SERVICE DE PHYSIQUE DES PARTICULES

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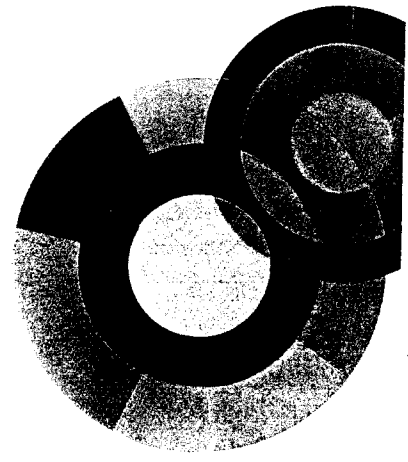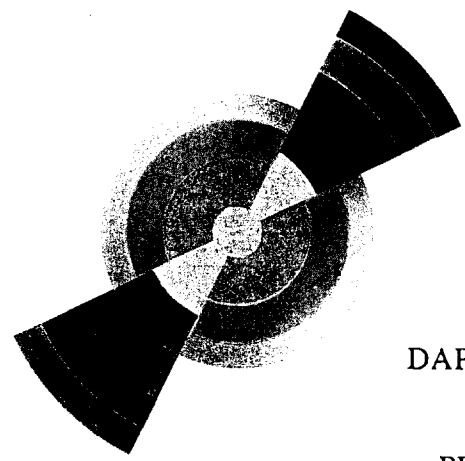
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Abstract

We describe the production of a 62 PBq (62×10^{15} Bq) source of ~ 750 keV neutrinos coming from the electron capture decay of ^{51}Cr. This is the first time that such a high-intensity, low-energy neutrino source has been produced. The rationale for having such a source is to check the overall procedures of the radiochemical solar neutrino experiment, GALLEX. The source was obtained by neutron activation of 36 kg of enriched chromium at the Siloé reactor at Grenoble. The enriched chromium (containing 38.6 % of ^{50}Cr compared to 4.35 % for natural chromium) was produced by the Kurchatov Institute in Moscow, in the form of CrO_3. It was then electrolyzed in Saclay to obtain chromium metal tubes, which were subsequently broken into coarse chips of typically 1 mm^3 volume. The chromium chips were put inside 12 special zircalloy irradiation cells that were then placed around the Siloé reactor core, which had been specially reconfigured for this irradiation. The irradiation lasted for 23.8 days. Then the activated chromium was transferred in a stainless-steel container into a sealed tungsten shield having a wall thickness of 8.5 cm. The ^{51}Cr activity at the end of the irradiation (EOB) has been measured to have a mean value of (62.5 ± 0.4) PBq, using several techniques : by neutronics and gamma scanning in the reactor shortly after EOB, with an ionization chamber, by calorimetry, and, after a considerable decay period, by gamma-ray spectroscopy and measurement of the non-radioactive ^{51}V daughter. The source resided in the center of the GALLEX detector at the Gran Sasso Underground Laboratory between June 23, 1994 and October 10, 1994, irradiating the gallium target with a neutrino intensity well above the solar neutrino background. The results of this full-scale test are in good agreement with the expected efficiency of the entire GALLEX experiment calculated as a product of the known efficiencies of the various parts of the experimental procedure.

1   Introduction.

Radiochemical neutrino experiments have low thresholds (some hundreds of keV) which make them invaluable tools for the detection of solar neutrinos. Their principle is simple : in inverse beta decay, a solar ν_e interacts with a given isotope, transforming it into the radioactive isobar of the next higher element; the small number of produced radioactive nuclei are extracted from the target, and their decay is observed in a proportional counter. Three such experiments are now running : the chlorine experiment [1] (reaction $\nu_e + {}^{37}Cl \rightarrow {}^{37}Ar + e^-$, threshold 814 keV), and two gallium experiments, GALLEX [2, 3] and SAGE [4] (reaction $\nu_e + {}^{71}Ga \rightarrow {}^{71}Ge + e^-$, threshold 233 keV). All three observe a deficit of solar ν_e compared to the predictions of solar models [5, 6]. Thus it is very important to find a convincing way to show the reliability of their results, before attributing these

deficits either to astrophysics or to particle physics.

During the development of the GALLEX detector, several tests were performed with the GaCl$_3$ solution, a) using Ge carriers and radioactive Ge isotopes to test the chemical procedures, and b) producing Ge radionuclides in situ by either radioactive decay or nuclear reactions to measure possible hot-atom effects that would occur as a result of neutrino capture in the solution. All of these tests [7] demonstrated that the removal of Ge radioactivity from the gallium target solution was quantitative. However, these tests could only approximate the conditions encountered during inverse beta decay in the GALLEX target. The only direct verification of the entire detection procedure is to produce an artificial neutrino source whose emission rate is well known and much more intense than the solar neutrino flux in the detector, to install it inside the target at the Gran Sasso Laboratory (LNGS) and to count the radioactive atoms which are then produced. A comparison of the measured and the expected ^{71}Ge production rate yields the efficiency of the detection procedure. During summer 1994, GALLEX became the first radiochemical experiment to perform this crucial test successfully [8].

This paper describes how the artificial neutrino source was produced. The nuclide used was ^{51}Cr ($T_{1/2} = 27.7$ d), obtained from neutron activation, in the Siloé reactor at Grenoble, of chromium enriched in ^{50}Cr . Its activity at the end of the neutron bombardment (EOB) has been measured, (62.5 ± 0.4) PBq, using several techniques [9]. The characteristics of the source are described in section 2, as well as the choice of the isotope, ^{51}Cr. The problem of impurities and of the shielding is discussed in section 3. The chromium preparation (production of the enriched chromium in the form of CrO$_3$, electrolysis to obtain chromium metal, formation of the target for irradiation) is described in section 4. The description of the dedicated Siloé reactor core, of the irradiation cells (called hereafter irradiators) and of the irradiation itself is given in section 5. Section 6 details the preparation of the source and section 7 explains the different methods which have been used for measuring the source activity. The transportation of the source between Grenoble and the Gran Sasso Laboratory and its installation in the center of the GALLEX detector are described in section 8. Future plans are briefly presented in section 9.

2 Characteristics of the artificial neutrino source and choice of the material.

Solar neutrinos have an energy spectrum ranging from 0 to about 14 MeV, with the major part of the flux (due to pp-fusion) occuring up to 420 keV (the mean energy of those which interact with a gallium target is of the order of 1 MeV). The main requirements for an artificial neutrino source for the GALLEX detector were the following : a) an energy spectrum (discrete lines or a continuous spectrum) as close as possible to the solar neutrino spectrum ; b) an initial neutrino flux that exceeds the flux coming from the Sun by about an order of magnitude, a necessary requirement since we aim for a statistical error of the same order as the experimental

errors in the solar runs; c) the possibility to measure the source activity with good precision, better than 2-3%; d) a facility to install the source in the center of the detector (there is a 50 cm diameter cylindrical thimble in the center of the GALLEX A-tank) and to shield against beta- or gamma-radioactivity, coming either from the decaying ^{51}Cr or from activated impurities in the source material.

The first idea of an artificial neutrino source to measure the efficiency of solar neutrino detection is due to Alvarez who proposed a ^{65}Zn source [10] producing 1.35 MeV neutrinos. Sources of lower energy neutrinos more appropriate for the efficiency calibration of GALLEX were later proposed. The most notable were ^{51}Cr (energy E up to 750 keV), proposed by Raghavan [11] in 1978 and ^{37}Ar (E = 814 keV), proposed later by Haxton [12]. We performed a systematic search for candidate nuclei using as criteria the activation cross section, isotopic abundance, and similarity of the neutrino spectrum to the solar spectrum. The search revealed ^{152}Eu (E = 1.86 MeV) as a possible source [13]. The final choice of ^{51}Cr was due not only to practical and economic considerations concerning the likelihood of successful isotopic enrichment and neutron activation but also because of the absence of higher energy gamma-rays that would cause heating and shielding problems.

A feasibility test performed in 1986 with natural Cr showed that an intense source of ^{51}Cr could indeed be produced in a nuclear reactor, but that enriched ^{50}Cr would be required as the target [14].

^{51}Cr is produced by neutron capture on ^{50}Cr (^{50}Cr + n → ^{51}Cr + γ) with high cross sections for thermal and epithermal neutrons (15.9 and 7.8 barns respectively) [15]. Its half-life is 27.706 ± 0.007 days [16]. ^{51}Cr decays by electron capture with a Q-value of 751 keV (see figure 1) to the ground state of ^{51}V (90.14% branching ratio [16]) and to its first excited state (9.86 ± 0.05%), which deexcites to the ground state with the emission of a 320 keV γ-ray. Aside from the low intensity internal bremsstrahlung (branching ratio 1.0×10^{-4}), the neutrino spectrum consists of four monoenergetic lines (746 keV (81%), 751 keV (9%), 426 keV (9%) and 431 keV (1%)). The mass difference between the two atomic states ^{51}Cr and ^{51}V is 751 keV but a few keV are taken by rearrangement X-rays and Auger electrons (5 keV for K-capture and negligible for L-capture, with L/K = 10%). The energy of the dominant 746 keV neutrinos is very similar to that of the solar neutrinos coming from the electron capture decay of ^{7}Be (E = 862 keV), and their energy is close to the mean energy of solar neutrinos seen by the gallium detectors. The 320 keV γ-radiation emitted simultaneously with the 426 or 431 keV neutrino is very easy to shield with a few mm of lead or tungsten. This γ-ray is also particularly useful since it allows an easy determination of the source activity (see section 7).

Natural chromium consists of 4 stable isotopes whose abundances are given in table 1, along with their thermal neutron capture cross sections. The isotope of interest, ^{50}Cr, represents only 4.35% of natural chromium. Neutron activation of ^{52}Cr and ^{53}Cr produces stable isotopes, while activation of ^{54}Cr produces radioactive ^{55}Cr. Since ^{55}Cr has a half-life of 3.5 min, the ^{51}Cr is the only radioactive isotope of chromium that is present during the test of GALLEX at LNGS.
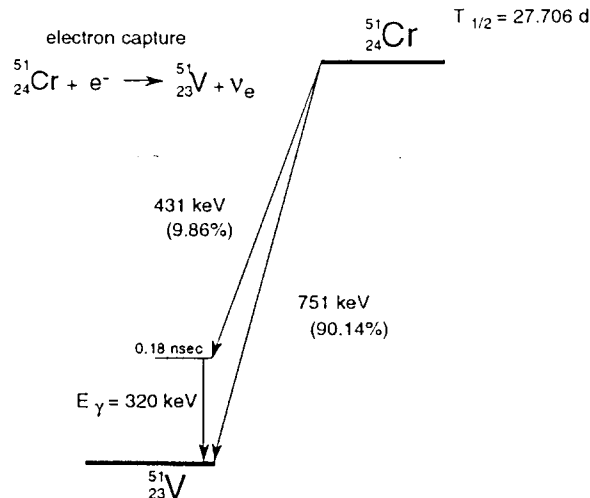
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electron capture

$^{51}_{24}Cr + e^- \longrightarrow\ ^{51}_{23}V + \nu_e$

$^{51}_{24}Cr$ $T_{1/2} = 27.706\ d$

431 keV
(9.86%)

751 keV
(90.14%)

0.18 nsec

$E_\gamma = 320\ keV$

$^{51}_{23}V$

Figure 1: *Characteristics of the decay of ^{51}Cr. The "751 keV" line combines the 746 and 751 keV lines and the "431 keV" line combines the 426 and 431 keV lines.*

As was indicated above, the low natural abundance of ^{50}Cr along with the high cross section of ^{53}Cr makes it technically impossible to reach the required activity level, ≈ 50 PBq, by using natural chromium as the target in the reactor irradiation [14]). Instead, chromium enriched in ^{50}Cr (and depleted in ^{53}Cr) is needed.

With such characteristics, it was calculated that 35-40 kg of enriched material could provide a neutrino source having the required activity. Such a large amount cannot be easily produced with a mass separator in a research laboratory; a dedicated isotope enrichment facility must be used. Accordingly, the enriched chromium was purchased from the Kurchatov Institute in Moscow (Russia). The isotopic composition of the enriched Cr is also given in table 1.

3   Impurities and shielding.

Apart from the neutrino, the most penetrating radiation emitted by ^{51}Cr is the 320-keV γ-line. The interaction length (l) of a 320-keV γ-ray in lead is only 1.1 mm, and a 3 cm lead shield would attenuate the radiation by 12 orders of magnitude. However, certain impurities with (n,γ) cross sections in the barn range, which would be activated to produce radionuclides with typical lifetimes of a few days, would pose a problem, even at concentration levels of parts per million (ppm) in the enriched chromium. Their contribution to the neutrino flux would be negligible, but they would be sources of penetrating γ-rays of ≥ 1 MeV (l$\geq$1.3 cm), with activity levels of tens of GBq after irradiation. All this strongly conditions the characteristics of the

	Isotopic composition of natural Cr	Isotopic composition of the enriched Cr used in GALLEX	Thermal neutron capture cross sections (barns) [15]
^{50}Cr	4.35 %	38.6 %	15.9
^{52}Cr	83.8 %	60.7 %	0.76
^{53}Cr	9.50 %	0.7 %	18.2
^{54}Cr	2.35 %	<0.3 %	0.36

Table 1: *Isotopic composition of chromium and thermal neutron capture cross sections (measured at 2200 m/s).*

shielding to be installed around the radioactive chromium. Moreover, optimization studies of the source experiment in the gallium tank by Monte-Carlo indicated the necessity of building as compact a source as possible. To accommodate the large volume of radioactive chromium in the existing thimble (inner diameter of 50 cm) in the GALLEX target tank, a shielding material with high stopping power is necessary. It was decided to construct a tungsten shield since 8.5 cm of tungsten (the thickness finally chosen) has the same stopping power as 14 cm of lead. A lead shield with its thicker walls would have required the source to be longer by 1.2 m, equivalent to a loss of 5 % of the activity of the neutrino-produced ^{71}Ge. The tungsten material was provided by the Plansee Company (Austria) and is referred to as "Densimet 1", whose composition is W (94.64 %), Ni (3.36 %), Fe (1.80 %), Cu (0.20 %), and density is 17.9 g/cm^3. The minimum shielding distance is 8.5 cm, giving an attenuation factor of better than 5×10^{-4} for a 1.5 MeV γ-ray.

We consider in the following discussion the radioactive chromium and its surrounding tungsten shield as a single object. The radiation dose rate emitted by the source, once inside the shield, has to satisfy several safety constraints. Although the dose rate admitted in a restricted area could be up to 25 μSv/h, safety authorities of the LNGS have recommended an activity limit of ≤ 200 μSv/h at contact with the shield and ≤ 2.5 μSv/h at 2 meters from the center of the source, in order to meet the requirements of the LNGS underground laboratory as a low level facility. In addition, transport regulations require a dose rate of ≤ 5 μSv/h at contact with the transport container. To match these requirements, impurities that could potentially pose problems had to be limited by specifying the maximum impurity concentrations that could be tolerated in the enriched chromium and by strictly controlling these impurities in the purchased material and during the subsequent processing steps. These specifications, listed in table 2, were deduced for each relevant nuclide from (a) its radiation characteristics (type, energy, branching, lifetime), (b) the ensuing absorption characteristics in tungsten, and (c) the corresponding thermal-neutron activation cross section and isotopic abundance of the precursor nucleus (see for example [17] for a preliminary study).

Special care was taken to reduce contamination by impurities in the various

Element	Radionuclide					Specification
	nuclide	half-life	i(%)	σ (b)	γ rays (MeV)	max. conc. (ppm)
Ga	^{72}Ga	14 h	39.9	4.86	2.201 (26 %) ~2.50 (20 %)	100
Na	^{24}Na	15 h	100	0.53	1.369 (100 %) 2.754 (100 %)	6
As	^{76}As	26 h	100	4.50	~2.10 (1 %)	10
La	^{140}La	40 h	99.9	8.93	1.596 (95.4 %) 2.347 (0.8 %) ~2.5 (3.5 %)	0.5
Cd	^{115m}Cd	44.6 d	28.7	0.34	1.290 (1 %)	400
Fe	^{59}Fe	45 d	0.28	1.28	1.291 (43 %)	5000
Sb	^{124}Sb	60 d	42.7	4.2	1.691 (47 %) 2.091 (6 %)	20
Tb	^{160}Tb	72 d	100	23.4	~1.3 (12 %)	8
Sc	^{46}Sc	84 d	100	27.2	1.120 (100 %)	1
Ta	^{182}Ta	115 d	100	20.5	~1.22 (55 %)	10
Zn	^{65}Zn	244 d	48.6	0.76	1.115 (50.6 %)	1000
Ag	^{110m}Ag	250 d	48.2	4.7	1.384 (26 %) 1.505 (12 %)	60
Co	^{60}Co	5.3 yr	100	37.2	1.332 (100 %)	8

Table 2: *Maximum allowed levels of critical impurities in enriched chromium (elemental concentrations in ppm or $\mu g/g$ of metallic chromium). The sequence of listing is with increasing half-lives of the radionuclides. The column labelled "i" gives the isotopic abundance of the parent isotope. The column labelled "gammas (MeV)" gives the energy in MeV of the main gamma lines, with the corresponding intensities between parentheses (the symbol, $\sim$, denotes the average energy for multiple γ rays with similar energies).*
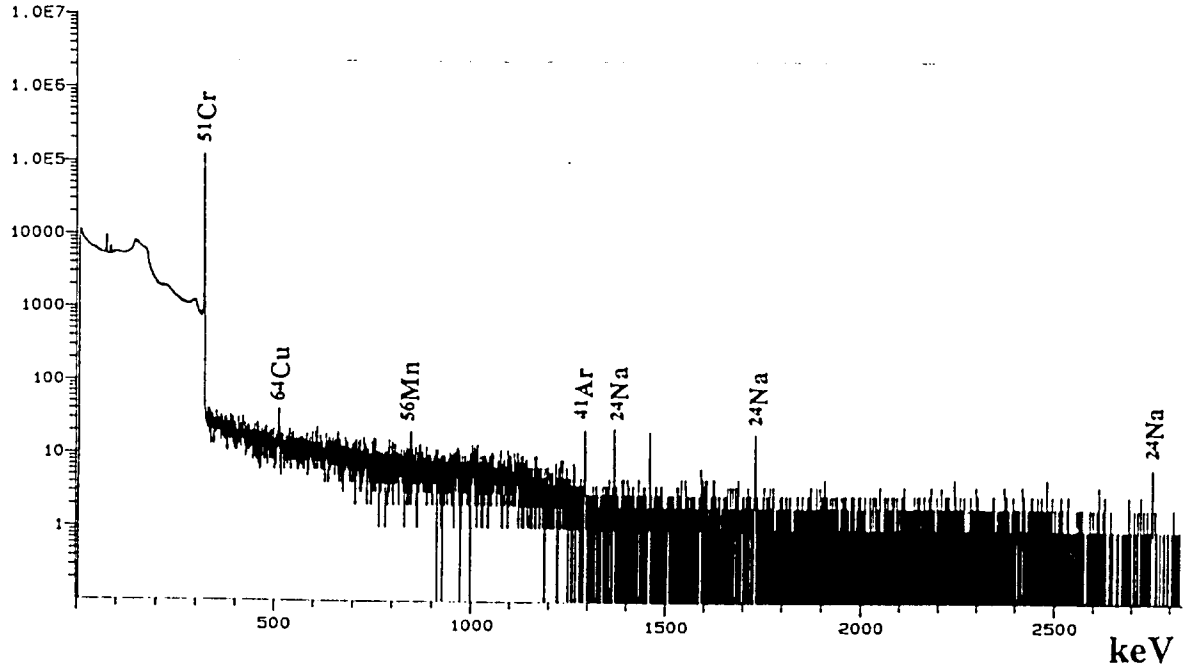
Figure 2: *Spectrum of a chromium sample after neutron activation.*

steps of the preparation of the source : chromium enrichment, electrolysis, chipping, manipulation. Controls by neutron activation analysis were routinely made at key steps in the production processes. A typical γ-ray spectrum of a chromium sample, measured after neutron activation with an intrinsic germanium detector, is shown in figure 2. The 320-keV γ-line corresponding to the ^{51}Cr is dominant; for this specific sample, some impurities of Cu, Mn, Ar and Na are visible.

The overall check of the impurities was made by γ-ray spectroscopy after the source was produced [18]. The long irradiation in Siloé of the full amount of enriched product made possible the very precise analysis of the impurities. Results are reported in section 6.

4 Chromium preparation.

4.1 Production of the enriched chromium.

Several methods were investigated for producing isotopically enriched chromium :
a) chemical separation of Cr isotopes is not practical since the respective isotopic

mass differences are very small; b) plasma centrifugation has been studied in detail [19], but did not succeed in producing significant amounts of enriched material; c) isotopic separation by ion cyclotron resonance [20] has also been studied and small amounts (few hundreds of mg) of enriched chromium have been produced at the laboratory scale [21], but the method was not used for the chromium source, mainly for economic reasons; d) gaseous ultracentrifugation [22].

The technique used by the Kurchatov Institute to enrich chromium for our experiment is gaseous centrifugation [23]. Isotope separation in a gas centrifuge can be achieved because the radial pressure gradient of the gas is dependent on its molecular weight. Heavier fractions of a mixture concentrate nearer to the outer wall than do the lighter fractions and thus can be separated. The primary equilibrium effect of isotope separation in a gas centrifuge depends on the angular velocity and on the absolute difference between the molecular weights of the isotopes. A typical centrifuge is shown schematically in reference [24].

For chromium enrichment, volatile chromyl fluoride, CrO_2F_2, was used. The first step involved synthesis by chemists of the Kurchatov Institute of 1500 kg of this hazardous, corrosive compound from chromium metal. Special attention was given to a study of the CrO_2F_2 decomposition at the centrifuge wall and the prevention of the formation of solid deposits that would impede the separation process. In order to provide the required radiochemical purity of the ^{50}Cr isotope, only new equipment was employed in the separation. Since a single gas centrifuge has a relatively small separating power, several hundred centrifuges were constructed and used [23] in a cascade mode for the production of the enriched chromium material (the cascade process is described for example in [24]). As a result, the isotopically enriched CrO_2F_2 is distinguished for its very high degree of chemical purity, especially with regard to non-volatile components. Three cascade lines at the facility were put into operation to produce about 3 kg of enriched chromium per month. About 1000 kg of CrO_2F_2 were finally used for producing 40 kg of enriched chromium, and the efficiency of the system was then estimated not to exceed 9 % [23]. As noted above, severe specifications on impurity levels were imposed by GALLEX on the enrichment process (see section 3 and table 2).

The enriched CrO_2F_2 was then hydrolyzed in water (in the form of ice) to form chromium trioxide, CrO_3 : $CrO_2F_2 + H_2O \longrightarrow CrO_3 + 2HF$. Delivery of this CrO_3 to Saclay lasted from August 1992 to February 1994.

CrO_3 is thermally unstable and thus cannot be used in a nuclear reactor. Another oxide, Cr_2O_3, was studied [25] because it is stable at high temperatures. However, there was concern about its ability to dissipate heat in the reactor environment, and about the wide range of oxide particle-sizes that was produced. Instead, we chose to transform the oxide into metallic chromium by electrolysis, taking special care to avoid introducing impurities during this step.

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4.2 Electrolysis.

After a successful prototype was developed at Saclay, 4 electrolysis cells were specially built to produce chromium metal from CrO_3. The cells were stainless steel coated with teflon, and the cathode consisted of tubes of ultra pure copper, 16 mm diameter; 5 such cathodes were installed in each cell (see figure 3). The anode was a titanium grid covered with platinum. The electrolyte was made from CrO_3 dissolved in ultrapure water with a concentration of 300 g/l (corresponding to a density of 1.218 g/cm^3). In each cell, which contains 7 liters of solution, 35 g of H_2SO_4 (65 %) were added to define appropriate acidity conditions for the electrolysis.

An automatic system controlled the levels, concentration, and temperature of the electrolyte as well as the current. The electrolyte temperature was 60°C and the current intensity, 70 A. The concentration of the electrolyte was continuously monitored and adjusted to be between 300 and 330 g/l. The solution levels were also routinely adjusted. For safety reasons, H_2 at the cathode and O_2 at the anode were continually flushed out of the cells, with a flow of nitrogen (70 l/h) on the H_2 side and a flow of air (300 l/h) on the O_2 side.

In the usual procedure, the cells were in continuous operation for three days and the chromium metal was deposited on the copper, making a uniform deposit $\approx 1,5$ mm thick. After 72 hours of continuous operation, 200 g of chromium were produced in each cell (~ 40 g per electrode).

The electrodes were then put in nitric acid (96 %) where the copper was completely dissolved, leaving a hollow chromium tube (110 mm long, 16 mm diameter). The chromium tubes were then washed with ultrapure water. Taking into account all these operations, about one kg of chromium metal was produced per week by the system of four cells.

4.3 Production of the chromium metal chips.

Next the chromium tubes were manually broken into small chips, using a dedicated chromium-coated hammer. This was done in a special ultraclean cell under inert atmosphere. The range of acceptable chip sizes was from 0.3 to 3 mm. Larger chips would prevent the desired representative sampling (see section 7) with moderate amounts of sample, while smaller chips would result in irradiated dust that would complicate handling of the source. Grains > 3 mm diameter were broken again with the hammer. Chips < 0.3 mm diameter, i.e., powder, were removed with a filter which, together with losses in the electrolysis, led to a diminution of the material employed by about 4 kg. The final chips were irregular in shape, about 1 mm^3 in volume. The weight distribution of the individual chips is shown in figure 4.

The final treatment included cleaning in distilled water, to remove the adherent fine chromium metal powder. Because chromium forms metastable hydrides during electrolysis, the chips were then outgased in an electric oven, under vacuum at 600 °C for 20 hours to remove the bulk of adsorbed hydrogen (3-4 cm^3 H_2/g Cr). The reason for this step was that sudden outgasing of hydrogen from the 36 kg of
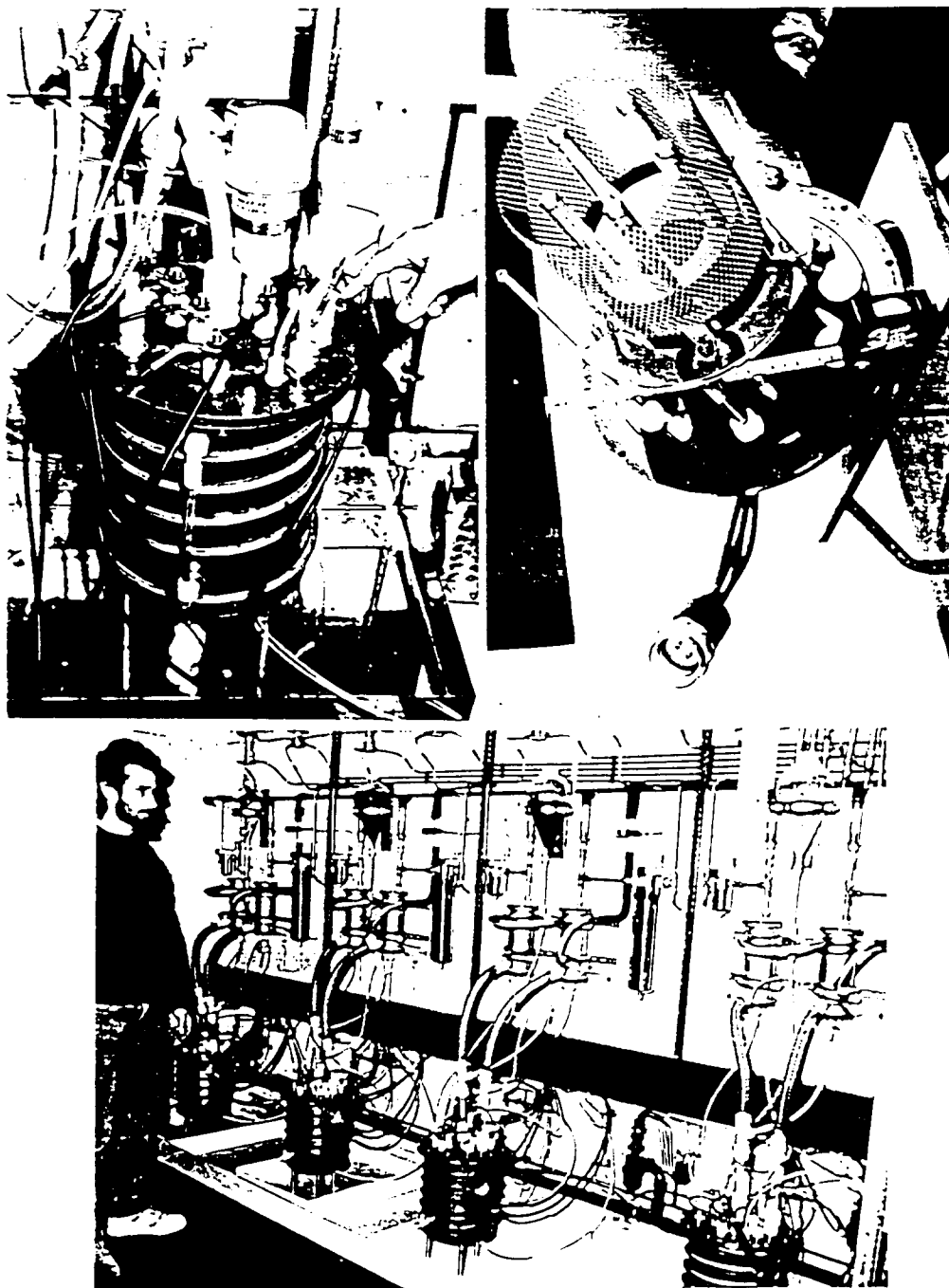
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Figure 3: *Photographs of the electrolysis device. Upper left shows an electrolysis cell. Upper right shows the electrodes (5 copper cathodes and 1 anode made of a platinum-coated titanium grid). The bottom photo shows the complete system of the four electrolysis cells.*
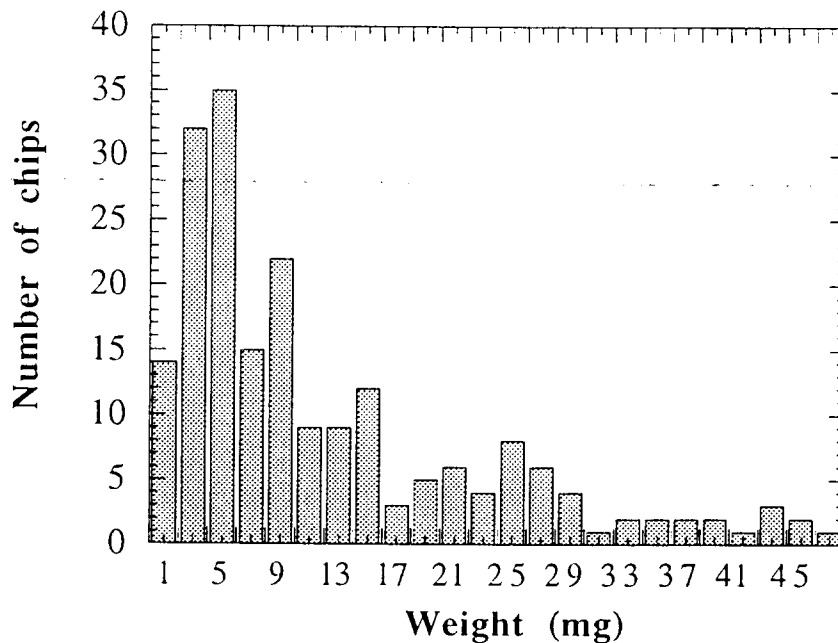
Figure 4: *Weight distribution of the chromium chips.*

chromium during the reactor irradiation could have been a safety hazard. After these procedures, 36 kg of chromium were available for the irradiation. The bulk density of the final product was about $3.6\,g/cm^3$. The thermal conductivity of the chromium chips was measured to be about $0.7\,W.m^{-1}.K^{-1}$ at $300\,^{\circ}C$.

5 Chromium irradiation.

5.1 Choice of the reactor.

The requirements for the production of an artificial neutrino source for GALLEX, i.e. irradiation of a large, voluminous amount of material with a high average thermal neutron-capture cross section in a high average thermal-neutron flux, considerably limit the choices of possible nuclear reactors. We also had to consider the ability to unload the irradiated material at the appropriate time, which not only requires an operating hot cell, but also a continuous irradiation period that is comparable to the 27.7 d half-life of ^{51}Cr. Our choice was limited to European reactors to minimize the problems in transporting the source from the reactor to the LNGS.

Power reactors generally are not suitable for this application because they have only small thermal neutron fluxes, no internal sites to place irradiation targets, and very long operating cycles. Among research reactors, high flux reactors, like ILL at Grenoble or Orphée at Saclay, have a very compact core and can only accommodate

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small irradiation samples. The HFR-Petten reactor in the Netherlands, the SM-2 reactor in Russia, as well as the Osiris reactor in Saclay, have a core enclosed inside an aluminum or zircalloy wall which limits the average flux of neutrons and the possible arrangement of fuel elements. The Essor reactor at Ispra, Italy, offers an even lower average neutron flux. The BR2 reactor at Mol, Belgium, although having a few irradiation sites with a high thermal flux (up to 9×10^{14} n/cm^2.s), has not enough space available to accommodate the entire 36 kg of chromium. In fact, only Siloé (Grenoble, France) is flexible enough to accommodate a customized irradiation of such a large target and to allow its rapid removal [26].

5.2   Siloé core.

Siloé is located at the Centre d'études nucléaires de Grenoble and operated by CEA (Commissariat à l'énergie atomique). It is a swimming pool reactor, with 35 MW thermal power. The core is immersed under 8 m of water, which serves as moderator, coolant, and shield. To produce a high activity source, the chromium irradiation requires a high neutron flux throughout the entire chromium target. The neutron flux is high inside the core itself, and decreases rapidly outside due to thermalization and capture in water. Optimization studies of the system (core + irradiators) have been performed to define the best configuration of the core that would produce an optimal neutron flux in the irradiation positions. Another constraint to consider was the possibility of having a reactor cycle several days longer than the usual 21-day period; the final requirement was to strictly respect the general operating and safety rules of the reactor.

The dedicated core for the chromium irradiation was specially adapted to contain 34 fuel elements, made with UAl_x enriched to 93 % in ^{235}U, and arranged in a checkerboard configuration, as shown in figure 5. Four elements were equipped with control rods, two for security and two to stabilize the power.

The full amount of chromium was too large to fit into the usual irradiation holes inside the core (some pieces of aluminum were put in these irradiation holes during our chromium irradiation, for neutron regulation purpose). Instead, the chromium had to be placed around the fuel elements, in 6 crenels built on the 3 available sides (the fourth one is a beryllium wall). A crenel (see figure 6) is surrounded by fuel elements on three sides prolongated by two beryllium elements having the same geometry. The beryllium elements have two positive effects : to increase the mean neutron flux by about 9 % and to reduce the flux decrease caused by neutron capture in the water. The overall symmetry of the disposition of fuel elements, beryllium reflectors and chromium targets (figure 5) makes the energy spectrum of neutrons similar from one crenel to another. The absolute flux intensity only varies from one position to another depending on the known burn-up of fuel elements used around a crenel; it varies slightly during irradiation depending on the height of the control rods. All irradiators are equipped with 3 temperature and pressure monitors; irradiator 1, which is represented in figure 6, has in addition collectrons which monitor the neutron flux.
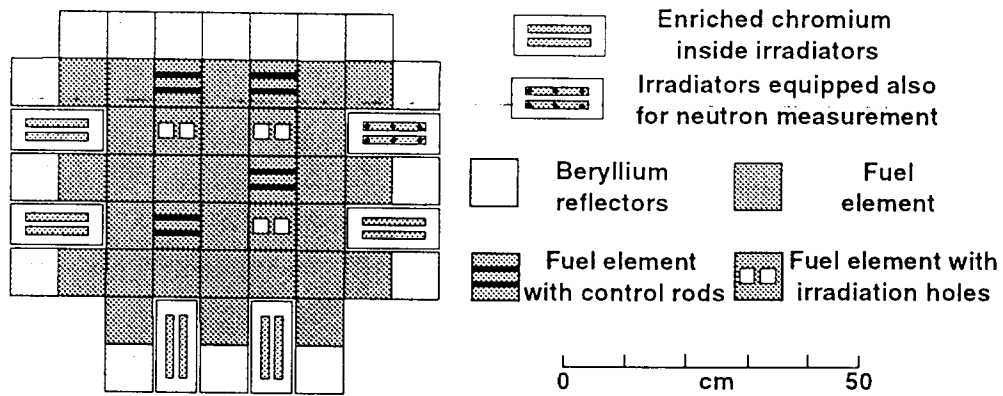
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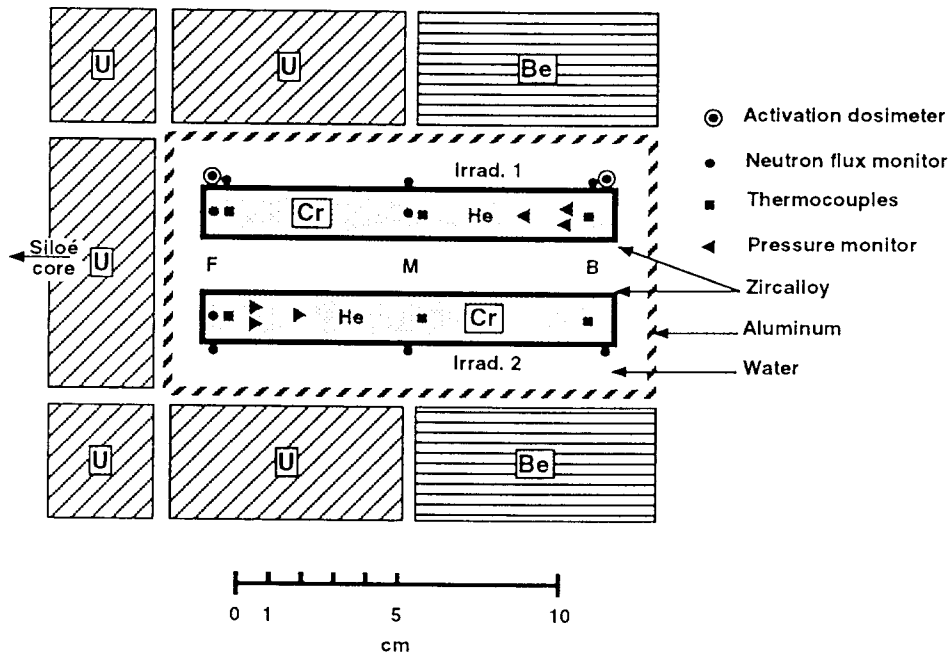Figure 5: *Special Siloé core configuration for chromium irradiation.*

Figure 6: *Detailed configuration of a crenel (located at the upper right side in figure 5), showing the beryllium reflectors and the irradiators 1 and 2. Small triangles and squares correspond to pressure and temperature probes respectively. F, M and B refer to front, medium and back positions.*

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The effective averaged thermal cross section of ^{50}Cr for the given spectrum of thermal and epithermal neutrons in Siloé is 17.2 barns.

5.3 The irradiators.

The chromium chips were placed in 12 special irradiators that were constructed of zircalloy in order to be transparent to neutrons as much as possible; two irradiators were placed in each crenel. Their geometry is optimized for the thermalization of the neutrons and the cooling of the irradiated material. The heat deposition, 32 kW per irradiator, comes mostly from photons from uranium fission. In any case the temperature of the water must stay below its boiling point and the temperature inside the chromium must stay well below the sintering temperature (although the melting point for the chromium is 1890 °C, the chips begin to sinter at 700 °C; sintering would interfere with the homogenization of the samples for determining the source activity). The best configuration consists of two thin parallelepipedic boxes (50 cm high × 12.6 cm long × 1.4 cm wide) separated by 12 mm of water (see figure 6). About 3 kg of chromium chips were put in each box. Since these boxes would be placed in Siloé under 8 m of water, they were filled with helium at 1.9 bar to offset the external water pressure and then were sealed.

5.4 Irradiation test.

An irradiation test was performed during August 1993. It consisted of irradiating 6 kg of natural metal chromium for 3 weeks, i.e. a duration similar to that of the planned ^{50}Cr irradiation. The chromium had been prepared by electrolysis and treated by the same techniques used for the enriched chromium. The two zircalloy irradiators used (~ 3 kg in each) were the first two built and were used again in the final irradiation. The core was not dedicated for this test, but the irradiators were installed in a crenel with beryllium reflectors on two sides.

Continuous measurements of temperature, helium pressure and neutron fluxes were made during this test, which has been used by the French safety authorities (Institut de Protection et Sûreté Nucléaire, IPSN) to certify the device (chromium chips and irradiators) and the irradiation process.

5.5 The irradiation of the enriched chromium.

The core was loaded with fuel elements to allow an irradiation time of 23.8 days, instead of the usual fuel cycle of 21 days. The chromium was exposed to an average perturbed neutron flux of 5.2×10^{13} n/cm^2.s, with large spatial variations (a maximum factor of 2) around this mean value that depended on the positions of the irradiators in the reactor; these variations reflect the vertical and radial neutron-flux gradients which in turn are influenced by the self-shielding of the chromium.

Figure 7 shows the vertical and radial distribution of this neutron flux, and figure 8, the distribution of the neutron flux as a function of time during the irradiation

inside irradiator 1. Figure 8 clearly shows the stability of the neutron flux during the entire course of the irradiation.

The irradiation started at 10 am on May 27, 1994, and lasted without any interruption to the end-of-bombardment (EOB $= t_0$), at June 20 at 6 am.

During the irradiation, there was continuous monitoring and control of the pressure inside the irradiators; the reactor would have been stopped if there were any indication of a large increase in internal pressure that could have caused an irradiator to rupture. The recorded curves (not shown) present a very smooth behaviour.

Similarly the temperature was continuously checked at several "hot points". A typical measurement is shown in figure 9. The behaviour is also very smooth and the temperature is very stable after the normal increase during the first day.

6   Preparation of the source.

The irradiated material was unloaded in a hot cell equipped with a complex tele-manipulation system at Siloé. The contents of each irradiator were poured through a tube into a cylindrical stainless steel container. After checking that all the chromium had been unloaded from the irradiator, the filled container was weighed. The operation was repeated for each irradiator, with 24 hours needed to empty the 12 irradiators. The cylindrical container was equipped internally with fixed blades which act as in a cement-mixer. After it was filled with all of the chromium, the container was installed on a tilted support and rotated along its axis clockwise and counterclockwise. The purpose of this procedure was to homogenize the chromium, to average out the neutron-flux differences experienced by the different irradiators (previous tests had shown that 10 minutes were sufficient to produce complete mixing in this device). So, after about 10 minutes of mixing, the container was put in a vertical position. A sampling rod with holes uniformly distributed along its length was then introduced in the container to take out 8 samples at different levels (see figure 10). Four such operations of mixing + sampling were made, giving 31 samples (one was lost during manipulation). This sampling procedure took 6 hours.

All of the chromium chips (minus the samples) were then placed in a sealed stainless-steel container and inserted into the tungsten shield (8.5 cm wall thickness). This shielding corresponds to more than 80 attenuation lengths for the 320-keV γ-line from the ^{51}Cr. As noted in Section 3, this large wall thickness was selected in order to satisfy health-physics safety requirements, assuming that the maximum levels of allowed impurities that produce MeV γ-lines would not exceed the specifications listed in table 2. (Without any impurities, 1.5 cm of tungsten would have been sufficient to shield the 320 keV γ-line.) The total weight of the source, including its tungsten shield, was 1150 kg.

The final operation was to remove the shielded source from the hot cell and to decontaminate its external surfaces of traces of the active chromium. All these operations with very active materials delivered a maximum dose rate of 0.1 mSv to the most exposed worker.
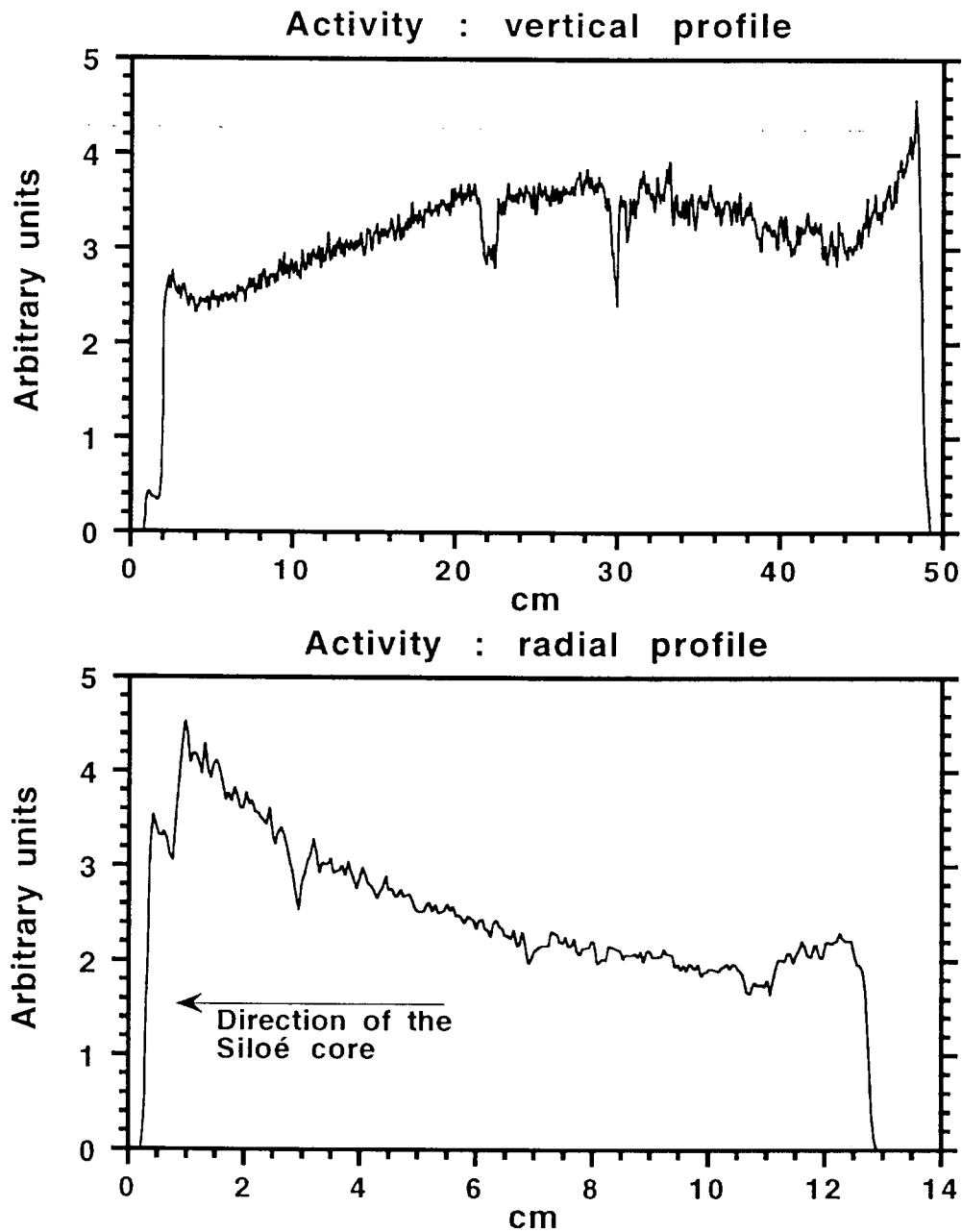
Figure 7: *An example of the vertical and radial distributions of the neutron flux at the instrumented crenel (irradiator 1, as shown in figures 5 and 6). These are obtained from the gammametric measurement of the activated chromium after the irradiation. The dips at ~20 and 30 cm in the vertical profile and at 3 cm in the radial profile are due to partial shielding by structural supports in Siloé.*
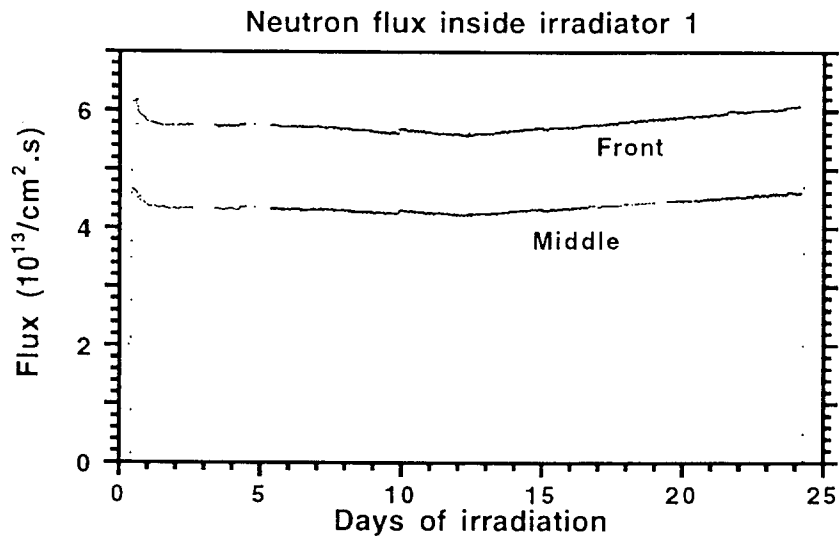
Figure 8: *Neutron flux as a function of time during the irradiation.*
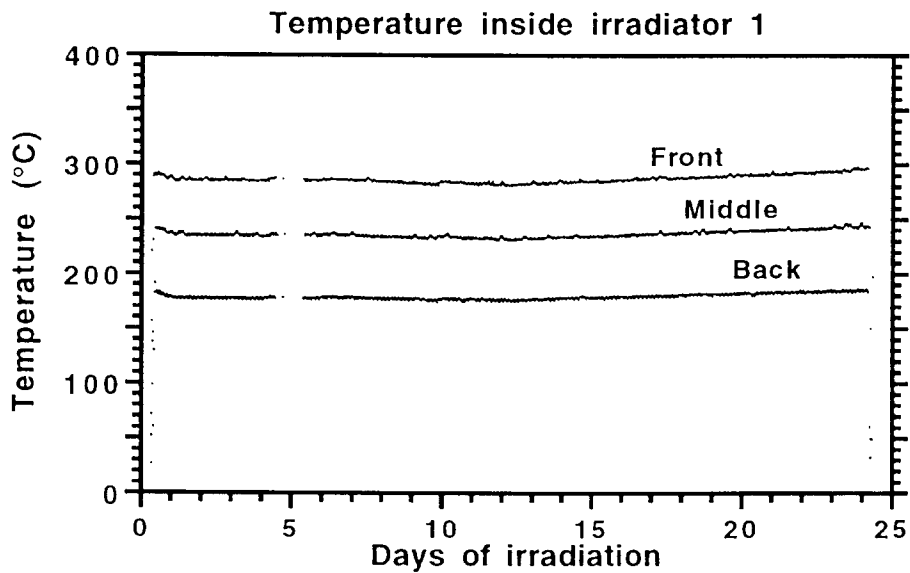

Figure 9: *Temperature inside the irradiator 1 during the irradiation. The three curves correspond to the positions F, M and B as shown in figure 6.*
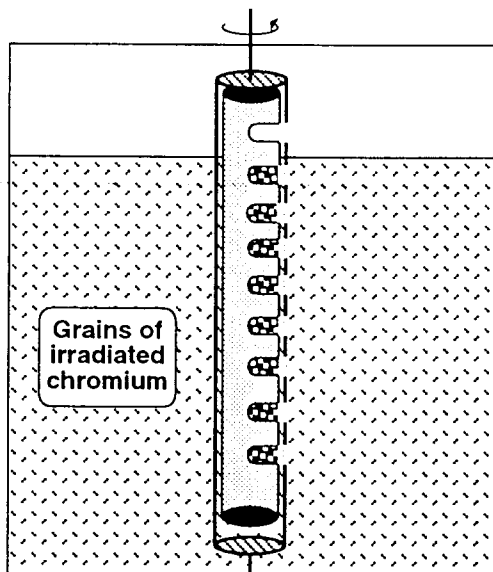
Figure 10: *Special rod used for sampling in the mixing device.*

The source configuration inside the shield is shown in figure 11 and a photograph is shown in figure 12 (the role of the visible copper cooling jacket is explained in section 8.2). Measurements of the external dose rates at any position on the surface of the tungsten shield yielded values smaller than 7 μSv/h [18], well below the 200 μSv/h-level recommended by the LNGS authorities. At 2 meters the dose rate was hardly measurable, but definitely less than 1 μSv/h. Gamma-ray spectroscopy performed outside the shield immediately after removal of the source from the hot cell showed activation of impurities of Na, Sc, Co, Cu, As, Ag, and Sb [18]. These data are reported in table 3. They are orders of magnitude below the acceptable levels, with the exception of Cu, which was probably introduced in the chromium during electrolysis. However, its contribution to the total external dose rate was negligible when the source was removed from the hot cell, due to the short half-life (12.7 h) of ^{64}Cu. A typical spectrum obtained with a Ge diode placed close to the source-plus-shield is shown in figure 13.

Since fast neutrons could produce ^{71}Ge nuclei in gallium, we searched for a flux of neutrons from the source after its removal from the gallium target. No excess of fast neutrons above the background was seen [18].

7 Measurement of the source activity.

The amount of irradiated chromium in the resulting neutrino source was somewhat less than the initial amount received from the Kurchatov Institute, because there were losses of material in succeeding processes, e.g., residual chromium remained in the electrolysis solution, small quantities of metal were left in the irradiators, and
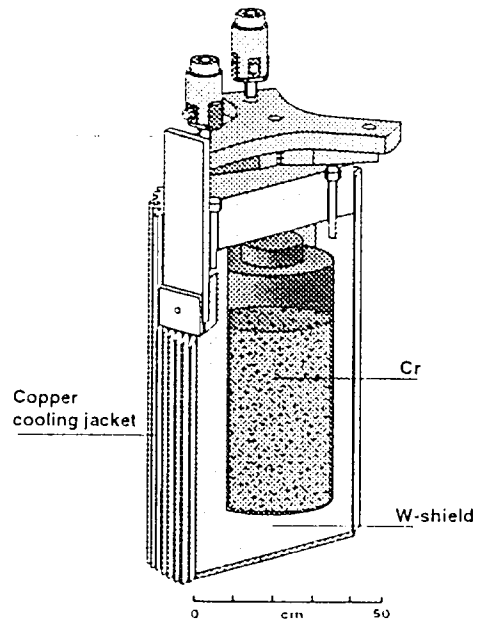
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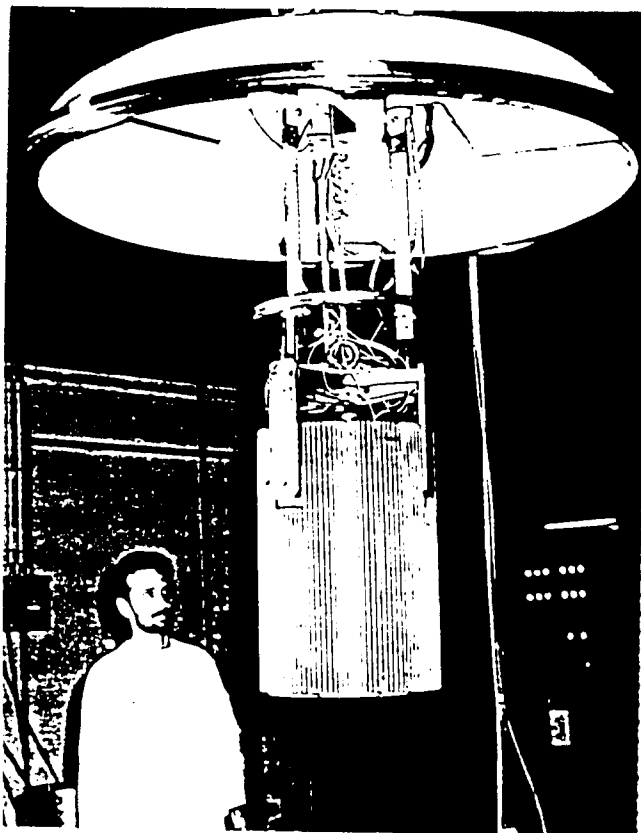Figure 11: *Schematic drawing of the Cr-source inside the W-shield.*

Figure 12: *Photograph of the Cr-source with its copper coooling jacket.*

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nucleus	half-life	energy (keV)	source activity (GBq) after irradiation	source activity (GBq) after removal
^{24}Na	14.8 h	1369	0.10 ± 0.02	
		2754	0.07 ± 0.01	
		average	0.08 ± 0.01	
^{46}Sc	83.9 d	1121	0.13 ± 0.02	0.025 ± 0.003
^{48}Sc	26.3 h	984	0.05 ± 0.03	
		1038	0.07 ± 0.03	
		1312	0.10 ± 0.01	
		average	0.095 ± 0.01	
^{60}Co	5.27 y	1173	0.02 ± 0.01	0.028 ± 0.003
		1333	0.04 ± 0.01	0.040 ± 0.005
		average	0.03 ± 0.01	0.031 ± 0.002
^{64}Cu	12.7 h	1346	210 ± 20	
^{77}Ge	11.3 h	2342	0.5 ± 0.2	
^{76}As	43.7 h	1213	6 ± 3	
		1216	3 ± 1	
		2096	1.5 ± 1.6	
		average	1.6 ± 0.2	
^{97}Zr	17.0 h	1750	0.40 ± 0.15	
^{110m}Ag	250 d	658	4 ± 2	
		764	10 ± 3	4 ± 1
		818		2.7 ± 0.5
		885	4.3 ± 0.5	4.2 ± 0.4
		938	4.2 ± 0.5	4.2 ± 0.5
		1384	5.0 ± 0.5	4.5 ± 0.4
		1476	5.1 ± 0.5	4.0 ± 0.4
		1505	4.3 ± 0.5	4.2 ± 0.4
		1562	4.0 ± 0.5	2.9 ± 0.5
		average	4.35 ± 0.25	4.0 ± 0.1
^{124}Sb	60.2 d	1368		0.18 ± 0.06
		1437	0.4 ± 0.15	0.21 ± 0.06
		1691	0.36 ± 0.05	0.12 ± 0.02
		1919		0.09 ± 0.03
		2040	0.4 ± 0.03	0.10 ± 0.03
		2091	0.51 ± 0.08	0.12 ± 0.01
		2185		0.07 ± 0.03
		2294		0.09 ± 0.03
		average	0.40 ± 0.03	0.10 ± 0.01

Table 3: *Measured activities of trace elements in the source just after insertion in the shield (June 26, 1994) and after removal from the gallium target (October 10, 1994).*
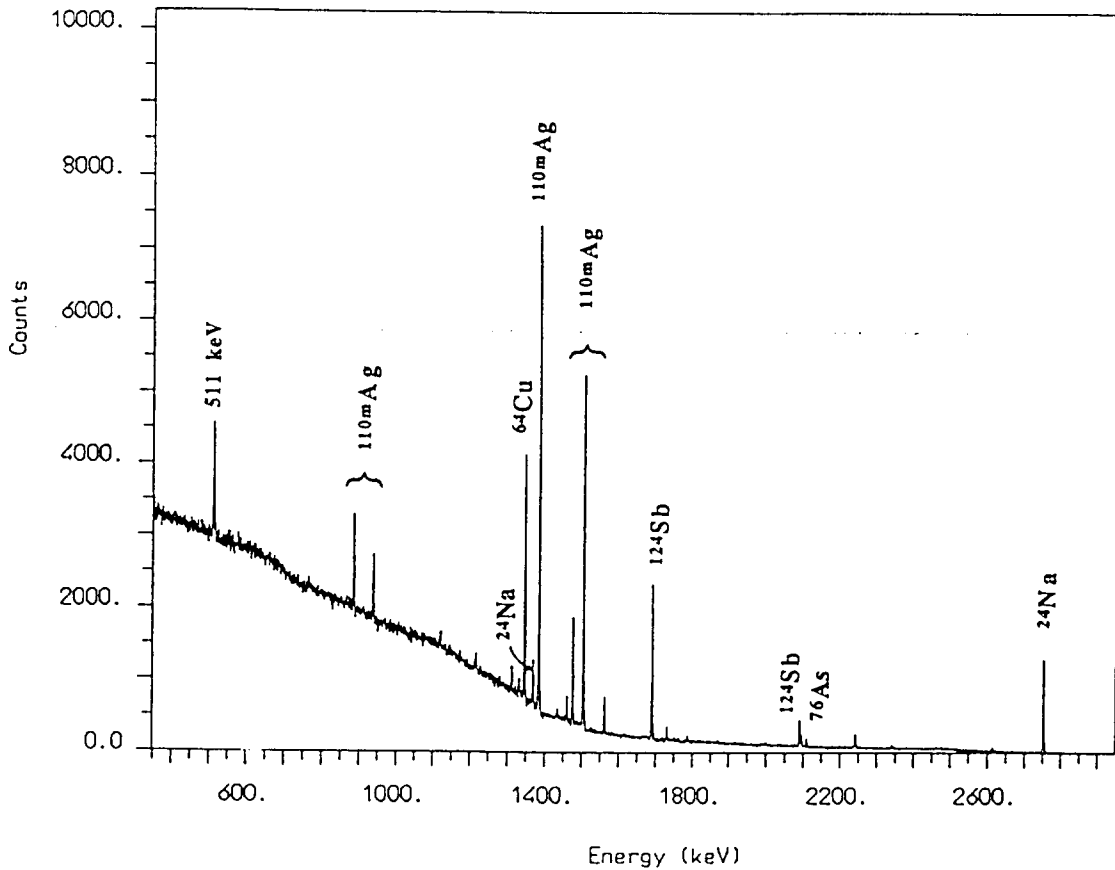
Figure 13: *Spectrum obtained with a Ge diode a few hours after removal of the source-plus-shield from the hot cell. The main γ-lines are indicated.*

samples were taken for activity measurements. The final amount in the source was $35\,530 \pm 10$ g of Cr. This net value was based on successive weighings in the hot cell of the mixing device, first filled with chromium and then emptied ($13\,710$ g). The activity of the source was then determined by several independent methods, as discussed below.

7.1 Sampling and direct activity measurements.

The granular form of the chromium metal was chosen to allow thorough mixing of the irradiated material in the hot cell, in order to average out the large neutron flux variations to which the chromium had been exposed. To verify the representative nature of this check, 31 samples were extracted from various positions in the mixture, as explained in section 6; their individual weights range between 0.5 and 1.0 g.

The activity of each very active sample (about 2 TBq/g) was measured at Saclay, using an ionization chamber [28] (see [27] for details on the technique). The chamber was calibrated with a standard ^{51}Cr source prepared by the French Bureau of Standards (Laboratoire Primaire des Rayonnements Ionisants, Saclay). We have performed a Monte-Carlo simulation of the source sampling procedure (see [31] for details) in which samples were generated under the assumption that there was perfect mixing of the grains and that the selected grains had a mass distribution identical with that measured before irradiation (see figure 4). The distribution of

the values of the specific activities of the Monte-Carlo samples was Gaussian, with a dispersion of 5.4%, consistent with the dispersion of the 31 samples of the real source (6.5% [28]). This study therefore demonstrates that the samples taken from the real source are representative. The variance of the 31 individual results is consistent with the assumption that the total of all these samples is representative of the full 35.53 kg of chromium.

After the ionization chamber measurements, these samples were totally dissolved in 10 ml of $7M$ H_2SO_4 at FzK-Karlsruhe, and diluted to 100 ml. Aliquots of these solutions were diluted by a factor of 10 000, for assay by high-resolution γ-ray spectroscopy with Ge crystals at MPIK-Heidelberg and FzK-Karlsruhe [29]. The volumes of the solution samples that were assayed were typically 1 ml. These samples were counted, beginning 10 months after EOB. At MPIK, a ^{51}Cr standard source in solution, obtained from Physikalisch-Technische Bundesanstalt Braunschweig (PTB = German Office for Physical and Technical Standards and Measures, at Braunschweig) was used to calibrate the efficiency of its detector; at FzK a reference standard was used (code QCY44 supplied by Amersham International, England), containing the nine radionuclides ^{109}Cd, ^{57}Co, ^{139}Ce, ^{113}Sn, ^{203}Hg, ^{85}Sr, ^{137}Cs, ^{60}Co and ^{88}Y (but not ^{51}Cr) in solution, with γ-ray energies ranging from 59.5 keV to 1836 keV. The advantage of using ^{51}Cr as a standard is that it eliminates any systematic error introduced by the uncertainty in the branching ratio of the 320-keV γ-ray, a feature exploited only by MPIK.

Eighty-ml portions of the 100-ml FzK solutions were shipped to BNL, where small aliquots (25-μl each) were taken and dried on aluminum cards to form counting samples that were very nearly point sources. Assay of the first of these Cr samples, by high-resolution γ-ray spectroscopy with a Ge detector, was begun 322 d after EOB. An absolute calibration of the Ge detector efficiency at BNL was made with a standardized ^{133}Ba radioactive point source from the U.S. National Institute of Standards (NIST). The four highest energy γ-rays emitted in the decay of ^{133}Ba (276, 303, 356, and 384 keV) bracket the 320-keV γ-ray of ^{51}Cr, providing a convenient means to calibrate the detector [30].

The results of the individual γ-ray measurements from the different laboratories are given in table 4, expressed as the disintegration rates of ^{51}Cr at EOB per gram of chromium, in units of TBq/g. Note in table 4 that, despite the differences in the experimental details used in γ-ray spectroscopy at MPIK, FzK, and BNL, the results are very similar.

The average specific activity of the 31 metal samples measured using the ionization chamber method is 1.71 ± 0.04 TBq/g [28] and, consequently, the total activity of the source is 61.3 ± 0.8 PBq at EOB. The total activity at EOB measured by the Ge crystals is 63.2 ± 0.9 PBq for Heidelberg, 63.1 ± 0.9 PBq for Karlsruhe [29], and 63.1 ± 1.0 PBq for BNL [30].

Sample number	Mass (g)	Ion. chamber	Ge MPIK	Ge FzK	Ge BNL	Vanadium
1.2	0.7053	1.850	1.891	1.893	1.922	1.815
1.3	0.7089	1.589	1.611	1.631	1.615	1.572
1.4	0.9399	1.848	1.898	1.933	1.914	1.875
1.5	0.7582	1.695	1.742	1.769	1.765	1.703
1.6	0.9459	1.728				
1.7	0.9869	1.724	1.744	1.755		
1.8	0.9689	1.692	1.743	1.775	1.726	1.682
1.9	0.8100	1.746	1.802	1.790	1.797	1.747
2.2	0.4334	1.903	1.958	1.938	1.946	2.100
2.3	0.4326	1.975	2.050	2.050	2.023	1.923
2.4	0.4710	1.772	1.843	1.792	1.829	1.773
2.5	0.6605	1.723	1.769	1.795		
2.6	0.7318	1.603	1.647	1.635	1.641	1.604
2.7	0.6753	1.763	1.814	1.854	1.798	1.825
2.8	0.6830	1.705				
2.9	0.6623	1.671	1.695	1.698	1.719	
3.2	0.3591	1.593			1.669	1.673
3.3	0.6679	1.517	1.592	1.616	1.605	1.545
3.4	0.6774	1.657	1.711	1.699	1.715	1.703
3.5	0.6449	1.644	1.693	1.708	1.712	1.699
3.6	0.7631	1.693	1.752	1.757	1.761	1.730
3.7	0.8138	1.715				
3.8	0.7442	1.712	1.777	1.747	1.754	1.700
3.9	0.7051	1.849	1.921	1.903	1.903	1.943
4.2	0.4733	1.764				
4.3	0.6542	2.082	2.112	2.115	2.137	2.155
4.4	0.8003	1.713	1.706	1.697	1.717	1.682
4.5	0.8388	1.723	1.730	1.764		
4.6	lost					
4.7	0.9831	1.676	1.707	1.720	1.708	1.682
4.8	0.8301	1.727	1.734	1.753	1.760	1.730
4.9	0.7058	1.616	1.609	1.627	1.639	1.610

Table 4: *Disintegration rates of ^{51}Cr (in TBq/g) at EOB of the 31 samples measured by the different methods. For columns 4, 5 and 6, Ge means gamma-ray spectroscopy with Ge diodes.*

mode	energy (keV)	branching fraction	<energy> (keV)
K	5.465	0.8068	4.409
K + γ	325.545	0.0882	28.698
L	0.628	0.0833	0.052
L + γ	320.708	0.0092	2.952
M	0.067	0.0113	0.001
M + γ	320.147	0.0012	0.398
total			36.510 ± 0.161

Table 5: *Ionizing radiation from the ^{51}Cr decay.*

7.2 Calorimetry.

The basic idea of the calorimetric measurement of the source strength is that the emitted γ, X, and Auger radiations will heat up the source and tungsten shield leading to a well-defined rate of temperature increase if the system is isolated from the environment. The energy going into atomic excitation of the daughter atom after electron capture is very close to the binding energy of the captured electron in the daughter atom [32]. We can thus calculate (table 5) the total energy released in the decay of ^{51}Cr (except for the neutrinos) from the decay branching ratio and the energy of the first excited state in vanadium [16], the L/K [33] and the M/L electron capture ratios, and the binding energies for K, L-I, and M-I electrons in vanadium [34]. No experimental information on the M/L electron capture ratio for ^{51}Cr was found in the literature; we therefore adopted 0.135 ± 0.035, a range which covers all experimental values from neighboring nuclei [35]. A total of (36.510 ± 0.161) keV is liberated per decay, corresponding to a heating power of (216.42 ± 0.95) W/PBq.

The calorimeter was calibrated by measuring the rate of temperature increase produced by a known input power provided by a calibrated resistance heater. The resulting value of the total heat capacity, C, of the source and tungsten shield was $(1.72 \pm 0.06 \times 10^5)$ J/K. Then, by comparing the rate of temperature increase measured for the source with the one previously obtained in the calibration, one can derive the power of the source. A source of 62 PBq results in a temperature increase of about 0.13 K/min. Though being less precise than the counting methods, the
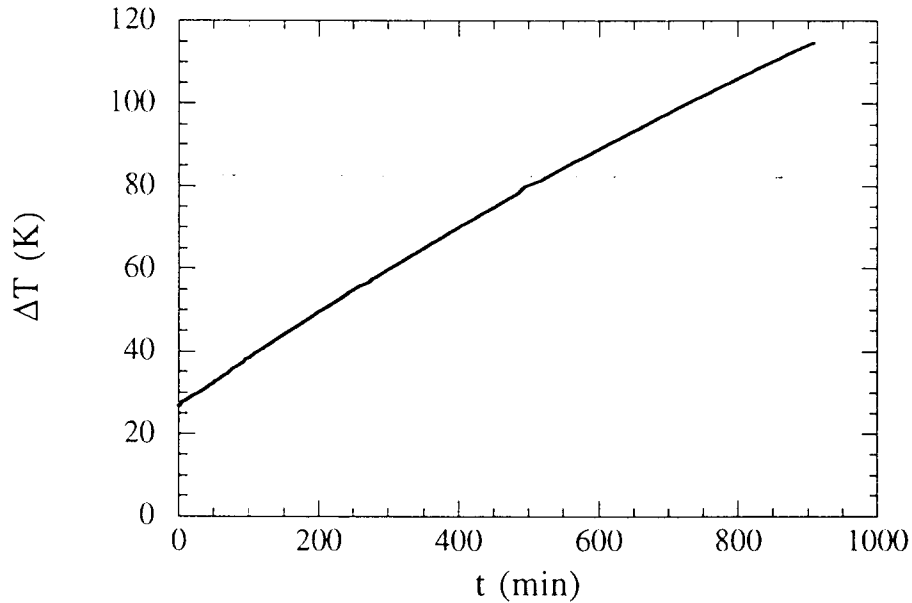
Figure 14: *The temperature of the combination of source and shield measured in the calorimeter as a function of time. (ΔT is the difference between the average internal temperature at any instant, t, and the external temperature).*

calorimetric method offers the advantage of not depending upon our ability to take representative samples.

The source and tungsten shield were suspended in the calorimetric vessel at the Siloé site. Good isolation of the system was assured by evacuating the vessel and by surrounding the tungsten with a radiation shield. The temperature of the tungsten was monitored with thermocouples placed on the surface and with one additional thermocouple in a hole inside the tungsten. The variation of the temperature, measured for the combination of source and shield, is shown in figure 14.

Figure 15a shows the rate of temperature increase for one of 7 runs done at Saclay with the known input power from the calibrated resistance heater and figure 15b, the analogous curve for data taken at the Siloé reactor with the power supplied by the source. Both curves demonstrate the influence of the heat leaks to the exterior.

The relevant parts of the curves were fitted to a straight line with the intercept yielding the ratio of power to heat capacity, P/C, and the slope yielding the inverse of the time constant of the system, τ. The heat capacity, C, is extracted from the known power in the resistance heater measurements. The source power, P, is then deduced at Siloé from the measured P/C value. A comparison of the data obtained with the resistance heater and at the reactor site leads to a measured value for the activity of (61.9 ± 3.0) PBq at EOB. The error (5%) reflects the dispersion of the heat capacities measured with the resistance heaters [36].
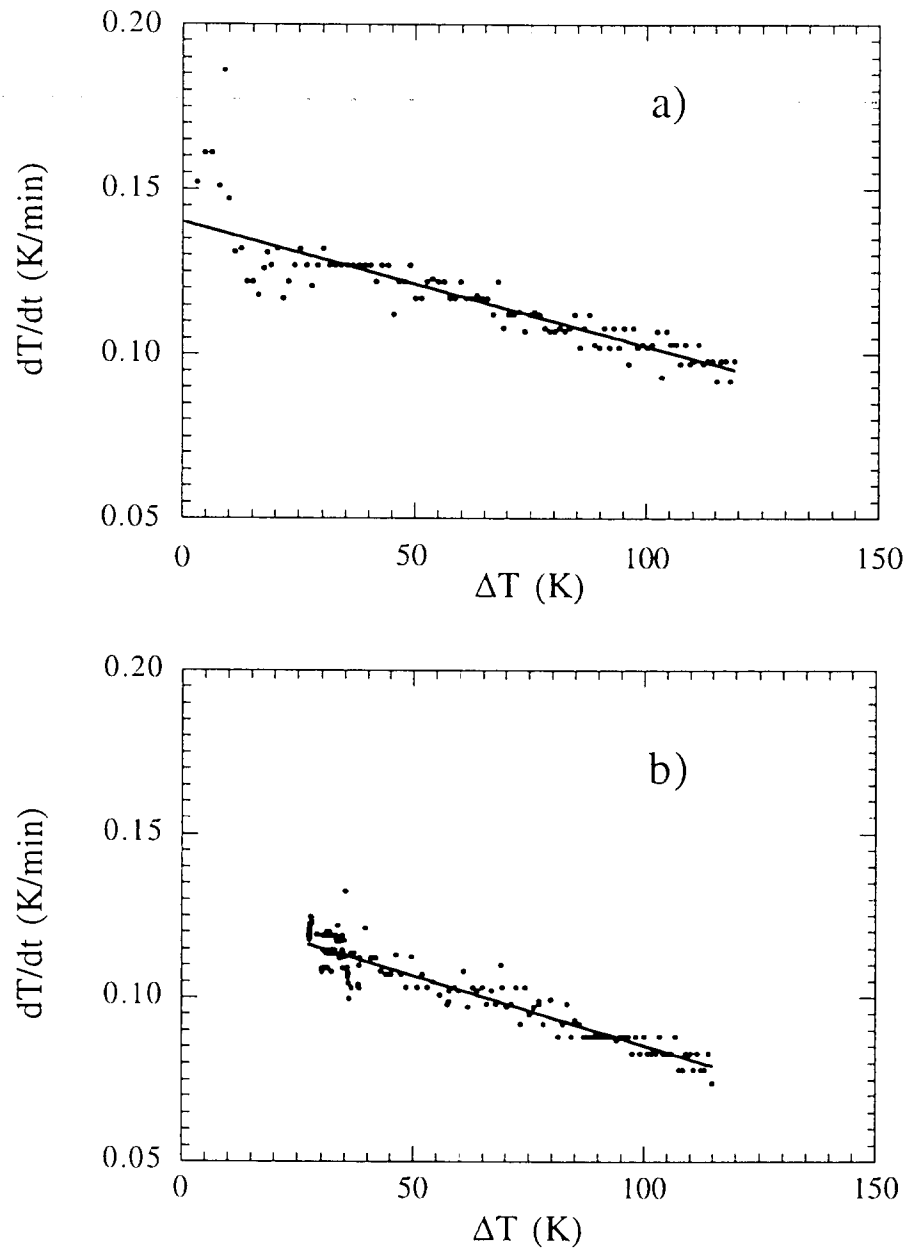
Figure 15: *a) dT/dt vs. ΔT for a measurement where the power is supplied a) by a resistance heater, and b) by the chromium source. The increase in a) at small ΔT reflects the heat deposit of the preheated chromium in the tungsten. This part of the curve is absent in b) at the reactor site since the start of measurements was delayed due to safety procedures.*

7.3 Neutronics and gamma-scanning.

The knowledge of the neutron flux and of the relevant capture cross section for producing ^{51}Cr provides another method for evaluating the activity of the source. Knowledge of the energy spectrum of the neutrons and of the energy dependence of the neutron capture cross section leads, by numerical solution of the neutron transport equation (Apollo-Marsyas and Neptune-Icare codes) for the Siloé configuration described in figure 5 [37], to the determination of the relative neutron flux as a function of position in the median plane of the reactor core. As explained in section 5.2, the symmetry of the core improves the precision of the computation. An on-line thermal neutron flux measurement in one crenel (irradiator 1) gives the overall normalization of the flux. The neutron flux distribution perpendicular to the median plane is deduced from the activity measured in the irradiators (see for example figure 7), as a function of the height of the chromium above the median plane. The value of the source activity deduced from these neutron measurements is (64.4 ± 5.2) PBq at EOB [37].

Gamma-ray scanning of the 320-keV γ-ray was performed in half of the irradiators (one in each crenel), in the so-called working pool of Siloé, during the transfer of the irradiators between their irradiation positions and the hot cell. This scanning was used to obtain the value of the total source activity independently of the neutronics measurements. A γ-ray spectrometer immersed in the working pool measured the vertical and radial photon distributions, as shown in figure 7. An absolute measurement of the activity was done at the center of each of the assayed irradiators. In each crenel, the activity of only one irradiator was measured, while the activity of the second irradiator was deduced from the activity of the measured irradiator, using a coefficient taken from the neutron flux calculations. This method gave (64.0 ± 5.2) PBq at EOB [37].

7.4 Measurement of the vanadium content.

Stable ^{51}V is the sole decay daughter of ^{51}Cr. In principle, measuring the number of atoms of ^{51}V that have grown into the Cr source after an interval of several ^{51}Cr half-lives then determines the number of ^{51}Cr atoms that were present at EOB. The well-known technique of neutron activation analysis (NAA), which is sensitive for vanadium at concentrations as low as ppb, was employed at BNL [30] to measure the ^{51}V contents of the Cr solutions that had been received from FzK (see Sect. 7.1 above).

However, this situation was aggravated by the fact that ^{51}V is not only produced from ^{51}Cr decay after EOB, but also from its decay during the lengthy Siloé irradiation. One could try to simplify matters by chemically removing this ^{51}V shortly after EOB to define a zero-time for the ingrowth of ^{51}V from ^{51}Cr decay. But such procedures would be difficult, requiring special equipment in hot cells to handle the intensely radioactive source.

By evaluating the well-known equations that govern the production and disap-

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pearance of ^{51}Cr and ^{51}V during the reactor irradiation, we demonstrated that this complicated chemical separation of vanadium from chromium was not necessary. These equations depend upon the length of the irradiation (assumed to be constant with time - see figure 8), the neutron flux, the neutron-capture cross sections of ^{50}Cr, ^{51}Cr and ^{51}V, and the radioactive decay rate of ^{51}Cr. For the conditions of the Siloé irradiation, the calculated result was that although the numbers of atoms at EOB of ^{51}Cr and ^{51}V changed with changing neutron flux, their ratio was a constant, 0.327, to better than 1 % over a factor of 10 change in flux.

This ratio is constant because the radioactive decay rate of ^{51}Cr is much larger than the neutron-capture rates in Siloé of ^{50}Cr, ^{51}Cr and ^{51}V. With these approximations, after an irradiation time t_{irr}, the ratio of the numbers N_V and N_{Cr} of ^{51}V and ^{51}Cr atoms at EOB is independent of the neutron flux :

$$\frac{N_V(t_{irr})}{N_{Cr}(t_{irr})} = [\lambda_1 \, t_{irr} \, \frac{1}{(1 - \exp^{-\lambda_1 \, t_{irr}})} - 1] = 0.327 \qquad (1)$$

where λ_1 is the ^{51}Cr decay constant.

After a sufficiently long time after EOB, τ, essentially all of the ^{51}Cr has decayed to ^{51}V, and :

$$N_V(\tau) = N_{Cr}(t_{irr}) + N_V(t_{irr}) = 1.327 N_{Cr}(t_{irr}) \qquad (2)$$

Independent of flux variations, the disintegration rate of the ^{51}Cr at EOB is simply related by a constant factor to the measured amount of ^{51}V in the source at τ [30].

Neutron irradiation of ^{51}V produces ^{52}V, which decays with a 3.746 min half-life and emits a 1434-keV γ-ray. One-ml samples of the Cr solutions from FzK were irradiated (about 1 year after the Siloé irradiation) in the 3-MW BNL Medical Research Reactor, at thermal neutron fluxes of $\approx 10^{13}$ n/cm^2.s simultaneously with comparators containing a known amount, 10 μg, of natural vanadium. Irradiation of the Cr unknown sample together with the vanadium comparator, followed by γ-counting of both samples under identical conditions, eliminates the need to know the values of the ^{51}V neutron-capture cross section, the neutron flux, and the detector counting efficiency. In this relative method, the ratio of the ^{51}V masses in the unknown and the comparator is equal to the ratio of their ^{52}V activities, taken at the end of the BNL reactor irradiation, as obtained by least-square analysis of the respective ^{52}V decay data.

The resulting NAA values were in the range of a few μg of vanadium per ml of the individual Cr solutions; these vanadium concentrations were all $\geq$600 ppm (μg of V per g of Cr). By comparison, separate NAA measurements of samples of the enriched Cr metal that had never been exposed to the Siloé neutron flux showed that they contained negligible levels of vanadium impurities, $\leq$5 ppm.

Eq. (2) can be transformed into a relation between the disintegration rate of the ^{51}Cr at EOB, $\lambda_1 \, N_{Cr}(t_{irr})$, and the measured ppm of ^{51}V. The derived disintegration-rate values in units of TBq/g (0.002579 $\times$ (number of ppm of ^{51}V)) are listed in the last column in table 4.

7.5 Mean value of the source activity.

The mean values obtained by the different independent methods (see Table 4) are in excellent agreement, as recapitulated in Table 6.

Method	Laboratory	Value (PBq)
Ionization chamber	Saclay	61.3 ± 0.8
Ge spectroscopy	Heidelberg	63.2 ± 0.9
Ge spectroscopy	Karlsruhe	63.1 ± 0.9
Ge spectroscopy	Brookhaven	63.1 ± 1.0
Calorimetry	Grenoble/Saclay	61.9 ± 3.0
Neutronics	Grenoble	64.4 ± 5.2
Gamma scanning	Grenoble	64.0 ± 5.2
Vanadium content	Brookhaven	62.3 ± 1.1
Weighted mean		62.5 ± 0.4

Table 6: *Summary of the different measurements of the source activity, quoted at EOB.*

The agreement of these results within their quoted errors allows us to calculate a mean value, where the individual values are weighted by the inverse of their respective dispersions (see last line in Table 6). The corresponding disintegration rate of the ^{51}Cr source at EOB in Siloé is 62.5 PBq, with a small error of ± 0.4 PBq (1.69 ± 0.01 MCi). To our knowledge, this is the strongest low-energy neutrino source ever produced. The individual errors include estimates of the systematic errors, but the methodology involved in the various determinations is significantly different, and does not guarantee the homogeneity of these systematics. A more realistic estimate of the error could be obtained from a more conservative calculation of the "maximum error" on the mean, which takes into account the range of individual results and which yields $^{+1.9}_{-1.2}$ PBq [9].

8 The source at Gran Sasso

8.1 Transport.

The transport of a radioactive material must follow the safety regulations of the International Atomic Energy Agency (IAEA, Vienna). For a source activity greater than 30 TBq, such as the present one, the safety authorities require that a special procedure must be arranged for the transport. Although the tungsten shield is sufficient to ensure biological protection and prevent any leakage of radioactive material, it does not fulfill other criteria related to a possible accident during transport, such as ability to withstand free-fall from a height of 9 m, ability to resist a fire with temperatures up to 800 °C for 30 min or to remain leak-free during immersion
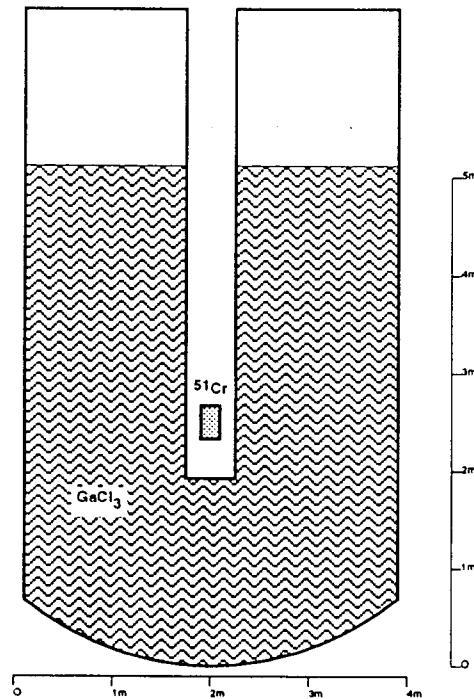
Figure 16: *Schematic drawing of the GALLEX A-tank containing the gallium chloride solution with the shielded chromium source inside the central reentrant tube.*

in water at a depth of 15 m for 8 hours. A standard container (called CC10) does exist which satisfies these criteria for transporting radioactive material. It consists mainly of stainless steel, but with an inner liner of wood and plaster. Its weight is 2700 kg. The solution adopted was thus to insert the source with its tungsten shield inside the CC10 container, where it was attached with wood supports. The entire assembly fulfilled all IAEA regulations. The dose rate on the external surface of the CC10 container was smaller than 1 μSv/h. A special truck, from an authorized nuclear transport company, carried the source from Grenoble to Gran Sasso in 15 hours, on June 23, 1994.

8.2   Installation and monitoring.

At the LNGS, the shielded source was removed from the CC10 container and immediately mounted in the central reentrant tube ("the thimble") of the GALLEX A-tank (see figure 16). The time between EOB and introduction of the source into the thimble was 3.69 days, equivalent to a decrease of 8.8 % in activity.

To avoid any heat transfer from the source to the gallium solution, which could change the kinetics of the germanium desorption [38], there is a thermal copper

shield cooled with tap water (at a flow rate of 60 l/h) between the source and the wall of the thimble (see figure 12).

The source experiment started on June 23, 10.36 pm (local time), when the ^{51}Cr neutrino source was inserted in its final position in the center of the GALLEX A-tank.

To allow for the decay of ^{51}Cr , exposures were shorter than for solar runs. The length of the exposures was varied so that more or less the same number of atoms of ^{71}Ge was extracted in each run ; in particular the first extraction was made only 3.35 days after the introduction of the source, but the last exposures lasted for 2 to 3 weeks.

8.3 Results.

Results have been reported elsewhere [8, 9] and can be summarized as follows : the ratio of the radiochemically determined activity from ^{71}Ge counting, $(60.4 \, ^{+6.7}_{-6.4})$ PBq [9], and of the directly measured activity as described in this paper, (62.5 ± 0.4) PBq, is 0.97 ± 0.11. This value provides a quantitative demonstration of the overall efficiency of the GALLEX experiment. It shows that the 40 % deficit of the solar neutrino flux observed by GALLEX as compared to the solar model predictions [3] cannot be attributed to experimental artifacts. In fact, the source experiment directly demonstrates the validity of the basic principles of the radiochemical methods used in our detector : some ten radioactive atoms produced by neutrino capture can be quantitatively removed from tons of target material, purified and counted.

9 The future.

A second source experiment was started by GALLEX in October, 1995. Its purpose is to reduce the error obtained in the first experiment ($\approx$10 %, see [9]) to a value of $\approx$7 %, which is similar to the expected value of the error on the GALLEX solar neutrino measurement at the end of the experiment (December 1996) when about 60 solar runs will be finished.

This second source experiment was begun by reirradiating the original enriched chromium metal from the first source. Less than 0.1 % of the ^{50}Cr had been transformed to ^{51}V during the first irradiation.

The enriched chromium will be available to interested scientists in the future to produce ^{51}Cr neutrino sources for future solar neutrino detectors, for example, Borexino, HELLAZ or Indium experiments, or for any other scientific purpose. However we should note that the Siloé reactor will not be available after the end of 1997. As said previously in section 5.1, the other reactors at present are not suitable for such an irradiation.

Acknowledgements

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