TELLURIC ACID IRRADIATION IN AQUEOUS SOLUTION

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INTRODUCTION

One of the most frequently used radioisotopes is I^{131} and thus special attention should be given to its production methods.

Production of I^{131} can be carried out by two processes, namely as fission products and from Tellurium activation based on the following reaction: $Te^{130}(n,\gamma)\,Te^{131} \xrightarrow[25m]{\beta^-} I^{131}$.

The former is used in large nuclear installations while by the second method Tellurium can be irradiated in several forms such as metallic Tellurium, Tellurium oxide and Telluric acid, this last target only being used when thermal fluxes inferior to 10^{13} $n/cm^2/sec.$ are available.

The production of the I^{131} based on irradiation of Telluric Acid presents as an advantage the simplicity of the chemical processing involved, thereby justifying its use whenever feasible. Experience has shown, however, that irradiation in fluxes higher than 10^{13} $n/cm^2/sec.$ gives rise to the formation of high consistency masses when Telluric Acid is irradiated in powdered form (1). This, then, hinders the chemical processing and consequently reduces the yield.

It must be stressed that this phenomenon is not only due to the temperature, for its occurence also appears in water-cooled irradiation elements, (2) in which the Telluric Acid temperature was verified to be lower than its melting or decomposition point.

In order that the production of I^{131} at the Instituto de Energia Atômica be kept at the desired level, a change of the tar-

get to metallic Tellurium or Tellurium Oxide would be required. This would mean a change in the chemical processing system (3). Evidently, another alternative would be to reduce the reactor flux and thus increase the irradiation time of Telluric Acid in powdered form. Such an alteration in the reactor operating program, besides being anti-economical, would jeopardize other uses such as beam-hole experiments, and the radioisotopes activations with high specific activity. As a result, Telluric Acid irradiation in solution was suggested.

Considerable controversy (4) abounds as to the convenience of irradiating solutions, for if irradiation is carried out in a closed container, problems arise related to pressure build-up due to the decomposition of the water. On the other hand, irradiation in an open air condition creates environmental contamination problems. The latter condition was decided on, using an atmospheric pressure system whereby the gases were conveniently filtered and directed to the off-gas system. Due to the solubility of the Telluric Acid, it was found that reasonable quantities could be irradiated in accordance with the reactor geometry. The concentration used was 400 g/liter.

IRRADIATION FACILITY ASSEMBLY

Initially the material to be used in the construction of the irradiation facility assembly had to be chosen and, for this, preliminary corrosion tests with the Telluric Acid solution were made. Among the materials tested, it was found that stainless steel SS316 and anodized aluminum were the most resistant.

Aluminum was preferred due to its absorption and activation cross-sections and also owing to its assembly simplicity. Subsequently, a container subjected to various irradiations was opened up (following a convenient decay time) in order to examine its internal condition in relation to corrosion, but nothing abnormal was observed.

Direct contact between the container with the Telluric Acid in solution and the swimming pool water is not advisable. For that

reason, the container was placed in a 3-inch aluminum tube containing water up to 30 cms. below the flange F. In irradiating position, this flange is located 1.9 mts above the upper core surface. Thus when the irradiation element is suspended for transfer of the solution to the transport container, the flange can be opened manually without any danger to exposure on behalf of the operators. Connected to the said flange are two 6.7 mts long aluminum tubes whose extremes remain at 1.5 mts from the surface of the water. Inside one of the tubes another plastic one passes, directly connected to the irradiation container. Consequently, both the surface of the water in the irradiation element as well as the Telluric Acid surface remain at atmospheric pressure, independently however. As the surface of the water in the tube is at 1.7 mts above the Telluric Acid surface, the hydrostatic pressure in the Telluric Acid solution is lower than the pressure of the water contained in the outer tube while the latter in turn is lower than that of the swimming pool water. Any eventual leakage either through rupture or deficient closing of the flanges will be inwardly directed thereby avoiding the contamination of the swimming pool water as also a possible effect of the reactor reactivity. The positive variation of reactivity resulting from the rapid replacement of the solution by water was calculated. The result indicated that said variation would always be inferior to 0.000$ $\frac{\delta k}{}$, which is consistent with the control rod range.

A carbon filter was placed at the end of the plastic tube and the air activity was measured.

The container and irradiation facility are shown in Fig.1. Two containers of 2.5" and 2" in diameter respectively are being used.

It is to be noted that the total volume occupied by gases resulting from the decomposition of the water would be only 600 cm^3, i.e., the internal volume of the Al tube (E) and the plastic tube up to the active carbon filter. Passing this filter, the gases are mixed with the air in the off-gas system.

During the preliminary tests, special care was provided to check the degree of purity of I^{131}. The presence of contaminations,

not found in the case of irradiation in powdered form, might be possible. Gamma spectrometric and chromatographic analyses were made, confirming (5) the same degree of purity.

A thermal flux depression resulting from the replacement of water by the Telluric Acid solution in the 2.5" diameter container was calculated by the Lewis method (6), providing a total perturbation of only 2% (f=0.98).

GAS EVOLUTION

Once the irradiation facility is established, the next step should be to identify and measure the activity of each of the radioisotopes carried by the gases and, at the same time, to measure the gas evolution resulting from the water decomposition, as also the variation of those parameters with the concentration, the quantity of material and the reactor power.

IDENTIFICATION

The elements most likely to appear would be I^{131}, A^{41} and Tellurium isotopes. An active carbon column to withold the I^{131} and the A^{41} was placed in a plastic tube exit. A gamma spectrometric analysis of the columns was made by a multichannel analyzer. The initial scanning showed several peaks besides I and A, in all probability Tellurium isotopes. The energy and intensity of the peaks were compared with the results of calculation of the Tellurium isotopes activation. According to the latter, the isotope with the highest activity to appear would be Te^{127}. In fact following the half-life in the corresponding peak, the expected value is found and by this process the identification of most of the peaks became possible.

In the initial irradiation tests, A^{41} was expected owing to the quantity of irradiated solution being small in relation to the volume of the container, air occupying the remaining volume.

FILTER EFFICIENCY

Filters of several sizes were used during the material collection period, it being noted that under identical exposure conditions the activity depended on the carbon quantity, whereby it was found that some activity continued escaping into the atmosphere.

Two columns were subsequently fitted, one after the other, and their gammaspectrometry performed (fig.2). It became evident that the first filter retained all the iodine yet only 50% of the Tellurium isotopes. To eliminate this, a Millipore filter was attached to the columns and followed by a further carbon column in which nothing was detected.

RATE OF GAS EVOLUTION

In order to determine the maximum amount of radioisotopes carried off by the gases, which depends on the evolution rate (7, 8), measurements were taken of said rate as functions of the reactor power and solution concentration, the aim in mind for these measurements thus being to acquire data of operational interest. The variation of the local thermal flux in small core reactors (e.g. IEAR-1) (9, 10), renders the reproducibility of the rate of gas evolution measurements difficult.

Besides the flux variation due to the positioning of the control and safety rods, the core arrangement, irradiation facilities, etc., the influence of local temperature is appreciable concerning gas evolution. Various measurements were made and a maximum 10% deviation was observed in gas evolution from one day to the next, resulting from the different positioning of the rods, notwithstanding the power and other conditions remaining constant.

For gas evolution measurement, the Telluric Acid irradiation container was connected through a plastic tube to a thin walled polyethylene container , immersed in a water tank especially designed to allow for the measurement of the displaced water volume, the latter being measured in function of time. A by-pass of the plastic tube is

fitted with a manometer for registering the internal pressure. For every measurement, the pressure difference between the internal system and the outside was approximately 15 mm Hg.

Directly under the free surface of the tank water, there is a supporting grid to avoid the polyethylene containing floating to the surface. The water column from the polyethylene container base to the water surface furnishes the pressure difference aforementioned (i.e. 20 cm of water average). No corrections were made to the gas evolution rate measurements due to that pressure. The measurement system used is extremely simple: however, it will only show relatively accurate results when the rate of gas evolution values are of the same order of magnitude as those indicated in this paper, or higher.

The rate of gas evolution commences to be measured after the reactor has been operating for at least an hour at constant power in order to obtain a steady state condition.

Initially, a number of measurements taken showed the linearity of the gas evolution rate with the volume of the solution. All later measurements were therefore taken in function of the volume.

Several measurements of the gas evolution rate for different concentrations from 10 g/l to 400 g/l were taken and the results shown in Fig.3. As far as it was possible, the existing conditions for the measurements referred to were identical. Under the same conditions aforementioned, a measurement of deionized water was taken and the resulting value found to be negligible in relation to the values found for the different solutions. The curve shows that for low concentrations the gas evolution rate is high, decreasing rapidly with the concentration increase until reaching a plateau. It is expected that the curve will initially rise until attaining a maximum, in as much as for pure water the rate value is much lower than the minimum scale on the graph. To attain an equilibrium rate of higher concentration (> 100 g/l), approximately one hour was required, yet with lower concentrations up to seven were necessary.

Measurements of gas evolution as a function of the reactor power were made. The reactor attained criticality at 500 Kw, the power later raised to 1 Mw, subsequently to 1.5 Mw and finally to 2 Mw. The power was kept constant for two hours at each level and the gas evolution measured. The results are shown in Fig. 4. For every measurement, only a small variation in local temperature and the position of the control rod was observed.

AIR ACTIVITY

It was judged necessary to determine the specific air activity mainly for health physics considerations as also to evaluate the loss of I^{131}.

The carbon filter activities were determined with the aid of a multichannel analyzer with a well-type crystal.

Initially, several I^{131} standard samples with different activities were prepared, and their spectrums compared with that of the carbon filter.

The filter spectrum was performed under given conditions and, subsequently, under the same conditions, the standard sample spectrum was performed. The counting times were compared in order that the 0.365 Mev. peaks might attain the same level, which was achieved with the "pre-set count".

The relation between the counting times was therefore equal to the relation between the activities of the standard and filter, thus the filter activity was easily determined. To avoid dead time corrections or peak displacements, several standards were used and the sample was compared with that which most approached its activity by visual evaluation.

Both the filter and the standard I^{131} samples were placed in similar containers and occupying equal volumes. The two containers were then counted in the same relative position.

By this procedure, the differences owing to absorption and

geometry were minimized.

The measurements for 300 g TA in 750 cm^3 were the following:

For the first three hours:	0.32 μc
For the fourth hour:	0.16 μc
From the fifth hour onward:	0.15 μc/hr.

Allowing that within the first three hours a certain time is necessary to attain a steady state condition, a mean value can be admitted as 0.15 μc/hr. for 300 g in 750 cm^3 for a 2 Mw operation. For an 8 hour operation, we should have approximately 1.1 μc of I^{131}. That activity dispersed in the swimming pool room results in a specific air activity of 2.3×10^{-9} μc/cm^3. The MPC for I^{131} is 10^{-9} c/cm^3, thus if all the radioactive material were to escape to the surroundings, no dangerous concentration would exist.

HANDLING AND TRANSPORT

After irradiation and a "convenient decay time", the element is hoisted by a crane to a suspended position that flange (E) to be opened manually (Fig. 5). The plastic tube is then disconnected from the container. A small diameter aluminum tube, connected to a plastic one, is introduced into the solution container, and the plastic tube is connected to the transport container. The solution is then removed by suction to the transport cask-shield (fig. 5). The latter is then withdrawn to the vicinity of the processing cell. The transfer to the flask is also through suction (fig. 6). During the transfer, the dose rate in the operator's position is of 10 mr / h and lasts for roughly 2 minutes, exposure, as a result being insignificant.

GENERAL CONSIDERATIONS

As previously stated, when the Telluric acid in powdered form was irradiated, conglomerations which reduced the processing efficiency were found. This at times took place with all the Tel-

luric acid irradiated, rendering it difficult to maintain a regular I^{131} production.

The average value of the activation efficiency was 0.04 mc of I^{131} per gram of irradiated Telluric acid and per Mwhr. By the present method, production is regular and the average value of the activation efficiency is 0.06 $\frac{mc}{g\ Mwhr}$, irradiation being carried in the same reactor core conditions.

A further advantage is the elimination of the sample aluminumcapsules for waste disposal following irradiation, as also the absence of Na^{24} from these aluminum capsules which increased the radiation level of the processing cell. Handling is simplified and the removal is by suction allowing the operators to work safely.

As previously stressed, the measurements indicated in item "Rate of Gas Evolution" were aimed at acquiring general data of operational interest. Thus, it was possible to extrapoliate data in order to estimate the total volume of gas evoluted under different time and power operating conditions, as also the quantity and concentration of the Telluric Acid solution. An example of a routine operation at this Institute follows: Irradiation of 300 g TA in a 750 cm^3 solution for 16 hours at 2 Mw. The total volume of gas evoluted is approximately 8 liters. The amount of I^{131} retained in the filter is near to 1.1 mc. The activity of the processed I^{131} is 640 mc. The total transfer time for the solution from the reactor core to the processing cell is an average 30 minutes. Besides the amount of I^{131} retained in the filter mentioned previously, other losses evidently take place during the removal by suction. Said iodine as well as other radioelements are then collected by other carbon filters found on the vacuum line.

One drawback is the formation of milk-white and extremely light precipitates in the solution during irradiation, periodically requiring the removal of these deposits by water cleansing.

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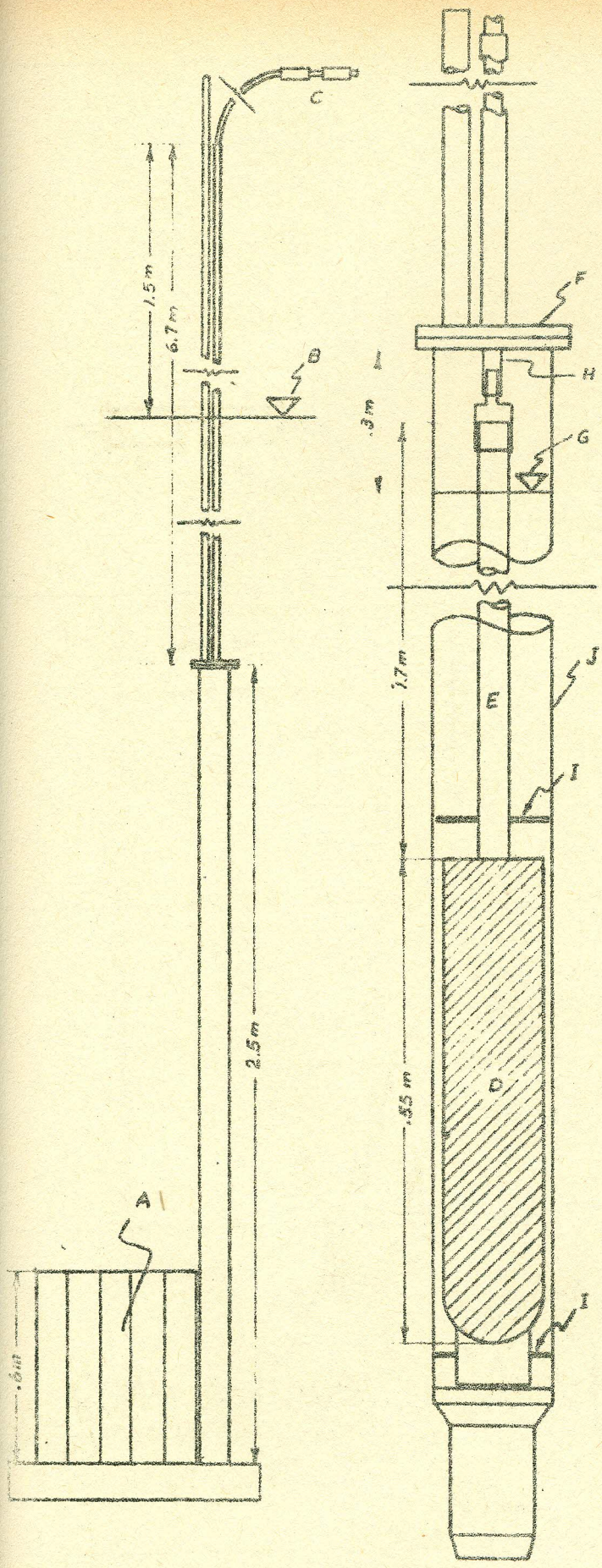

Fig.1. Irradiation element assembly.

A - Reactor core
B - Pool water level
C - Activated carbon filters
D - Solution container, 2" or 2.5" diam.
E - Aluminum tube, 0.5" diam.
F - Liners
G - Water level
H - Plastic Tube
I - Screw fastened rubber gasket filled flange.
J - Outer aluminum tube, 3" diam.

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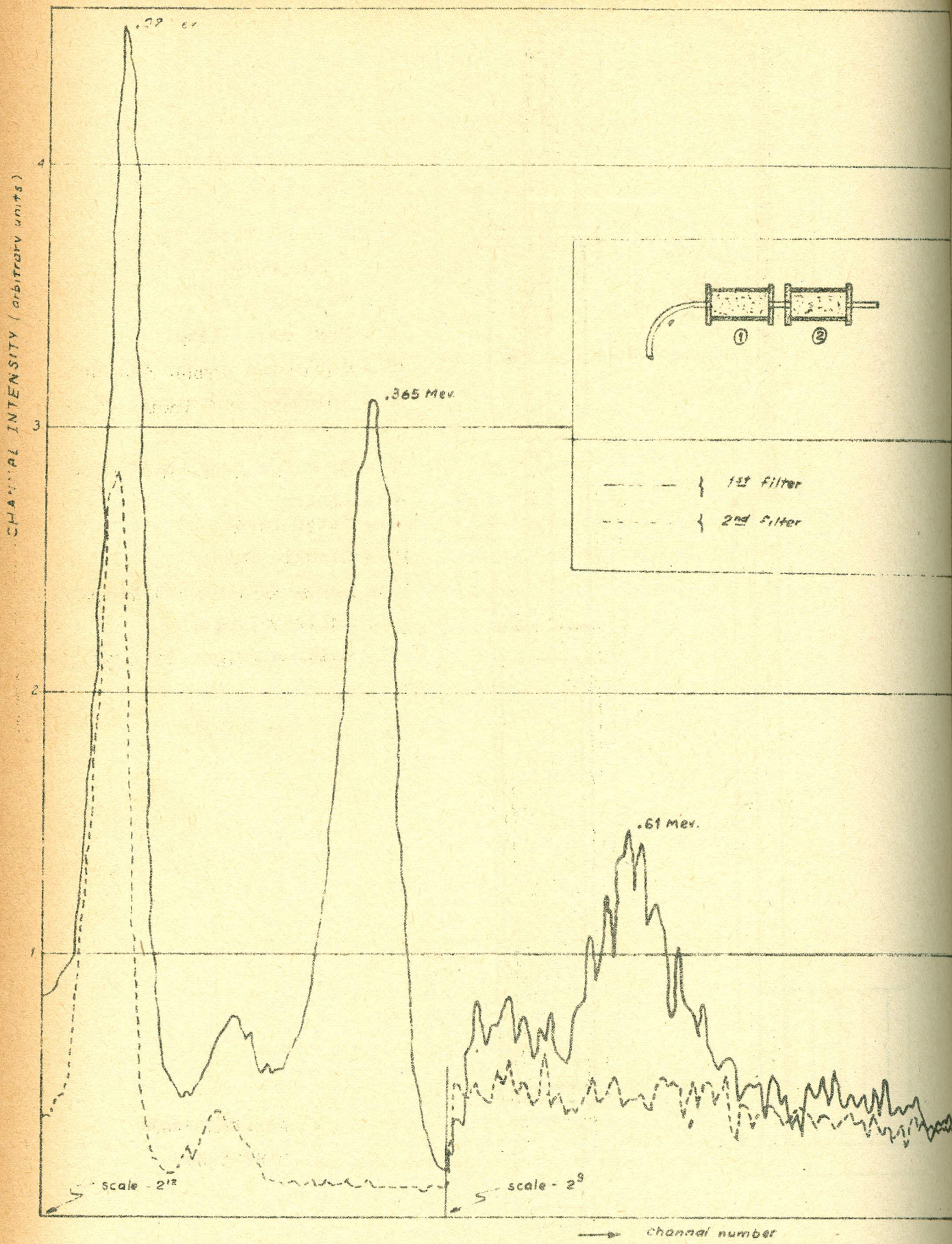

fig. 2. Gamma spectra showing the filter efficiency for I^{131}.

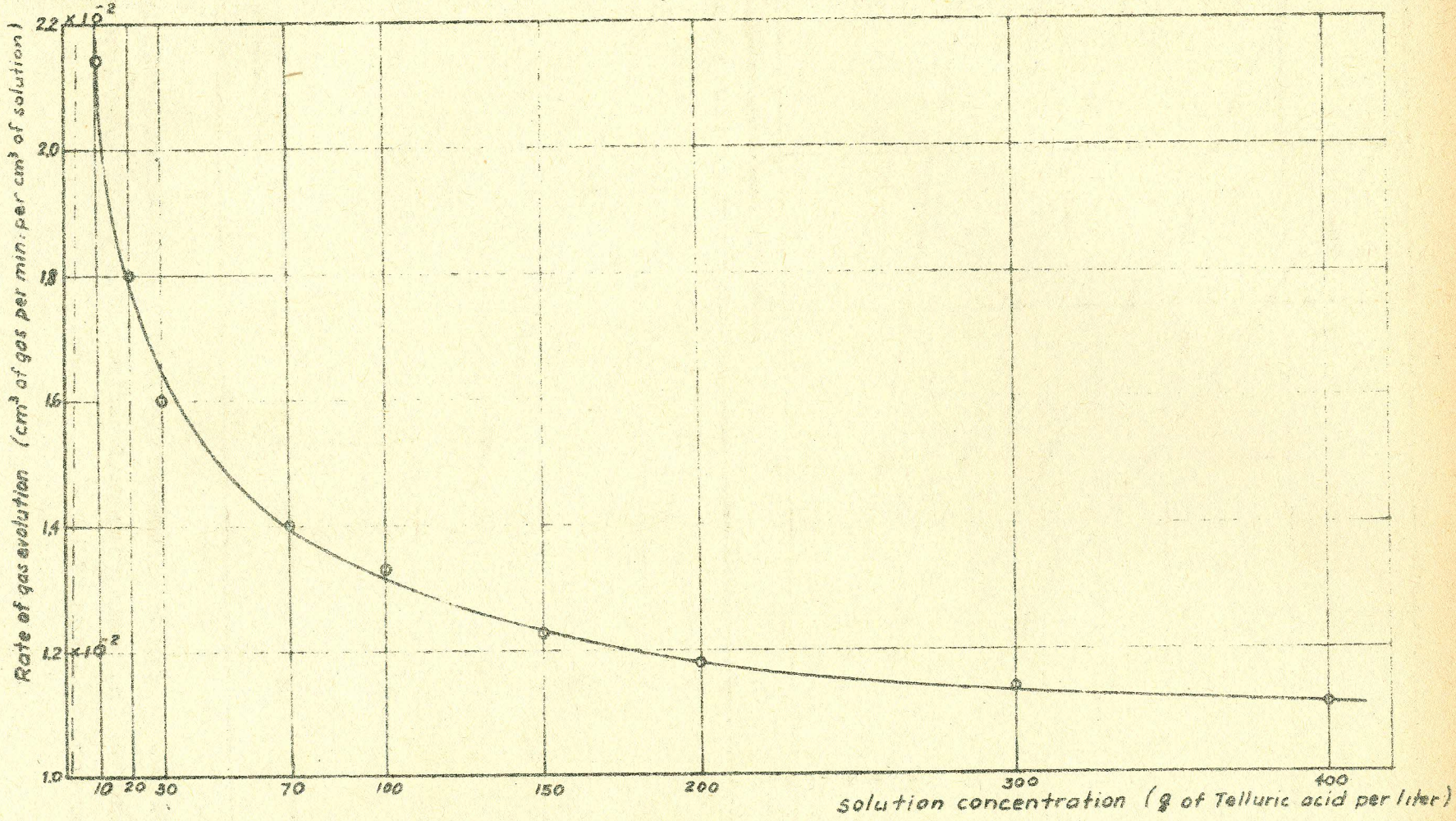

fig. 3. Rate of gas evolution x solution concentration. The average thermal neutron flux is 9×10^{12} at 2 Mw. Reactor core position 68.

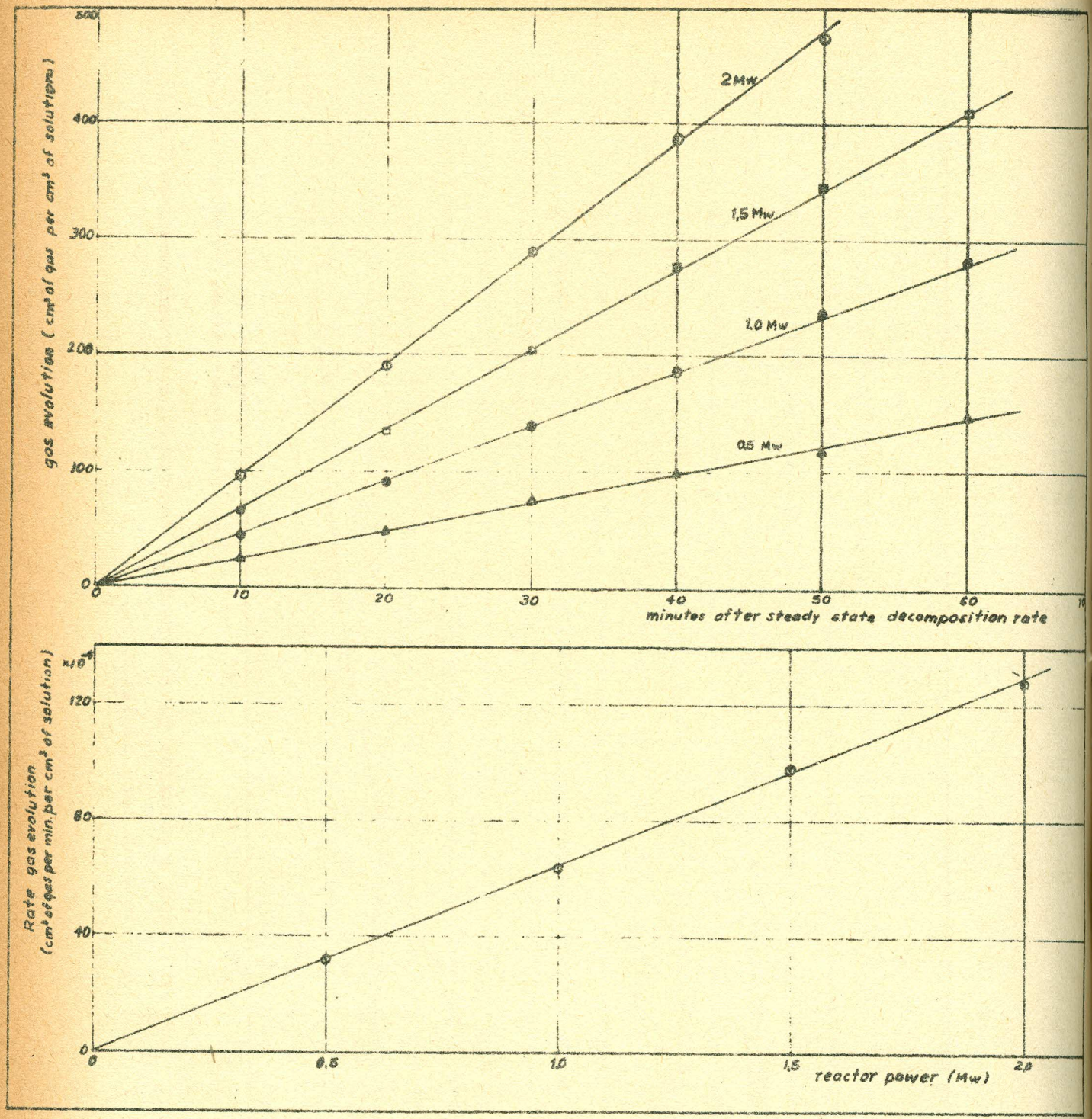

fig. 4. Rate of gas evolution x Reactor power. 300 g of Telluric acid in 750 cm^3 of solution. The average thermal neutron flux is 10^{13} $n/cm^2/sec$ at 2 Mw. Reactor core position 52.

For technical reasons it was impossible to reproduce Figures 5 and 6.