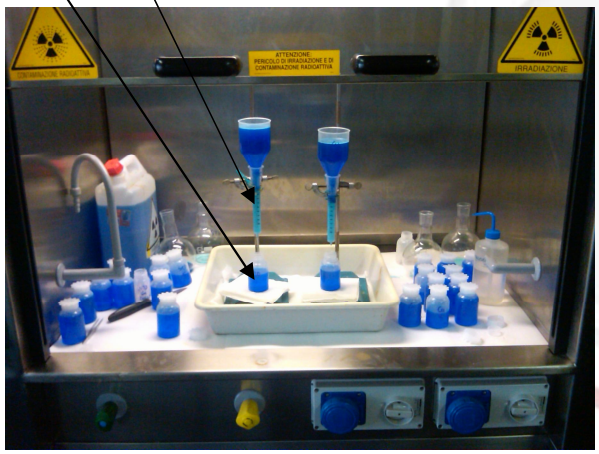
^{232}Th in Copper: Recovery and Evaluation of Retention Factor for ^{233}Pa

Retention Factor: 98.9%

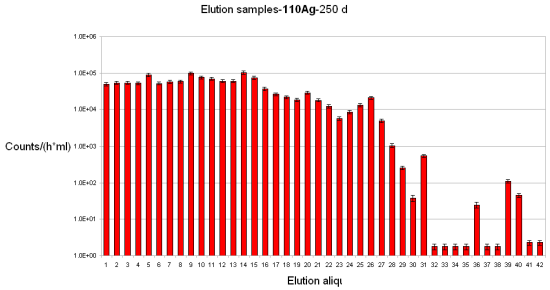
Recovery: 85-95%

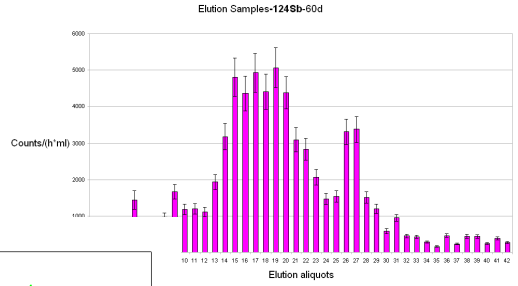


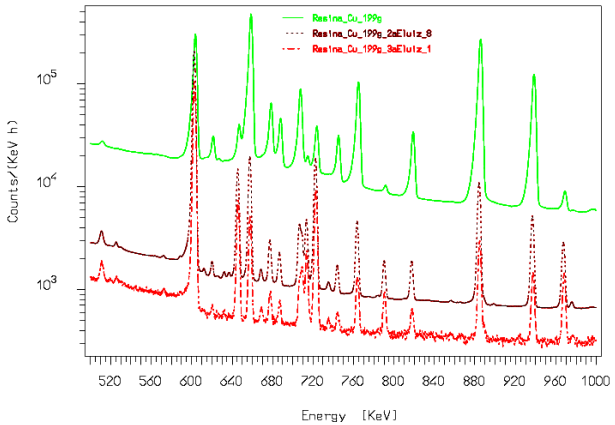


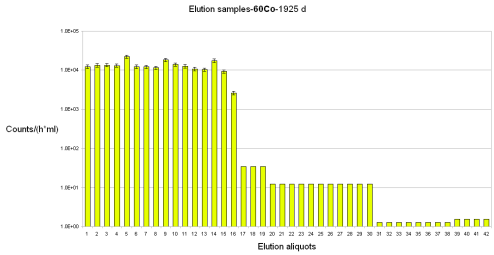


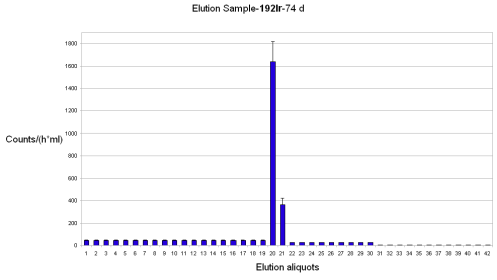
^{232}Th in Copper: Background reduction

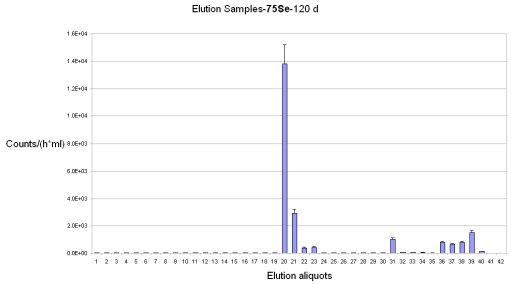












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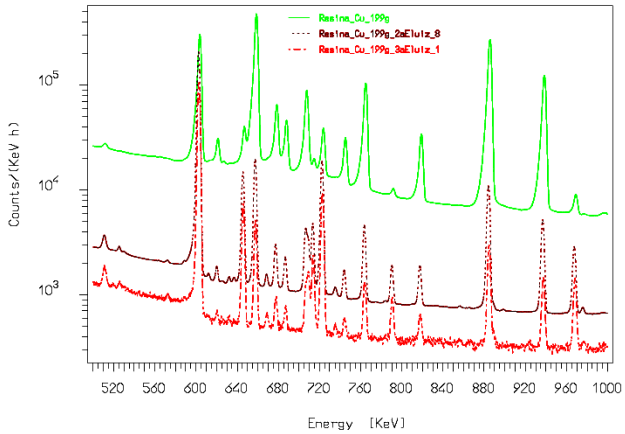
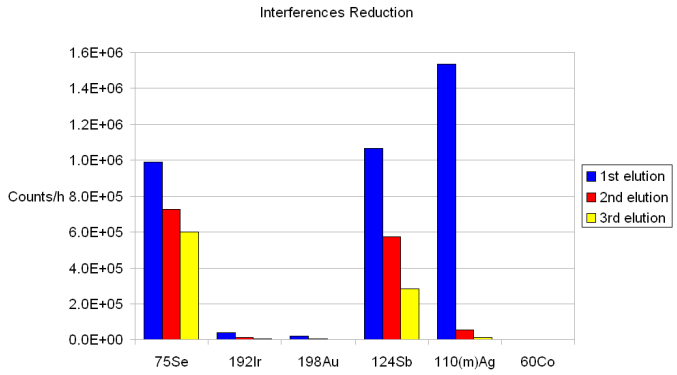
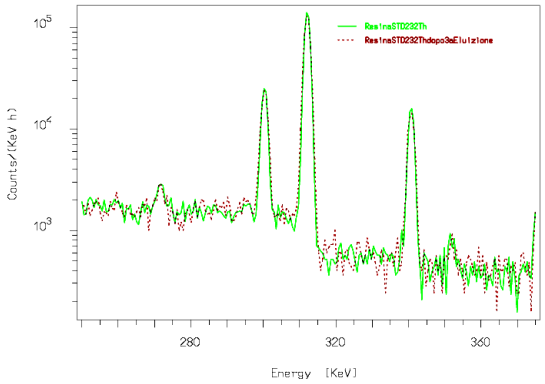


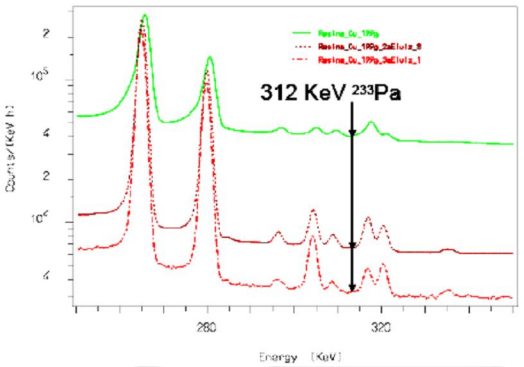


Reduction of 99% ^{60}Co,
98% ^{110m}Ag,
89% ^{192}Ir,
73% ^{124}Sb,
39% ^{75}Se

^{232}Th in Copper: Detection Limit ^{232}Th in Copper



Column with ^{233}Pa before and after elution with HNO_3, HCl 4M

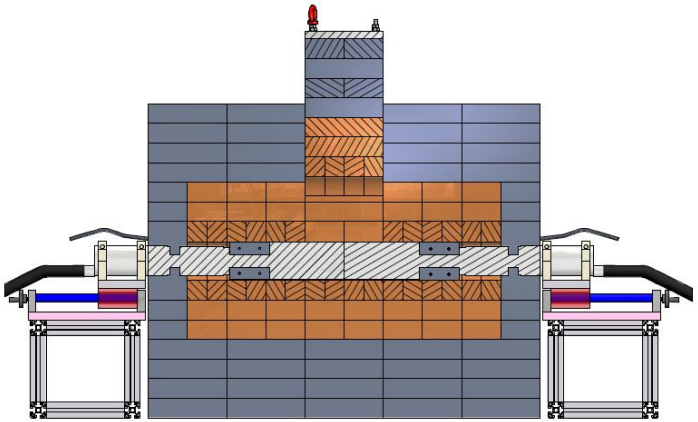


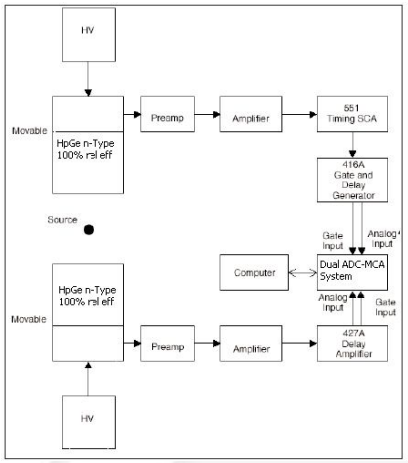
Column with Sample

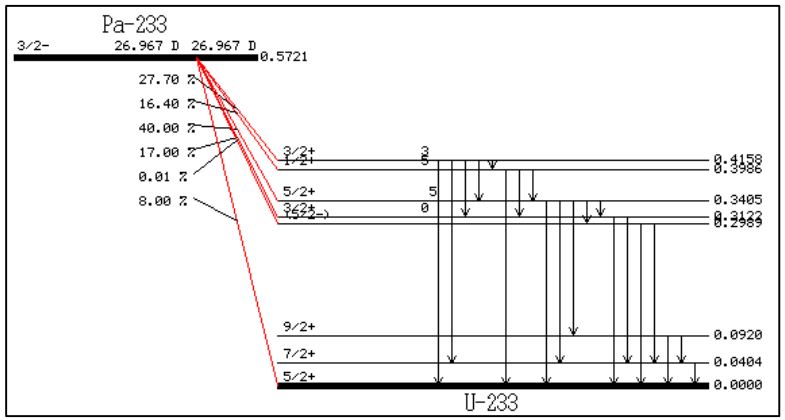
DL: 4.88×10^{-13} g/g

Is the best DL in NAA for Copper Sample

Prospects: γ-γ coincidence







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Introduction: NAA working activity

People involved:

A. Borio, A. Salvini
LENA and University of Pavia
M. Clemenza, E. Previtali
University of Milano-Bicocca and INFN Milano Bicocca

Formal agreement between:

University of Milano-Bicocca
University of Pavia
LENA Laboratory
INFN
for R&D on high sensitivity NAA