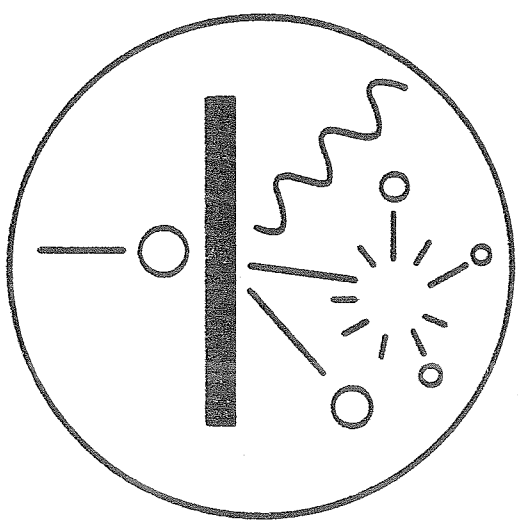
INTERNATIONAL NUCLEAR

TARGET DEVELOPMENT SOCIETY

NEWSLETTER



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Editorial

We would like to renew our request to our Membership to send us short reports on their recent activities for publication in the INTDS Newsletter.

Also information on publications made elsewhere and letters which are of general interest for our Members as well as questions are welcomed.

Thanking you in advance for your contribution.

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1. Rolling of Cd plunger targets on Au backing

A trick has been found to make Cd plunger targets (0.5 mg/cm^2) on Au backing (2mg/cm^2). If they are rolled straightforward starting with 4 mg/cm^2 Cd and 20 mg/cm^2 Au they get wrinkles after an hour and it is no longer possible to stretch them in such a way that they become really flat. We were able to make stable targets by at first rolling them to double the required thickness and leaving them in that stage for some hours. The best way is to put them between the new steel sheets for the next rolling process before waiting this time because otherwise it can happen that they form a very tiny roll which is difficult to unroll. When they are rolled to their final form after that they do not change any more.

2. Other targets made by rolling

Mo 0.6 mg/cm^2 (it has been found that it is important to roll it with a very low velocity to avoid pinholes), Cd 0.5 mg/cm^2 on Ta 3.4 mg/cm^2 (starting with 4 and 21 mg/cm^2 and low pressure until it sticks together), Pb 5 mg/cm^2 (from 20 mg/cm^2 between teflon sheets), Zn 2 mg/cm^2. There have been made also 1.7 mg/cm^2 Ni targets and Pd 5 mg/cm^2 on sandwich backings of 70 mg/cm^2 Bi and Cu of the same thickness starting with 2, 170, and 150 mg/cm^2 of material (there was 1 mg/cm^2 In evaporated on the Cu layer). All three foils are rolled together in 2 or 3 steps. To get a good sticking it seems to be important that the Ni or Pd is freshly made by rolling and that the surface of the Bi is rubbed by some paper to remove any oxide.

3. Selfsupporting targets made by evaporation

The following targets have been made by evaporation from an electron gun on a Betainmonohydrate layer and floated of in water:
Al 8 µg/cm^2, Si 5 µg/cm^2, Se 20 µg/cm^2, Ni 20 µg/cm^2, ZnS 30 µg/cm^2, and PbF_2 100 µg/cm^2.

Preparation and Characterization of a set of $^{235}UF_4$ reference fission foils

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I. INTRODUCTION

Considering the importance of the ^{235}U (n,f) cross section as a reference cross section and its direct importance in reactor applications, the knowledge of this cross section with 1 % accuracy is required. One of the contributing components of the uncertainty is the absolute mass of ^{235}U. Therefore an intercomparison of fission samples obtained from various laboratories was organized by W.P. Poenitz and J.W. Meadows (3). For this purpose, CBNM prepared a series of accurate and high quality reference layers of $^{235}UF_4$ on stainless steel substrates.

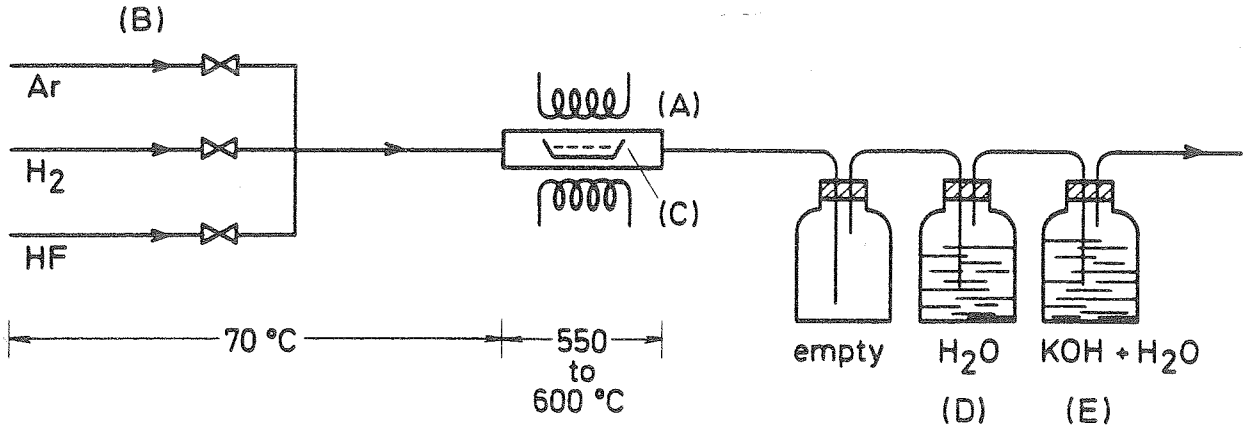
II. PREPARATION OF UF_4

The starting material consisted of highly purified U_3O_8, NBS standard reference material U-970, certified for use as an isotopic standard.

The certified isotopic composition is (1) :

U 234	:	1.6653 ±	0.0017	at %
U 235	:	97.663 ±	0.003	at %
U 236	:	0.1491 ±	0.0005	at %
U 238	:	0.5229 ±	0.0006	at %

This material was transformed into UF_4 by direct hydrofluorination (2). Anhydrous UF_4 results from the reaction of U_3O_8 with anhydrous HF at 550° C to 600° C, under addition of H_2 gas. The equipment used (Fig. 1) consisted essentially of a tube furnace (A) and gas lines (B) with valves and gas cylinders for HF, H_2 and Ar. All gas lines, valves and fittings were made from monel with gaskets from teflon in order to prevent reaction with HF. All gas supply tubes (B) were heated to 70° C to avoid condensation of HF. The hydrofluorination was carried out in a Pt-boat (C). The reaction time was about 1 hour.



(A) Tube furnage
(B) Gaslines
(C) Pt-boat
(D) Bubbler with H_2O
(E) Bubbler with KOH solution

Figure 1 : Hydrofluorination apparatus

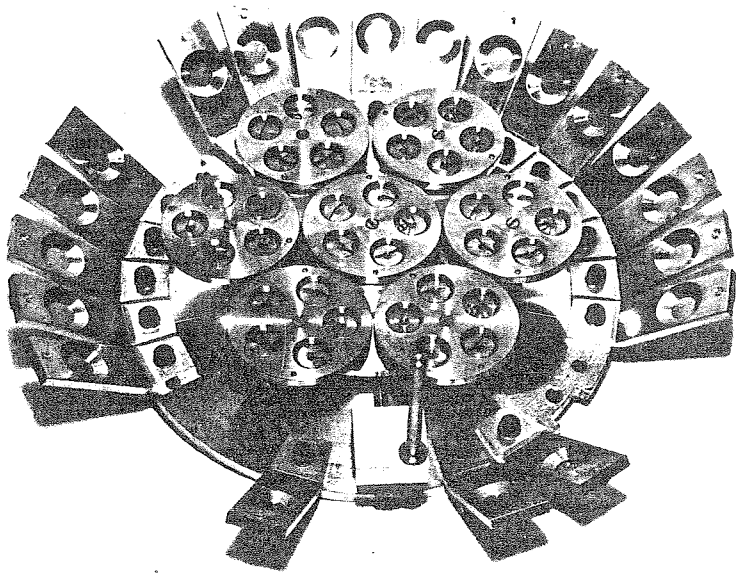


Figure 2 : Multi-substrate holder

III. VACUUM DEPOSITION OF UF_4

UF_4 was used as the source material for vacuum deposition. The layers were prepared by evaporation in vacuum (10^{-3} Pa) from a resistance heated Ta-crucible.

3/4" stainless discs were placed at about 25 cm from the evaporation source, in a rotating substrate holder (Fig. 2). The evaporated spot was 1/2". Deposits of about 100 and 200 $\mu g/cm^2$ were prepared.

After evaporation, the isotopic compostion was checked again with the following results :

U 233	:	< 1.3 ppm		
U 234	:	1.6642 ±	0.0030	at %
U 235	:	97.6540 ±	0.0075	at %
U 236	:	0.1495 ±	0.0025	at %
U 238	:	0.5323 ±	0.0010	at %

The slight difference between both isotopic measurements is not significant as far as the specific activity of the material is concerned :

Sp. Act. based on NBS I.A. : 3.9007 ± 0.0103 (0.26%)

Sp. Act. based on CBNM I.A. : 3.8982 ± 0.0118 (0.30%)

Sp. Act. (CBNM) / Sp. Act. (NBS) = (0.9994 ± 0.0040) %

IV. CHARACTERISATION OF THE DEPOSITS

Two sets of five deposits (100 and 200 $\mu g/cm^2$) were measured in such conditions to achieve the highest accuracy on two samples of each thickness.

The samples were characterized by relative alpha counting of all samples in a low geometry and subsequent destructive analysis by isotope dilution mass spectrometry (IDMS) of some of the samples. IDMS yields the number of atoms with a known accuracy. Thus the experimental determination of a calibration factor (cps/μg ^{235}U) was possible, allowing the characterisation of the remaining deposits with an accuracy depending on a) the precision of low geometry alpha-counting of each target b) the precision of the experimental calibration factor, c) the accuracy of IDMS.

Uncertainties on the half-lives of the different isotopes of uranium (mainly ^{234}U) thus become unimportant in these determinations of the of uranium 235.

a. Relative alpha counting

The relative alpha activity was measured using a 600 mm^2 Si surface barrier detector, an aperture of 20 mm diameter, and a source-to-aperture distance of 50.00 mm.

The targets were loaded on a turn-table, and positioned one by one in the axis of the detector by an x and ρ movement of the table controlled by a microcomputer. Variations in height due to this movement have been determined to be $\leq$ 0.03 mm. Hereby introduced errors are covered by the uncertainty on the experimentally determined alpha-counting/IDMS ratio.

The spectrum of the detected alpha particles was registered on a 512 channel analyzer. The energy drift was verified at each measurement. The relative activity was determined by observing the counts between well fixed channels (corresponding to a lower energy of ca. 1.0 MeV and an upper one of ca. 6.3 MeV). They were corrected for an experimentally determined background, which was the mean of 11 countings covering a period of 5 weeks. Furthermore each target was counted 6 times in 2 different positions on the turntable.

b. Isotope dilution mass spectrometry

IDMS was carried out by spiking each sample with an approximately equal amount of natural uranium.

A gravimetric solution was prepared by dissolving NBS standard reference material SRM 960, uranium metal, in nitric acid.
A known number of atoms of this natural uranium was added to each individual deposit as a fraction of this solution. Complete dissolution and isotopic homogeneization was achieved subsequently. Then a chemical purification was performed by ion exchange : after chemical conditioning the solution was transferred to a column, loaded with Dowex 1 x 4 (100 - 200 mesh) resin, and eluted with 8 M HNO_3. After evaporation to dryness the fraction was redissolved in 1 M HNO_3 to yield a solution with a concentration of 10 mg U/ml.
Dilution ratio measurements were carried out by double filament thermal ionization mass spectrometry on 2 µg samples. The ratio $^{235}U/^{238}U$ was corrected for an experimental isotope fractionation of 0.1 %.

The isotopic analysis was carried out by measuring the $^{235}U/^{238}U$ ratio, as well as the $^{234}U/^{235}U$ and $^{236}U/^{235}U$ ratios on 10 µg samples. The $^{235}U/^{238}U$ ratio measurement was done by magnetic field peak-switching and using a vibrating read amplifier, voltage to frequency conversion and data taking by a H.P. 9830 computer. The $^{234}U/^{235}U$ and $^{236}U/^{235}U$ ratios were measured by manual magnetic scanning and using a strip-chart recorder equipped with an expanded scale device.
All results were corrected for bias using the NBS uranium isotopic standard reference materials.
^{233}U could not be detected at a detection limit of 1 ppm.

c. Experimental results

The results of the calibration procedures are summarized in Table I. These results take into account a background correction of (0.0035 ± 0.0003) cps.

Target	Relative alpha counting (cps)	IDMS (µg ^{235}U)	cps/µg ^{235}U
35N-31	4.564 ± 0.004	118.57 ± 0.29	0.03849
32	4.575 ± 0.005	119.01 ± 0.29	0.03344
37	9.400 ± 0.010	245.51 ± 0.62	0.03829
40	9.404 ± 0.008	244.56 ± 0.62	0.03845

Calibration factor : $\bar{x}$ = 0.03842 +0.00009 (n = 4) (0.24 %)

Table I : Calibration of rel. alpha counting with IDMS

On the basis of this calibration factor the following results were obtained for the delivered targets :

Target	Certified values (µg ^{235}U)
35N-33	119.01 ± 0.43
34	119.07 ± 0.43
35	118.79 ± 0.43
35N-36	244.07 ± 0.88
38	245.29 ± 0.88

Table II : Results certified on the basis of rel. alpha counting and IDMS (target 35N-39 was used to control the isotopic composition)

Target	Abs. alpha counting	Rel.α-counting+IDMS	IDMS	α-counting (3)	Fission ratio mass (3)
35N-31	119.0 ± 1.0	-	118.57 ± 0.29	-	-
32	119.3 ± 1.0	-	119.01 ± 0.29	-	-
33	119.2 ± 1.0	119.01 ± 0.43	-	119.4 ± 0.5	119.6 ± 0.4
34	119.3 ± 1.0	119.07 ± 0.43	-	-	-
35	119.0 ± 1.0	118.79 ± 0.43	-	-	-
36	244.5 ± 2.1	244.07 ± 0.88	-	244.8 ± 1.0	245.6 ± 0.7
37	245.1 ± 2.1	-	245.51 ± 0.62	-	-
38	245.8 ± 2.1	245.29 ± 0.88	-	-	-
40	245.2 ± 2.1	-	244.56 ± 0.62	-	-

Table III : Intercomparison of results (in μg ^{235}U)

V. COMPARISON WITH OTHER RESULTS

The relative countings used above were also exploited to obtain comparative figures on an independent basis. The count rates could be used as such, as the cut-off below 1 MeV was negligible (< 0.01 %) as were corrections for self-absorption, back-scattering and pile-up. Corrections for dead-time were carried out automatically, but are also small (≤ 0.04 %). The geometry factor was calculated to be 104.16. The uncertainty on this calculation was verified to be $\pm$ 0.8 %. The half-lives used to calculate the specific activity of the material are :

U 234	:	$(2.454 \pm 0.006)\ 10^5$ yrs
U 235	:	$(7.037 \pm 0.011)\ 10^8$ yrs
U 236	:	$(2.342 \pm 0.003)\ 10^7$ yrs
U 238	:	$(4.468 \pm 0.005)\ 10^9$ yrs

The difference in results between the two methods is 0.2 %, which is largely inside the uncertainties of both methods. They are furthermore in good accordance with alpha counting measurements and fission ratio measurements with samples of various origins carried out at Argonne National Laboratory (3).

REFERENCES

(1) NBS, Certificate of Analysis, Standard Reference Material U-970 (30.7.1970)

(2) J. Van Audenhove et al., N.I.M. 167, 61-63 (1979)

(3) W.P. Poenitz and J.W. Meadows, Argonne Nat. Lab., private communication

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