Radiochemical cross sections measurements on nuclear reactions induced with 14.5 MeV neutrons.

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RADIOCHEMICAL CROSS SECTIONS MEASUREMENTS ON NUCLEAR REACTIONS INDUCED WITH 14.5 MeV NEUTRONS

THESIS

Presented in candidature for the degree of
MASTER OF SCIENCE

in the

UNIVERSITY OF DURHAM

by

FLORIN BUNUS, ing. chim. (St. Cuthbert's Society)

MEMORANDUM

This thesis is the result of the research work carried out at the Londonderry Laboratory for Radiochemistry of Durham University between October 1961 and October 1962 under the supervision of Mr. G.R. Martin, B.Sc., A.R.C.S., F.R.I.C., Reader in Radiochemistry.

The results of the present work were obtained by the author and no part of this thesis has previously been submitted for a degree in this or any other university.

Florin Bunus

ABSTRACT

A Radiochemical study of cross section measurements in nuclear reactions with fast neutrons (14 MeV) has been carried out based on an activation technique.

As a source of 14 MeV neutrons the D + T nuclear reaction has been used. The deuterons were accelerated in a Cockroft Walton installation and Titanium (or Zirconium) tritiated targets have been used.

As a neutron flux monitor the $Fe^{56}(np)Mn^{56}$ nuclear reaction has been chosen.

The (np),(nd) and $(n\alpha)$ reaction in Tungsten target has been investigated and a chemical method of separation between the elements involved (W, Ta, Hf) has been developed.

Activity measurements were made with an end window gas flow proportional counter calibrated by reference to a 4π gas flow proportional counter. Tantalum and Hafnium induced activities were always counted in oxide form.

A discussion of the results obtained is given and compared, when possible, with other similar data.

In a subsequent study (np) and (nd) nuclear reactions in a Mercury target resulting in very short activities of Au^{202} (25 sec.) and Au^{203} (55 sec.) have been investigated using a 100 channel pulse height analyser (associated with a scintillation counter) in addition to beta counting technique.

To determine the emission of Au^{202} and its half life and $(n\alpha)$ reaction in Thallium target resulting in the same Au^{202} has been investigated.

Here also a discussion of the results obtained is given.

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INTRODUCTION

A great deal of information concerning nuclear structure has been obtained by examination of cross sections (σ) as a function of incident neutron energy. Nuclear cross section is defined as an area, but its size is of the order of nuclear area only for high energy neutrons when $\lambda \ll R$ and $\pi R^2 \approx 3.10^{-24}$ or 3 barn. The total cross section is of the order of few barns, but partial cross sections differ for different types of reaction and element (1) (2) (3) (4).

At lower neutron energies when $\lambda > R$ wave properties appear and the cross section gets larger than the value mentioned, (may be thousands of times higher).

The simplest method to measure the total cross section is by transmitance. ($\sigma_{total} = \sigma_{non\ elastic} + \sigma_{elastic}$). Generally a total cross section will be a summation of other partial cross sections ($\sigma = \sigma_1 + \sigma_2 + \sigma_3$). The total cross section obeys the 1/v law but usually many resonances are superimposed. For light elements resonances are widely spaced so they can be observed at high energies (in MeV region). For heavy elements the resonances are closed spaced and they are observed only at low energies.

Each resonance represents an excited state of the compound nucleus formed after the addition of a neutron.

Each such state or energy level has its properties deduced from these resonances. At higher neutron energies apparently the resonances disappear but this is quite unreal because of the increasing resolving power, the smooth curve is formed by resonances spaced closer together. The width of levels increases with increasing neutron energy and gets closer spaced, eventually they overlap when continuum is attained. In this region the cross section determination gives an idea of nuclear levels(3).

The excitation energy in the compound nucleus is present in definite states (levels). The formation of compound nucleus is very probable or its formation cross section is high when incident neutron has exactly the energy to form the compound nucleus in one of its many states. Later a neutron or gamma ray is emitted and the nucleus is found in a true "bound state" (which cannot emit neutrons). This bound state has a probability to emit γ quanta (falling to a lower final state).

The theoretical treatment of various cross sections as a function of energy is based on nuclear models that allow its calculation.

A nuclear theory has been developed in order to predict neutron cross sections in terms of a nuclear model. Starting with this point of view a model is proposed and is compared with the experiment. In this case a nuclear model is used to predict the cross section and an improvement of the theory is looked for. A good example when measurements played an important role in improving the theory is the behaviour of fast neutron cross section which led to the inclusion of nuclear transparency in the model.

The compound nucleus theory, when an incident particle bringing its binding energy + kinetic energy is strongly absorbed in the nucleus, sharing its energy with all the nucleons, long living and forgetful of mode of formation, was very close with reality. It has recently been found that this model of compound nucleus (analogous to the liquid drop) is not entirely correct. The bombarding particle may travel a long distance (several nuclear diameters, before being absorbed. This transparency proved to be very successful in many cases. Weisskopf estimates the cross sections for this model of partial transparency. In this model many nuclear levels are filled, the incident neutron may travel a long path before being absorbed(3). If neutron energy increases, the chance to be absorbed gets higher because more energy levels are available to it. Experiments proved that at 15 MeV the nucleus is more black, but above 15 MeV the decreasing interaction cross section out-weighs the increasing level density and the nucleus becomes more 'transparent'. At present these interactions (elementary particle cross sections) using free particles can be measured.

So far it has been found from experiments that incident neutrons do not amalgamate with the rest of nucleus when its energy is very low (Pauli principle), nor when it is very high due to weak interactions.(1),(3) Actual theory refers to both models.

A model is very important to predict a cross section and a cross section to test the model. As was mentioned much of the information about nuclear structure comes from the study of the variation of cross section with the energy of incident neutrons.

But for slow neutrons this variation is very irregular and the nuclear excitation levels can be estimated from the fine structure of the excitation curve.(2) To estimate the fine structure monoenergetic sources of neutrons of desired energy are required.(preferable adjustable).

Sources of monoenergetic neutrons

Monoenergetic sources of neutrons of sufficient intensity of low energies are based on two techniques:

- time of flight (neutron velocity selector, mechanically chopped neutrons).

- crystal spectrometer velocity selector.

These two methods succeed in selecting neutrons of defined energy from a broad energy spectrum. This method applies up to a few KeV.(1)(3)(4)

Other sources of higher monoenergetic neutrons are based on nuclear reactions with light nuclei. Sources to provide monoenergetic beams are generally those charged particle reactions producing neutrons in which input energy is kept low enough to avoid production of excited states. The best reactions are with Hydrogen isotopes.

Even in the case that neutron energy spectrum is not monoenergetic, time of flight technique may be used in the range mentioned or electronic pulse methods of filtration in MeV range.

Initially Ra-α-Be sources of neutrons have been used(3)(4) but such determinations are useless since the neutron spectrum is wide. (And over Z = 10 the Coulomb barrier becomes important for α's of 5 MeV).

- Accelerators as: Van der Graaff, Cockroft-Walton, cyclotron, syncrocyclotron, cascade generators have been used to accelerate the bombarding particles (p, d,) for this purpose.

The most useful reactions are those with Li, T for (pn) and D and T for(dn). Sometimes photoneutrons from H(γn) or $Be^9(\gamma n)Be^8$ have been used. Monoenergetic neutrons from $Li^7(pn)Be^7$ have been obtained but the negative Q value of this implies a threshold energy of 1.88 MeV for incident protons.

The $H^2(dn)He^3$ has a positive Q value (3.25 MeV) so neutrons are produced at low bombarding energies. Since no excited state of He^3 is present, monoenergetic neutrons up to 7 MeV can be produced (4). The neutrons from $Li^7(dn)Be^8$, $Be^9(d,n)B^{10}$ are always contaminated with neutrons of lower energies due to Be^8,B^{10} nuclei being left in excited states.(3)(4) For $T(pn)He^3$ there is a threshold of 1.019 MeV due to negative Q.

$T(dn)He^4$ can produce neutrons from 12-20 MeV. It is an extremely useful source of high energy neutrons since the reaction has a very high, broad resonance with a peak at 109 KeV deuterons, also a good yield of high energy neutrons is obtained with low energy incident particles. A very important fact is the total absence of excited states of He^4 so the neutrons are monoenergetic (14 MeV). For a thick Tritium gas target and 600 KeV deuterons the yield is 5 x 10^8n/sec per μ amp (for Tritium adsorbed in thick Zirconium targets). With 200 KeV deuterons the yield is 10^8 n/sec per μ amp.(4) For these low incident particle energies, neutrons emitted at 90° are used since their energy is 14.1 MeV (4) and insensitive to incident deuterons energies.

Paul and Clarke(6) used this neutron source successfully in measuring activation cross sections for fifty seven elements.

This study includes reactions as: $(n\gamma)$; (np); $(n\alpha)$; (n,2n).

Neutron flux measurement

In absolute fast neutron nuclear reaction cross section measurements, the precise neutron flux determination is an important factor on account of the impossibility of detecting neutrons directly. The indirect methods based on secondary effects have been used.

- A method known as the associated particle technique is based on measurement of charged particles emitted in the nuclear reaction, for example: $T+d = He+n$, in this case α particles are detected.(3)(4) So α particles from a precisely determined solid angle are counted with a proportional or scintillator counter.

- Another method is based on scattering of neutrons by protons. The protons of recoil from a hydrogenous material are counted. A knowledge of n-p scattering cross sections is required and it is made precisely by the transmission technique.(3)(4) Here also the geometry plays an important role and solid angles must be well known. The neutron source must be punctiform, this involves appreciable distances between source and sample and since neutron flux decreases in accordance with $1/d^2$, a good geometry is attained at the expense of neutron flux intensity. It is a limiting factor for both methods in usual laboratory conditions. We cannot afford the flux reduction when the neutron source produces about

$10^9 n/cm^2 sec$ especially for low cross sections.

- The third method of secondary standard is adequate for small nuclear reaction cross sections.(4)(16).

In this method the activity induced in the sample is compared with that induced in a reference exposed to the same flux.

So from the known cross section of the reference element the unknown one can be deduced but the precise determination of the first one is an important factor which affects the results.

The reference cross section must be established either by recoil particle or the associated particle method. In one method (4) (13) (7) a sample as a thin disc is sandwiched by two thin reference discs. After irradiation the activity of each reference disc is counted and a mean value of it may be related to the neutron flux. The method is reliable when all discs are thin. (In order to calibrate this activity in terms of neutron flux a separate experiment was carried out when neutron flux from the target was found by counting the associated α's). This description is not suitable when the sample is powder as very often happens. For this reason another method has been developed (16) and it was suitable for our work. In this method the sample and the reference substance are well mixed and after irradiation the sample is separated from the reference element by a suitable procedure and counted separately. This method developed at Durham has the following advantages:

- Sample is irradiated in powder form.

- Fluctuations in neutron flux during irradiation play no role because both sample and reference, being homogenously mixed are submitted to the same neutron flux.

- No precise geometry is necessary for the same reasons.

- The sample may be placed close to the target so maximum neutron flux is attained.

- Sample in powder form facilitates the chemical dissolution after the separation of sample-reference.

As a consequence of these advantages the reference element used as a secondary standard must respect the following criteria:

a) To have a high cross section determined exactly.

b) It should be easily separated from the sample to allow short lived activity to be counted.

c) It should have a convenient half life to permit a later counting.

d) It should have a quite high threshold energy to discriminate against the lower energy neutrons.

In literature (16) (13) the following reactions have been used as standards:

$Fe^{56}(n,p)Mn^{56}$

$Al^{27}(n,\alpha)Na^{24}$

$Cu^{63}(n,2n)Cu^{62}$

In our work when sample is used in the powder form the best standard is given by the first reaction because it allows rapid separation. The value for the cross section assumed from literature is 124 mb but a more reliable value is about 100 mb obtained in Durham using the associated particle method. (Based on measurements of very small volumes of He 10^{-8} cm^3.)

Fast neutron cross section determinations

The first important fast neutron cross section determinations were obtained by Paul and Clarke.(6) Their experimental results were compared with theoretical ones which were calculated on the basis of evaporation theory (5) due to Blatt and Weisskopf. They found that for (n,2n) reactions the theoretical predictions were in close agreement with experimental results but for other types as (n,p) $(n\alpha)$ especially for heavy nuclei the experimental values are much higher. In their work as neutron source the $T(dn)He^4$ reaction was used. Making speculations on the reasons for deviations of (n,p); (n,α) results, from evaporation theory and the fitting of some results they assume that in those cases when compound nucleus is formed the evaporation theory is approximately correct and the deviations are due to other process of interactions between incident neutron and only one proton or alfa particle which are emitted before much energy sharing takes place.

The nuclear reactions studied were due to (np); $(n\alpha)$; and (n,2n).

In that concern the radiative capture cross sections for 14.5 MeV neutrons an important study (7) has been made with about 30 nuclides. The cross sections found were of the order of 5 mb except for the lightest of the nuclides examined.

The value of 5 mb found for radiative capture cross sections for 14.5 MeV neutrons is higher than one predicted from the theory of the compound nucleus. The low values observed near the magic numbers (neutron) are due to low level densities of nuclides with a closed neutron shell structure.

In a later study (8) the authors using 14.5 MeV neutrons have determined the cross sections for (np) and $(n\alpha)$ in the mass range $100 < A < 250$. They found that (np) cross sections fell gradually from 10 to 1 mb in this mass range, if Paul and Clarke (6) found an increase in cross sections it should have been determined by the fact that most of their measurements were restricted to nuclides with mass less than 100.

In this range their supposition is correct.

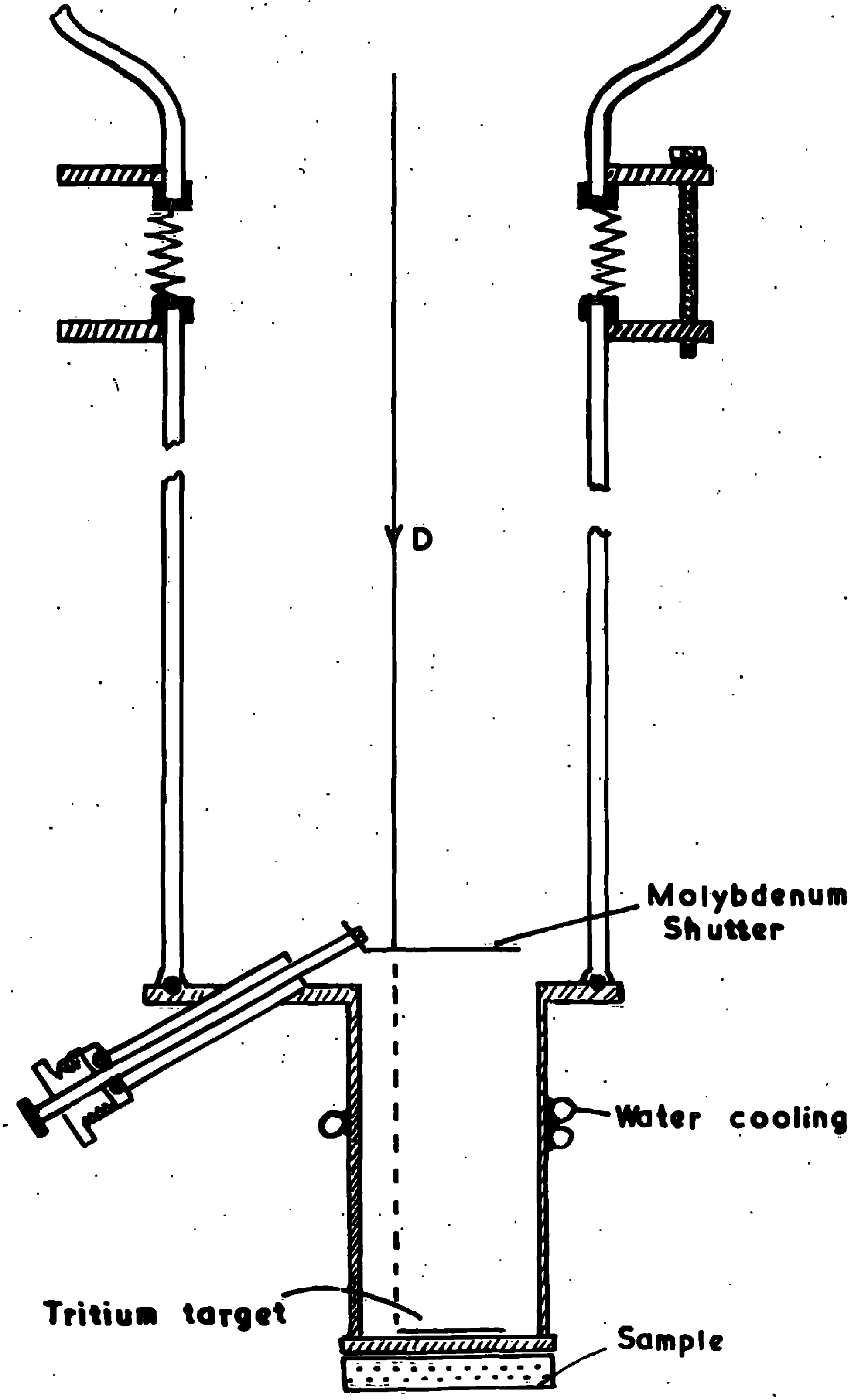
In 1957 Brown and Muirhead (9) explained (np) cross sections due to Paul and Clarke if protons can also be emitted by direct interaction process. In this case the incident neutron interacts with a proton which may be in the body of the nucleus (contrast with surface theory) and this type of process is complementary to the compound nucleus mechanism.

For target elements with mass number greater than 100 the Coulomb barrier still has little effect on the emission of highly energetic protons obtained from direct interaction with fast neutrons (14.5 MeV) but proton emission from a compound nucleus will be virtually prohibited by the Coulomb barrier, so this last contribution can be neglected.

There are some previous measurements for (np) and (nα) cross sections (10) (11) (12) and the authors have included them in their study. Their conclusion for (np) reaction for heavy nuclides is a downward trend from 10 to 1 mb over the mass range 100-250 and there is no trend from o to 100 mass range where values are from 10 to 400 mb. The authors have calculated for each experimental result the theoretical cross section predicted by direct interaction theory of Brown and Muirhead and they inclined for this theory in 100-250 mass range.

FIGURE 1

The target assembly


D
Molybdenum Shutter
Water cooling
Tritium target
Sample

For ($n\alpha$) reactions there is no satisfactory theoretical treatment.

There is very little experimental study for (n,pn) and (n,np) reactions with heavy nuclides targets.(14) (15). In one study (13) the authors have measured (nd) cross section with 14.5 MeV neutrons of five heavy nuclides using activation method and have compared their results with theoretical ones predicted by direct interaction theory. Of course by activation methods one cannot distinguish between (n,pn); (n,np) or (nd) reactions.

For the five nuclides that have been measured at this neutron energy (13-21 MeV) it seems that the cross sections determined are very dependent of neutron energy. They found that Brown and Muirhead direct mechanism satisfy the experimental results.

Apparatus and experimental procedure

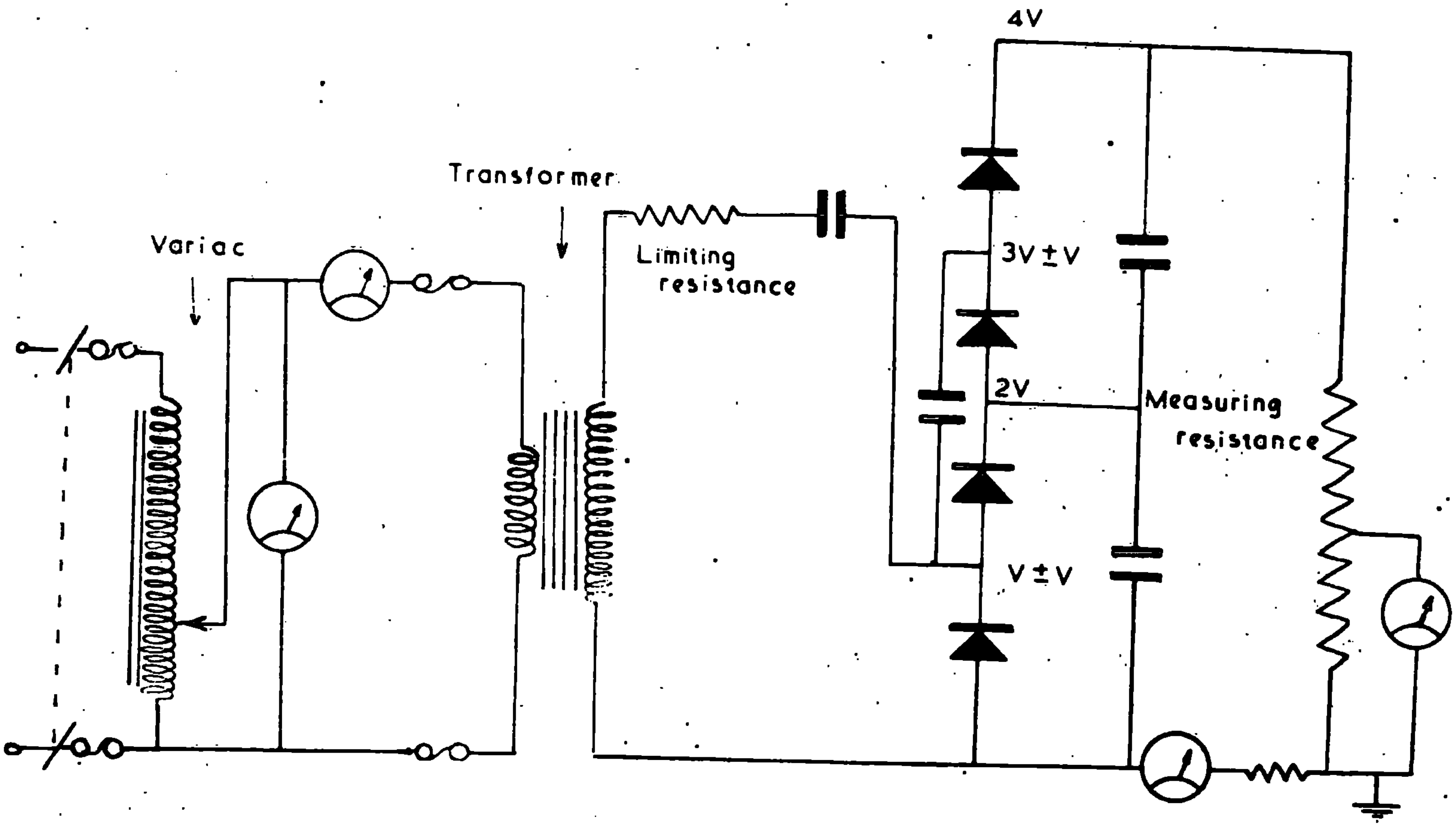
1) Neutron source

As we have mentioned the nuclear reaction $T(dn)He^4$ is used to obtain neutrons of 14.5 MeV. Tritium is adsorbed in a thin Titanium foil supported by a Copper disc. Tritium activity is about one Curie (0.3 ml S.T.P.) Each disc is divided into four segments and were fixed on the brass target block.

In Fig. 1 it is given the description of the target

<u>FIGURE 2</u>

Voltage-Quadrupling circuit



VOLTAGE-QUADRUPLING CIRCUIT.

assembly.

Very close the sample to be irradiated is fixed.

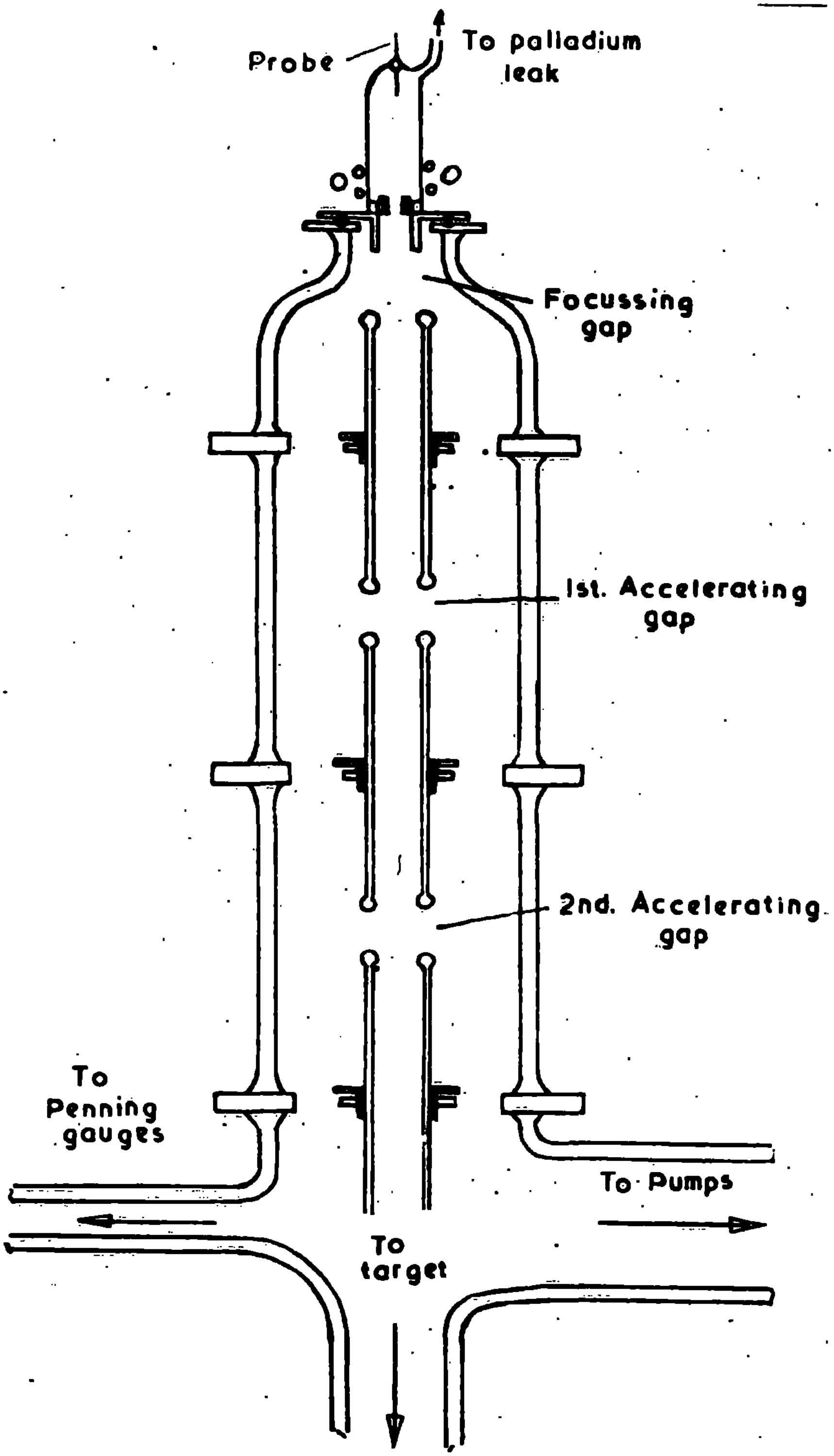
Bombarding particles are deuterons accelerated in an accelerating tube, which hit the target producing fast neutrons.

This nuclear reaction has a high wide resonance for 109 KeV deuterons, so the deuteron beam must have a suitable energy which is produced with an accelerating tube of a Cockroft-Walton accelerator. It consists of a quadrupling circuit described in Fig. 2. The 240 V a.c. with a variac is feeding the primary circuit of a transformer which can supply 100 KV output in its secondary circuit. This is fed in the quadrupling circuit (rectifiers, condensers) and the high d.c. voltage is feeding the gaps of accelerating (linear) tube, Fig. 3.

The deuterium is produced by the electrolysis of heavy water and the gas is supplied to the top ion source equipment of the accelerating tube. Ionisation is produced with high frequency electrodeless discharge, and the ions extracted by a potential difference applied for this purpose. The deuterons are accelerated between the gaps due to d.c. high voltage applied from the accelerating equipment described. (quadrupling circuit).

FIGURE 3

Accelerating tube



ACCELERATING TUBE.

The gap between successive sections has both focussing and defocussing action on the deuterons. Entering the gap is focussed and leaving it is defocussed but focussing action is much stronger since deuterons move slower when entering the gap. The cylindrical accelerating electrodes are enclosed in a glass tube connected with a vacuum pumping system.

The target assembly is situated in the middle of the target room to avoid scattering neutrons (walls) entering the sample. The target assembly is provided with a Molibdenum shutter which can be open to permit the deuterons to hit the target.

Deuterons have 130 KeV energy and the beam produced 200 µamp.

The neutrons produced are not exactly monoenergetic, their energy is nevertheless defined between 14.5-14.9 MeV limits.

Fluctuations in neutron flux result from variations in magnitude and position of the deuteron beam and from the gradual ejection of tritium atoms from the target under bombardment; the accumulation of decomposed pump oil on the target surface also reduces the neutron yield.

We have also to take into consideration the build up of deuterium in the tritium target and with the time neutrons of 2.5 MeV are produced by $H^2(dn)He^3$ nuclear reaction but their contribution can be neglected in our case. (Low activity induced in the sample so only new tritium target was used).

Unless fortuitously, the half-lives of monitor and reaction product are the same, it is necessary that the variation of neutron flux during the time of irradiation must be exactly known.

These fluctuations were monitored by a proton recoil scintillation counter (high biased to eliminate lower energy neutrons). The neutron counting is made at intervals shorter than half life of the induced activity. This fluctuation in the neutron flux must be known for the study of fast neutron cross sections.

2) Counting equipment and auxiliary facilities

(a) Geiger-Müller (end window) counter.

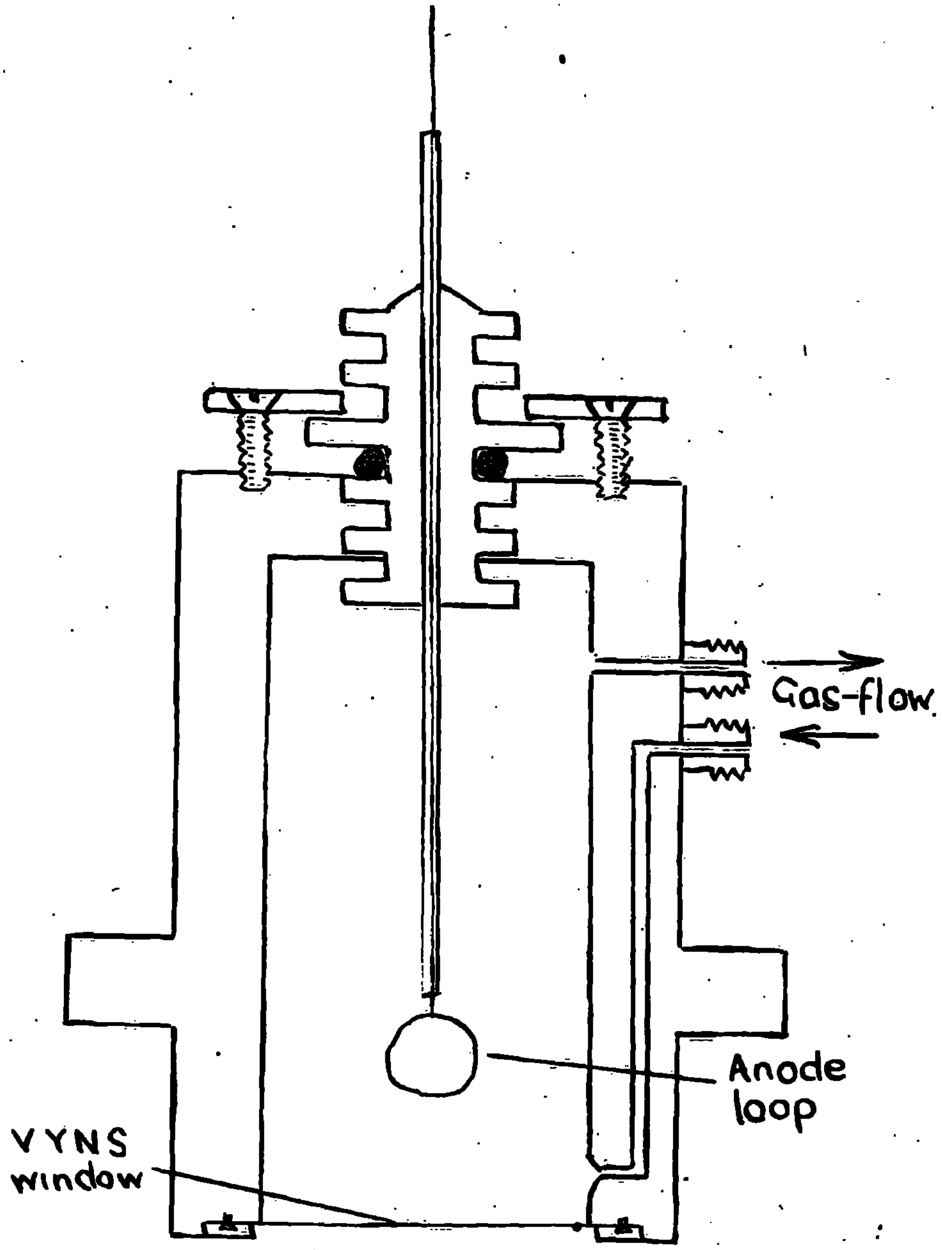
It was used for informative determinations concerning radioactive isotopes determinations, added as tracers in performing a chemical method. The counter characteristics: HT 1650 V, a plateau with a slope 2%, dead time 500 μsec. The average background (lead castle) 20 c/min.

(b) Halogen quenched liquid counter.

This has been used for the determination of Mn^{56} activity obtained with fast neutrons in: $Fe^{56}(np)Mn^{56}$ when Fe has been used as a reference element in a mixture with the sample.

FIGURE 4

The end window proportional counter


Gas-flow
Anode loop
VYNS window

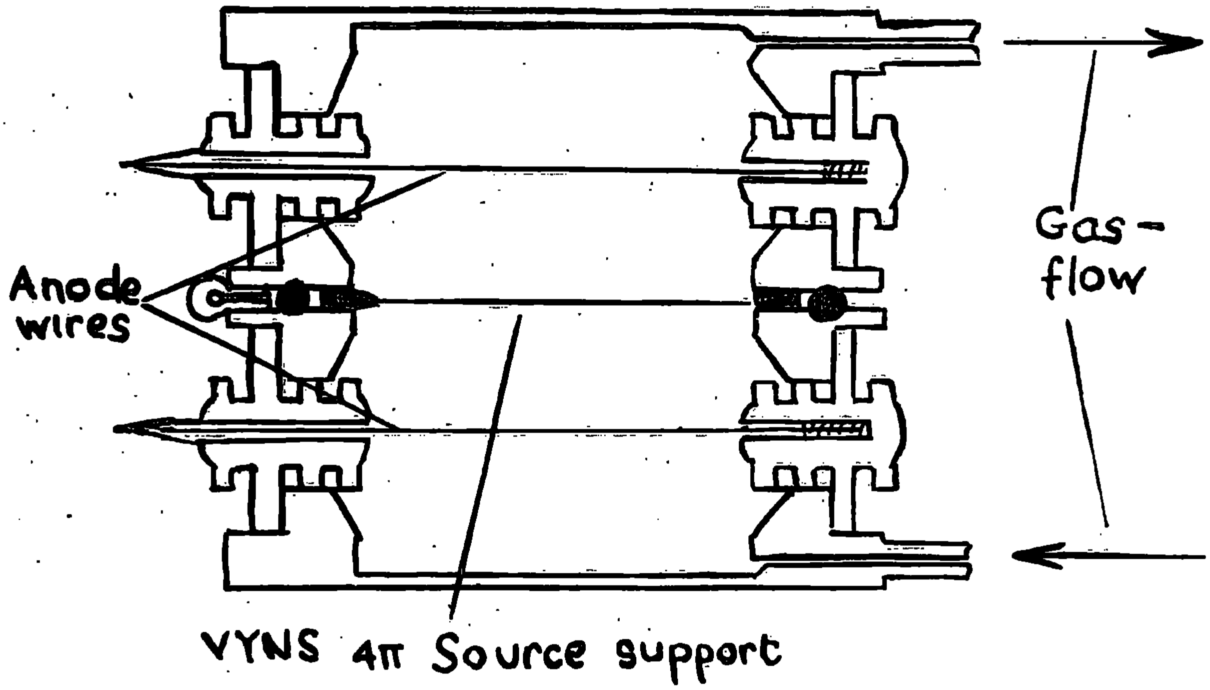
It was calibrated with a 4π gas flow proportional counter for Mn^{56} and its efficiency found to be 8.635% and optimum HT 350 V with a good plateau of several hundred volts. Its dead time was 500 μsec and for this reason always a dead time correction was necessary having to deal with high activities. Background in the lead castle had values between 13-15 c/min.

(c) The end window gas flow proportional counter for β rays.

It has a cylindrical form, made from brass with an internal diameter of 2.6 cm and at its end with a very thin window, a plastic film (VYNS, 70-80 $\mu g/cm^2$ thickness) one side gold coated to be conductive. Inside the counter, centrally situated is an anode loop ⅜ inch diameter made from constantan (0.001 inch diameter). It was soldered in a thin Nickel tube insulated from the counter walls with a teflon plug Fig. 4, and also provided with a gas inlet and outlet. The flow gas is a mixture of 90% argon and 10% methane purified not to contain even traces of oxygen, H_2O etc. Its flow rate must be about 0.5-0.75 ml per second (16) and care should be taken to avoid overpressure (slightly over atmospheric pressure) because of the fragile window. This counter needs a HT 1800 V with about 300 V long plateau, its paralysis time 50 μsec and the background in a lead castle 12-15 counts/minute.

FIGURE 5

4π proportional counter


Anode wires
Gas-flow
VYNS 4π Source support

This counter has very reliable properties, giving reproducible results. The end window proportional counter has been used for current activity determinations and was calibrated (efficiency) with a 4π proportional counter.

(d) 4π proportional (gas flow) counter.

It was used for absolute activity measurements (disintegration rate) of radioisotopes selected to calibrate the end window proportional counter. It is very advantageous because all β rays are emitted in the counter volume and are counted.

Even emission of γ rays (following β's) or anihilation radiation being emitted within the dead time interval are not counted. Similar with other secondary effects as β's scattering etc. So its 100% efficiency for β rays make it to be very useful for disintegration rate measurements.

Pate and Yaffe (17) have described the characteristics obtained in a series of determinations with an improved technique.

In our studies, the 4π counter used, consisted of two cylindrical parts hinged together Fig. 5, provided in an equatorial plane with the source support. The two pieces are then separated by the source, but source support permits the gas (CH_4 10% + argon 90%) to flow through the two chambers (groove at the edge). The counter is provided with two anode wires (one for each part) fixed in good insulator at the two ends (Teflon). The counter plateau was never less than 400 V and HT 1750 V, its paralysis time 50 μsec. Background in lead castle 35 c/min.

When the source is changed, the counter is open and after each operation there is a time of 10 minutes until the counter regains its sensitivity.

In normal conditions ionisation produced gives a unit probability response but a small fraction of energy (of β rays) will not give 1 response. This fraction increases as the maximum energy of the spectrum decreases. One principal factor which may affect response probability is the source. The source support is a very thin VYNS plastic film (17) (16) supported by Aluminium rings (2.6 cm. diameter).

This Aluminium frame is fixed in between the two counter chambers.

The VYNS film (20 $\mu g/ cm^2$) is one side gold coated, to make it conductive. The preferable gold thickness 2 $\mu g/cm^2$ and may be determined spectrophotometrically. A thicker film may affect β rays of low energy.

Radioactive source must be deposited on the central area of the film. The best position described by an area with 1 cm. diameter. The radioactive residue to be of the order of 10 μg or even less for the same reasons (for low energy β emitters).

The preparation of source support (VYNS film) used in our study is given by Pate and Yaffe.(17) In our work we have prepared a stock solution by dissolving 1 weight resin in 2 weights solvent. Before use it is diluted; 2 volumes of the stock solution with 1 volume of solvent.

When thin saturated solution of VYNS in cyclohexanone is spread over the surface of water, the cyclohexanone is dissolved and a VYNS film is left on the surface. So this film should be placed on the Aluminium support and later gold coated in vacuum. On the opposite side one drop of radioactive solution should be placed but previously to allow a uniform spreading, one insulin drop (1 mg insulin dissolved in 10 ml water + 1 drop HCl conc.) with an appropriate equipment allow to wet the central area.

So with insulin as spreading agent a drop of radioactive solution, dried permits a uniform central distribution of the activity.

(e) Scintillation counter (NaI crystal activated with Tl) has also been used in our determinations.

The scintillating crystal (either a plain cylinder or a "well crystal" as appropriate) is optically coupled to an electron multiplier photo-tube.

(f) 100 channel pulse height analyser (associated with a scintillation counter).

The unit sorts pulses into 100 channels each with a storage capacity of 16,383 ($2^{14}-1$). The pulses produced by the detector are sorted into channels as a function of their peak voltage. The pulses are stored in a delay line and the same time displayed on a cathode ray tube screen. The storage unit is a Nickel delay line (allow 14 binary digits in each channel then 14 x 100 = 1400). Channel width is selected between two values; $\frac{1}{3}$ V and 0.1 V.

Pulses with the same amplitude will be directed into the same channel and stored.

A back bias enables the input pulse to be biased between 0 and 50 V.

On the screen there are 100 vertical rows of 14 dots arranged that lower voltages are on the left and higher on the right hand side of the tube. Values of the dots in vertical direction are from bottom to the top 2^0, 2^1, 2^2, 2^3....

To estimate the channel number the possibility to shift the X deflection exists every fifth channel.

The unit is also provided with coincidence and anticoincidence facilities.

The unit includes also a timing equipment which permits counting runs to be carried out for a preset live time and compensates for analyser dead time and no correction for this is required. The time interval chosen depends on activity and at higher activity is longer because the analyser is "dead" for a larger fraction of the time. The Kicksorter is provided with a unit which allows the data stored to be printed in octal notation on roll paper and full automatic recording under the control of the live time integrating time unit.

The unit is also provided with a manual control. When channel information has been recorded, if automatic control operates, the analyser and timing scaler begin counting again for the same time interval prefixed and followed by another recording a.s.o. It is also provided with controls which make it possible to limit the number of channels printed to those of immediate interest, thereby reducing the print-out time.

For each determination, initially, the scintillating crystal is calibrated with standard radioisotopes and a peak activity versus channel number permits an estimation of sample isotope energy.

The very short half lives isotopes activities cannot be printed since printing time for 100 channels last 90 sec.

For this purpose a camera was arranged to be actuated by the timer and it records in two seconds on film the spectrum displayed on the cathode tube screen. When the camera shutter is closed automatically the 100 channels

Kicksorter start counting again, the timer operates the shutter, when it is open the counting is stopped and spectrum displayed on the screen is photographed.

(g) When proportional end window counter has been used to follow a decay curve for a long time (40 hours) an auxilliary recording equipment connected with the scaler have permitted a preset number of counts to be recorded on a roll paper the same time with minutes time intervals (pulses from a timer unit).

14 MeV neutron cross sections in Tungsten

In this study a Tungsten target has been selected to make an exact determination of fast neutron cross sections for nuclear reactions possible. The same time a Tungsten target permits an (nd) reaction study where the experimental data are quite absent.

In our experiments the target has been used in a powder form for reasons we have just mentioned in a previous chapter.

Iron granules homogeneously mixed with the target element has been used as reference element. The sample mixture in a polythene tube was placed near target assembly.

When optimum deuteron beam is obtained, the Molibdenum shutter was opened and fast neutrons are produced.

During the time of irradiation (1 hour) the neutron counting rate is recorded at 10 minutes time intervals.

The following more probable nuclear reactions (theoretically) with the natural isotopic mixture of Tungsten:

(W^{180} 0.14%, W^{182} 26.2%, W^{183} 14.3%, W^{184} 30.7%, W^{186} 28.7%)

(n,2n)

W^{180} (n,2n) W^{179} (40 min, IT 7 min); W^{182} (n,2n) W^{181}(145d); W^{186} (n,2n) W^{185}(70d)

(n,p)

W^{182}(n,p) Ta^{182}(115d, IT 16 min); W^{183}(n,p) Ta^{183}(5d); W^{184} (n,p) Ta^{184}(8.7h); W^{186}(np) Ta^{186}(10 min)

(n,α)

$W^{184}(n\,\alpha)\,Hf^{181}$ (45 d); $W^{186}\,(n\,\alpha)\,Hf^{183}$ (64 min)

(nd)

W^{186} (nd) Ta^{185} (49 min)

We only mention the radiative capture possibility $(n\,\gamma)$ when the following isotopes are obtained: W^{181}, W^{183} (5 sec), W^{185}, W^{187} (24 h).

In our study we shall exclude the Tungsten isotopes because low activity as well as very complex decay scheme for many isotopes, very long or very short half lives, and (n,2n) combined with $(n\,\gamma)$ reactions make it practically impossible.

The nuclear reactions taken into consideration are (np); (nd); $(n\,\alpha)$. For relatively short irradiations (1 hour) only Ta^{184}, Ta^{186} as (np); Ta^{185} as (nd) and only Hf^{183} as $(n\,\alpha)$, will be significant. All other nuclear reactions give products of half life so long that no significant activity is to be expected.

To verify this supposition we have found from a preliminary irradiation (without a chemical separation) that a decay curve shows only a short component of 50 min and a long one of 8 hours half lives. This has been done with an end window proportional counter. The short component of 50 minutes cannot be ascribed to Ta^{185} 49 min only because

Ta^{186} 10 min and Hf^{183} 64 min.

Using a single channel pulse height analyser and waiting about 70 minutes after the end of irradiation when all Ta^{186} 10 min is gone, we tried to separate Ta^{185} 49 min of Hf^{183} 64 min. We have done a calibration of scintillating crystal with the three peaks of Se^{75} and from nuclear data we have chosen the 0.175 MeV, γ quanta of Ta^{185}, (with appropriate channel limits). A similar decay curve has been obtained but it was quite impossible to be resolved.

From these determinations it seemed clear that a chemical separation between the three elements W, Ta, Hf was a necessity, and of course proportional counter is preferable because β rays are detected with higher efficiency and precise calibration and reliable measurements are possible.

All three radioisotopes involved in our study are β and γ emitters.(38)

Ta^{184} β^- of 1.06 (25%); 1.26 (72%); 1.36 MeV (3%).
Ta^{185} β^- 1.48 (5%); 1.72 (95%) complex γ spectra
Hf^{183} β^- 1.4 (100%).

Chemical methods associated with fast neutron cross section determinations in Tungsten target

In bombarding Tungsten target with fast neutrons Tantalum and Hafnium isotopes are built up as well as radioactive Tungsten isotopes. Their chemical separation must be performed in order to estimate the radioisotopes obtained.

It is also very important to ensure such a chemistry that the separated isotope should be in a pure chemical form at last or should not contain other radioactive contaminant elements and exchange equilibrium must be established between carrier and activity. It is preferable that the chemical yield should be high but it is not essential since finally a chemical yield determination is made.

The first step after irradiation is the magnetic separation of Iron granules from the Tungsten target (powder).

The next step is the disolution of the sample in a convenient reagent ensuring complete solution of Tungsten and the same time of Hafnium and Tantalum.

Because Hafnium and Tantalum radioisotopes are in tracer quantities the reagent used should already contain Hafnium and Tantalum carriers.

A very important factor is the time interval in which the chemical separation is performed having in view that the shortest half life taken into consideration is 49 min.

Starting with these points of view we shall discuss briefly some of the existing methods from the literature.

Tungsten metal is only superficially attacked by acids, aqua regia but its rapid dissolution is obtained with a mixture of acids as $HF + HNO_3$ or $HClO_4 + H_3PO_4$. (18)

Hafnium is acid resistent being attacked only by aqua regia, HF and better by a mixture of $HF + HNO_3$.(21) Hafnium dioxide (HfO_2) which is used in our experiments as carrier is slowly attacked by $HF + HNO_3$ mixture.

Tantalum is insoluble in aqua regia but readily dissolved in a mixture of $HF + HNO_3$.(18)

The solution chemistry of Hafnium is not well known and considerable confusion exists in the literature regarding the ionic species in the aqueous solution. This depends on the fact that Hafnium ions undergo hydrolysis and polymerisation in aqueous solution, which is strongly dependent on pH.(19) The only important valency is +4.

Hafnium has some very insoluble compounds as phosphate, iodate, cupferrate etc. The hydroxide is precipitated with ammonia. Barium and HF give barium fluorohafnate ($BaHfF_6$) which is insoluble in excess fluoride and affords a good separation from Tantalum.

The usual form in gravimetric analysis procedures is ignited HfO_2 following the precipitation as hydroxide ($HfO_2.XH_2O$),(22) cupferate etc. Mandelic and Flavianic acids have also been used.(20)

In addition to hydrolysed species, Hf forms complex ions with many reagents. The fluoride is the most stable complex. It also forms chelate complexes with acetylacetone, TTA, cupferron, etc.

It is important to underline that Hf tracer is strongly co-precipitated with most precipitates in acid solutions not containing complex-forming ions.

Hf with alkali fluoride in aqueous solution can give also these complexes: K_2HfF_6 and K_3HfF_7.(23)

Tantalum(24) in aqueous solution exists only in the form of complex ions. Tantalic acid is readily hydrolysed. Alkaline hexatantalates are soluble. Its important valence state is +5.

Tantalum may be precipitated with some tungsten by tannic acid from tartrate or oxalate solution. From a tantalate solution hydrated oxides are precipitated when acid is added.

Cupferron has been extensively used to precipitate Ta from tartrate or oxalate solution.

The usual gravimetric procedure is precipitation as hydrated oxide, tannate or cupferrate followed by ignition to the pentoxide.

Only in the presence of complex forming anions (tartrate, oxalate, fluoride) is tracer Tantalum stable in solution.

In absence of this, tracer Ta may be carried by such precipitates as MnO_2, TeO_2, dust, silicagel etc.

Tantalum with HF forms only TaF_7^{--} complex (24) and with a fluoride (KF) in HF solution K_2TaF_7 (23) slightly soluble in cold water. The existence of a compound of the type $TaF_8.^{---}$ is known only with NaF.(23)

The same type with Tungsten is also possible but only with KF,(K_3WF_8); it has a high solubility.(23).

A number of chelate complexes are also known.

Some of these chemical properties of different compounds have been successfully used in obtaining a chemical separation among the W,Ta,Hf elements.

Some precipitation methods have been used for this separation:

One of them (18) is based on pronounced differentiation of chemical behaviour between some elements in oxalate solution so Tantalum is more readily precipitated than Hafnium with tannin. This method is not always adequate: it depends on the relative amounts of the elements involved and Tungsten, if present, is carried by the Tantalum.

The method is tedious and the time required is much too long.

A separation between Ta and W with silicic acid is described in (25) but completeness of separation is affected by the presence of Hf or Zr.

In a new method (26)(19) where W, Ta, HfO_2 have been dissolved in a mixture of HF + HNO_3, and later the solution was made 3N in HNO_3 + HF and Ba^{++} ions added, Hafnium was precipitated as $BaHfF_6$ and separated. In a next step K_2TaF_7 has been precipitated with KF.

If tungstic acid was used as a target this was dissolved in NaOH and the Ta induced radioactivity coprecipitated with ferric hydroxide by the addition of $FeCl_3$ solution.(30) The precipitate was dissolved in HCl and a second precipitation by the addition of inactive sodium tungstate (hold back carrier) was necessary.

This method does not effect a Hafnium separation.

Other methods are based on solvent extraction technique.

Methyl isobutyl ketone (hexone) has been used to extract Ta fluorides in the presence of H_2SO_4,HCl,HNO_3,HF.(24) Hexone HF-HNO_3 system may be used to extract Ta in presence of other elements including Hf. The same reagent (hexone) used in the system hexone-HF-H_2SO_4-NH_4F will extract Tantalum but not Hafnium.(27)(28) Unfortunately, the hexone method is affected by the presence of Tungsten.(29), and for this reason this method is excluded.

Butylphosphoric acid method extracts Ta as well as Hf in the same time. (It is also excluded).

It has been shown that Ta could be extracted from 6M HCl -(1-3)M,HF with di-isobutyl carbinol. Zr (or Hf) remains in the aqueous phase.(24)

For Hafnium some of the previous reagents (hexone, butylphosphoric acid) may be used but without attaining a complete separation of Tantalum.

Trialkylphosphine oxides (as tri-n-actyl-phosphine oxide) in cyclohexane completely extract Hafnium.(19)

But solvent extraction methods are affected by the presence of large amounts of Tungsten and do not always give a complete separation between Hf and Ta. To improve the separation re-extraction is necessary and sometimes a long contact between phases is required to attain equilibrium.

Ion exchange methods afford the most satisfactory means of separating many elements including Hafnium and Tantalum when time factor can be neglected.

The fluoride complexes usually are not adsorbed on a cation exchange column. Mixture of Hf and Ta can be separated following adsorbtion on the cation exchange column by selective elution (oxalic acid etc.)(32)

The anion exchange resins are useful when fluoride complexes are formed (avoiding hydrolytic polymerisation of Hf).

The separation of Hf-Ta was effected on a Dowex-2 resin column by elution with HCl of appropriate concentrations.(33)

Ta exhibits a strong tendency to colloid formation and to ensure true solution strong complexing agents are used (F^- or $C_2O_4^{--}$) and are suitable for Ta separation from other elements. The colloidal nature of Hf and Ta may be exploited, because colloids are not adsorbed on cation exchangers whereas ionic elements are.(34)

A study of separation, and distribution coefficients for different elements (including W, Hf, Ta) between the ion exchange resin Deacidite and Zeo Carb 225 and up to 10 M, HCl (1M HF) solution have been determined (35), and a scheme of separation is proposed.

With an anion exchange chromatographic method (36) a separation of Ta of other elements were possible.

The ion exchange methods are also affected by large amounts of Tungsten in the presence of small amounts of Ta and Hf. To perform a separation between Ta and Hf a previous elimination of W should have been done. Even when a high degree of separation would be possible, the long chemical method associated with it does not permit the application of these methods due to relatively short half life of 49 min. (Ta^{185}).

Chemical methods used in present work

A) Chemical separation of elements involved in present study.

In finding a chemical method we had to take into account two facts of great importance:

1) A complete chemical separation between Tantalum and Hafnium is required, even traces of contaminants are not allowed.

2) The time of chemical separation to be as short as possible because of the 49 min. half life of Ta^{185}, and the low activities involved.

Starting with the methods we have just enumerated and withdifficulties we have mentioned, we have concluded that most suitable method seems to be one described as a precipitation method.(26)

In its initial form this method was not suitable for our purposes and some modifications have been found necessary.

In this description as given in (26) W, Ta, Hf were dissolved in HF and HNO_3. Carriers were added and solution was made 3N in HNO_3 + HF. Ba^{++} ions to precipitate $BaHfF_6$ completely were added. After centrifugation the solution was saturated with KF or KHF_2 to precipitate K_2TaF_7 (white).

In order to check the efficiency of the method Feigl Spot

Test technique was unsuccessfully used on the separated fractions.(37) Test for Hf in presence of other metals with specific reagents as β nitrosoαnaphtol, alizarin, morin, p-dimethylaminoazophenyl arsonic acid, is hindered by the presence of F^- ion.

The test for W (even no importance) based on catalysis of Titanium (Ti^{+++})-malachite green reaction has been used when malachite green is slowly reduced at colourless compound in the absence of W, (but Ti^{+++} act rapidly as a reducer in the presence of W as a catalytic accelerator). This reaction was masked by the presence of NO_3^- and F^-.

We proposed to check the usefulness of the method with use of the radioisotopes of Ta and Hf.

We have sent Ta and HfO_2 samples to Harwell when Ta^{182} and Hf^{181} radioisotopes have been produced by slow neutron irradiation in a nuclear reactor.

These radioisotopes (Ta^{182} 115d) Hf^{181} 45d were added as tracers in our study. After irradiation these samples were both dissolved in a mixture of $HNO_3 + HF$, and kept in polythene bottles in strong HF media to avoid colloidal form. From these suitable dilutions were made and these secondary solutions (HF media also) were used as tracers. (Ta^{182} as 0.84 $\mu c/ml$ and Hf^{181} 0.86 $\mu c/ml$).

In our preliminary study Ta and HfO_2 were dissolved in a mixture of HF + HNO_3 (carrier amounts of 10-50 mg) and only one tracer was added. In a next step 5 g Tungsten (powder) was dissolved in this solution and the chemical method was applied. If Hf^{181} has been used as tracer both separated fractions for Hf and Ta have been checked for radioactivity. The same method has been employed with Ta^{182} tracer.

The most important parameters which play a role in the separation of Hafnium and Tantalum are: HF/HNO_3 ratio, HF + HNO_3 dilution factor after the dissolution of the target, quantities of Hafnium and Tantalum added as carriers.

In our experimental study we have found that the attack of W + Ta + HfO_2 is best accomplished when HF/HNO_3 ratio is about 2.5 (a sufficient excess in HF is required). For the next step a dilution is required. In the Table 1 we give the experimental results and the role of dilution effect on the activity in separated fractions when one tracer was added and carrier quantities were maintained invariable. (Hf^{181} was used as tracer).

TABLE 1

No.	HF conc. ml	HNO_3 conc. ml	H_2O ml	W g	Ta mg	HfO_2 mg	Hf^{181} activity In Hf fraction	In Ta fraction
1	15	6	50	5	26	12	9860	1130
2	15	6	40	5	26	12	9972	874
3	15	6	30	5	26	12	10100	340
4	15	6	20	5	26	12	11121	23
5	15	6	20	5	26	12	11442	8
6	15	6	20	5	26	12	10840	34

It was not our intention to give absolute activity measurement, no calibration of the end window proportional counter has been made for this purpose. This was a purely informative way to estimate the best range of dilution, so fluctuation in the counting rate is expected.

These series of determinations are quite conclusive, and we should use a dilution of 1:1. An explanation can be formulated that at higher acidity there is a complete separation of Hafnium from Tantalum and that at higher HF dilution a hydrolysis or incomplete precipitation should be possible.

From the separated fraction, the counting measurements were made on Tantalum and Hafnium oxides respecting the geometry, sample weight etc. In a later discussion we shall describe the method used.

Tantalum undergoes a very similar behaviour (when Ta^{182} tracer has been used) but sometimes upto 5%-8% from Tantalum activity is carried by Hafnium as $BaHfF_6$ but it is not an impediment since a simple washing of this compound with HF removes completely Tantalum contaminant activity.

When Tantalum carrier amount increases, the tracer activity carried by Hafnium decreases. This activity dependence is shown in Table 2. In last two columns is given counting rate of HfO_2 and Ta_2O_5 separated from initial solution.

TABLE 2

No.	H F ml	HNO_3 ml	H_2O ml	W g	Hf mg	Ta mg	Ta^{182} activity In Hf fraction c/min	Ta^{182} activity In Ta fraction c/min
1	15	6	20	5	12	8	853	7124
2	15	6	20	5	12	10	723	6811
3	15	6	20	5	12	20	260	7040
4	15	6	20	5	12	30	129	7140
5	15	6	20	5	12	50	66	7523
6	15	6	20	5	12	100	60	7289

It looks advantageous to use an increased carrier amount but it is limited by sample weight allowed to be counted and it should be in the range of 10-50 mg.

There are also fluctuations in counting rate, we have given an explanation and we shall add also errors which are due to fluctuations in chemical yield of Tantalum.

For Hafnium tracer having in view a 100% chemical yield we have not found a similar behaviour (in the range 8-50 mg HfO_2). For Tantalum there is a chemical yield dependence of Ta concentration in solution due to the appreciable solubility of the K_2TaF_7.

This contaminating activity in the Hf fraction is easily removed by washing the $BaHfF_6$ precipitate with HF aqueous solution.

From this experimental results we have defined the following chemical method for Hf and Ta separation.

We shall complete the dissolution of 10-20 mg HfO_2 and 20-30 mg Ta in 15 ml HF + 6 ml HNO_3 conc. (HfO_2 + Ta are used as carriers). The complete dissolution of HfO_2 is quite slow (4 hours).

It is preferable for this solution to be prepared 24 hours before the irradiation. After irradiation of the sample (5 g W) was carried out for an hour, and Iron granules separated (electromagnet), the W target sample is dissolved rapidly in the acid solution containing carriers. 20 ml distilled water is added (about 1:1 dilution), the beaker is strongly cooled with ice and 50 mg of $Ba(NO_3)_2$ solution is added

(1 ml). The solution is stirred and continuously cooled for 5 minutes. In this condition $BaHfF_6$ complete precipitation is obtained, it is transferred to a lusteroid tube, centrifuged and $BaHfF_6$ separated. The solution with Ta and W is saturated with KF and partial precipitation of Ta as K_2TaF_7 is achieved (a chemical yield from 30-60% is obtained). The precipitate is separated by centrifugation.

Glass has been avoided in all chemical operations.

In a series of preliminary tests 50 mg $Ba(NO_3)_2$ has been proved to be a sufficient reagent for complete precipitation of the Hf. The concentration of 50 mg/ml was used to avoid unnecessary dilution of the solution.

A larger excess than 50 mg must be avoided since complication in next chemical step has been found.

$BaHfF_6$ after separation is washed many times with diluted HF (1:1) and traces of Ta removed.

In their initial form Hf as $BaHfF_6$ and Ta as K_2TaF_7 cannot be used for counting rate measurements for two reasons:-

1) The larger quantities than 20-40 mg from these compounds are obtained. For instance K_2TaF_7 is carrying about 10 times more KF and precipitate weight is between 200-400 mg. So large amounts cannot be used for counting measurements.

2) For counting rate measurements the samples must always be in a simple chemical form which give reproducible results.

Since the ratio between reagent carried (KF) and the compound (K_2TaF_7) varies appreciably from one sample to another, counting efficiency (scattering) is affected.

Since Ta and Hf have very close atomic weight (mass number of their stable isotopes in the range 177-181) we have found that the best way for counting rate measurements is to bring the samples in oxide form. It also gives the lowest source weight (highest specific activity) possible.

$BaHfF_6$ is easily dissolved in a mixture of HNO_3-H_3BO_3 (8-10 M HNO_3 saturated with H_3BO_3), NH_4OH is added and $HfO_2 X H_2O$ precipitated, centrifuged and washed with water. It is filtered, washed with acetone, dried and heated. HfO_2 in powder form is transferred on a special vacuum funnel with acetone and samples for counting rate measurements 10-40 mg (dried also) are obtained.

K_2TaF_7 is dissolved in a few drops of HF and water, and $Ta_2O_5 X H_2O$ is precipitated with ammonia. It is also washed, dried, heated and after suspending in acetone, Ta_2O_5 samples are prepared in the same way as for Hf. (They do not exceed 40 mg.).

It is preferable (in total absence of HF) to use glass filter paper when very good, uniformly distributed, flat, non-breaking samples are obtained.

The chemical yield is not 100% in both cases, because with Tantalum, we can never attain this yield, and with Hafnium some losses are inevitably introduced by the need to work quickly.

After counting rate measurements have been done, we must estimate the chemical yields for our further calculations.

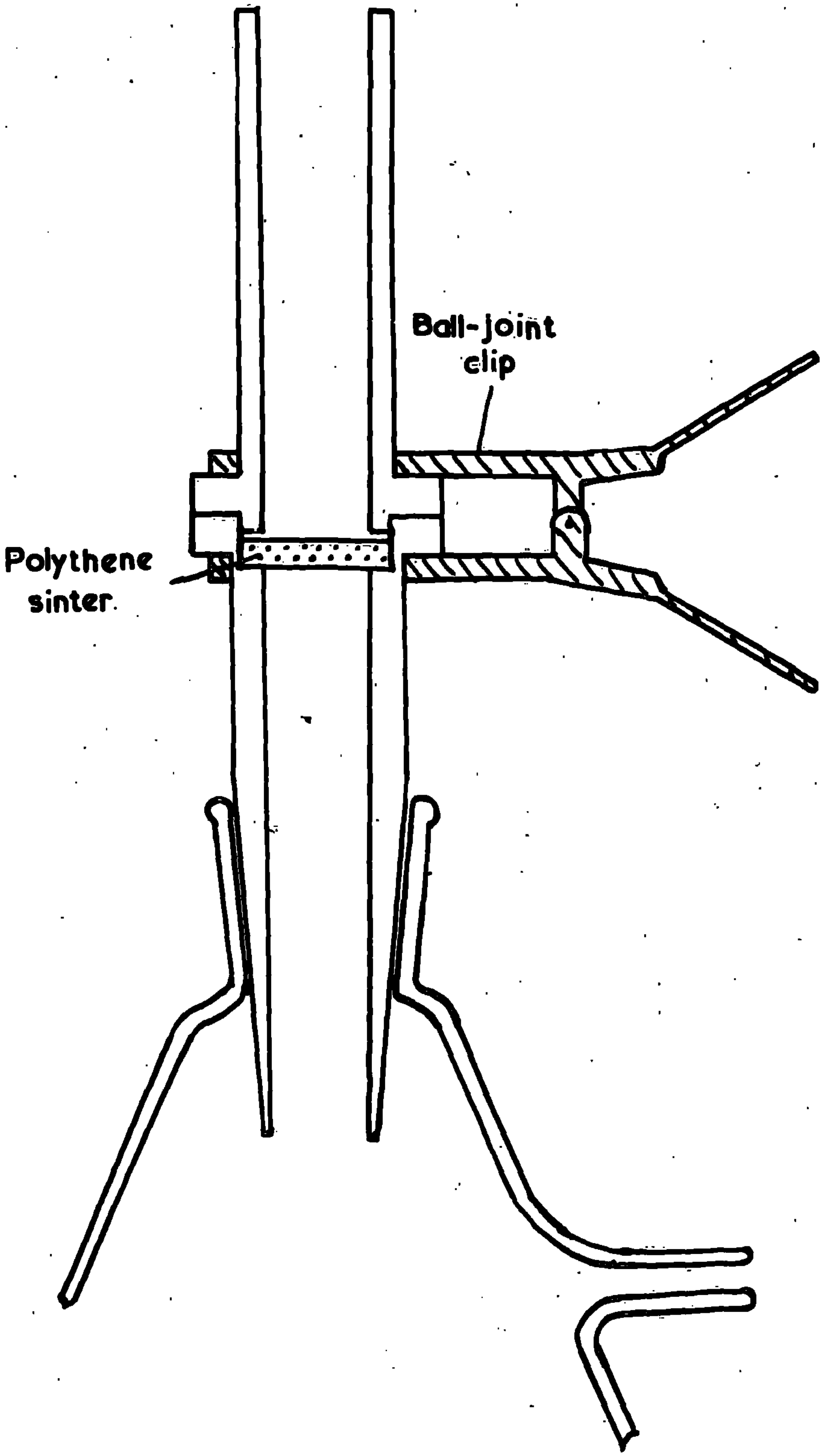
A gravimetric method has been found very suitable for this purpose.

HfO_2 is dissolved in a mixture of HF + HNO_3 conc. (idem Ta_2O_5). A check for Ba^{++} was made but its complete absence was proved. Boric acid is added to complex F^- ion (don't seem to be necessary) and ammonia added to precipitate $HfO_2 X H_2O$. This is well washed, dried and ignited in a crucible and its weight determined.

This method was applied in both cases and was checked with known quantities of the elements in the chemical way described and proved to give always satisfactory results.

FIGURE 6

Demountable filter-funnel


Ball-joint clip
Polythene sinter

B) Chemical methods associated with the end window gas flow proportional counter calibration

a) Solid sample preparation for counting rate measurement.

The active precipitate, dried, is uniformly dispersed in acetone and collected on glass filter paper fixed in a special demountable funnel under vacuum. (Fig. 6) The sample uniformly distributed on the filter paper area is transferred to an Aluminium tray. The sample is dried and its weight is accurately determined with precision of 0.1 mg, and counted with reproducible geometry.

b) Absolute disintegration rate determination of P^{32} used as standard solution.

From nuclear data(38) all nuclear reactions under consideration give β^- emitters with energies within the range 1.3-1.7 MeV (75-100% within this range). It is known (J.H. Davies[16]) that the efficiency of the end-window proportional counter used is very nearly constant for $E > 1.0$ MeV., and for convenience ^{32}P was selected for calibration measurements. It was first shown that $^{32}PO_4^{\equiv}$ (carrier free) is quantitatively carried on precipitates of $HfO_2 \cdot X\, H_2O$. It is the ideal procedure since both sample and standard are then in the same chemical form, and scattering of β rays in both samples is identical.

In preliminary experiments, it was found that in ammoniacal solution in the presence of Hf which precipitates as $HfO_2 X H_2O$, P^{32} is not completely precipitated and the activity has always been found in solution in a proportion of 10-15%. Phosphoric acid has been used as carrier, experiments with other elements as Ba^{++}, Ca^{++} (excess) which give very insoluble phosphates (in ammoniacal medium) as Hafnium does, has been used with the same results.

We have concluded that P^{32} is not entirely in the pentavalent form. To ensure a complete precipitation of P^{32} with Hf only P^{32} in pentavalent form must be used. It was found also that traces of HF (Explanation of Hf Chemistry) affect the P^{32} precipitation.

From these conclusions it was necessary to add some non isotopic carrier for P^{32}, (the smallest amount possible) to precipitate it in a form when P^{32}(+ 5 state) is found in precipitate, redissolve it and determine the disintegration rate in this solution.

Hf or Ta are the best carriers for this purpose. We have chosen Hafnium and HfO_2 is dissolved only in a mixture of HF + HNO_3. From this solution $HfO_2 X H_2O$ is precipitated with ammonia and since the presence of HF should be avoided the precipitate is separated by centrifugation, redissolved in HNO_3 (freshly precipitated $HfO_2 XH_2O$ is dissolved in HNO_3

cf. $Ta_2O_5X\ H_2O$ which is dissolved only in HF, it is a reason to use Hf as carrier for this purpose) reprecipitated etc. many times and finally the HF-free Hafnium solution is added to the P^{32} solution and the $^{32}PO_4^{\equiv}$ is co-precipitated with $HfO_2X\ H_2O$ by addition of ammonia. This is washed and redissolved in HNO_3 and this solution contains P^{32} only in +5 state. Other tests with this solution proved that all activity is easily carried down by a HfO_2 precipitate (with ammonia).

This P^{32} solution was used for 4π proportional counter measurements of disintegration rate.

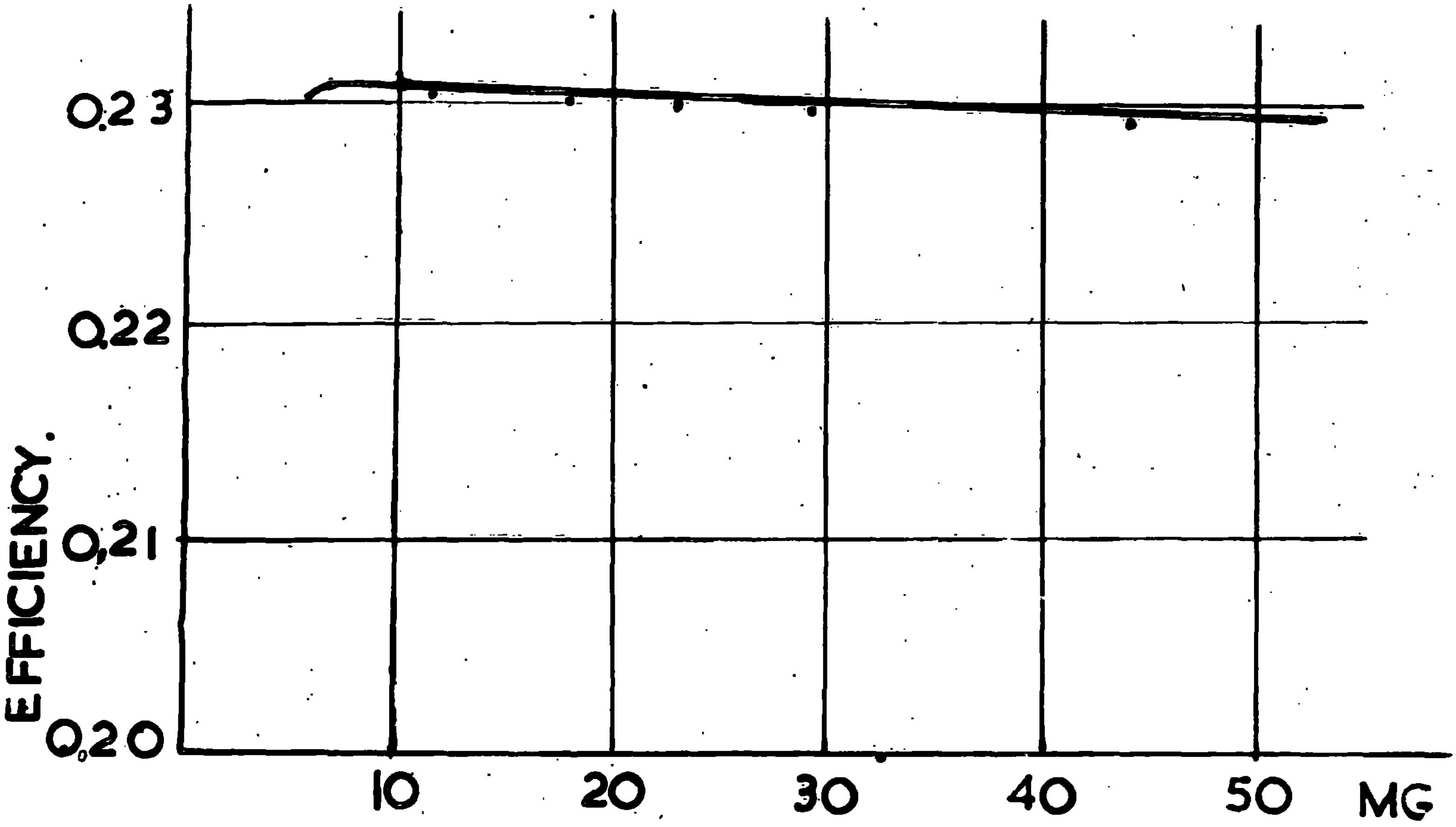
On the opposite side of gold coated VYNS film in Aluminium frame, a drop of insulin (as described in a previous chapter) was carefully placed on 1 cm diameter circle in the centre of the film.

Later a weighed volume of radioactive solution (by difference from a polythene flask) to ensure an activity of 10,000-15,000 c/min. at a residue of 10-15 μg was placed on the film and was dried (infra-red lamp) with great care to avoid breakage of the film.

The Aluminium frame was placed in equatorial position in the 4π gas flow proportional counter and counting rate (disintegration rate) was measured. Five similar films were prepared, and with all corrections fulfilled (decay time,

FIGURE 7

A self absorbtion curve


EFFICIENCY.
0,23
0,22
0,21
0,20
10
20
30
40
50
MG

dead time, background etc.). The specific activity found was 308 dis/mg. The precision between the different samples was in the range 308 ± 1 dis/mg.

C) Efficiency of the end window gas flow proportional counter

Aliquot part of P^{32} standard solution (308 dis/mg) is added to an acid solution containing 120 mg HfO_2 (HF free prepared as before). $HfO_2\,XH_2O$ is precipitated (including P^{32}) and filtered. The activity in solution after the separation of $HfO_2.XH_2O$ is checked with a liquid counter but no radioactivity was found. (Background).

The $HfO_2\,XH_2O$ precipitate is washed, dried and calcinated. At the end of calcination, it is cooled and weighed. All chemical operation until calcination was achieved has been done with care to avoid any losses. HfO_2 obtained, in powder form (ground) is well stirred with acetone and divided in five portions containing HfO_2(and P^{32}) within the range 6-44 mg. So sources of various thickness were prepared with the same standard filter funnel technique and counted with the same geometry (with end window proportional counter). Counter efficiency is given by $e = \frac{\text{Observed counting rate}}{\text{disintegration rate}}$. A plot of e as a function of the weight of the source is given in Fig. 7(for one counter used) and this curve used to reduce observed counting rates to absolute disintegration rates.

This self-absorption curve was plotted for sources in the range 6-44 mg and counter efficiency varied very little with the source thickness in this range on account of the high energy of the P^{32} β rays.

This plot gives us an exact determination of end window proportional counter efficiency (in this energy range) as a function of sample weight. Because counter calibration was made with HfO_2 sources (containing P^{32}) and in the experimental determinations the samples were always prepared as HfO_2 or Ta_2O_5 a high accuracy in estimating counter efficiency is expected when other conditions (geometry etc.) are respected.

D) Chemical methods associated with the reference element

For the reference element it was found convenient to use a halogen quenched liquid counter with capacity 10 ml.

It was calibrated with Mn^{56} standardised with a 4π counting technique.

The iron used as a reference element with a granule size of 0.5 mm had a purity of 99.8%. The iron was used in a homogeneous mixture with the target. After irradiation it was separated from the sample using an electromagnet. After the separation, the iron (0.5 g) was dissolved in 10 ml of a standard acid mixture (50% 5N, HNO_3 + 50% 5N, H_2SO_4)

containing a little manganese carrier. The solution was made up to 12 ml in a graduated flask and 10 ml from this was transferred with a pipette to the liquid counter.

It is a standard method used in this laboratory for this purpose.

Principles of calculation

Some basic principles should be respected in cross sections calculations.

We have mentioned that a reference nuclear reaction has been used in our determination and this is given by the Iron granules in a homogeneous mixture with W target, $Fe^{56}(np)Mn^{56}$. Here we assume that both reference element (Fe) and the sample (W) are exposed all the time to the same neutron flux. This assumption is correct but the reference reaction cross section must be exactly known (its determination is made with absolute neutron flux measurement).

Radioactive isotopes produced in sample and reference, decay at different rates and corrections should be made.

The radioactive elements are produced, (when an accelerating machine is used), at an irregular rate of (40) R(t) nuclei/sec where R(t) is a function of time. It is due to neutron flux fluctuations with the time.

The rate of formation is $\frac{dN^1}{dt} = N\sigma\phi$ equ.(1) the time interval dt. Here N = number of target atoms, σ = cross section in cm^2 and ϕ = neutron flux/cm^2sec.(39)

From equation (1) we can write $dN^1 = N\sigma\phi \, dt$.

For an irradiation extending over a finite time interval this expression must be integrated to obtain the total number of N^1 (product atoms) at the end of irradiation:

$$N_o^{1} = \int_0^{\tau} N\sigma\phi\, e^{-\lambda^{1}t}dt \qquad \text{equation (2)}$$

In this equation (39) an allowance has been made for atoms decaying during the irradiation.

I^1 is absolute disintegration rate $I_o^1 = N_o^1\lambda^1$.

In equation (2) N is constant in this time interval, σ and λ are also constant but ϕ is variable. We can write:

$$I_o^1 = N\sigma\lambda^1\int_0^{\tau}\phi\ e^{-\lambda^1 t}dt \qquad (39)$$

The number of active nuclei present at the time τ can be calculated by considering the number formed during a short interval of time dt in the past (40) so finally we have

$$I_o^1(\tau) = N\sigma\lambda^1\int_0^{\tau}\phi\ e^{-\lambda^{1}(\tau-t)}dt \qquad \text{equation (3)}$$

We have mentioned that a neutron monitor measured the relative flux Φ. This is related to the actual flux ϕ at the sample position by $\phi = \Phi\,\eta$.

Then eq. (3) become: $I_o^1(\tau) = N\sigma\lambda^1\eta\int_0^{\tau}\Phi\ e^{-\lambda^1(\tau-t)}dt$

Because the reference element is placed in such a position (homogeneous mixture) as both the reference and target are intercepted by the same flux η will be the same for both. If r refers to reference element we can write:

$$\frac{I^1}{I_r} = \frac{N\sigma\lambda^1\int_0^{\tau}\phi\ e^{-\lambda^1(\tau-t)}dt}{N_r\sigma_r\lambda_r\int_0^{\tau}\phi\ e^{-\lambda_r(\tau-t)}dt} \qquad \text{equation (4)}$$

A numerical method, where the integrals are replaced by a summation, is more convenient but a finite time interval in the abscisae should be assumed and should be quite small compared with the half life:

When these conditions are respected we can write:

$$\frac{I'}{I_r} = \frac{N\sigma\lambda' S}{N_r\sigma_r\lambda_r S_r}$$

Summation: Where S denotes the $\sum_{t=0}^{t=\tau} (\Phi\, e^{-\lambda(\tau-t)} \cdot \Delta t$

and a similar relation for S_r is easily deduced replacing λ with λ_r. In this relation σ is unknown.

The activities introduced in this equation are determined by extrapolation of their decay curves to the zero time. Since detection coefficients (counter efficiency) for these nuclides are C and C_r then activities A_0 and A_r are related with the disintegration rate by:

$A_0 = \lambda c\, N_0$ or $A_0 = cI$ and introducing these notations

$$\frac{A'_0}{A_r} = \frac{c\, N\sigma\lambda' S}{c_r N_r\sigma_r\lambda_r S_r} \qquad \text{equation (5)}$$

From this equation we can easily evaluate σ which is unknown, the other factors being well defined as before mentioned.

Experimental procedure and results of irradiation

Mixtures usually contained 5 g of Tungsten (powder) and 0.5 g iron granules. A homogeneous mixture is obtained when two components are carefully mixed for 30 minutes.

The mixture is transferred to a polythene box, which is provided with a very thin cover. The small cylindrical box is completely filled with the mixture and no empty space between mixture and cover is allowed to ensure a better neutron flux when the box is placed close under the tritium target.

The polythene thin cover protects the mixture from contamination with recoil atoms from the brass target assembly.

The time of irradiation is one hour. Longer irradiation intervals must be avoided since longer-lived undesirable activities are built up.

Previously an acid solution containing HNO_3 + HF,Ta and Hf carriers has been prepared.

At the end of irradiation the mixture is transferred to a paper sheet and with an electromagnet the Iron is separated and Tungsten weighed and dissolved in acid solution containing Hf and Ta carriers.

Dissolution of W is made with care because strong acid attack of the metal. The chemical method described in a previous chapter is applied and Hf separated as $BaHfF_6$, redissolved in HNO_3 (8-10 N) saturated with H_3BO_3, and HfO_2xH_2O precipitated with ammonia, washed, dried weighed and counted.

For Tantalum activities another mixture of W + Fe is used. After irradiation, separation of iron with electromagnet dissolution in acid solution, and Hf separation the Ta is precipitated as K_2TaF_7 by addition of KF. The precipitate is redissolved in HF aq., and $Ta_2O_5.xH_2O$ is precipitated with ammonia, washed, dried, weighed and counted.

The samples used for counting rate determinations are prepared with care to avoid accumulation of materials so after drying, (strong heat) the HfO_2 or Ta_2O_5 is transferred with acetone on a funnel. Dispersion must be uniform and samples for counting are in the range 6-44 mg weight.

It is necessary for the sample to be heated and then transferred as a slurry in acetone to avoid non-uniform distribution of the sample and breaking (always observed with water).

When $W^{186}(n\alpha)Hf^{183}T_{\frac{1}{2}} = 64$ min) is studied a decay curve in a time interval of 6-8 hours is checked. (when possible).

For $W^{184}(np)Ta^{184}T_{\frac{1}{2}} = 8.7$ h and $W^{186}(nd)Ta^{185}T_{\frac{1}{2}} = 49$ min the longest half life is taken into consideration and a decay curve for about 40 hours (sometimes longer) is followed, it depends on activities involved which are quite low.

All decay curves were made from the data obtained with an automatic recording unit and a timer. The counting rate was recorded on a paper tape concurrently with the time intervals. (A preset number of counts are recorded and the unit time intervals).

The activities were extrapolated to t=0 (end of irradiation) and counting efficiency determined as a function of sample weight from calibration curves. Corrections for chemical yield were also made.

Iron granules were also dissolved in an acid solution, as we have mentioned, and Mn^{56} activity counted with the liquid counter whose efficiency had previously been determined as 8.635%.

Corrections for, dead time, decay, background etc. were made and the activity at t = 0 determined.

During irradiation of the mixture (W + Fe) the neutron flux Φ is checked at intervals of 10 minutes, so $\Phi . e^{-\lambda(\tau - t)}$ and also $(\phi \, e^{-\lambda_r(\tau - t)})$ is determined assuming τ = 60 and t is taken in the middle of each interval, and finally the summation over all intervals which gives S and Sr is obtained.

σ_r is taken 100 mb and was the most reliable value determined for $Fe^{56}(np)Mn^{56}$ in this laboratory. (42)

For the calculation of number of atoms in the target as well as in the reference element isotopic abundance has been taken into consideration. Disintegration constant were obtained from nuclear data and expressed in minutes^{-1}.

In the following tables we shall give the results obtained for each determination.

First determination

for nuclear reactions: $W^{184}(np)Ta^{184}$ and $W^{186}(nd)Ta^{185}$.

No.	Specification	Symbol	Units	Value	Remarks
1	Tungsten weight	W	g	4.8255	
2	W^{184} abundance	W^{184}	%	30.7	(41)
3	Chemical yield	η	%	44.1	
4	Counting rate	A	counts/ min	501	A at t=0 corrected for chem. yield
5	Counting rate for reference	A_r	"	2.546×10^4	
6	Sample weight (for counting) as Ta_2O_5		mg	35.2	(prop. counter 53)
7	End window prop. counter efficiency	C	%	33.15	
8	Liquid counter efficiency	C_r	%	8.635	
9	No. of W^{184} atoms in the target	N		$8.05 \times 10^{-3} \times \mathcal{N}$	(Avogadro number)
10	Iron weight	Fe	g	0.5012	
11	Fe^{56} abundance	Fe^{56}	%	91.66	(41)

(continued)

12	No. of Fe^{56} atoms	Nr		$8.23x10^{-3}.\mathcal{N}$	
13	Reference reaction cross section	σ_r	mb	100.	
14	Ta^{184} dis. constant	λ	min^{-1}	$1.33x10^{-3}$	(41)
15	Mn^{56} dis. constant	λ_r	min^{-1}	$4.47x10^{-3}$	
16	Integral value for Ta^{184}	S		$3.19x10^{4}$	
17	Integral value for reference	S_r		$2.92x10^{4}$	
18	Nuclear reaction cross section	$\sigma_{(np)}$	mb	1.66 from	eq. 5

1	W^{186} abundance	W^{186}	%	28.7	(41)
2	No. of W^{186} atoms in the target	N'		$7.5x10^{-3}x\,\mathcal{N}$	
3	Counting rate	A'_o	counts/min	409	at t=0 corrected for chemical yield
4	Disintegration const.of Ta^{185}	λ'	min^{-1}	1.441x10-2	
5	Integral value for Ta^{185}	S'		$2.2545x10^{4}$	
6	Nuclear reaction cross section	$\sigma_{(nd)}$	mb	0.185	eq. 5

Second determination

for nuclear reactions: $W^{184}(np)Ta^{184}$ and $W^{186}(nd)Ta^{185}$

No.	Specifications	Symbol	Units	Value	Remarks
1	Tungsten weight	W	g	4.7242	
2	W^{184} abundance	W^{184}	%	30.7	
3	Chemical yield	η	%	45.2	
4	Counting rate	A	counts/ min	260	t = o corrected for yield
5	Counting rate (reference)	A_r	"	1.855×10^{4}	
6	Sample weight as Ta_2O_5		mg	19.2	(prop. counter 90)
7	End window prop. count. efficiency	C	%	23.05	
8	Liquid counter efficiency	C_r	%	8.635	
9	No. of W^{184} atoms in the target	N		$7.87\times10^{-3}\times\mathcal{N}$	($\mathcal{N}$= Avogadr number)
10	Iron weight	Fe^{56}	g	0.5036	
11	Fe^{56} abundance	Fe^{56}	%	91.66	
12	No. of Fe^{56} atoms	N_r		$8.25\times10^{-3}\times\mathcal{N}$	
13	Reference reaction cross section	σ_r	mb	100	
14	Ta^{184} dis. constant	λ	min^{-1}	1.331×10^{-3}	
15	Mn^{56} dis. constant	λ_r	min^{-1}	4.47×10^{-3}	
16	Integral value for Ta^{184}	S		5.06×10^{4}	
17	Integral value for reference	S_r		4.579×10^{4}	

(continued)

18	Nuclear reaction cross section	$\sigma_{(np)}$	mb	1.68
1	W^{186} abundance	W^{186}	%	28.7
2	No. of W^{186} atoms in the target	N'		$7.3\times10^{-3}\,\mathfrak{N}$
3	Counting rate	A'	counts/ min	213
4	Disintegration const. of Ta^{185}	λ'	min^{-1}	1.441×10^{-2}
5	Integral value for Ta^{185}	S'		3.377×10^{4}
6	Nuclear reaction cross section	$\sigma(nd)$	mb	0.20

Third determination

for nuclear reactions: $W^{184}(np)Ta^{184}$ and
$W^{186}(nd)Ta^{185}$

No.	Specifications	Symbol	Units	Value	Remarks
1	Tungsten weight	W	g	4.9124	
2	W^{184} abundance	W^{184}	%	30.7	
3	Chemical yield	η	%	38.81	
4	Counting rate	A	counts/ min	472	(t=0 corrected for η)
5	Counting rate (reference)	A_r	"	$3.2001\text{x}10^{4}$	
6	Sample weight as Ta_2O_5		mg	21.50	(counter 90
7	End window prop.count.eff.	C	%	23.05	
8	Liquid counter efficiency	C_r	%	8.635	
9	No. of W^{184} atoms in the target	N		$8.1810\text{x}10^{-3} \times \mathcal{N}$.	
10	Iron weight	Fe	g	0.5011	
11	Fe^{56} abundance	Fe	%	91.66	
12	No. of Fe^{56} atoms	N_r		$8.200\text{x}10^{-3} . \mathcal{N}$.	
13	Reference reaction cross section	σ_r	mb	100	
14	Ta^{184} disint. constant	λ	min^{-1}	$1.331\text{x}10^{-3}$	
15	Mn^{56} disint. constant	λ_r	min^{-1}	$4.47\text{x}10^{-3}$	
16	Integral value for Ta^{184}	S		$2.999\text{x}10^{4}$	
17	Integral value for reference	S_r		$2.729\text{x}10^{4}$	
18	Nuclear reaction cross section	$\sigma_{(np)}$	mb	1.70	

(continued)

1	W^{186} abundance	W^{186}	%	28.70
2	No. of W^{186} atoms in the target	N'		7.65×10^{-3} π
3	Counting rate	A'	counts/min	384
4	Disintegration const. of Ta^{185}	λ'	min^{-1}	1.441×10^{-2}
5	Integral value for Ta^{185}	S'		2.067×10^{4}
6	Nuclear reaction cross section	$\sigma_{(nd)}$	mb	0.197

First determination

for nuclear reaction: $W^{186}(n\alpha)Hf^{183}$

No.	Specifications	Symbol	Units	Value	Remarks
1	Tungsten weight	W	g	4.7990	
2	W^{186} abundance	W^{186}	%	28.7	
3	Chemical yield	η	%	40.1	
4	Counting rate	A	counts/min	1200	t=0 corrected for η
5	Counting rate (reference)	A_r	"	3.6310×10^{4}	
6	Sample weight as HfO_2		mg	15.3	
7	End window prop count eff.	C	%	33.92	
8	Liquid counter efficiency	C_r	%	8.635	
9	No. of W^{186} atoms in the target	N		$7.471 \times 10^{-3} \cdot \mathcal{N}$	($\mathcal{N}$= Avogadro No.)
10	Iron weight		g	0.5084	
11	Fe^{56} abundance		%	91.66	
12	No. of Fe^{56} atoms	N_r		$8.341 \times 10^{-3} \cdot \mathcal{N}$	
13	Reference reaction cross section	σ_r	mb	100	
14	Hf^{183} dis. constant	λ	min^{-1}	1.082×10^{-2}	
15	Mn^{56} dis. constant	λ_r	min^{-1}	4.471×10^{-3}	
16	Integral value for Hf^{183}	S		3.4211×10^{4}	
17	Integral value for reference	S_r		4.091×10^{4}	
18	Nuclear reaction cross section	$\sigma(n\alpha)$	mb	0.464	

Second Determination

for nuclear reaction $W^{186}(n\alpha)Hf^{183}$

No.	Specification	Symbol	Units	Value	Remarks
1	Tungsten weight	W	g	4.8736	
2	W^{186} abundance	W^{186}	%	28.7	
3	Chemical yield	η	%	54.51	
4	Counting rate	A	counts/min	349	
5	Counting rate (reference)	A_r	"	$9.625x10^{3}$	
6	Sample weight as HfO_2		mg	14.1	
7	End window prop. counter efficiency	C	%	33.95	
8	Liquid counter efficiency	C_r	%	8.635	
9	No. of W^{186} atoms in the target	N		$7.611x10^{-3}$, $\mathcal{N}$	
10	Iron weight		g	0.5017	
11	Fe^{56} abundance		%	91.66	
12	No. of Fe^{56} atoms	N_r		$8.21x10^{-3}$, $\mathcal{N}$	
13	Reference reaction cross section	σ_r	mb	100	
14	Hf^{183} dis. constant	λ	min^{-1}	$1.082x10^{-2}$	
15	Mn^{56} dis. constant	λ_r	min^{-1}	$4.471x10^{-3}$	
16	Integral value for Hf^{183}	S		$1.913x10^{4}$	
17	Integral value for reference	S_r		$2.321x10^{4}$	
18	Nuclear reaction cross section	$\sigma(n\alpha)$	mb	0.492	

Third determination

for nuclear reaction $W^{186}(n\alpha)Hf^{183}$

No.	Specification	Symbol	Units	Value	Remarks
1	Tungsten weight	W	g	4.9240	
2	W^{186} abundance	W^{186}	%	28.7	
3	Chemical yield	η	%	46.12	
4	Counting rate	A	counts/min	453	
5	Counting rate (reference)	A_r	"	$1.9773\text{x}10^{4}$	
6	Sample weight as HfO_2		mg	11.40	
7	End window prop.count.efficiency	C	%	23.10	
8	Liquid counter efficiency	C_r	%	8.635	
9	No. of W^{186} atoms in the target	N		$7.672\text{x}10^{-3}$,	$\mathcal{N}$
10	Iron weight		g	0.5118	
11	Fe^{56} abundance		%	91.66	
12	No. of Fe^{56} atoms	N_r		$8.375\text{x}10^{-3}$,	$\mathcal{N}$
13	Reference reaction cross section	σ_r	mb	100	
14	Hf^{183} dis. constant	λ	min^{-1}	$1.082\text{x}10^{-2}$	
15	Mn^{56} dis. constant	λ_r	min^{-1}	$4.471\text{x}10^{-3}$	
16	Integral value for Hf^{183}	S		$4.0960\text{x}10^{4}$	
17	Integral value for reference	S_r		$4.9630\text{x}10^{4}$	
18	Nuclear reaction cross section	$\sigma_{(n\alpha)}$	mb	0.472	

Discussion of the results obtained

A summary of these results are given in the following table.

Nuclear reaction: $W^{184}(np)Ta^{184}$

No. of determination	fast neutron cross section (mb)
1.	1.66
2.	1.68
3.	1.70
Mean Value	1.68 ± 0.02

Nuclear reaction $W^{186}(nd)Ta^{185}$

1.	0.185
2.	0.200
3.	0.197
Mean Value	0.193 ± 0.007

Nuclear reaction $W^{186}(n\alpha)Hf^{183}$

1.	0.464
2.	0.492
3.	0.472
Mean Value	0.476 ± 0.014

All these calculations of fast neutron cross section were based on the assumption of reference reaction cross section $Fe^{56}(np)Mn^{56}$ of 100 mb.

In the last table we give a mean value of specified cross section and a maximum possible spread of the results is given (estimated limits of error).

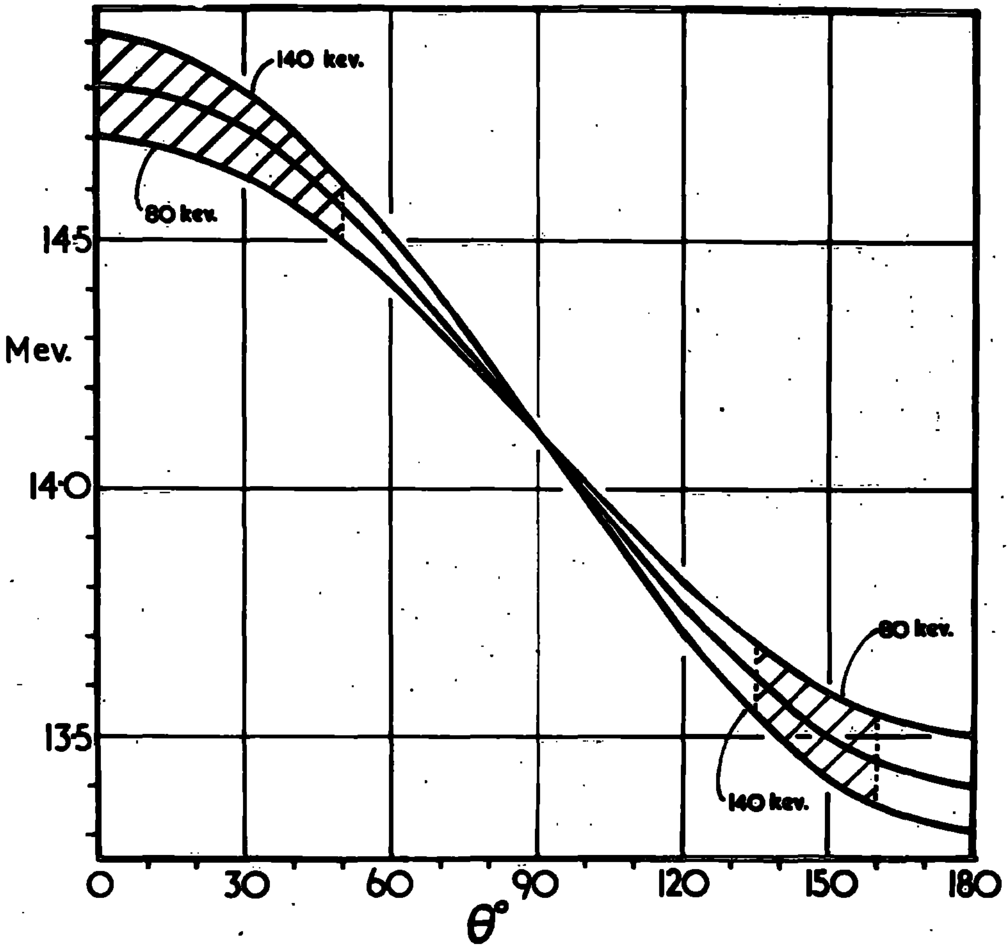
This spread can be accounted for errors involving statistical fluctuations since activities measured were not high enough (due to low cross sections), some errors (quite small) from chemical yield determinations. A small source of errors in estimating the neutron flux. Sometimes neutron detectors efficiency changed but since a reference activity has always been checked these errors can be neglected.

Counter calibration has been made with great care, using radioactive solution standardized with 4π technique, so we can estimate a high precision (1-2% errors) in counter efficiency, and systematic errors are very low. This is an important improvement since in many other works this accuracy was not attained and systematic errors due to counter calibration played an important role.

In our experiments deuteron energy was always the same, and sample was very close placed to the source of neutrons so that a quite large solid angle was subtended. Because (a small) the dependence of neutron energy with angle of

FIGURE 8

Neutron energy as a function of angle of emission



NEUTRON ENERGY AS A FUNCTION
OF ANGLE OF EMISSION.

emission (Fig. 8) we define the neutron energy to be within the range 14.7 ± 0.2 MeV in our experiments. Some studies do not specify the neutron energies, this fact is not trivial because often the cross section may change by a factor of two in an interval of 0.5 MeV in experimental results but is expected that theoretical calculations should give better results than a factor of two. (13)(43)

As we have mentioned in a previous chapter the statistical theory of the compound nucleus has failed to explain the 14.5 MeV neutron cross sections and the experimental results for (np) and ($n\alpha$) are much greater than the theoretical results calculated with this theory.

Brown and Muirhead theory is based on direct interaction, when incident neutron interacts with a proton which is inside nucleus (difference of surface theory). The emitted protons, difference of compound nucleus, have a high energy peak with a maximum of energy of incident neutron + Q (energy released) in the reaction.

In the direct interaction theory, the target elements with high mass number, the high Colomb barrier has little effect on proton emission. In the statistical theory of compound nucleus the proton emission is strongly prohibited by Coulomb barrier and its contribution is negligible to total (np) reaction.

All recent studies, and fast neutron cross section measurements proved that direct interaction theory for (np) reactions explain satisfactorily the experimental results.

So with this theory, a calculated cross section for $W^{184}(np)Ta^{184}$ is given (8), 2.2 mb. In the same work the author gives an experimental value of 4.75 mb so the ratio 4.75/2.2 is about 2.20. In our experiments we have obtained 1.68 ± 0.02 so the ratio 1.70/2.2 $\simeq$ 0.78.

Our value is closer to theoretical one and the theoretical interpretation of experimental results of different authors should be studied with care since sometimes for the same nuclear reaction $W^{184}(np)$ the values within 14-3 mb range are given (44). The usual experimental errors are due to neutron flux determination and counter calibration. In our experiments these errors are negligible, since the sample and reference are intercepted by the same neutron flux, and end window proportional counter calibration was very exactly determined.

The activation method used cannot distinguish for (nd) reactions, the other alternative (n,pn) and (n,np). There are very few experiments to deal with this sort of nuclear reaction because very small cross section and sometimes very short half lives.

In fact very few elements have isotopes where an (nd) reaction should be possible without interference with an (np) reaction producing the same radioisotope.

These requirements are fulfilled by Pd^{108}, Te^{128}, Te^{130}, Ce^{142}, Hg^{204}, W^{186} (Hg^{204} gives a short half life Au^{203} 55 sec.)

In the case of (n,np) reaction the mechanism is explained by initial emission of a neutron directly (or from a compound nucleus) followed by emission of a proton from the residual nucleus. Emission of proton is possible only if binding energy of neutron in residual nucleus (after neutron emission) is higher than proton one. The (n,pn) mehhanism is explained (13 by an (np) reaction followed by a neutron emission (directly or from residual compound nucleus).

Because we cannot distinguish, as mentioned, between (nd), (n,np); (n,pn) reaction we shall treat them as (n,pn) to compare our results with an experimental and theoretical work obtained by Coleman et al.(13). In this work the authors have done a study of (n,pn) reaction on W^{186} and neutron cross section is given as a function of incident neutron energy.

They have found that a strong dependence of (n,pn) cross section as a function of neutron energy exist and the following values 0.11 ± 0.05 and 0.3 ± 0.15 for neutrons of 14.5 and 14.85 MeV are given. Since our results are within the same limits 0.193 ± 0.007 we can easily explain it having in view a spread in neutron energy of 14.7 ± 0.2 MeV, and our results are very comparable with those obtained by this study.

The theoretical cross section based on direct interaction theory of Brown and Muirhead within approximately the same neutron energy range (14.5-15.0 MeV) gives 0.02 and 0.05 mb respectively.(13).

Theoretical results are based only on (n,pn) nuclear reaction but we have mentioned that activation methods cannot distinguish between the three possibilities which can exist altogether. Having in view these considerations and comparing theoretical and experimental results we can assume that Brown-Muirhead theory explains this behaviour.

For (n,α) reaction cross section the failure of statistical theory of compound nucleus seems to be evident from Paul and Clarke's study(6) especially within the range of heavy elements where ratio of $\sigma_{obs.}/\sigma_{calculated}$ (evaporation theory) is very large.

Our value of 0.476 ± 0.014 mb is found to fit very well with others experimental results within (or very near) this mass number range. Coleman gives some results from 0.4-1 mb. in this mass number range.(8) But for (nα) reaction no clearly defined trend has been found as, for example, with (np) reaction so any comparison must be done with care. But it is evident that a direct interaction mechanism must be involved although a detailed theoretical treatment is not yet possible.

Because mean free path within the nucleus of an α particle from $(n\,\alpha)$ reaction with fast neutrons is smaller than that of a proton it is supposed that $(n\,\alpha)$ reaction originates in the nuclear surface.(8)

14 MeV neutron cross sections in mercury target

The scope of this work was to determine those cross sections in mercury target where literature data are absent. Here also the theoretical possibility for an (nd) reaction without interference with an (np) giving the same product exists. (Similar to Tungsten).

The natural mixture of Mercury stable isotopes is as follows:

Hg^{196}0.15%; Hg^{198}10.0%; Hg^{199}16.9%; Hg^{200}23.1%; Hg^{201}13.2%; Hg^{202}29.8%; Hg^{204}6.8% (41)

Because no stable isotope of Hg^{203} exists there should be no interference of an (np) reaction with an (nd) reaction in Hg^{204}.

In fact the following nuclear reactions with fast neutrons (14.5 MeV) in Mercury targets are expected, taking account only of those reactions giving radioactive products:-

(n γ) reactions:

$Hg^{196}(n\gamma)Hg^{197}$; $Hg^{198}(n\gamma)Hg^{199}$; $Hg^{202}(n\gamma)Hg^{203}$; $Hg^{204}(n\gamma)Hg^{205}$

(n,2n)

$Hg^{196}(n,2n)Hg^{195}$; $Hg^{198}(n,2n)Hg^{197}$; $Hg^{200}(n,2n)Hg^{199}$; $Hg^{204}(n,2n)Hg^{203}$

(np)

$Hg^{196}(np)Au^{196}$; $Hg^{198}(np)Au^{198}$; $Hg^{199}(np)Au^{199}$; $Hg^{200}(np)Au^{200}$; $Hg^{201}(np)Au^{201}$; $Hg^{202}(np)Au^{202}$;

(n,pn),(n,np) and (n,d)

Here we expect only the existence of $Hg^{204}(nd)Au^{203}$ having in view the isotopic abundance and half life of product nucleus.

(nα)

$Hg^{198}(n\alpha)Pt^{195}$; $Hg^{200}(n\alpha)Pt^{197}$; $Hg^{202}(n\alpha)Pt^{199}$

For our study we have chosen the following nuclear reactions: $Hg^{202}(np)Au^{202}$ and $Hg^{204}(nd)Au^{203}$.

The Au^{202} half life is not exactly known and an approximation of 25 sec. is given by F.D.S. Butement (45) its decay scheme is also unknown. In the same study using enriched Hg^{204} Butement has found for Au^{203} a half life of 55 sec.

An approximation of max. β energy of 1.9 MeV is given (absorbtion in Al) and of γ rays emitted 0.69 MeV. (for Au^{203}). The estimation of ratio between β^- and γ emission in Au^{203} is unreliable.

Because the short half lives of 25 sec. (not exactly known) and 55 sec., of product nuclei, we have chosen a duration of irradiation of 2 minutes.

In such a short time the short activity from nuclear reactions: $Hg^{202}(np)Au^{202}$ (and $Hg^{204}(nd)Au^{203}$ (55 sec.) are most probable but also other activities with longer half life but with higher cross section should be obtained.

Experiments on Mercury targets. (gamma measurements)

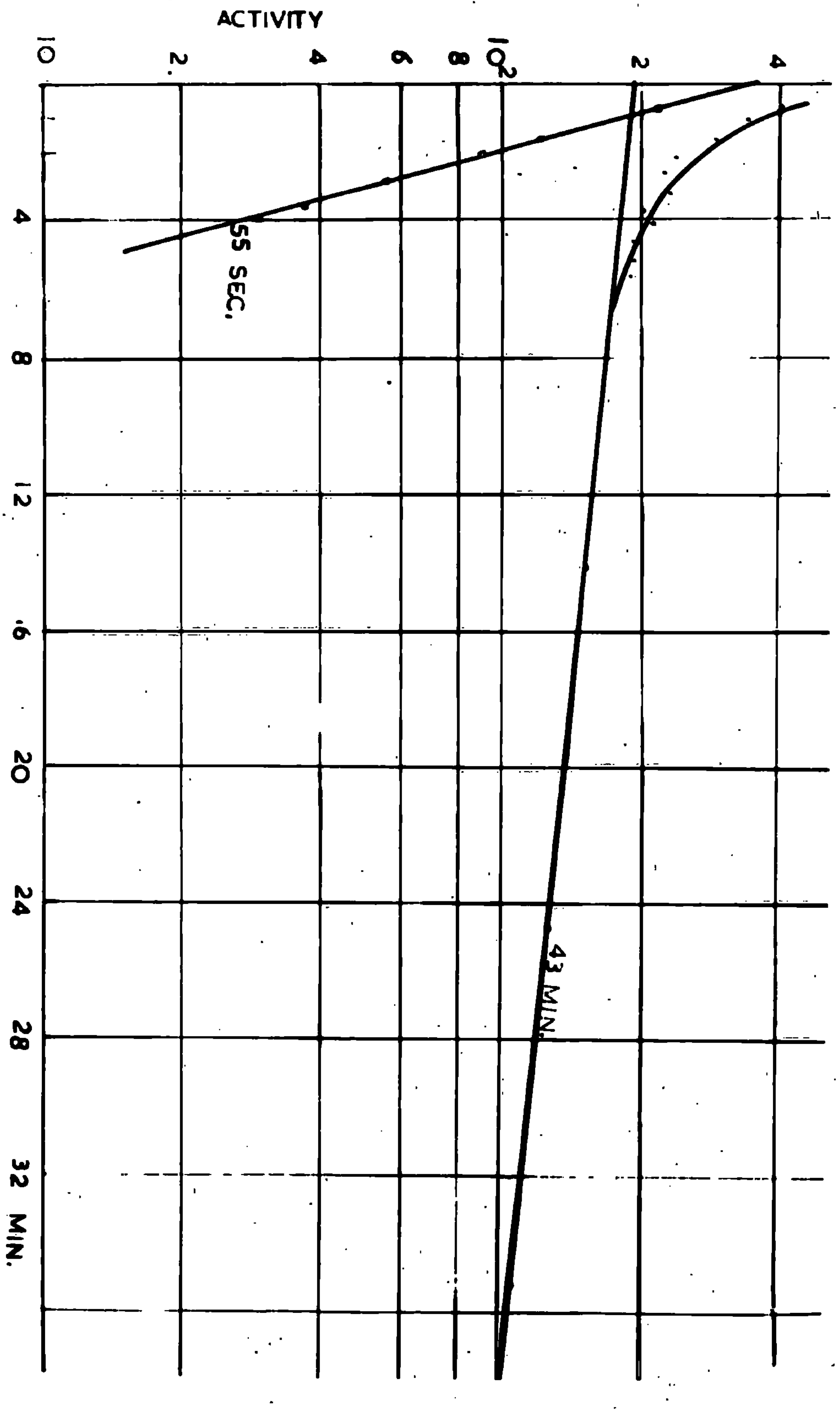
In four experiments 65 g Mercury samples in cylindrical polythene boxes (1.3 cm. diameter and 6 cm height were irradiated in neutron flux with new Tritium targets for two minutes.

30 seconds after the end of irradiation the sample was placed in a well crystal scintillation counter for counting rate measurements of gamma rays. This counter was associated with a 100 channel pulse height analyser.

Having in view the energy of gamma quanta of Au^{203} (55 sec.) of 0.69 MeV a calibration of the counter with Cs^{137} with γ ray of 0.662 MeV was made. A calibration for Se^{75} gamma quanta (0.14; 0.27; 0.40 MeV) was also taken into consideration as a function of channel number. From these data with appropriate back bias (the same as for calibration), the channel set limits, and time interval prefixed, the automatic run system (and printing) was set on. Because of the short half life of 55 sec. the counting interval was as short as possible (0.3' in this experiment) and channel limits set so that only 10 channels were printed out, taking 20 seconds (counting system stopped). Channel width was 0.1V. The channel limits were chosen to include the peak of 0.69 MeV. For the long lived component a time interval of 100'

FIGURE 9

Measurements of γ activity in Mercury


ACTIVITY
10
2.
4
6
8
10²
2
4
4
8
12
16
20
24
28
32 MIN.
55 SEC.
43 MIN.

was fixed. Because the activities are quite high the live time measured by the timing unit does not correspond exactly with the clock time so a stop watch was used and time intervals for counting, and printing (when counting equipment is stopped) were measured. The accumulated counts were printed in groups of 10 channels (set limits 10-19 with bias 73) on roll paper in octal notation and the Kicksorter was running for about 2-3 hours. The results printed on roll paper per group of ten channels were transformed into decimal system and added together. With these results and time intervals (middle) from the stop watch, the decay curves were plotted (counting rate/10 channels versus time).

In one of these four experiments, for long lived component with time interval 100, bias 10, channel set limits 100 the whole spectrum was recorded to find out the existence of higher or lower energies.

From these four plots (decay curves) a very precise 55 sec. component was found the same time with a long component (42-44 min.). An example is given in Fig. 9.

For long component we assume the presence of Hg^{199} (metastable state of 43 min with gamma quanta of 0.159 and 0.368 MeV) because (n,2n) reaction have much higher cross section than (np) reaction resulting Au^{200} (48 min).

Of course the presence of Au^{200} cannot be excluded even if no gamma quanta with higher energies than 1 MeV was found (Au^{200} has also gamma quanta with energies 1.23 and 1.36 MeV) for many reasons:

- Crystal efficiency is lower at higher energies.
- (n,2n) may be with 2 or 3 orders higher than an (np) reaction.
- From nuclear data only 24% from γ quanta has 1.23 MeV and absorbtion in Mercury is not negligible.

All these factors are reducing very much the contribution of gamma rays from Au^{200}. It is also evident that a chemical separation between Hg and Au is necessary.

To explain the existence of Hg^{199} component which has no gamma quanta with energy above 0.4 MeV when set limits (and back bias) were fixed in 0.69 MeV region, we must assume that at high activity when two neighbouring peaks (one at 0.37 MeV and another at 0.69 MeV) exists, the tail from the first one could be detected in the second one (10 channels), or it is possible as the two γ quanta from Hg^{199} 0.16 and 0.37 MeV to build up (cascade) and to enter the 0.69 MeV region.

For all four plots if we take the ratio between the two extrapolated activities at t = 0 we find R ≡ 1.78 - 1.88 values (refers to 10 channels) but with first five channels only, this ratio is higher 3.15 (activity of short component/activity of long one.

In these determinations a well crystal has been used when efficiency is much improved compared with a flat crystal. The resolution of a flat crystal is better than other one because higher probability of photo effect. In a larger crystal the Compton effect as escape effect is lower and resolution is better. These two effects are opposed.

In another experiment when irradiation lasted 12 minutes to verify the long lived components, by calibrating the well crystal with Se^{75} two peaks of the same half lives (two parallel lines) 43 minutes and corresponding to 0.16 and 0.37 MeV has been found.

So we can assume precisely that the long component is Hg^{199}.

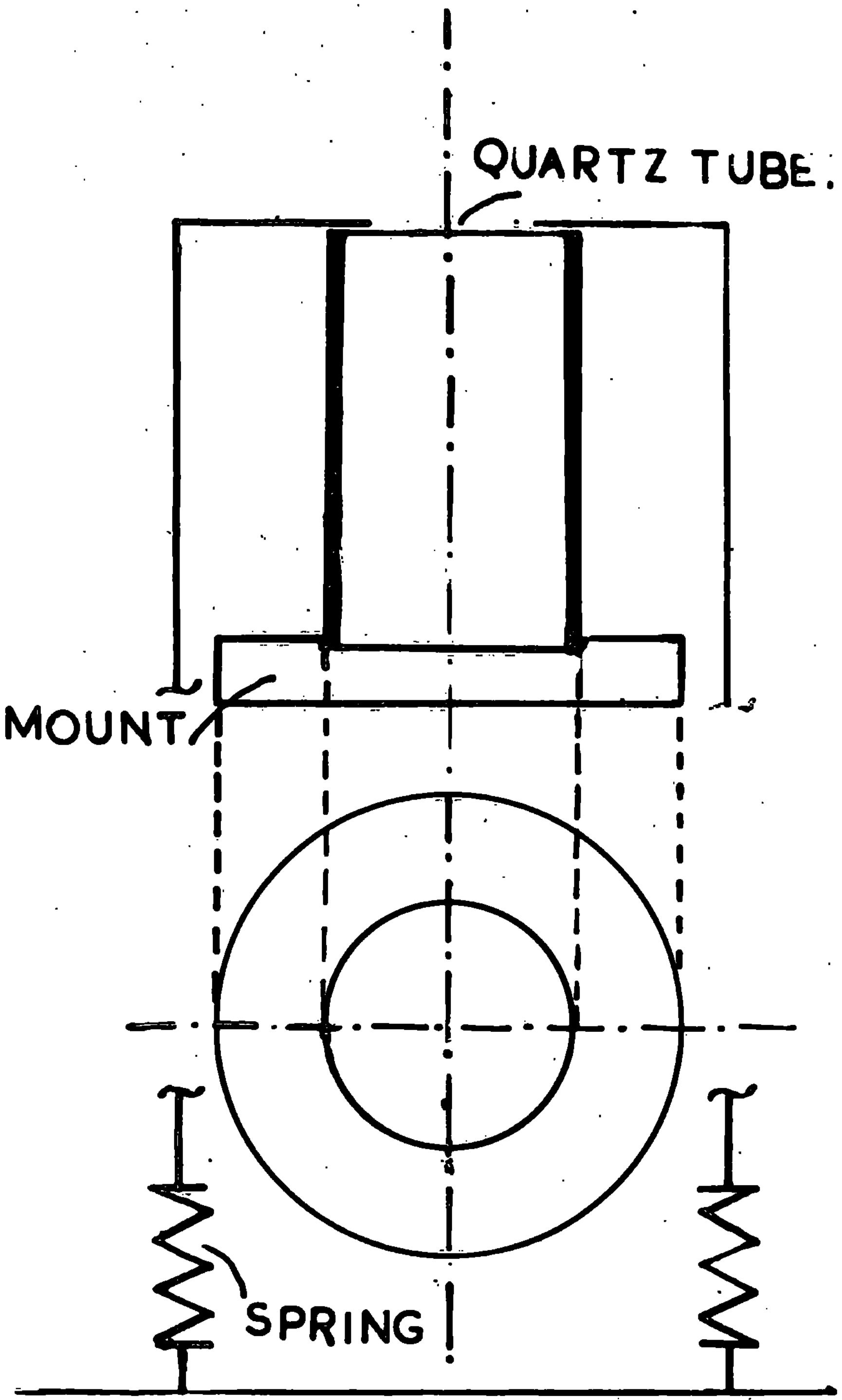
In an experiment with a flat crystal the activity was quite low (low efficiency) but even in this case no gamma quanta above 0.7 MeV has been obtained (improved resolution).

Because we cannot assume the complete absence of Au^{200} (48 min) however undetectable in Hg medium a chemical separation needs to be considered.

Because efficiency of a flat crystal (for γ's) is very low (1-2%) in the further experiments we shall use a proportional counter (the same type as used in a previous chapter) which

FIGURE 10

Apparatus for rapid Au-Hg separation


QUARTZ TUBE.
MOUNT
SPRING

has an efficiency for β's (within this energy interval $>$ 1.8 MeV) of approximately 30%. Both Au^{202} (25 secs.) and Au^{203} (55 sec.) emit β^- rays with energy above 1.8 MeV.

From Dr. Butement's study (45) an estimation of 1 γ quanta to 10 β particles is given and this factor is also favourable to β counting.

For β counting a complete separation between Au and Hg should be done and having in view the very short half lives a rapid separation is necessary.

Au-Hg separation

- It was made by rapid boil off of Mercury after irradiation with fast neutrons, when residue contain only Gold and incidentally Platinum from (nα) reactions.

Here are two factors to be considered:

- It is desirable that the amount of Mercury to be larger to get higher induced activity.
- The larger the amount of Mercury the longer the distillation time.

From these two opposite considerations, we have estimated that 10 g of Mercury is a reasonable amount which can be distilled in 30 seconds.

A very simple apparatus was used for this purpose (Fig. 10).

\- It consists of a flat metallic mount on which a copper tray was placed in a central area. On the tray circomference a 4 cm high quartz cylinder tube with 2.5 cm. diameter is close placed and fixed with two springs.

After irradiation (2 min.) the Mercury is quickly placed on preheated copper tray, and start boiling instantaneously (the mount is heated all the time). The quartz tube prevents Mercury jumping out.

In 30 seconds all Mercury is gone and Gold (and Pt) residue is placed under proportional counter for activity measurement.

β activity measurements on Gold

As we have mentioned an end window gas flow proportional counter for β rays has been used.

The electronic equipment consisted of a scaler controlled by a timing unit. With a prefixed time interval the scaler is stopped and started automatically. A camera actuated by the same timing unit takes pictures on the roll film in the very short intervals when scaler (and counting equipment) is stopped. On the film is recorded the no. of counts for prefixed intervals the same time with the time intervals. From these films the counting rate can be easily calculated per each time interval.

From the end of irradiation until sample starts to be counted elapses 60-90 sec.

Here also we have to use short time intervals for initial activity when short components exists (12 sec. intervals, of which for 10.8 secs. the equipment counts and for 1.2 sec. it is stopped for recording) and later when short components are gone we shall use 4 minute time intervals (2.4 sec. is dead) to record long lived components.

It is very important to establish the long lived components, and from a separate decay curve of this we have obtained a slightly deviated straight line with a half life of approx. 45 min. This fact proves the existence of the following nuclear reactions for the long lived components:
$Hg^{200}(np)Au^{200}$(48 min); $Hg^{201}(np)Au^{201}$ (26 min) and $Hg^{202}(n\alpha)Pt^{199}$ (30 min. We can assume at least the existence of Au^{200} (48 min) and one (or both) of other.

The first (np) reaction should have a higher cross section and has a more abundant target nuclide and it is natural for it to predominate.

It is not very easy to subtract the long activity from the total one in order to estimate the short lived components because it is a mixture of rather similar long lived activities.

A very successful technique applied in Durham is based on "Bunney plot" which permits the calculation of the two activity components at the time t = 0. To use this method we must know the half lives for the two components. In our case we assumed $T_{\frac{1}{2}}$ = 48 min. and 28 min. (average between 26 min. and 30 min.)

Bunney plot use the equation $I = I_1 e^{-\lambda_1 t} + I_2 e^{-\lambda_2 t}$, which is divided with $e^{-\lambda_1 t}$ and the following equation is obtained:

$$I\, e^{\lambda_1 t} = I_1 + I_2 e^{(\lambda_1 - \lambda_2)t}$$

If we plot $Ie^{\lambda_1 t}$ (ordinate) versus $e^{(\lambda_1 - \lambda_2)t}$ (absscisa) we shall obtain a straight line (Bunney plot) which at intercept with the ordinate should give I_1^o and when t = 0 or $e^{(\lambda_1 - \lambda_2)t} = 1$ we have $I = I_1^o + I_2^o$ and I_2^o is easily defined.

And finally from $I = I_1^o e^{-\lambda_1 t} + I_2^o e^{-\lambda_2 t}$, where t is the time corresponding with the middle of interval in counting measurements, subtracting I from total activity we shall find the decay curve for short component.

Because this resulting curve is not a straight line we assume that a mixture of the two components of Au^{202}(25 sec?) and Au^{203}(55 sec.) exists. The first activity is formed in an (np) reaction and the second in (nd).

We have defined, from γ-ray spectrometry, the existence of 55 sec. activity but we expect that cross section for (np) reaction to be higher than (nd) one.

For the short components decay curve we can also apply a Bunney plot to find I_1^0 and I_2^0 but half life of Au^{202} is only rather approximate known (45) and little information exists about its decay scheme.

Because even for Au^{203} (55 sec), in γ ray spectrometry determination, a doubt could arise if a γ ray emission from Au^{202} (? 25 sec.) exists it might interfere with Au^{203}. (55 sec.) We have proposed to check its emission as well as its half life (for Au^{202}). This study cannot be done with a Mercury target but it should be possible with Thallium in checking an $(n\alpha)$ reaction which gives the same Au^{202} without interference with Au^{203}.

14 MeV neutron bombardment of Thallium target

In this case being also interested in short lived activity the time of irradiation was from 30 to 60 seconds.

In this condition the following nuclear reactions with the two Tl stable isotopes (Tl^{203} 29.5% and Tl^{205} 70.5%) are likely to be significant:-

$(n\,\gamma)$; $Tl^{205}(n\,\gamma)Tl^{206}$(4.2 min., β^{-} = 1.51 MeV no γ)
(n,2n); $Tl^{203}(n,2n)Tl^{202}$(12d, K,L,e^{-}, γ = 0.44 MeV)
(n,p): $Tl^{205}(np)Hg^{205}$ (5.1 min, β^{-} = 1.8 MeV, γ = 0.20 MeV)
$(n\,\alpha)$; $Tl^{203}(n\,\alpha)Au^{200}$ and $Tl^{205}(n\alpha)Au^{202}$.

Thallium thin discs were used as targets and flat crystal scintillation counter associated with 100 channel Kicksorter.

Instead of printing out device, which is too slow, a camera actuated by the timing unit was used. It takes pictures. The spectrum is displayed on the photo tube screen (this equipment has been described in a previous chapter).

For short initial activities very short time intervals were used, and later when the short component is gone the time intervals were much larger.

Tl foils are very easily oxidised in air, and during a fast neutron irradiation N^{16}(7.4 sec.) is built up from $O^{16}(np)N^{16}$. Checking the decay curve from the film (which was read with a projector) N^{16} has been always found interfering with any possible 25 sec. activity. By a rapid wash in water of Tl sample after irradiation and using a 3" diameter flat (cylindrical) crystal no peak with short (25 sec.) half life has been found. The same results were obtained with Tl lump in well crystal.

From these experiments a well defined peak with half life 12d and γ energy corresponding to 0.44 MeV prove the existence of $Tl^{203}(n,2n)Tl^{202}(12d,\ K,L,e^{-}\ \gamma = 0.44\ \text{MeV})$ nuclear reaction with high cross section.

Taking into consideration the short half life the rapid transport of Tl disc from the accelerating tube to the counting equipment was made with a rabbit fixed at the target assembly, and released when necessary. When the rabbit with Tl disc is released it falls in a pipe and transported by vacuum to the counting equipment.

Because no γ peak with short half life has been obtained we conclude that from $Tl^{205}(n\alpha)Au^{202}$ no γ ray (detectable) can be ascribed to Au^{202}. In our previous study on Hg targets we can assume that only 55 sec. Au^{203} was the short component of the decay curve and no interference with Au^{202} should be possible.

To continue our study we must find the half life of Au^{202}. It is possible to produce it from the $(n\alpha)$ reaction just mentioned, in Tl metal (disc) as target.

For this study it is also preferable to use the end window gas flow proportional counter with the same electronic equipment (scaler, timer) as in a previous work and with camera actuated by the timer.

The rapid transport of the Tl sample is made by the rabbit. (2-3 sec.)

In our determinations, to improve the statistics usually 3 or 4 irradiations of 1 min. were made and their activities added together. In an initial exponential curve the time for corresponding counting rates is not very important but for long lived activities their summation is made at the same time.

So the decay curve is to be split in two, one for long lived activities and the other for total activities.

For long lived decay curve a component of 4.5 min. has also been found and this fact suggests that besides $Tl^{203}(n2n)Tl^{202}$ the other nuclear reaction as: $Tl^{205}(n\gamma)Tl^{206}$ (4.4 min) and $Tl^{205}(np)Hg^{205}$ (5.1 min) are obtained.

The summation of the long lived activities including 12d and 4.5 min components) from three decay curves, corrected for different time intervals used in the initial measurements of short lived activities, was subtracted from the total activity at corresponding time.

Corrections for background have always been done.

A decay curve (straight line) for Au^{202} has been obtained.

Using the same target several times, Tl^{202}(12d) is built up so a new Tl disc is required. The same time to improve the statistics 4 Tl discs (4 irradiations) and two end window gas flow proportional counters fixed in parallel (the same plateau) were used to obtain improved efficiency. To reduce the 12d component all 4 samples were irradiated for only 30 sec.

The same calculation procedure has been used, having in view the summation of 4 decay curves (separated for initial total activity and final long lived activity, with respective time interval and background corrections). Finally from the total long lived curve, the summation of the two long lived components was subtracted from total initial activity and short lived component (straight line) was obtained.

In all these determinations because statistics could not be very much improved and from difficulties arisen from very short Au^{202} half life we have found for Au^{202} a half life of 22 ± 4 sec.

Results and conclusion

We have established that the half-life of Au^{202} is 22 sec (with ~ 20% error) and that there is no significant γ emission from this isotope. The short-lived (~ 55 sec.) 0.69 MeV γ activity must therefore be ascribed to Au^{203} produced by Hg^{204} (n,pn). Unfortunately the decay scheme of Au^{203} is not well established, and it is hardly possible to base a cross-section measurement on this observed γ activity.

Subtraction of the longer-lived components from the Au + Pt fraction of irradiated mercury gives a short-lived component of apparent half-life lying between 22 and 55 sec. Application of the "Bunney plot" technique to this component enabled us to resolve it into components with these two half lives, and comparison with the activity of Au^{200}(48') - for which the cross section for production by Hg^{200}(np) is already well known - enables us to establish cross sections for the two reactions involved: Hg^{202} (n,p) Au^{202} (25") and Hg^{204} (n,pn) Au^{203} (55"). The data are summarised below:

Table 1

No.	Specifications	Symbol	Units	Value	Remarks
1	Cross section for Hg^{200}(np)Au^{200}	σ 48 min.	mb	3.63	(8) lit.
2	55 sec Au^{203} activity at t=0	I^{o} 55 sec.	c/10.8 sec.	260	Bunney plot

(continued)

3.	25sec. Au^{202} activity at t=0	$I^{o}_{25\,sec}$	c/10.8 sec	3500	Bunney plot
4	48min. Au^{200} activity at t=0	$I^{o}_{48\,min}$	c/10.8sec.	183	"
5	Au^{202} disintegration const.	λ_{25sec}	$sec.^{-1}$	2.77×10^{-2}	Nuclear data
6	Au^{203} disintegration const.	λ_{55sec}	$sec.^{-1}$	1.26×10^{-2}	"
7	Au^{200} disintegration const.	λ_{48min}	$sec.^{-1}$	2.4×10^{-4}	"
8	Abundance of Hg^{200}	a Hg^{200}	%	23.1	"
9	Abundance of Hg^{202}	a Hg^{202}	%	29.8	"
10	Abundance of Hg^{204}	a Hg^{204}	%	6.8	"
11	time of irradiation	t	sec.	120	given
12	cross section for $Hg^{204}(nd)$ Au^{203}	σ_{55sec}	mb	0.6	calculated
13	cross section for $Hg^{202}(np)$ Au^{202}	σ_{25sec}	mb	1.56	"

σ_{55sec} and $\sigma_{25\,sec}$ has been calculated from a very simple formula based, as mentioned on σ_{48min}, the activities involved at t=0, the abundances and λ_{25sec}, λ_{55sec}. With the notations adopted in the previous table we can write

$$\sigma_{55sec} = \sigma_{48min} \times \frac{I^{o}_{55sec.}}{I^{o}_{48min.}} \; \frac{(1-e^{-\lambda_{48}t})}{(1-e^{-\lambda_{55}t})} \; \frac{a\ Hg^{200}}{a\ Hg^{204}} .$$

σ_{25} has been calculated with a similar formula.

All these results are based on experimental determinations of Coleman et al.(8) of $\sigma_{48min} = 3.63$. If we assume the theoretical result for σ_{48min} calculated with Brown-Muirhead direct interaction theory (when $\sigma_{48} = 3.0$ mb) we find, 0.495 mb and 1.29 mb for $Hg^{204}(nd)Au^{203}$ and $Hg^{202}(np)Au^{202}$ respectively.

Comparing these results with other similar (nd) cross section determinations it is slightly higher, but if we take into consideration the possible neutron energy spread 14.7 ± 0.2 MeV and the fact that these cross sections depend strongly on neutron energy we can partially explain this behaviour. We should also bear in mind the rather large uncertainty in the value to be taken for the Au^{202} half-life.

Generally speaking all experimental results on (nd) reactions give higher values for cross sections in comparison with theoretical values obtained by direct interaction theory because the last ones are based only on (n,pn) interpretations when the other possibilities such as: (n,np); (nd) exist.

We cannot compare our results with others similar on Hg target because the (np) and (nd) nuclear reactions involved here are not studied at all in the literature even important parameters as half life (for Au^{202}) or decay scheme (Au^{203}) are unknown.

We are inclined to accept this quite high value for (nd) cross section resulting in Au^{203} because the high activities of 55 sec component observed with γ counting equipment as well as with β^- in the limits permitted by the imprecision of measurements of Au^{202} half life. At the same time a note of caution must be introduced. No information is available about Au^{204} (the product of the $Hg^{204}(n,p)$ reaction), although β -decay systematics would suggest that its half life is very short (perhaps only a few seconds). The possibility cannot be excluded, however, that it has a half-life near enough to that of the 55 sec. Au^{203} to introduce some contribution to the activity ascribed to that nuclide. It might be noted that Au^{204} has neutron number 125, (adjacent to a closed neutron shell) and even if its ground state has a short half-life it would not be unlikely to find a meta-stable state with appreciable life-time.

We have found difficult an exact determination of Au^{202} half life using Mercury target because the very short half life involved as well as the many stable isotopes of Mercury resulting in many radioactive components. The same Au^{202} obtained in an $(n\alpha)$ reaction in Thallium as mentioned is complicated by the presence of Oxygen.

The (np) cross section, resulting in Au^{202}, gives an acceptable result, comparable with other nuclear reactions with target elements within the same mass range. If this experimental result obtained in our work of 1.56 mb is compared with the value predicted by Gardner in (44) the value of

1.40 mb, is in very close agreement with our results. The ratio of these two values; $1.56/1.40 \equiv 1.11$.

These measurements confirm again the direct interaction mechanism between neutron and target nucleus for the (np) reaction as well as for (nd) one within the limits mentioned.

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