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Reinhard von Ammon,
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Abstract

The overall procedure of the GALLEX-experiment for the measurement of solar neutrinos was checked by an artificial neutrino source consisting of the nuclide ^{51}Cr. A very high activity of this nuclide was produced by neutron-activation of 35.6 kg of metallic chromium enriched in ^{50}Cr to 38.6 %. It decays with a halflife of 27.7 days by electron capture emitting neutrinos of comparable energy to solar neutrinos.

For this check-experiment which was carried out twice, the activity of the ^{51}Cr-source must be known as exactly as possible. The activity-determination was carried out independently by several groups of the GALLEX-collaboration using different methods. At FzK, two methods were applied: dissolution of samples of the irradiated chromium metal in 3 M sulfuric acid and γ-spectrometry of the 320 keV line of ^{51}Cr; and, after almost complete decay of the ^{51}Cr, determination of ^{51}V, the stable decay product of ^{51}Cr, by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) and by Atomic Absorption Spectroscopy (AAS). These methods are described.

The accuracy of γ-spectrometry (±1.5 %) is somewhat better than that of atomic spectroscopy (± 3.0 %). According to these measurements the activity of the second source (average of ^{51}Cr- and V-measurements: 70.6 Pbq) turned out to be higher than that of the first source (64.6 Pbq) by 8.5 %. The results are discussed.

Zusammenfassung

Bestimmung der Aktivität der ^{51}Cr-Neutrinoquelle für das GALLEX-Experiment

Es wurde ein Funktionstest des Gesamt-GALLEX-Experiments zur Messung der Sonnenneutrinos mit einer künstlichen Neutrinoquelle aus ^{51}Cr durchgeführt. Eine sehr hohe Aktivität dieses Radionuklids wurde durch Neutronenaktivierung von 35.6 kg metallischen Chroms, das zu 38.6% an ^{50}Cr angereichert war, erhalten. Es zerfällt durch Elektroneneinfang mit einer Halbwertszeit von 27.7 Tagen und emittiert dabei Neutrinos mit Energien, die denen der Sonne vergleichbar sind.

Für diesen Funktionstest, der zweimal durchgeführt wurde, muß die Aktivität der ^{51}Cr-Quelle so genau wie möglich bekannt sein. Die Aktivitätsbestimmung wurde von mehreren Gruppen der GALLEX-Kollaboration unabhängig voneinander mit verschiedenen Methoden durchgeführt. Im FzK kamen zwei Methoden zur Anwendung: Auflösung des bestrahlten Chrom-Metalls in 3 M Schwefelsäure und einmal γ-Spektrometrie der 320 keV-Linie des ^{51}Cr, sowie, nach fast vollständigem Zerfall des ^{51}Cr, Bestimmung von ^{51}V, dem stabilen Zerfallsprodukt von ^{51}Cr, mittels ICP-AES (Atom-Emissionsspektroskopie mit induktiv gekoppeltem Plasma) und AAS (Atom-Absorptionsspektroskopie). Diese Methoden werden beschrieben.

Die Genauigkeit der γ-Spektrometrie (±1.5%) ist etwas besser als die der Atomspektroskopie (±3.0%). Die Aktivität der zweiten Quelle ergab sich mit 70.6 PBq um 8.5% höher als die der ersten Quelle (64.6 PBq) (Mittel aus ^{51}Cr- und V-Bestimmungen). Die Ergebnisse werden diskutiert.

Contents

1. Introduction

In the GALLEX experiment the flux of solar neutrinos is measured by the reaction

$$^{71}\mathrm{Ga} + \nu_e \longrightarrow {}^{71}\mathrm{Ge} + e^- \qquad (1)$$

in a detector consisting of 101 t of a concentrated (8 M) $GaCl_3$-solution [2]. A large effort was devoted to suppress any side reactions also producing ^{71}Ge, e.g. by a p,n-reaction caused by impurities or by myons from the cosmic radiation [3]. In addition, each third solar neutrino run of four weeks exposition time is accompanied by a blank run of very short exposition time (12 h), in order to detect such reactions.

A still better, actually essential check of the whole procedure of the experiment is the insertion of an artificial neutrino source of known strength into the detector tank. The neutrino flux of this source would have to be much higher (at least ten times at the beginning) than the neutrino flux of the sun at the earth, because the latter cannot be switched off.

^{51}Cr was chosen as the nuclide for this purpose [4]. It decays by electron capture (fig. 1) to ^{51}V with a half-life of 27.706 days by reaction 2:

$$^{51}\mathrm{Cr} + e^- \longrightarrow {}^{51}\mathrm{V} + \nu_e \qquad (2)$$

The neutrino spectrum consists of four monoenergetic lines (746 keV, 81%; 751 keV, 9%; 426 keV, 9%, and 431 keV, 1%) and is thus very similar to the mean energy of the solar neutrinos seen by the GALLEX detector. 90.14% of the ^{51}Cr decay to the ground state of ^{51}V, whereas 9.86% decay to an excited state deexciting to the ground state by emission of a 320 keV γ-ray. This can be used for determining the activity of a sample.

^{51}Cr was produced by neutron activation of ^{51}Cr in the Siloé-reactor at Grenoble. In order to achieve a ^{51}Cr-activity of the source strength necessary, 36.0 kg of metallic chromium enriched to 38.59% of ^{51}Cr (compared to 4.35% in natural chromium) were used. The enrichment was carried out at the Kurchatov Institute (Moscow) by gaseous centrifugation of volatile CrO_2F_2. This compound was subsequently hydrolyzed to CrO_3 and then - in Saclay - electro-precipitated in H_2SO_4-solution on copper electrodes as chromium metal. The isotopic and chemical composition of the chromium is detailed in table 1 [7]. This material, obtained in the form of coarse chips of about 1 mm^3 volume was irradiated in the reactor for a period of three to four weeks. At the end of bombard-ment (E.O.B.) the source was placed into a stainless steel vessel shielded by 8.5 cm of tungsten and shipped to the Gran Sasso Laboratory (LNGS). There it was placed into the thimble in the center of the target tank (fig. 2) for about four months.
In order to know the neutrino flux coming from this source, the activity of ^{51}Cr must be measured as exactly as possible.

This was done directly at the reactor site by measuring the neutron flux (neutronics) and calculating the activity with the cross-section for the n,γ-reaction of ^{50}Cr, by γ-scanning of the 320 keV γ-ray of the source, and by measuring the temperature increase in a calorimeter of known heat capacity (4, 6).

In addition, samples of the source material were taken immediately after irradiation. It was attempted to make these samples as representative for the whole source as possible [6].

The activity of these samples was measured at Saclay using an ionization chamber technique [4, 6]. Thereafter they were shipped to the Forschungszentrum Karlsruhe (FzK) for further chemical treatment and analysis.

While the source stayed in the target tank, a series of about ten runs of varying exposition time was carried out [4].

This whole procedure was carried out twice (the two experiments are furtheron called „source 1“ and „source 2“ [7]).

In the following sections, the dissolution of the samples is described as well as the subsequent analysis of the resulting solution (γ-spectrometry, determination of chromium by ICP-AES, and vanadium by ICP-AES and AAS). The results are discussed.

2. Dissolution of the Chromium Samples

Although chromium is a fairly non-noble metal (electrochemical potential Cr/Cr^{+3}: -0.744 Volt, close to zinc), its dissolution in non-oxidizing acids like H_2SO_4 is inhibited by passivation. Depassivation can be accelerated strongly by heating. Even then, the inhibition time may take hours or even days. Addition of small amounts of a reducing agent like metallic zinc or hydroxylamine usually initiates the dissolution immediately.

2.1 Source 1

In order to have reproducible conditions, we carried out the dissolution of the samples of source 1 in 3 M H_2SO_4 under heating to approximately 80°C and with addition of 0.2ml of 2.5 M hydroxylamine-hydrochloride solution. Dissolution of the 1g-samples in 20 ml of acid was complete after about three hours. Only the first four small samples which - because of the comparatively short cooling time - had to be treated in a hot cell where no heating device was available, were dissolved with addition of a grain of zinc metal. Since Zn^{+2} -ions might disturb other analytical determinations in the resulting solutions we refrained from using this reducing agent furtheron.

The 20 ml-solutions were filled up to 100 ml. This „master-solution“ (0.6 M H_2S0_4) was further diluted by a factor of 10^3 for analysis of Cr and by a factor of 10^4 for γ -measurements. H_2SO_4-concentration was kept 10^{-3} M for both measurements.

Aliquots of the master solution were also sent to MPI Heidelberg and to BNL (Brookhaven, U.S.A.) for parallel analyses.

2.2 Source 2

The larger sample of source 2 (41.4 g) was divided in the hot cell into three samples between 8 and 19 g. These were dissolved only by heating without addition of a reducing agent. A cooler on top of each 1 l-dissolver flask was meant to prevent any spilling of solution in case of a very vigorous reaction.

Dissolution in 200 ml of 3 M sulfuric acid started with one sample almost immediately, with another sample about 30 minutes after a temperature of about 70° C was reached, and with the third one about five hours after start of heating. The dark green color indicated the formation of Cr^{+3}-ions.

The dark color makes it difficult to visualize the complete end of dissolution, especially in a hot cell. This led us, with two of the three samples of source 2, to finish the dissolution after four hours, especially since no gas formation was visible any more. (In several test dissolutions of unirradiated chromium samples four hours had been sufficient for complete reaction as evidenced by analysis).

The 200 ml of solution were filled up to the one-liter-mark and well mixed. A 5 ml-sample was then taken and a stopcock put on top of the flask.

The third flask was left standing undiluted over the weekend. To our surprise, the two closed flasks were broken afterwards and their content was lost. Apparently we had stopped the reaction too soon and dissolution slowly proceeded whereby an overpressure of hydrogen had developed in the small gaseous volume in the neck of the flasks. Possibly also radiolysis played a role.

The third sample in the open flask was diluted as the others and a 5 ml-sample of the resulting „master-solution“ (0.6 M H_2SO_4) was taken. Aliquots of the 5 ml-samples were further diluted by a factor of $8{\cdot}10^3$ - $2{\cdot}10^4$ for the analytical measurements.

Aliquots were also sent to MPI Heidelberg and to BNL (Brookhaven, U.S.A.).

3. Determination of the Total Chromium Content

In order to check the dissolution yield of the chromium metal, the diluted solutions were analyzed for chromium by ICP-AES (Inductively Coupled Plasma Atomic Emission Spectroscopy). The measurements were carried out at a wavelength of 267.716 nm, at a atomizing pressure of 200 kPa and with a stabilizing time of 20 sec (spectrometer type Liberty 150, Varian).

3.1 Calibration

Each sample was measured three times and calibrated immediately after the third measurement. Two standards were made by dissolving metallic chromium (99.99%, Roth) in H_2SO_4 (0.8114 μg/ml and 1.198 μg/ml). A calibration curve for various dilutions of one of the standards is shown in fig. 4.

In order to increase the reliability of the measurements, calibrations were also carried out with two commercial standards (Merck and Johnson), both with a concentration of 1.0000 μg/ml and a H_2SO_4-acidity of 0.01 M. These standards agreed with the calibration curve (fig. 4) within ±1.0%.

The volumes of the pipettes used for sampling and diluting were calibrated by weighing. The standard deviation of ten consecutive weight determinations was ±0.33%, thus proving again that the error introduced by the large dilution was negligible.

3.2 Source 1

The results of the 27 samples of the first source are shown in table 2. The acidity of the diluted samples was $6 \cdot 10^{-3}$ M. The percentages of dissolution (referred to the chromium amount weighed in) are given in column 4. The mean dissolution yield percentage (101.1 ± 1.5%) proves that dissolution was complete in all samples (see also fig. 5).

3.3 Source 2

The Cr-analyses of the samples of source 2 (table 3a) were lower than the expectation value from the sample weight thus confirming our suspicion that dissolution had been incomplete. This was proven by sample 2 which, after a standing time of four weeks in the hot cell at rom temperature showed a Cr-yield of 100% within the uncertainty of the method (±3%) (table 4).
The sampling of the „master solution" was done with various pipets of different size (aliquots), thus proving that the error of dilution was negligible compared to the error of analysis (tables 3 and 4).

4. Determination of the Gamma-Activity of ^{51}Cr

4.1 Source 1 [4, 6]

Theoretically, the γ-activity after irradiation is given by the activation formula

$$A = \sigma \cdot \Phi \cdot N\,(1 - e^{-\lambda \cdot t})\ [\mathrm{Bq/g}] \qquad (3)$$

Here, σ, the cross section of the reaction = $17.2 \cdot 10^{-24}$ cm^2,

Φ, the average neutron flux of the reactor = $5.2 \cdot 10^{13}$ n/cm$^2 \cdot$ s,

N, the number of ^{50}Cr-atoms in 1 g of chromium = $N_L \cdot H/G$,
where N_L, the Loschmidt-number = $6.02 \cdot 10^{23}$,
H, the ^{50}Cr - abundance in the enriched chromium = 0.386,
G, the average molar weight of the sample = 51.235,
λ, the decay constant of ^{51}Cr = $2.896 \cdot 10^{-7}$ sec^{-1}, and
t, the irradiation time = $2.056 \cdot 10^6$ s.

With these numbers, eqn. 3 renders an expectation value for the specific activity of **A = 1.82 $\cdot 10^{12}$ Bq/g.**

Except for four samples which were dissolved in a hot cell and were measured 3.5 months after end of bombardment (E.O.B.) (halflife of ^{51}Cr: 27.706 days) all samples were treated and measured after about 7 months outside of the hot cell. The activity had fallen down by then by more than two orders of magnitude.

After dilution of a 1 ml-aliquot of the master solution by a factor between $8 \cdot 10^3$ and $2 \cdot 10^4$, the γ-spectrum of one ml of this solution in a 5 ml sample tube with a screw cap was measured on a germanium (puriss.) detector (model 7229P-7500-3019, Canberra) with a relative efficiency of 32%. Its energy resolution is 1.80 keV at an energy of 1.33 MeV, and 0.923 keV at an energy of 0.122 MeV (data given by the supplier).
The intensity of the 320 keV γ-line of ^{51}Cr (emission probability 9.83%) was determined.

The detector was calibrated with the mixed standard No. QCY44 (Amersham) containing the radionuclides ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{203}Hg, ^{85}Sr, ^{137}Cs, ^{60}Co and ^{88}Y with γ-energies ranging from 88.0 keV to 1836 keV. The uncertainties of this standard at the different energies vary between ±1 and ±5%. According to this calibration the efficiency of the detector for the γ-line at 320 keV for the sample in 15 cm distance was 0.191%; the energy dependence of the efficiency is shown in fig. 6.

All samples were measured for 1000 sec, the error of the count rate was always ±1%. A typical γ-spectrum which was recorded eight months after E.O.B. is shown in fig. 7, including the 320 keV-line of ^{51}Cr and lines of the longer living impurities ^{75}Se (halflife 119.64d) and ^{110m}Ag (halflife 249.9d). The concentration of these nuclides at E.O.B. on the average amounts to 0.8 ppb (^{75}Se) and 1.2 ppb (^{110m}Ag). Obviously the concentration of their activation precursors must have been far below the detection limit of standard analytical methods (see e.g. table 1).

The data were evaluated with the program Spectran-F (Canberra). Activity values were referred to the date of E.O.B. (6 a.m., June 20, 1994).

Since the Cr-analysis had proven that dissolution of all 27 samples was complete (table 2), the γ-activities measured had not to be corrected. The individual error of the samples is relatively large (±6.8 %) which is caused by the poor statistics of the relatively coarse grains of the material and their non-uniformity (each sample contained approximately 100 grains). The standard deviation of the measurements alone should be much smaller (±1-2%).

The average result **(1.777 · 10^{12} Bq/g) corresponding to 63.12 PBq of the whole source (35530 g**) compares very well with the results of other groups, obtained either by γ-spectroscopy or by other methods [4, 6]. It also compares quite well with the expectation value (1.82 · 10^{12} Bq/g).

4.2 Source 2 [7]

The irradiation time was 26.526 days, i.e. longer than with source 1 by 2.73 d or 11%. In addition to that, the neutron flux during irradiation was 5.6 · 10^{13} n/cm^2·s or higher by 7%. Thus, a higher expectation value of the activity of the source results, namely:

$$A = \sigma \cdot \Phi \cdot N\,(1 - e^{-\lambda \cdot t}) \qquad (3)$$

$$A = 17.2 \cdot 10^{-24} \cdot 5.6 \cdot 10^{13} \cdot 4.5369 \cdot 10^{21} \cdot 0.4850 \qquad (3.1)$$

$$\mathbf{A = 2.12 \cdot 10^{12}\ Bq/g} \qquad (3.2)$$

Inspection of table 3 reveals that the experimental numbers are indeed higher than the activity of the first source, although the expectation value is not quite reached. The activity values which can be considered best and most reasonable are those of table 4, where no correction for an uncomplete yield of chromium had to be applied. The average of three determinations is **(1.985 ± 0.03)·10^{12} Bq/g**.

This value can best be compared to sample 2 of table 3 which, after correction for uncomplete (93.4%) dissolution gives **(1.966 ± 0.06) ·10^{12} Bq/g** (the larger error results from the correction for incomplete dissolution). The average of both numbers is **(1.9755 ± 0.05)** · **10^{12} Bq/g or, for the whole source, (70.19 ± 1.76)** · **10^{15} Bq** (the total weight of source is 35.60 kg, i.e. heavier than source 1 by 70 g).

If the average of all three samples weighted according to their weights is taken (see tables 3 and 4), a **specific activity of 1.938 TBq/g is obtained or an overall source strength of 69.00 PBq.**

5. Evaluation of the ^{51}Cr-Activity by Determination of its Daughter ^{51}V

^{51}Cr decays by electron capture to stable ^{51}V (fig. 1). Thus, after complete decay of ^{51}Cr, the ^{51}V-content should be directly proportional to the activity of ^{51}Cr originally present. However, three particularities have to be taken into account: first, it has to be ascertained that no vanadium is present at the beginning, or that its content is known precisely.

Second, the ^{51}Cr which has decayed already during irradiation has to be considered; and third, the vanadium-content after complete decay of the ^{51}Cr activated in source 2 is the sum of the ^{51}Cr-activities of both irradiations.

5.1 Activation and Decay of ^{51}Cr During Irradiation

The total activity of ^{51}Cr formed until E.O.B. in the Siloé-reactor, including the ^{51}Cr already decayed is given by the activation formula

$$A(^{51}\mathrm{Cr}) = \sigma \cdot \Phi \cdot N \cdot \lambda \cdot t \qquad (4)$$

where the symbols have the meaning described in section 4.1. Here the last term in eqn. 3, $(1 - e^{-\lambda \cdot t})$ is replaced by $\lambda \cdot t$, because <u>all</u> the ^{51}Cr formed decays to the stable end-product ^{51}V to be analyzed.

5.1.1 Source 1

From equation 4 an expectation value for the ^{51}Cr-activity of

$A(^{51}\mathrm{Cr}) = 2.4153 \cdot 10^{12}$ Bq/g is obtained, corresponding

to the number of ^{51}Cr- or ^{51}V-nuclei of

$$N(^{51}\mathrm{Cr}{=}^{51}\mathrm{V}) = \frac{A}{\lambda} = 0.8343 \cdot 10^{19}/\mathrm{g}$$

and the mass ratio of ^{51}V of

$$m(^{51}\mathrm{V}) = \frac{N(^{51}V) \cdot M}{N_L} = 7.066 \cdot 10^{-4}\ \mathrm{g/g} \text{ or } \mathbf{706.6\ ppm.}$$

Here M = molar weight of ^{51}V in g.

The ^{51}Cr-activity still present at E.O.B. is less than that given by eqn. 4, namely the activity given by eqn. 2.

As derived in section 4.1, the expectation value for this activity is $A(^{51}\mathrm{Cr}) = 1.82 \cdot 10^{12}$ Bq/g.

The ratio $A_{tot}(^{51}\mathrm{Cr})/A(^{51}\mathrm{Cr}) = \lambda \cdot t/(1 - e^{-\lambda \cdot t}) = 1.327$ indicates that during the irradiation time of 23.8 d roughly 1/3 of the ^{51}Cr formed has already decayed [8].

5.1.2 Source 2

The ^{51}V-content after complete decay of source 2 can be calculated accordingly. With the altered values for Φ and t eqn. 4 yields

$A(^{51}Cr) = 17.2 \cdot 10^{-24} \cdot 5.6 \cdot 10^{13} \cdot 4.5369 \cdot 10^{21} \cdot 2.8956 \cdot 10^{-7} \cdot 2.2918 \cdot 10^{6}$

$= 2.90 \cdot 10^{12}$ Bq/g

Accordingly $N(^{51}Cr{=}^{51}V) = 1.0015 \cdot 10^{19}/g$

and **$m(^{51}V) = 848.5$ ppm.**

This value has to be added to the ^{51}V-concentration formed during the irradiation of source 1, so that a total vanadium-concentration results after complete decay of the ^{51}Cr of source 2 of **1 555.1 ppm or 0.155 % by weight.**

5.2 Determination of Vanadium

The vanadium content of the solutions was determined by two methods: Atomic Absorption Spectrometry (AAS) and Inductively Coupled Plasma Atomic Emission Spectrometry (ICP-AES).

5.2.1 Atomic Absorption Spectrometry (AAS)

The spectrometer used was a type AA-400 (Varian). Measurements were carried out under the following conditions: wave length 318.5 nm, slit 0.2 nm, lamp current 7 mA, sensitivity $28 \cdot 10^{-12}$ g. The graphite tube technique was applied, whereby the graphite was treated with a Pd-Mg modifier at 150°C. Incineration temperature was 1600°C and the atomizing temperature 2700°C.

Each sample was measured ten times with a standard deviation of generally around ±1%.

Since it is essential that the standard is measured in a matrix identical with that of the sample, the following standard solutions were prepared from the commercial vanadium-standard (50 ng/ml; Merck):

Cr 120 μg/ml, H_2SO_4 0.03 M (source 1); Cr 120 μg/ml, H_2SO_4 0.01 M (source 2).
The calibration curve obtained with various amounts of one of these standard solutions (source 1) is shown in fig. 9.

5.2.2 Atomic Emission Spectrometry with Inductively Coupled Plasma (ICP-AES)

The spectrometer used was a type Liberty 150 (Varian). Measurements were carried out at a wavelength of 309.311 nm, an atomizing pressure of 200 kPa and with a stabilizing time of 20 seconds. Each sample was measured ten times with a standard deviation around ±1%.

5.3 Results

5.3.1 Source 1

The vanadium data for the 27 samples obtained with AAS and ICP-AES are presented in table 5. The results of both methods are very well consistent within ±0.3%. The standard variation of the ICP-AES-data (average: 719.6 ppm ±8.5%) is somewhat higher than of the AAS-data (722.2 ppm ±6.7%), but both numbers compare well with the variation of the γ-spectrometric measurements (see section 4.1).

The average vanadium mass ratio as determined by both methods is:

720.9 ppm ± 7.6%.

This value is to be compared to the theoretical expectation value of **706.6 ppm** (section 5.1.1) and to the average experimental γ-spectrometric value of 1.777 TBq/g (section 4.1) = 519.8 ppm $\cdot$1.327 = **689.8 ppm.** This means that the vanadium-result is higher than the γ-spectrometric result by 4.3% and higher than the theoretical prediction by 2.0%.

In fig. 8 the vanadium-data of all 27 samples are compared graphically with each other and with the γ-spectrometric data.

The activity value for source 1 at E.O.B., as converted from the vanadium-content is **(1.857 ± 0.06) TBq/g**. For the whole source this amounts to **(66.0 ± 2.0) PBq**.

5.3.2 Source 2 [7]

Three different dilutions of sample 2 were measured with ICP-AES. The results are listed in table 6. The average value is **1521.5 ppm**. This vanadium-content is the sum of sources 1 and 2. So the value obtained for source 1 (720.9 ppm) has to be subtracted to obtain the vanadium formed in source 2. It amounts to 800.6 ppm.

At the time of analysis, only 98.54% of the ^{51}Cr had decayed. The final, corrected value thus is **812.5 ppm**.

Again, this value is to be compared with the theoretical expectation value of 2.119 TBq/g and the average experimental γ-spectrometric value of 1.974 TBq/g or 1.938 TBq/g, if the weighted average of all samples of source 2 is taken (see section 4.2.2).

To convert these ^{51}Cr-activity numbers to vanadium-concentrations, the altered conditions of source 2 have to be taken into account: the longer irradiation time and the higher neutron flux cause the factor $F = \lambda \cdot t/(1 - e^{-\lambda \cdot t})$ to increase to 1.368.

Thus, the theoretical expectation value of the vanadium-content is **847.9 ppm**, and the experimental γ-activity-values correspond to vanadium-concentrations of **789.9 ppm or 775.5 ppm, respectively**. So, the experimental vanadium-value (812.5 ppm) is higher than the γ-activity-values by 2.8 or 4.5%, respectively, but lower than the theoretical value by 4.2%.

The ^{51}Cr-activity of source 2, as calculated from the experimental vanadium-determination (812.5 ppm) is

(2.030 ± 0.06) TBq/g and, for the whole source (35.6 kg) (72.28 ± 2.2) PBq.

6. Comparison of the Two Sources

As discussed in sections 4.1 and 4.2, the expectation value for the activity of source 2 is higher than that of source 1 because of the higher average neutron flux and the longer irradiation time. The values calculated (eqn. 3) are 1.82 TBq/g and 2.12 TBq/g, respectively, differing by 14.2 %. Experimentally, such a large difference could not be verified: If the results of γ-spectrometry and V-determination are averaged, 1.817 TBq/g (source 1) and 1.984 TBq/g (source 2) are obtained with a difference of 8.4%.
For comparison, an overview of all relevant results is given in table 7.

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W. Hampel, G. Heusser, J. Kiko, T. Kirsten, M. Laubenstein, E. Pernicka, U. Rönn, M. Sann, R. Wink, M. Wojcik
Max-Planck-Institut für Kernphysik (MPIK), Postfach 103980, D-69029 Heidelberg, Germany
R. v.Ammon, K.H. Ebert, T. Fritsch, D. Heidt, E. Henrich, L. Stieglitz, F. Weirich
Institut für Technische Chemie, Forschungszentrum Karlsruhe (FzK), Postfach 3640, D-76021 Karlsruhe, Germany
M. Balata
INFN - Laboratori Nazionali del Gran Sasso (LNGS), S.S. 17/bis km 18+910, I-67010 L´Aquila, Italy

E. Bellotti, C. Cattadori, O. Cremonesi, N. Ferrari, E. Fiorini, L. Zanotti
Dipartimento di Fisica, Universita di Milano e INFN - Sezione di Milano, Via Celoria 16, I-20133 Milano, Italy
M. Altmann, F. v.Feilitzsch, R. Mößbauer
Physik Department E15, Technische Universität München (TUM), James-Franck Straße, D-85748 Garching b. München, Germany
G. Berthomieu, E. Schatzman
Observatoire de la Cote d'Azur, Departement Cassini, B.P. 229, 06004 Nice Cedex 4, France
I. Carmi, I. Dostrovsky
Department of Environmental and Energy Research, The Weizmann Institute of Science (WI), P.O. Box 26, 76100 Rehovot, Israel
C. Bacci, P. Belli, R. Bernabei, S. d'Angelo, L. Paoluzi
Dipartimento di Fisica, II Universita di Roma „Tor Vergata" e INFN - Sezione di Roma 2, Via della Ricerca Scientifica, I-00133 Roma, Italy
A. Bevilacqua, M. Cribier, L. Gosset, J. Rich, M. Spiro, C. Tao, D. Vignaud
*DAPNIA, CEA Saclay, F-911*91 Gif-sur-Yvette Cedex, France
J. Boger, R.L. Hahn, F.X. Hartmann, J.K. Rowley, R.W. Stoenner, J. Weneser
Brookhaven National Laboratory (BNL), Upton, NY 11973, USA

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Captions of the Figures and Tables

Fig. 1: Decay scheme of ^{51}Cr.

Fig. 2: Scheme of the GALLEX A-tank containing the $GaCl_3$-detector-solution (53.5 m^3) and the ^{51}Cr-source inside the central tube.

Fig. 3: Calibration curve for the determination of chromium by ICP-AES (267.716 nm).

Fig. 4: Chromium-yields of the dissolution of the 27 samples of source 1.

Fig. 5: Efficiency of the Ge-detector depending on the γ-energy (distance of sample from detector: 15 cm).

Fig. 6: Typical γ-spectrum of a ^{51}Cr-sample, after eight months of standing time; lines of ^{75}Se (119.64 d) and ^{110m}Ag (249.9 d) are also seen.

Fig. 7: Comparison of the data on vanadium-analysis (AAS and ICP-AES) and γ-activity of the 27 samples of source 1.

Fig. 8: Calibration curve for the determination of vanadium by AAS.

Table 1: Isotopic and chemical composition of the chromium source [5].

Table 2: Chromium-concentration and γ-activity of the 27 samples of source 1.

Table 3: Chromium-concentration and γ-activity of the three samples of source 2 (H_2SO_4-concentration: 10^{-2} M).

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Table 5: Vanadium-concentration of the 27 samples of source 1 as analyzed by AAS and ICP-AES.

Table 6: Vanadium-concentration of the three samples of source 2 as analyzed by ICP-AES.

Table 7: Overview of the results on the ^{51}Cr-source-strength of both chromium sources, obtained with γ-spectrometry and with determination of vanadium.

Isotopic composition of chromium (%)

Isotope	^{50}Cr	^{52}Cr	^{53}Cr	^{54}Cr
natural	4.35	83.8	9.5	2.35
source	38.6	60.7	0.7	<0.3

Chemical composition of the chromium of the source

Element	Al	B	Ca	Cu	Fe	K	Mg	Mn	Na	Ni	P	S	Si	Ti	Zn
µg/g	70	4	125	6	50	15	10	20	<10	2.5	3	20	50	6	5

Table 1: Isotopic and chemical composition of the chromium source [5]

Sample No.	Weight [g]	Conc. of Solution [µg/ml]	Cr-Yield [%]	γ-Activity [TBq/g]		
				Individual Samples	Mixtures	
1/2	0.7055	7.18	101.8	1.893	1.816	1.800
1/3	0.7086	7.11	100.3	1.631		
1/4	0.9395	9.66	102.8	1.933		
1/5	0.7574	7.72	101.9	1.769		
1/7	0.9855	9.84	99.8	1.735		
1/8	0.9689	9.93	102.5	1.775		
1/9	0.8100	8.25	101.8	1.790		
2/2	0.4338	4.50	103.8	1.938	1.845	
2/3	0.4327	4.47	103.4	2.050		
2/4	0.4708	4.86	103.3	1.792		
2/5	0.6601	6.71	101.6	1.795		
2/6	0.7322	7.43	101.5	1.635		
2/7	0.6740	6.92	102.6	1.854		
2/9	0.6617	6.71	101.4	1.698		
3/2	0.3590	3.66	102.0	1.660	1.778	
3/3	0.6680	6.67	99.8	1.616		
3/4	0.6768	6.95	102.7	1.699		
3/5	0.6499	6.61	101.7	1.708		
3/6	0.7626	7.94	104.1	1.757		
3/8	0.7440	7.81	103.9	1.747		
3/9b	0.7041	7.31	103.9	1.903		
4/3	0.6546	6.68	102.0	2.115	1.792	
4/4	0.6087	6.24	102.6	1.697		
4/5	0.8387	8.51	101.4	1.764		
4/7	0.9825	10.17	103.5	1.720		
4/8	0.8325	8.49	102.0	1.753		
4/9b	0.7065	7.22	102.2	1.627		
		Mean	**102.2 ±1.5%**	**1.776**		

Table 2: Chromium-concentration and γ-activity of 27 samples of source 1

Sample No.	Dilution [ml/ml]	γ-Activity [TBq/g]	Cr-Yield [%]	γ-Activity corr. [TBq/g]
1 (14.1135 mg/ml)	1 : 100; 1 : 100 $= 1 : 10^4$	1.867	–	
	0.1 : 100; 5 : 100 $= 1 : 2 \cdot 10^4$	1.849	99.07	
	0.06 : 100; 10 : 100 $= 1 : 1.67 \cdot 10^4$	1.856	97.80	
	0.05 : 100; 10 : 100 $= 1 : 2 \cdot 10^4$	1.870	98.63	
Mean		**1.860**	**98.50**	**1.889**
2 (19.1914 mg/ml)	1 : 100; 1 : 100 $= 1 : 10^4$	1.842	–	
	0.1 : 100; 5 : 100 $= 1 : 2 \cdot 10^4$	1.829	93.93	
	0.06 : 100; 10 : 100 $= 1 : 1.67 \cdot 10^4$	1.829	93.93	
	0.05 : 100; 10 :100 $= 1 : 2 \cdot 10^4$	1.849	92.46	
Mean		**1.837**	**93.44**	**1.966**
3 (8.1315 mg/ml)	1 : 100; 1 : 100 $= 1 : 10^4$	1.829	–	
	0.1 : 100; 10 : 100 $= 1 : 10^4$	1.795	92.97	
	0.06 : 100; 20 : 100 $= 1 : 8.33 \cdot 10^3$	1.795	93.19	
	0.05 : 100; 20 : 100 $= 1 : 10^4$	1.807	93.95	
Mean		**1.806**	**93.37**	**1.935**

Table 3: Cr- concentration and γ-activity of the three samples of source 2; (H_2SO_4-concentration: 10^{-2} M).

Sample No.	Dilution [ml/ml]	γ-Activity [TBq/g]	Cr-Yield [%]	γ-Activity corr. [TBq/g]
2 (19.1914 mg/ml)	1:100; 1:100 $= 1 : 10^4$	1.995	101.3	
	0.1 : 100; 10:100 $= 1 : 10^4$	1.969	101.4	
	0.05:100; 10:100 $= 1 : 2 \cdot 10^4$	1.990	100.2	
Mean		**1.985**	**101.0**	**1.985**

Table 4: Cr-concentration and γ-activity of sample 2 of source 2 after four weeks of standing (H_2SO_4-concentration: 10^{-2} M).

Sample No.	Weight [g]	Vanadium- Content			
		AAS		ICP- AES	
		[ng/ml]	[ppm]	[ng/ml]	[ppm]
1/2	0.7055	54.8	776.7	53.5	758.2
1/3	0.7086	47.7	673.2	46.6	658.3
1/4	0.9395	72.2	768.5	74.2	789.5
1/5	0.7574	55.2	728.8	52.2	689.7
1/7	0.9855	71.1	721.5	69.5	705.2
1/8	0.9689	68.8	710.1	66.6	688.0
1/9	0.8100	59.7	737.0	57.6	711.5
2/2	0.4338	35.0	806.8	35.7	823.2
2/3	0.4327	35.1	811.2	38.0	878.0
2/4	0.4708	35.8	760.4	35.7	758.1
2/5	0.6601	47.5	719.6	45.4	688.5
2/6	0.7322	47.5	648.7	48.0	654.9
2/7	0.6740	47.6	706.2	47.6	706.2
2/9	0.6617	43.9	663.4	45.6	689.1
3/2	0.3590	25.3	704.7	24.1	672.4
3/3	0.6680	44.4	664.7	44.1	659.6
3/4	0.6768	48.2	712.2	47.1	696.2
3/5	0.6499	45.8	707.7	44.7	688.4
3/6	0.7626	53.9	706.8	54.8	718.1
3/8	0.7440	54.3	729.8	54.2	729.0
3/9b	0.7041	55.2	784.0	55.0	781.7
4/3	0.6546	55.5	847.8	57.2	874.1
4/4	0.6087	43.1	708.1	42.7	701.3
4/5	0.8387	58.6	698.7	58.4	696.8
4/7	0.9825	67.8	690.1	67.2	683.5
4/8	0.8325	58.7	705.1	59.0	708.2
4/9b	0.7065	48.5	686.5	46.5	657.9
	weighted	**average**	**722.2**		**719.6**
average	**of both**	**methods**	**720.9 ppm**		

Table 5: Vanadium-concentration of the 27 samples of source 1

Sample No.	Dilution [mg/ml]	Vanadium- Content	
		[µg/ml]	[ppm]
2 (19.1914 mg/ml)	1:100; 1:10 = 1:10^3	28.50	1485.0
	0.1:100 = 1:10^3	30.16	1571.5
	0.05:100 = 1:2 · 10^3	28.94	1508.0
Mean		29.2	1521.5
V-content of 1st source (720.9 ppm) subtracted		**800.6 ppm**	
corr. for in-complete decay of ^{51}Cr (98.54%)		**812.5 ppm**	

Table 6: Vanadium-concentration of sample 2 of source 2, as determined by ICP-AES

	Source 1		Source 2	
	[TBq/g]	Whole source [PBq]	[TBq/g]	Whole source [PBq]
Theory	1.82	64.7	2.12	75.5
γ-Spectrometry	1.777	63.1	1.938	69.0
V-Analysis	1.857	66.0	2.030	72.3
Average of both methods	1.817	64.6	1.984	70.6

Table 7: Overview of the results on the ^{51}Cr-source-strength of both chromium-sources, obtained with γ-spectrometry and determination of vanadium.

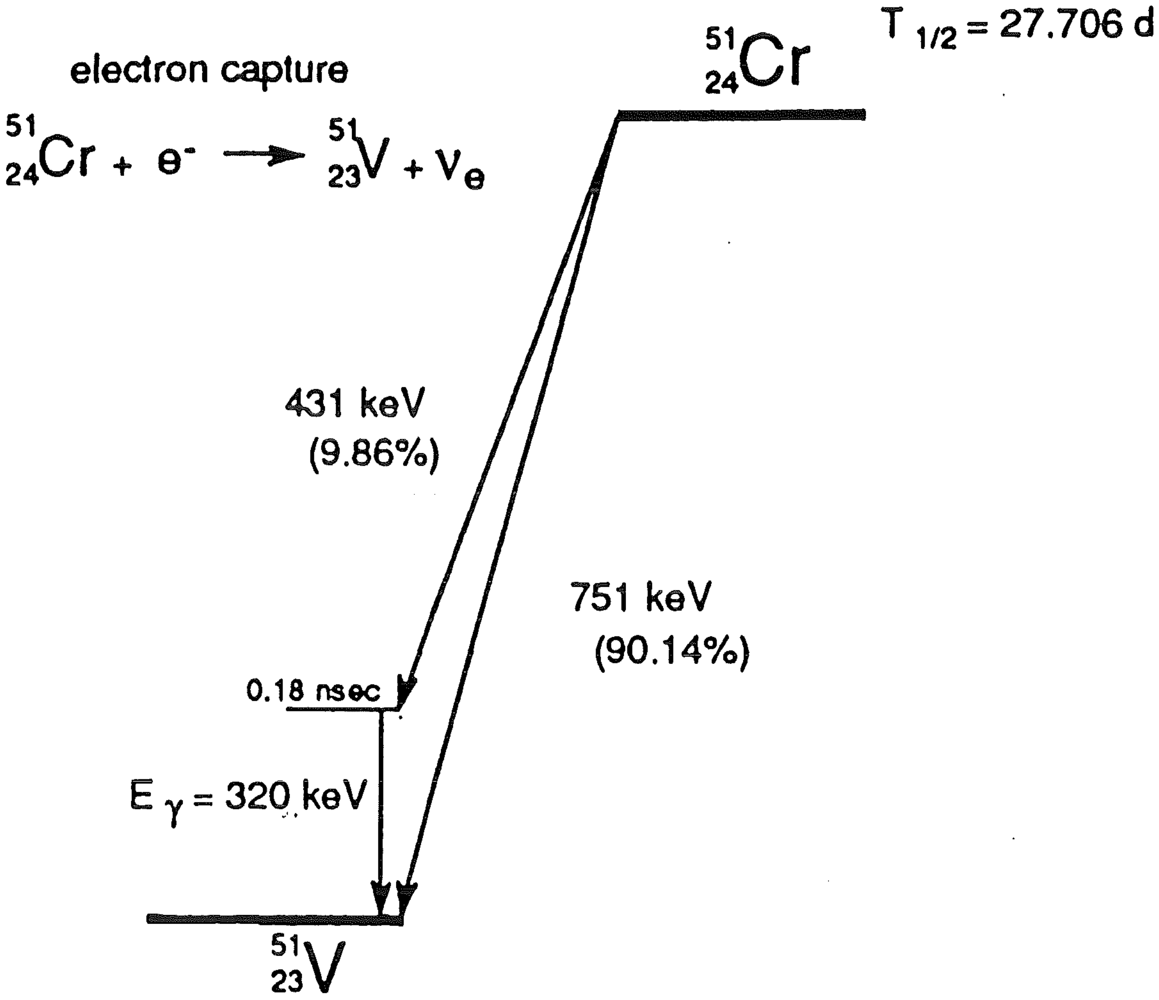


Fig. 1: Decay scheme of ^{51}Cr.

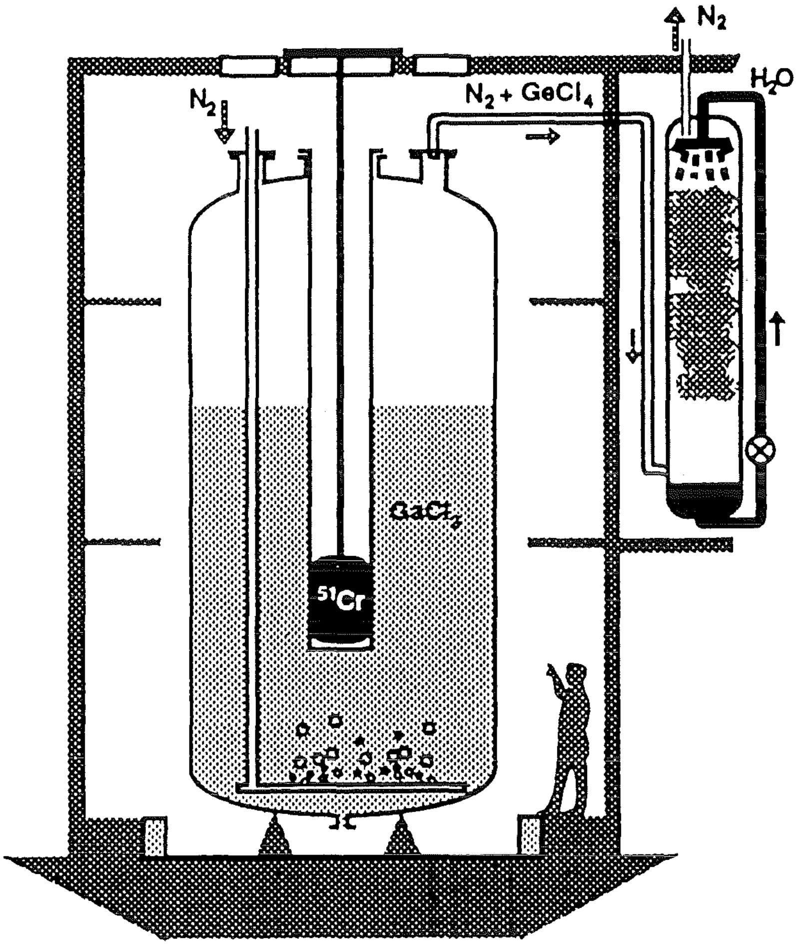


Fig. 2: Scheme of the GALLEX A-tank containing the $GaCl_3$-detector-solution (53.5 m^3) and the ^{51}Cr-source inside the central tube.

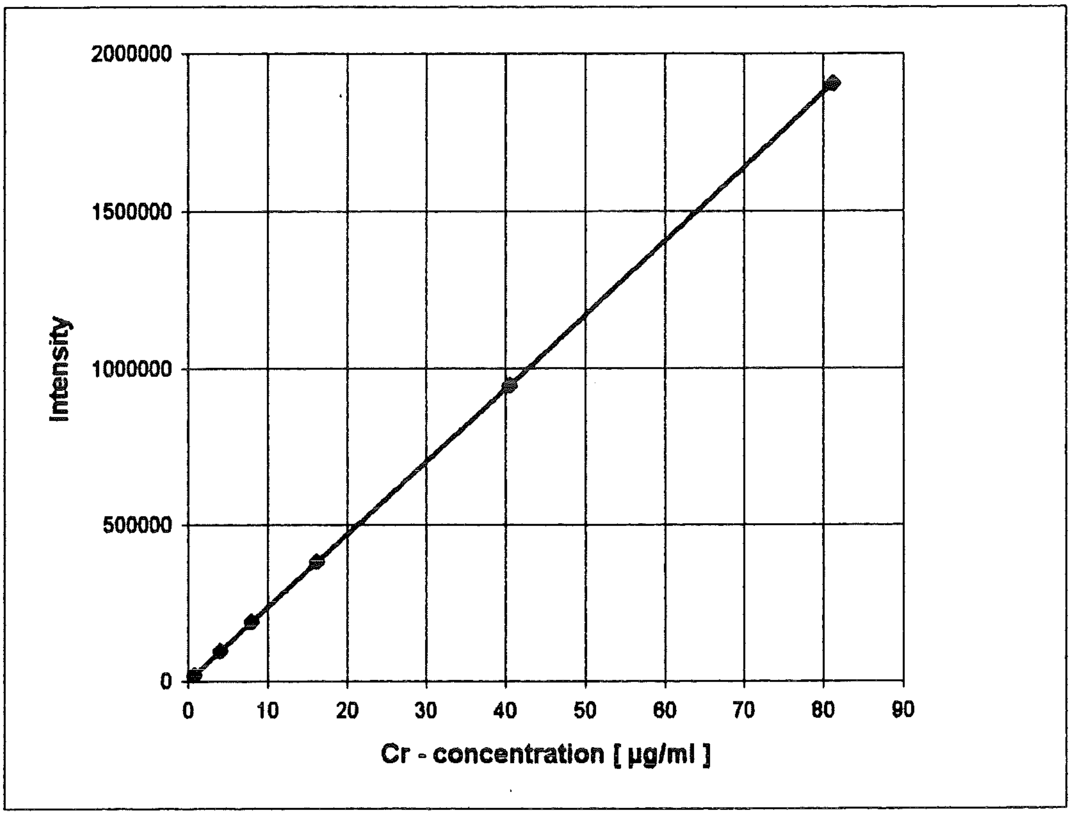


Fig. 3: Calibration curve for the determination of chromium by ICP - AES (267.716 nm)

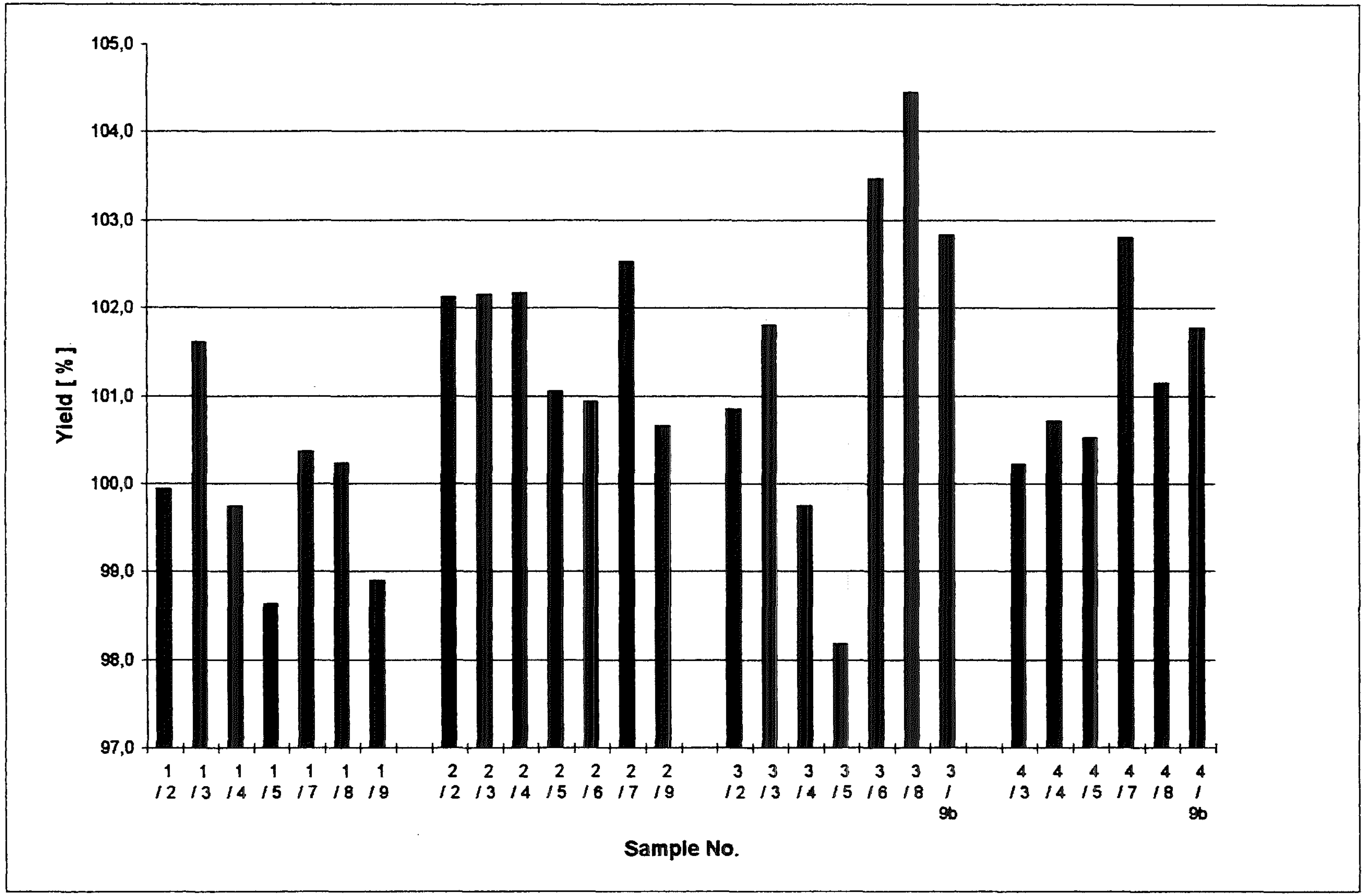


Fig. 4: Chromium - yields of the dissolution of the 27 samples of source 1.

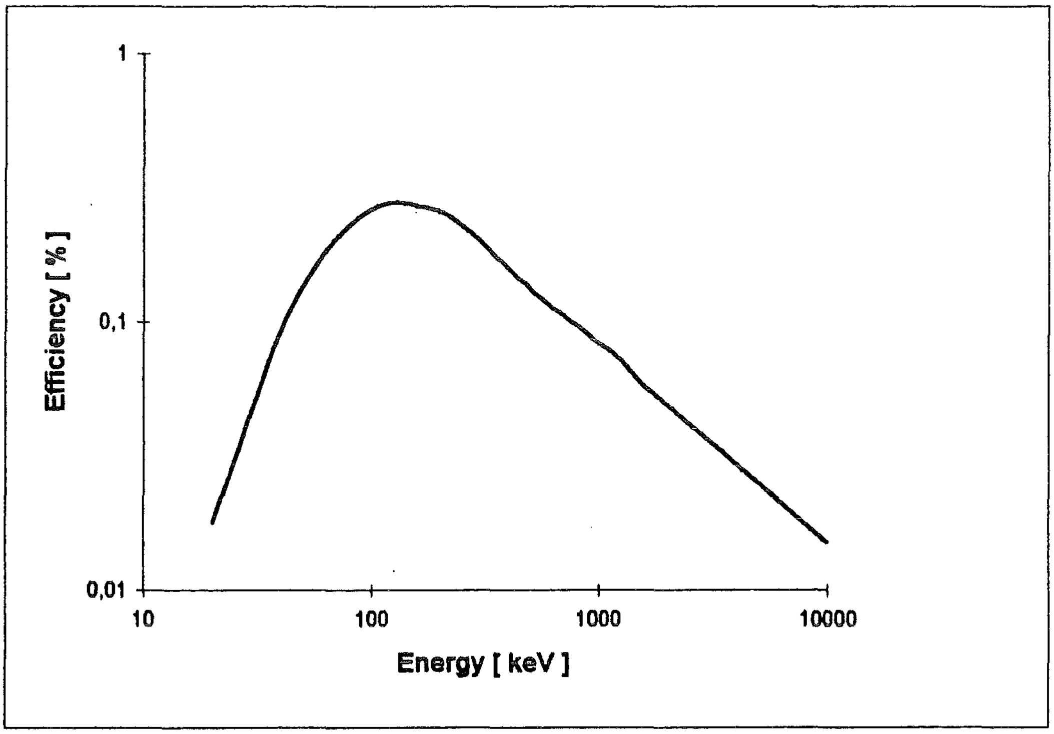
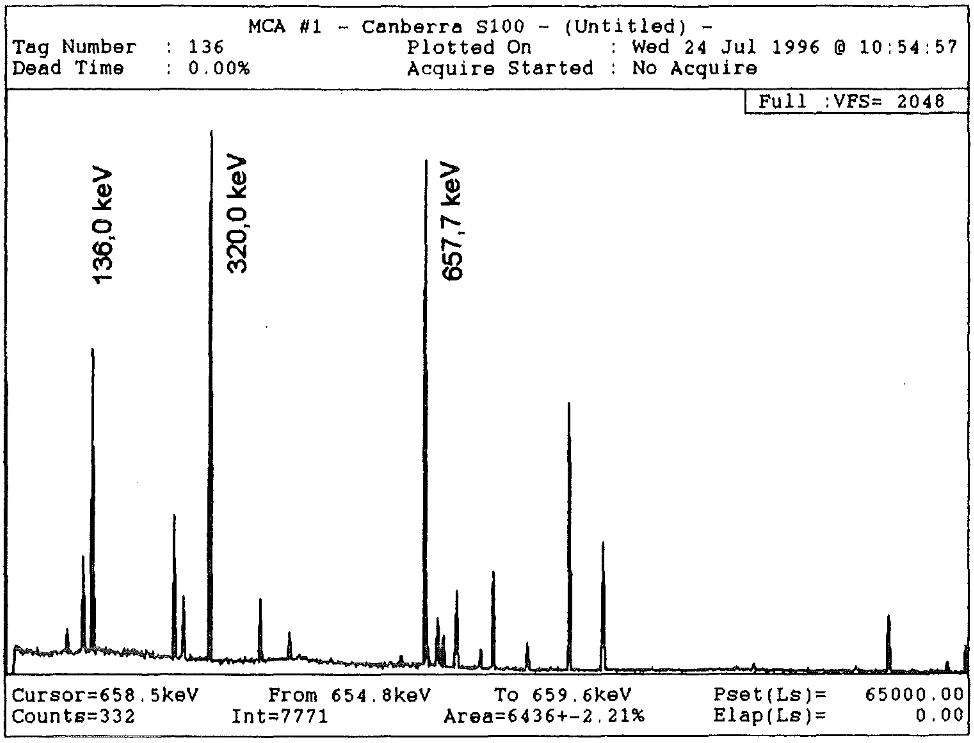


Fig. 5: Efficiency of the Ge - detector depending on the γ - energy
(distance of sample from detector : 15 cm)



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Tag Number  : 136               Plotted On      : Wed 24 Jul 1996 @ 10:54:57
Dead Time   : 0.00%             Acquire Started : No Acquire
                                                             Full :VFS= 2048
Cursor=658.5keV      From 654.8keV      To 659.6keV      Pset(Ls)=   65000.00
Counts=332          Int=7771       Area=6436+-2.21%      Elap(Ls)=       0.00
```

Fig. 6: Typical γ- spektrum of a ^{51}Cr - sample after eight months standing time.

The strongest γ- energies and their emission-propabilities:

Se - 75	121,1 keV	17,32%
	136,0 keV	59,00%
	264,6 keV	59,10%
	279,5 keV	25,20%
Ag - 110 M	657,7 keV	94,65%
	884,6 keV	73,40%
	937,2 keV	34,60%
	1384,3 keV	24,70%
Cr - 51	320,0 keV	9,83%

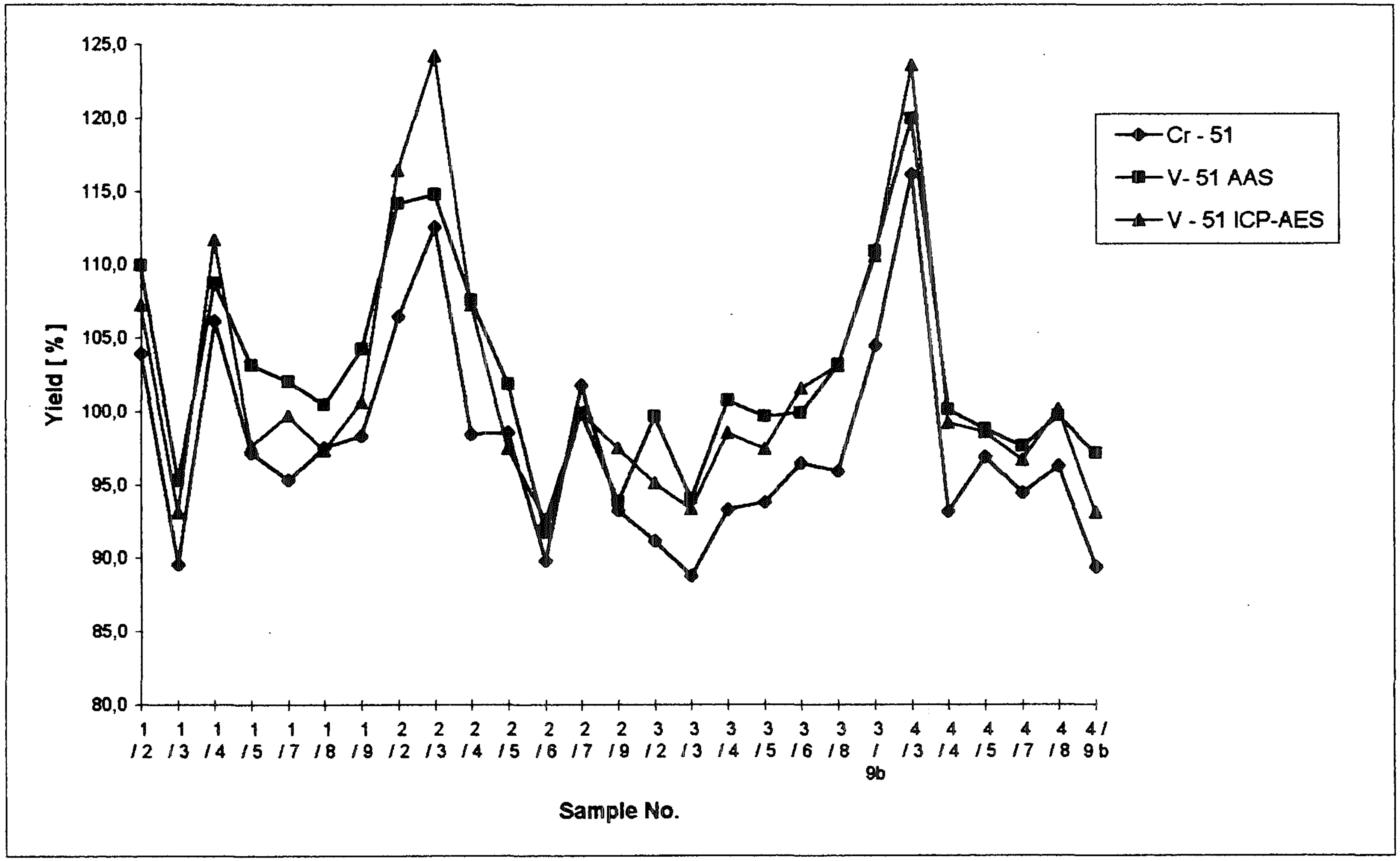

Fig. 7: Comparison of the data on Vanadium - analysis (AAS) and (ICP - AES) and γ - activity of the 27 samples of source 1.

The yields refer to the theoretical expectation values.

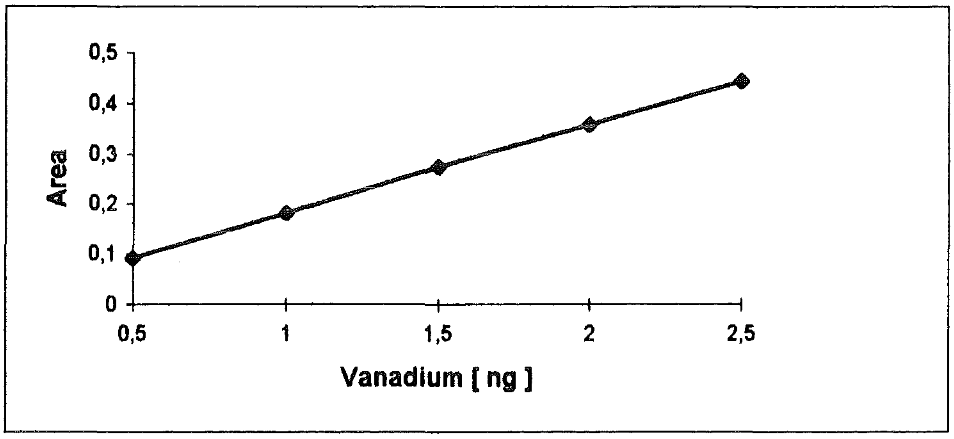

Fig. 8: Calibration curve for the determination of vanadium by AAS.