The Half-Life of ^{60}Fe: Status

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TOI: $T_{1/2}(^{60}Fe)$: (1.49 ± 0.27) Mys

Zn57 40 ms (7/2-) ECp	Zn58 65 ms 0+ EC	Zn59 182.0 ms 3/2- ECp	Zn60 2.38 m 0+ EC	Zn61 89.1 s 3/2- EC	Zn62 9.186 h 0+ EC	Zn63 38.47 m 3/2- EC	Zn64 0+ 48.6	Zn65 244.26 d 5/2- EC	Zn66 0+ 27.9
Cu56	Cu57 199.4 ms 3/2- EC	Cu58 3.204 s 1+ EC	Cu59 81.5 s 3/2- EC	Cu60 23.7 m 2+ EC	Cu61 3.333 h 3/2- EC	Cu62 9.74 m 1+ EC	Cu63 3/2- 69.17	Cu64 12.700 h 1+ EC,β-	Cu65 3/2- 30.83
Ni55 212.1 ms 7/2- EC	Ni56 6.077 d 0+ EC	Ni57 35.60 h 3/2-	Ni58 0+ 68.077	Ni59 7.6E+4 y 3/2- EC	Ni60 0+ 26.223	Ni61 3/2- 1.140	Ni62 0+ 3.634	Ni63 100.1 y 1/2- β-	Ni64 0+ 0.926
Co54 193.23 ms 0+ * EC	Co55 17.53 h 7/2- EC	Co56 77.27 d 4+ EC	Co57 271.79 d 7/2- EC	Co58 70.82 d 2+ * EC	Co59 7/2- 100	Co60 5.2714 y 5+ *	Co61 1.650 h 7/2- β-	Co62 1.50 m 2+ * β-	Co63 27.4 s (7/2)- β-
Fe53 8.51 m 7/2- * EC	Fe54 0+ 5.8	Fe55 2.73 y 3/2- EC	Fe56 0+ 91.72	Fe57 1/2- 2.2	Fe58 0+ 0.28	Fe59 44.503 d 3/2- β-	Fe60 1.5E+6 y 0+ β-	Fe61 5.98 m 3/2-,5/2- β-	Fe62 68 s 0+ β-
Mn52 5.591 d 6+ * EC	Mn53 3.74E+6 y 7/2- EC	Mn54 312.3 d 3+ EC,β-	Mn55 5/2- 100	Mn56 2.5785 h 3+ β-	Mn57 85.4 s 5/2- β-	Mn58 3.0 s 0+ * β-	Mn59 4.6 s 3/2-,5/2- β-	Mn60 51 s 0+ * β-	Mn61 0.71 s (5/2-) β-
Cr51 27.702 d 7/2- EC	Cr52 0+ 83.789	Cr53 3/2- 9.501	Cr54 0+ 2.365	Cr55 3.497 m 3/2- β-	Cr56 5.94 m 0+ β-	Cr57 21.1 s 3/2-,5/2-,7/2- β-	Cr58 7.0 s 0+ β-	Cr59 0.74 s β-	Cr60 0.57 s 0+ β-

Only one measurement

(specific activity and absolute AMS measurement)

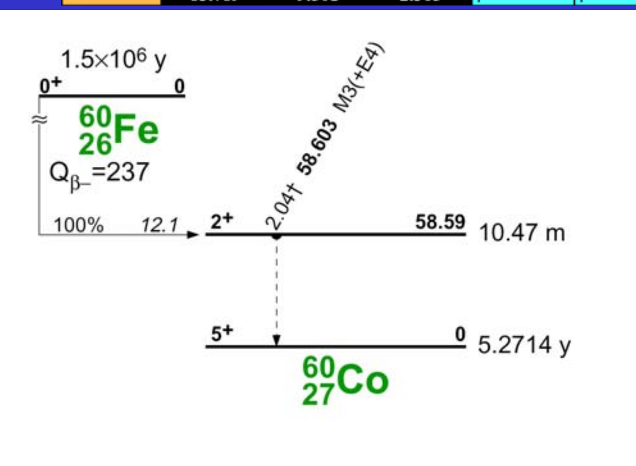
Kutschera et al., NIM B5(1984)439

Previous measurement:

Roy and Kohman

(1957 bzw. 1968)

3×10^5 a (± 300%)



Theory:

2nd forbidden decay, low end point

-> broad range for $T_{1/2}$

Kutschera et al., 1984

^{60}Fe: spallation of Cu with 191 MeV p $\Rightarrow \approx 10^{15}$ at

after 1 year: extraction of iron with a (remote) multi-stage anion exchange separation; adding 1.4 mg of iron $Fe(OH)_3 \downarrow$; weighing for determination of stable iron.

dilution of the sample material: 90% of 2 ml solution $\hookrightarrow$ 100 ml; deviation colorimetric vs. gravimetric 30%
monitor: ^{59}Fe (γ activity, $T_{1/2}$=44.5 d)

γ-ray activity of the daugther ^{60}Co:
first attempt - ^{60}Co from direct production
$\hookrightarrow$ chemical separation of Co
$\hookrightarrow$ measurement of the grow-in of the ^{60}Co activity using a Ge(Li) low-level counting facility
$\Rightarrow$ Fit of grow-in (only 1332 keV line used for analysis)

Determination of ^{60}Fe/Fe with AMS:
^{60}Fe/Fe: $(9.54 \pm 1.40) \cdot 10^{-8}$ (absolute measurement)
$N_{^{60}Fe} = (3.99 \pm 0.71) \cdot 10^{14}$ atoms
Result: $T_{1/2} = 1.49 \pm 0.29 \cdot 10^{6}$ yrs

$\Rightarrow$ **A New Attempt!**

How to measure a long half-life?

$$\Delta A = A_1 - A_2 = A_1 \cdot (1 - e^{-ln(2)\cdot\frac{\Delta t}{T_{1/2}}}) \approx 0$$

$$\Delta N = N_1 - N_2 = N_1 \cdot (1 - e^{-ln(2)\cdot\frac{\Delta t}{T_{1/2}}}) \approx 0$$

if $\Delta t << T_{1/2}$

$$-\frac{\mathrm{d}\,N}{\mathrm{d}\,t} = N(t) \cdot e^{-\lambda t}$$

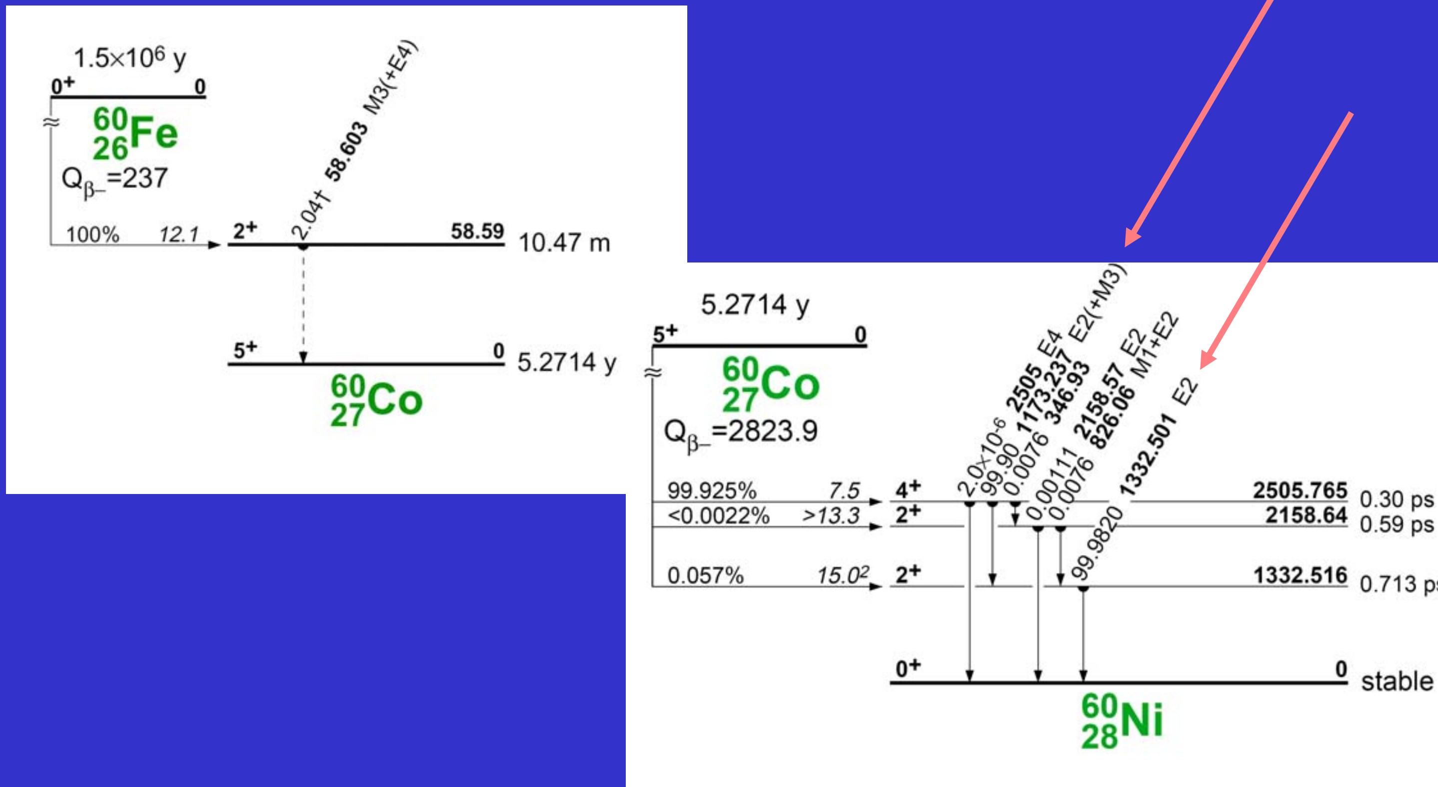
$$A = \lambda \cdot N = \frac{ln(2)}{T_{1/2}} \cdot N$$

More realistic:

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

Build-up of the ^{60}Co activity

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$



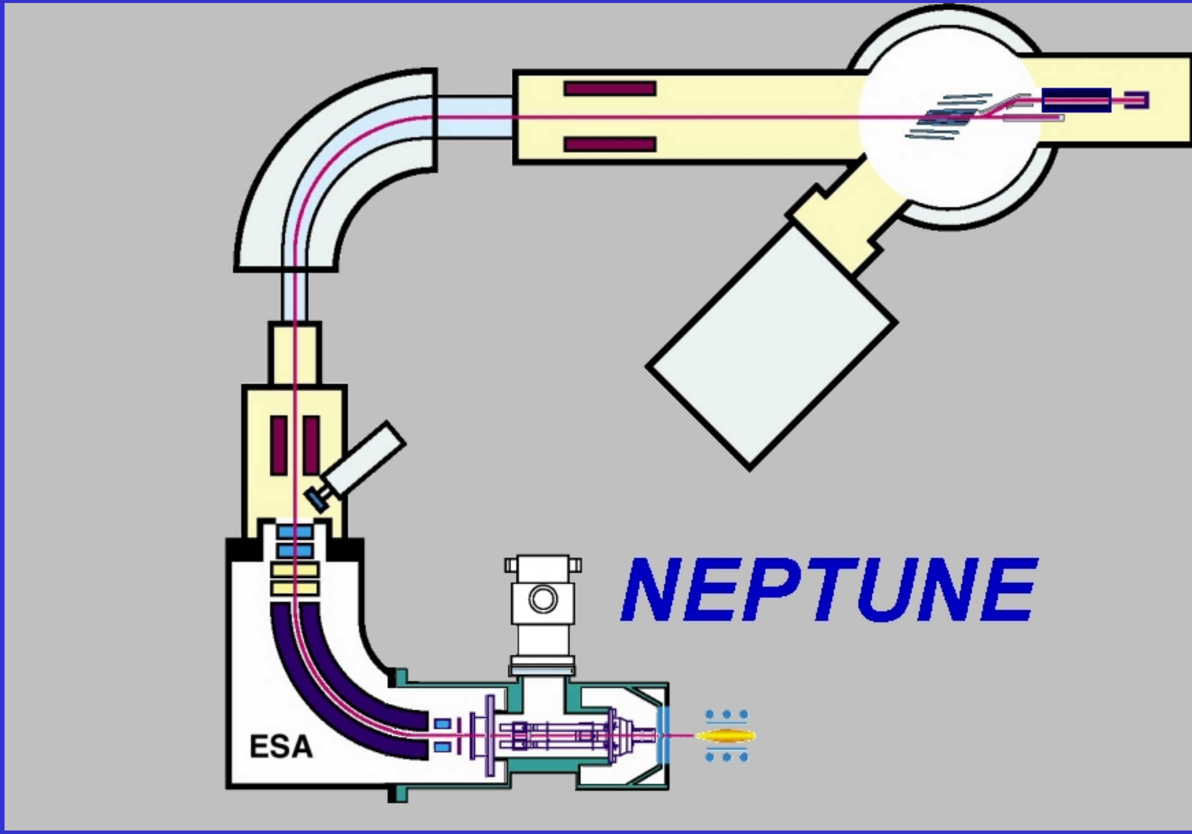
$$A_{^{60}Co} = N_{^{60}Fe} \lambda_{^{60}Fe} \cdot (1 - e^{-\lambda_{^{60}Co} \cdot t})$$

$$\approx N_{^{60}Fe} \cdot \lambda_{^{60}Fe} \cdot \lambda_{^{60}Co} \cdot t \quad \text{for } t << T_{1/2,^{60}\mathrm{Co}}$$

$$\approx N_{^{60}\mathrm{Fe}} \cdot \lambda_{^{60}\mathrm{Fe}} \quad \text{for } t >> T_{1/2,^{60}\mathrm{Co}}$$

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

Multicollector inductively coupled plasma mass spectrometry (MC-ICP-MS)



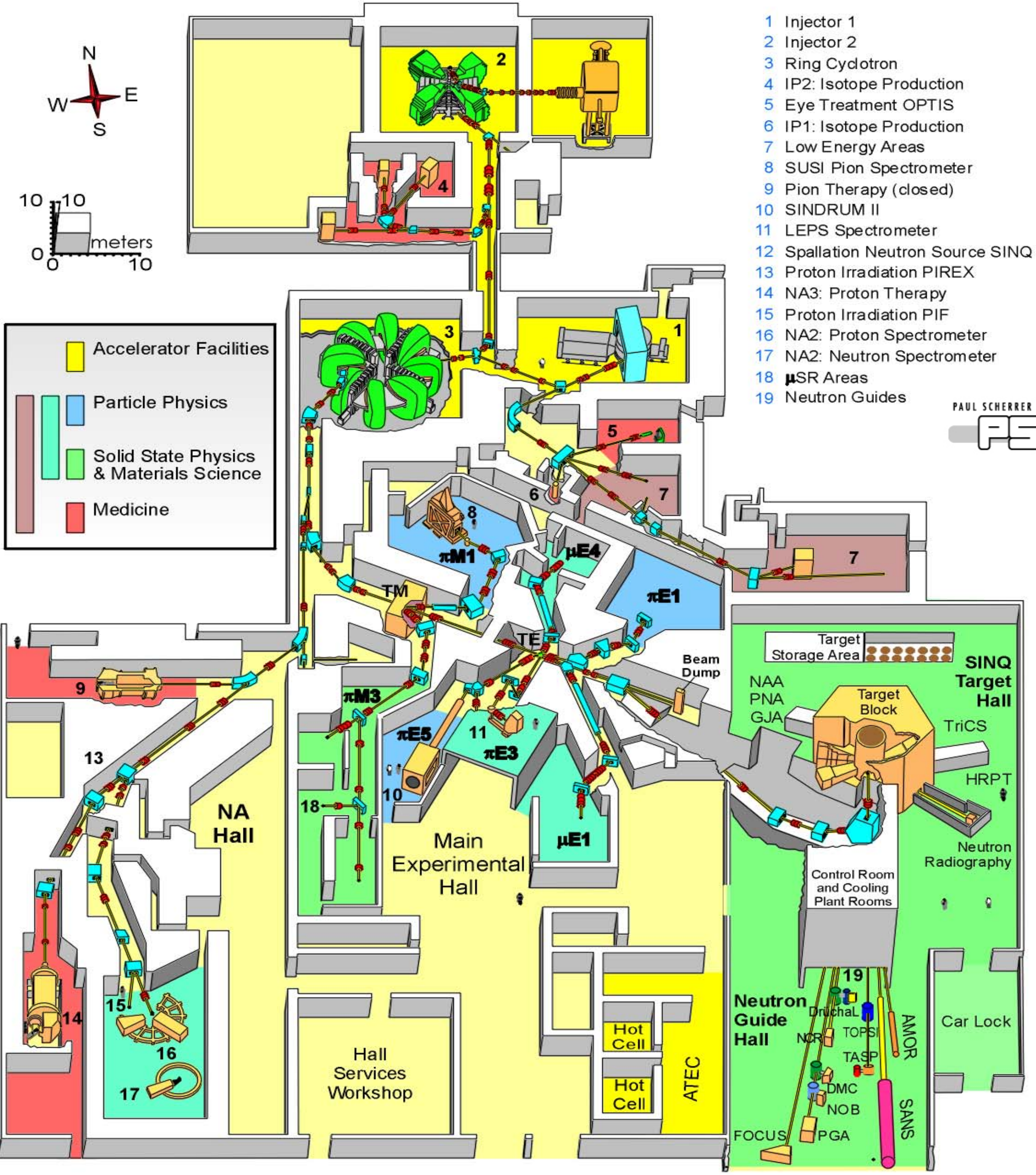
We need ^{60}Fe nuclei!

^{60}Fe:

Produced under stellar conditions !

Only ejected by SN !

But where to get it on earth ?



BMA-Target, Beam dump and shielding (Pion therapy station, 590 MeV protons)

Material:
Copper

Irradiated from 1980 to 1.09.1992

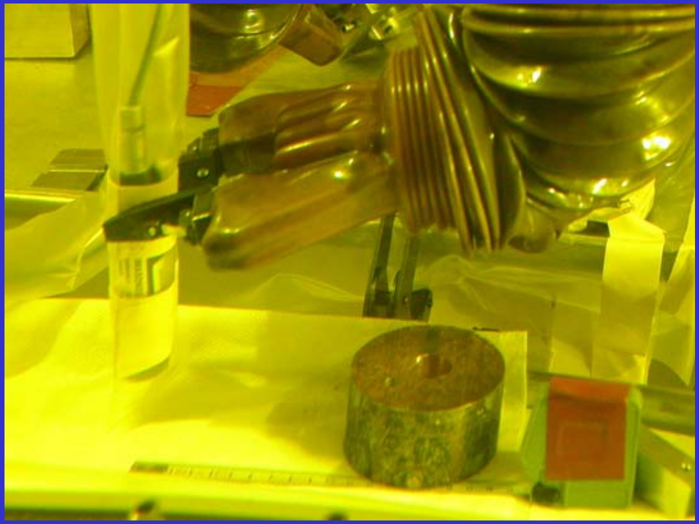
Copper beam dump (BMA1) from PSI (~ 10 kg)

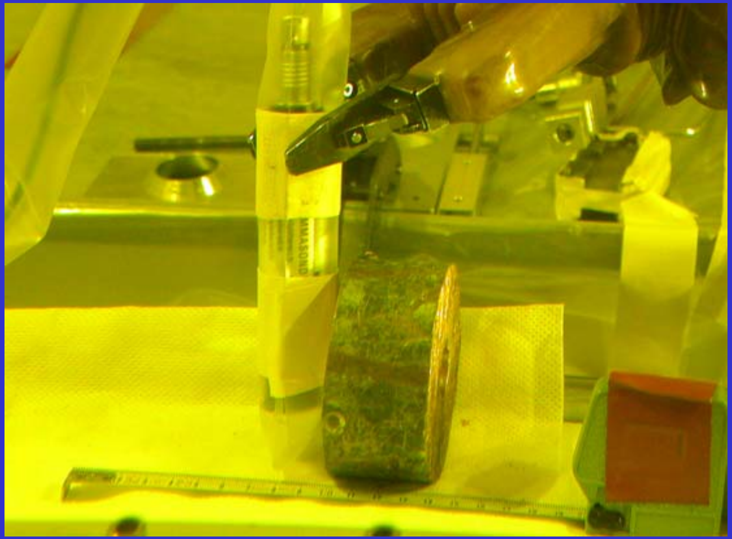
590 MeV p, 160 mAh (3.6 x 10^{21} protons)

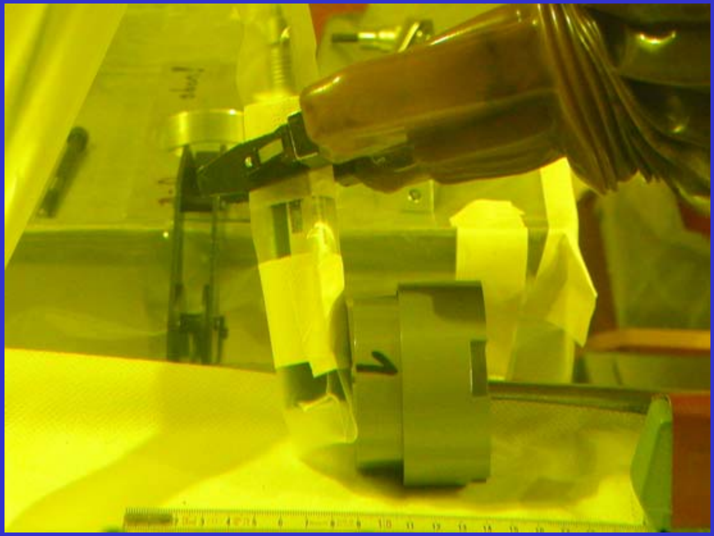
		Zn60 2.38 m 0+ EC	Zn61 89.1 s 3/2- EC	Zn62 9.186 h 0+ EC	Zn63 38.47 m 3/2- EC	Zn64 0+ 48.6	Zn65 244.26 d 5/2- EC	Zn66 0+ 27.9	Zn67 5/2- 4.1	Zn68 0+ 18.8
		Cu59 81.5 s 3/2- EC	Cu60 23.7 m 2+ EC	Cu61 3.333 h 3/2- EC	Cu62 9.74 m 1+ EC	Cu63 3/2- 69.17	Cu64 12.700 h 1+ EC,β-	Cu65 3/2- 30.83	Cu66 5.088 m 1+ β-	Cu67 61.83 h 3/2- β-
Ni56 6.077 d 0+ EC	Ni57 35.60 h 3/2-	Ni58 0+ 68.077	Ni59 7.6E+4 y 3/2- EC	Ni60 0+ 26.223	Ni61 3/2- 1.140	Ni62 0+ 3.634	Ni63 100.1 y 1/2- β-	Ni64 0+ 0.926	Ni65 2.5172 h 5/2- β-	Ni66 54.6 h 0+ β-
Co55 17.53 h 7/2- EC	Co56 77.27 d 4+ EC	Co57 271.79 d 7/2- EC	Co58 70.82 d 2+ * EC	Co59 7/2- 100	Co60 5.2714 y 5+ *	Co61 1.650 h 7/2- β-	Co62 1.50 m 2+ * β-			
Fe54 0+ 5.8	Fe55 2.73 y 3/2- EC	Fe56 0+ 91.72	Fe57 1/2- 2.2	Fe58 0+ 0.28	Fe59 44.503 d 3/2- β-	Fe60 1.5E+6 y 0+ β-	Fe61 5.98 m 3/2-,5/2- β-			
Mn53 3.74E+6 y 7/2- EC	Mn54 312.3 d 3+ EC,β-	Mn55 5/2- 100	Mn56 2.5785 h 3+ β-	Mn57 85.4 s 5/2- β-	Mn58 3.0 s 0+ * β-	Mn59 4.6 s 3/2-,5/2- β-	Mn60 51 s 0+ * β-			

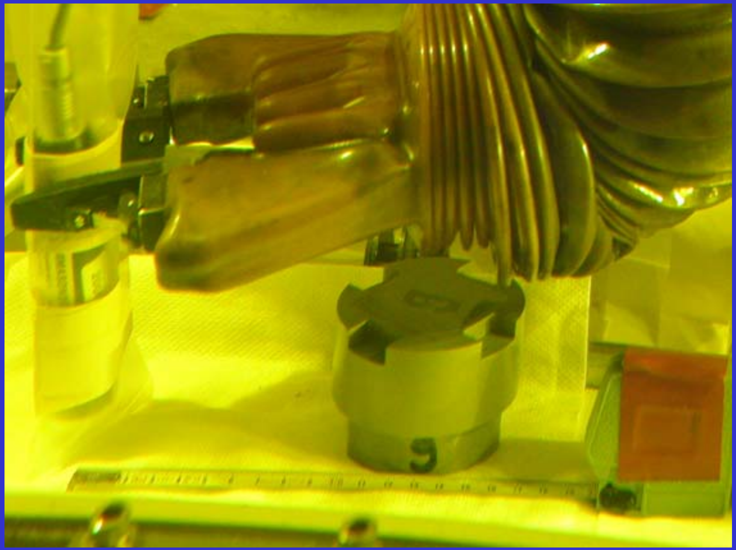
Hot material: e.g. ^{60}Co ~ 7 x 10^9 Bq

Handling of the Copper Beamdump at the hot chemistry (PSI)









Chemical separation of ^{60}Fe

Measurement of the grow-in of ^{60}Co
→ very good chemical separation from Co necessary

- Dissolution of 3.86 g Cu (beam dump) in 7 M HNO_3
- Evaporation to dryness
- Dissolution in 7 M HCl
- + 5 mg Fe^{3+} and 5 mg Co^{2+} as carrier
- Extraction with diisopropylether
- Aqueous phase: Ni, Co, Cu, organic phase: Fe
- Back extraction with 0.1 M HCl, repetition of procedure
- Additional purification by precipitation of $Fe(OH)_3$
- Result (by July 2005): ~ 10^{15} ^{60}Fe atoms, ^{60}Co reduced by more than 10^7
- But still: ^{55}Fe (~ 36 MBq !); $T_{1/2}$ = 2.73 yr

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

Calibration of the detector:

Avoiding geometrical corrections etc.

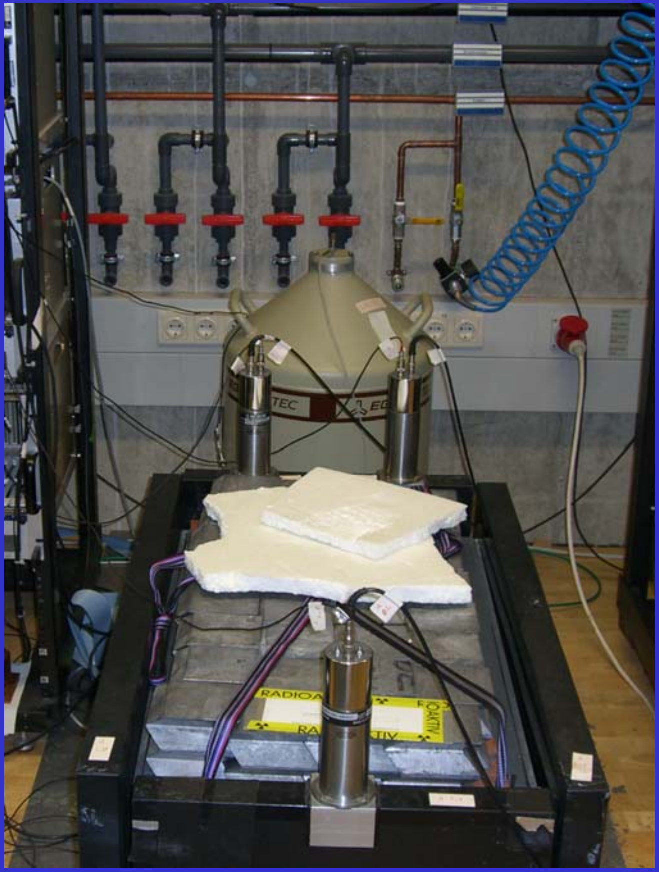
Calibration source (^{60}Co) with the same geometry:

5ml 0.1 nHCl

101.99 (± 1.5%) Bq ^{60}Co

(all uncertainties 1 sigma)

→ Germanium detector



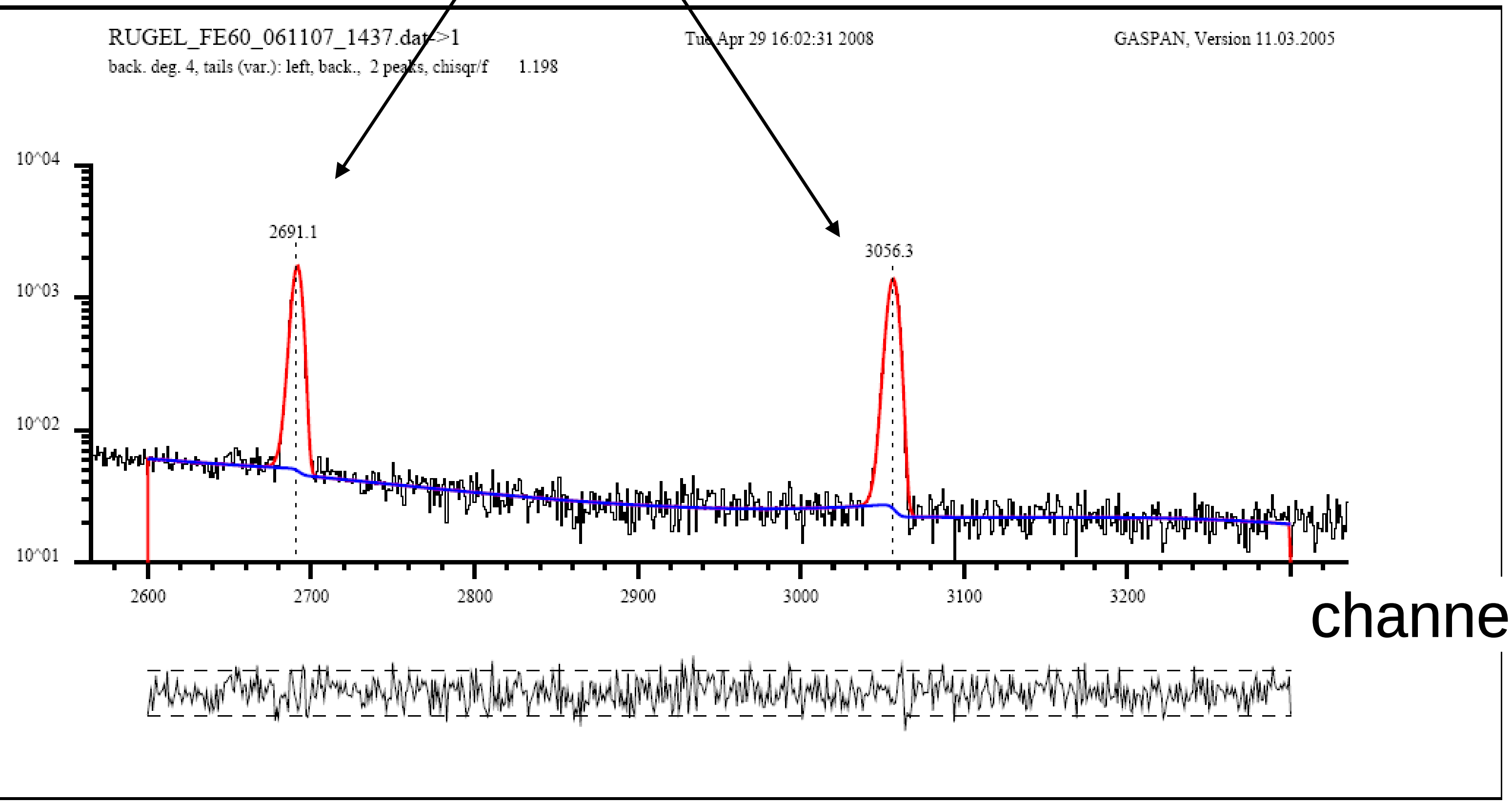
$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

^{60}Co

12710±130 cts

11450±120 cts

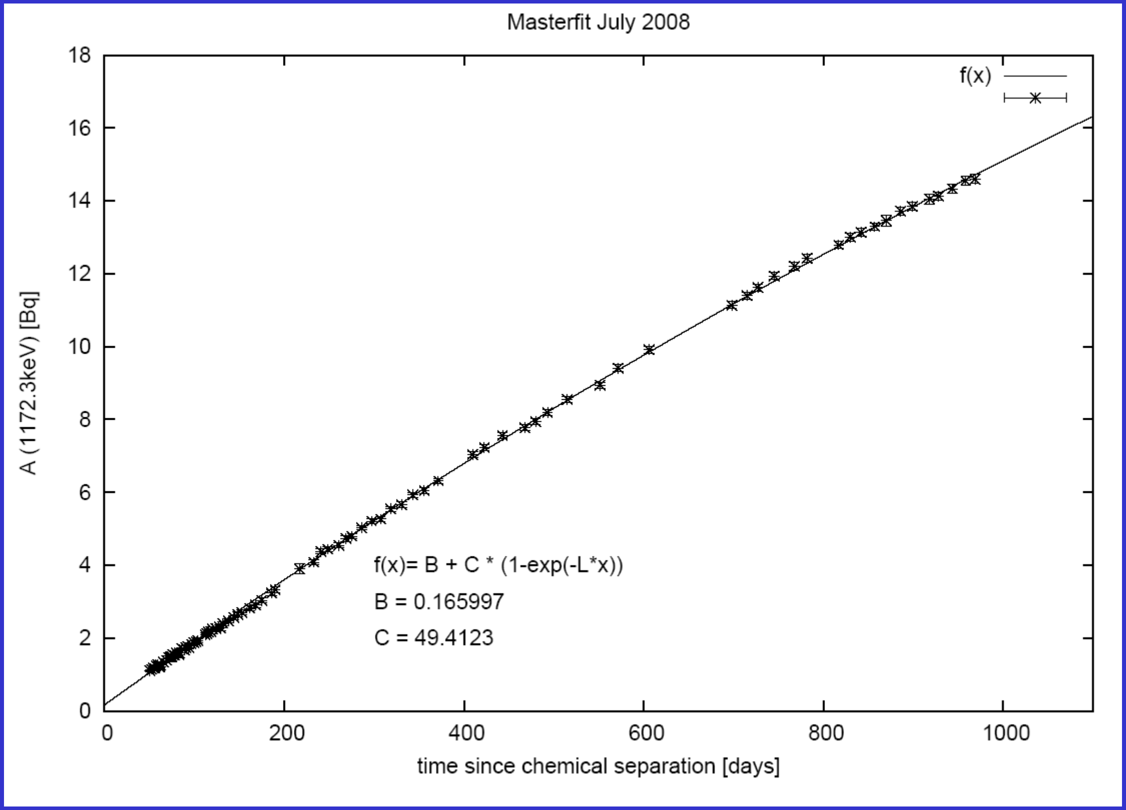
Counts log scale



channel

Build-up of the ^{60}Co

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$



T= 0

^{60}Co ~ 0.2 Bq

Chemical reduction at least 10^7 !

Saturation activity

49 Bq

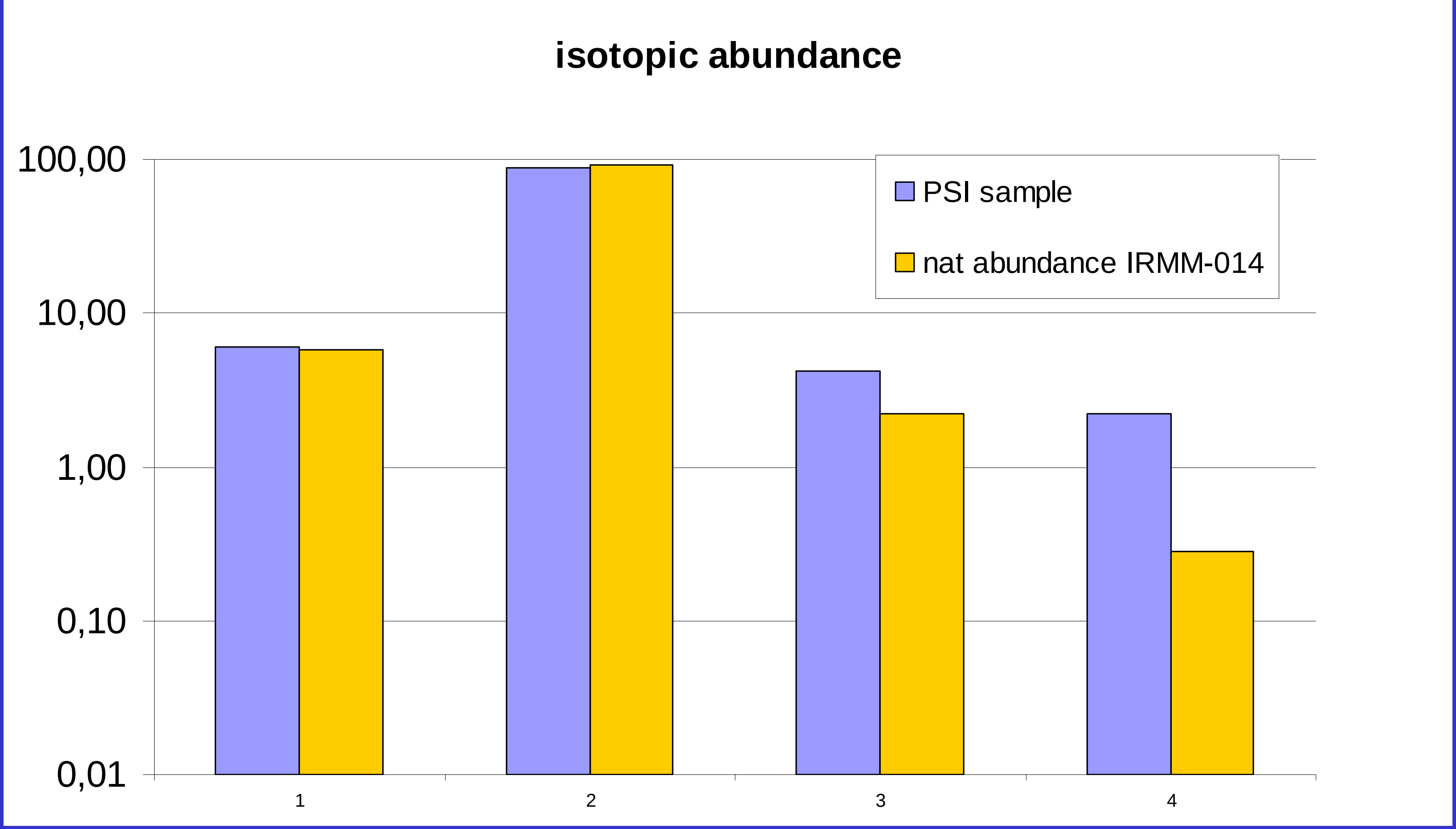
$$A_{^{60}Co} = N_{^{60}Fe} \lambda_{^{60}Fe} \cdot (1 - e^{-\lambda_{^{60}Co} \cdot t})$$

$$\approx N_{^{60}Fe} \cdot \lambda_{^{60}Fe} \cdot \lambda_{^{60}Co} \cdot t$$

for $t << T_{1/2,^{60}\text{Co}}$

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

ICP-MS measurement



^{54}Fe ^{56}Fe ^{57}Fe ^{58}Fe

PSI sample ratios checked with model calculations

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

MC-ICP-MS measurement

Isotopic dilution with a ^{57}Fe spike:

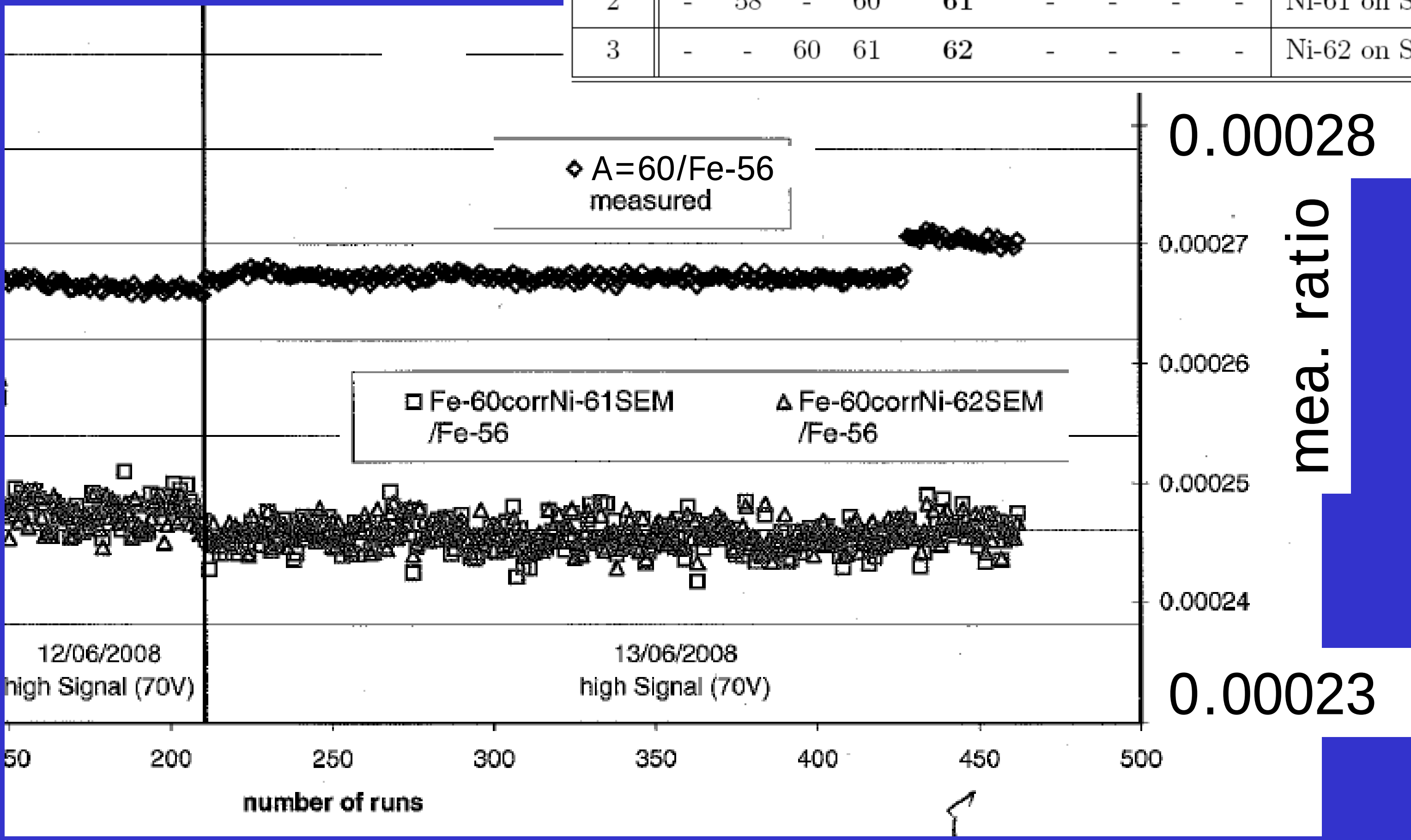
isotope	^{54}Fe	^{56}Fe	^{57}Fe	^{58}Fe
percentage [%]	-	3.11	95.10	1.79

Spiking $r = \frac{57}{56}$	c [µg/g]	Δc [µg/g]	c_N [N ^{60}Fe/g]
1.50	585.7	3.7	$1.2016 \cdot 10^{15}$
1.35	585.5	3.7	$1.2012 \cdot 10^{15}$
1.25	585.5	3.7	$1.2013 \cdot 10^{15}$
0.95	585.8	3.7	$1.2019 \cdot 10^{15}$
0.75	585.6	3.7	$1.2016 \cdot 10^{15}$

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

ICP-MS measurement

Line	L4	L3	L2	L1	C/SEM	H1	H2	H3	H4	Comment
1	-	54	55	56	57	58	60	61	-	
2	-	58	-	60	**61**	-	-	-	-	Ni-61 on SEM
3	-	-	60	61	**62**	-	-	-	-	Ni-62 on SEM



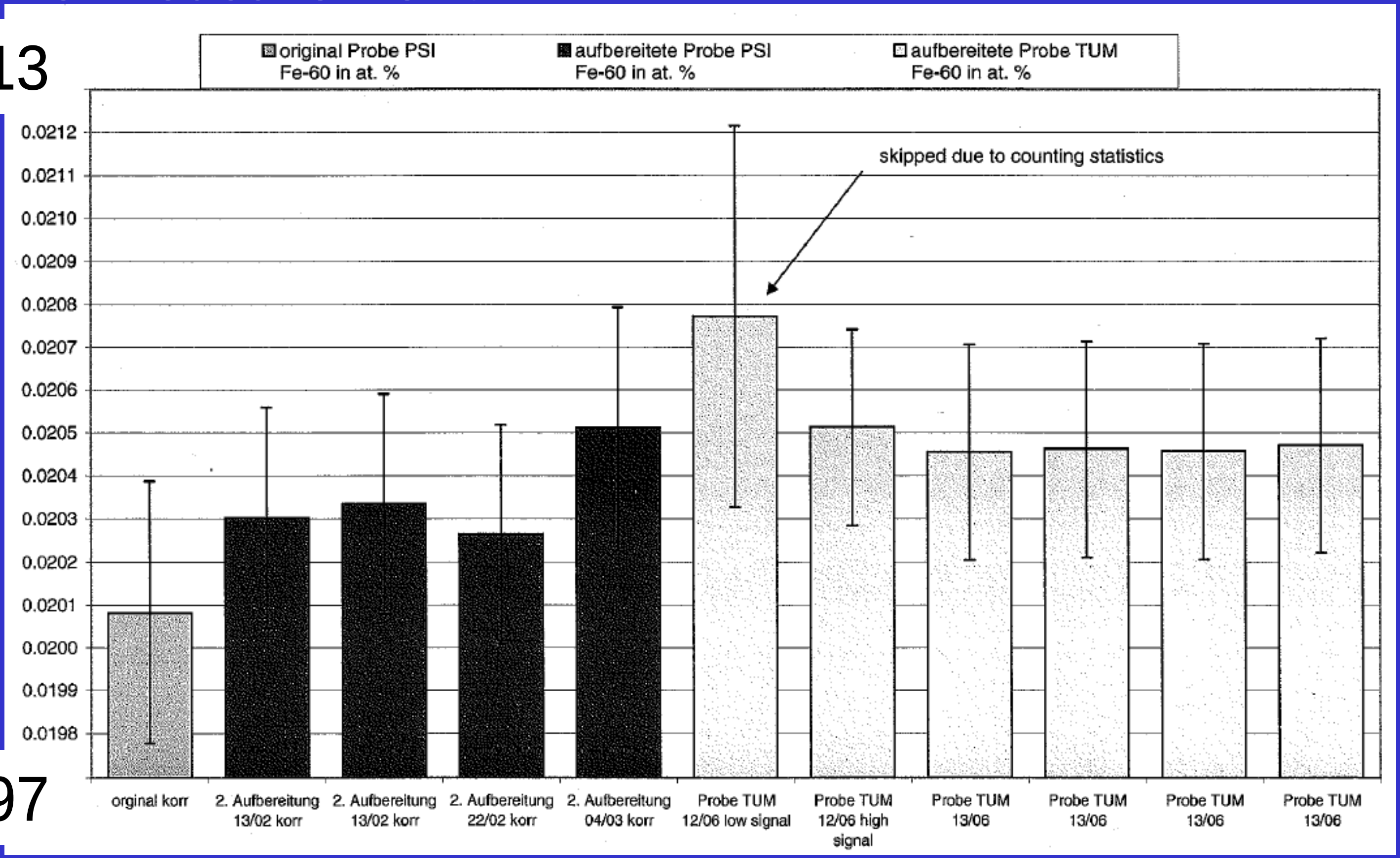
Corrections for: isobar ^{60}Ni, instrumental background; measured in different samples, many runs – consitent results!

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

ICP-MS measurement

0.0213

ratio $^{60}Fe/Fe$



0.0197

ratio $^{60}Fe/Fe$: 2.042×10^{-4}

Combination

$$A = \lambda \cdot N_{rad} = \frac{ln(2)}{T_{1/2}} \cdot \frac{N_{rad}}{N_{stable}} \cdot N_{stable}$$

Preliminary result:

?.? x 10^6 yrs

TOI: $T_{1/2}$(^{60}Fe): (1.49 ± 0.27) Mys