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PRODUCTION OF CARRIER-FREE Na-24 BY IRRADIATION OF PURE Mg MATRICES IN POOL-REACTORS

by

R. MALVANO, U. ROSA, P. GROSSO
(SORIN)

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Report prepared by SORIN
Centro Ricerche Nucleari, Saluggia - Italy

Euratom Contract No. 058-64-6 RISI

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Brussels, December 1966

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A radiochemical method based on the electrodeposition of the Na^{24} activity on a Hg cathod was developed : 0.5-g amounts of Mg can be processed in 4 hours, with nearly quantitative recoveries (93 ± 5 %) and high decontamination factors from the matrix (1.5 to 5×10^3). The procedure is described and the results of some production experiments at mC level are reported. The

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SUMMARY

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A radiochemical method based on the electrodeposition of the Na^{24} activity on a Hg cathod was developed : 0.5-g amounts of Mg can be processed in 4 hours, with nearly quantitative recoveries ($93 \pm 5\,\%$) and high decontamination factors from the matrix (1.5 to 5×10^3). The procedure is described and the results of some production experiments at mC level are reported. The specific activity obtained when using 99.99 % Mg was found to be in the order of 90 C/g, and the radiochemical purity about 99.9 %, for 4-day irradiations at 2×10^{13} fast neutron/cm^2 sec, taking into account a 15-hour decay (treatment and delivery time).

1. INTRODUCTION (*)

The short-lived radionuclide Na^{24}, $T_{\frac{1}{2}}$=15.0 h, is sometimes preferred to 2.6-y Na^{22} as radioactive tracer, particularly in some medical and biological applications. Carrier-free Na^{24} at mC levels may be produced in pool-reactors by irradiating both Na - free Mg or Al in positions of high fast-neutron flux, through the reactions $Mg^{24}(n,p)Na^{24}$ and $Al^{27}(n,\alpha)Na^{24}$.

The main problem connected with the production of carrier-free Na^{24}, is to avoid contamination by inactive sodium. Furthermore, the separation procedure must be relatively fast and sufficiently simple to be compatible with the remote-handling.

In the present paper a separation procedure by electrolysis on mercury cathode is described.

2. EXPERIMENTAL AND RESULTS

Na^{24} production in a pool-reactor

When the RS 1 Avogadro reactor is working at its normal power level of 5 MW, fast (>1 MeV) fluxes as high as 4 x 10^{13} n/cm^2.sec are available in the central position of the core; flux values in the order of 2 x 10^{13} n/cm^2 sec may be reached in some more easily available positions in the core surroundings. In these conditions, when irradiating up to saturation Mg or Al matrices, 13 to 26 or 7.5 to 15 mC of carrier-free Na^{24} can be produced per gram, as evaluated on the basis of the cross section data

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(*) Manuscript received on November 10, 1966 reported in the literature [1] and experimentally checked for some irradiation positions (see Table I). The experimental values for the mean cross-sections for a fission spectrum were obtained by irradiating Cd-lined Mg and Al samples, using as reference the reactions $Fe^{56}(n,p)Mn^{56}$, $Fe^{54}(n,p)Mn^{54}$ and $Al^{27}(n,p)Mg^{27}$ (0.97, 53 and 3.5 mb, respectively [2]). The fast flux spectrum in the checked positions was assumed to be similar to a fission spectrum, and the absolute measurements were performed by means of a 200-channel pulse-analyser Laben C-31, equipped with a 3" x 3" NaI(Tl) scintillation detector accurately calibrated (within ± 2%).

Radiochemical separation of Na^{24} from the matrix

Very little is reported on the literature about electrochemical methods for extremely diluted Na solutions, the only work described being the one developed by Chemla and Pauly for the separation of Na^{22} from cyclotron-irradiated Mg matrices [2]. These Authors were able to recovery about 95% of the Na^{22} activity, after a ten-hour electrolysis of 0.2g Mg dissolved in HCl, by using a particular arrangement which allowed a simultaneous Na recovery into H_2O from the amalgam.

Anyway, a poor reproducibility, low yields and too prolonged operation times resulted when some preliminary experiments were carried out on 0.5-g Mg samples. Similarly, the use of a conventional electrolysis cell led to unsatisfactory results. This fact was attributed to the possible interference of Cl_2 on the Hg cathodic surface: a sintered glass (or quartz) diaphragm was therefore used, and much better results were obtained.

Our electrolysis cell, shown in Fig. 1, is somewhat different from that described by Chemla and Pauly, as a diaphragm was inserted to separate the cathodic from the anodic compartment, and some modifications for routine remote-handling procedures were made. The recovery of the final Na^{24} solution and the rejection to the waste of the electrolyzed solution can be simply controlled by the Hg level, by raising the Hg reservoir outside the lead shield.

In order to optimize the procedure, some experiments were carried out in a cell similar to the one shown in Fig. 1, by using carrier-free Na^{22} or Na^{24} at high specific activity as tracer (μC level).

The Na electrodeposition was observed to begin taking place when in the cathodic-zone pH has reached the approssimative value of 1, and the complete separation is accomplished at pH values of 6-7. The use of other acids than HCl as matrix dissolver is therefore excluded. Since all HCl is electrolyzed before the beginning of the Na separation, the minimum amount of HCl was used for the dissolution of the matrix, in order to reduce the overall operation time.

Owing to the increasing of pH values in the cathodic compartement during the electrolysis, precipitations occur and the Na deposition rate is reduced; moreover, the formation of insoluble deposits is to be avoided in routine procedures. The use of complexing agents in order to keep the matrix dissolved during the whole processing time was considered; citric acid was therefore added, as other commonly used complexing agents such as EDTA, oxalic and tartaric acid were not taken into account, as being major sources of Na contamination.

An experimental run, using 0.5 g Mg in 30 ml solution to simulate the matrix, was carried out in order to test the optimum citric acid concentration (see Fig. 2): low citric acid concentrations, in fact, result in more or less abundant precipitates, while too high concentrations do buffer the solution at low pH values and the deposition time is remarkably increased. A 11.5% concentration of citric acid was found to be adequate for reasonably fast and quantitative separation (93 ± 5%, 4 hours).

The effect of the Mg concentration was similarly checked, by mantaining unchanged a concentration of 11.5% for citric acid (see Fig. 3).

With regard to the Al matrix, some experiments were also performed; the results illustrated in Fig. 4 show that only smaller Al amounts (about 0.2g) can be treated, and precipitation of insoluble hydroxide occurs. This fact can be referred to the amphoteric character of the Al ion.

Since for Al the Na^{24} production rate is coeteris paribus lower than for Mg(0.61 ± 0.04 produced Na^{24} activity ratio), and, moreover, the dissolution in HCl is much more difficult and the separation less satisfactory, the use of Mg as matrix was finally adopted.

All the experiments were carried out with the following experimental conditions: 30 ml electrolytic solution; 20 ml H_2O for Na recovery from the amalgam; applied voltage 30 V; initial current intensity 400 mA, initial current density 4 A/dm^2. When the Na separation takes place, after the Cl_2 gas evolution the current falls to lower values (about 250 mA). The Cl' concentration is initially nearly stoichiometrically equivalent to the Mg concentration (free HCl~0.05 N).

The outer walls of the bottom of the cell, kept in contact with Hg, are coated with silicone grease.

The recovery yields for the developed procedure, as checked through Na^{22} or Na^{24} measurements, was found to be $93 \pm 5\%$ as an average of 21 experiments in standard conditions (0.5- g Mg as $MgCl_2$ in 30 ml of 11-12% citric acid; 4-h electrolysis).

The decontamination factor from the Mg matrix was determined by means of neutron-activation analysis on 10-ml aliquots of the recovered solution. Irradiations of samples and Mg standard solutions were carried out in the pneumatic irradiation facility of the reactor (thermal flux about 10^{13} n/cm^2 sec), for 30-sec periods. The characteristic photopeaks of 9.5-min Mg^{27} were measured with a 200 channel pulse-analyser, equipped with a 3" x 3" NaI(Tl) crystal, and the background of longer-lived Na^{24} and Na^{22} (when present as yield monitor) was subtracted by means of decay curves.

A decontamination factor of $3.3 \pm 1.8 \cdot 10^{3}$ was found, as an average of 7 independent experiments.

Evaluation of the specific activity

On account of the wide possibility of inactive Na contamination, an accurate evaluation of the Na concentration in the final solution must be performed.

Reagents and materials were therefore analysed for their Na content, by means of neutron-activation methods. The technique consisted in irradiation in the pneumatic facility of samples and standard for 10 - 15 minutes, and in γ -spectrometry measurements with a 200 channel pulse-analyser.

In the case of HCl samples some hour waiting time was needed to allow the 38-min Cl^{38} decay. In order to evaluate the Na contribution from the walls of the electrolytic cell, the recovery water after 4-hour electrolysis of known amounts of HCl with determined Na contents, was submitted to the analysis. The Na content in three type of silicone was moreover determined, although some tests demonstrated that less than 3% Na was recovered in water contacted with silicone during 6 hours. In Table II the results of Na determinations are listed. No significant differences were noted between the contributions from a glass and a polymethylmethacrylate cell, the relatively high results for the latter type being possibly to be attributed to the probably impure cement used for the construction. Since the Na content in HCl and citric acid is low, no further reagent purification was attempted.

The analysis of the material to be used as a matrix is more complicated as 370 to 530 ppm of Na, are simulated by the (n,p) reaction on Mg^{24} depending on the irradiation position used (thermal-to-fast flux ratio 5 to 3.5).

A destructive procedure, based on Na-trace separation prior to irradiation, has been therefore developed [3].

A total amount of 34.8 ± 2.8 μg Na were found to be present in the recovery final solution, when treating 0.5g of Johnson and Matthey 99.99% Mg: if conditions corresponding to 4-day irradiations at fast fluxes of 2×10^{13} n/cm^2 sec, and 15-h decay are assumed, specific activities in the order of 90 C/g Na can be expected.

3. EVALUATION OF THE RADIOCHEMICAL PURITY

Mg matrices may contain thermal-neutron activable impurities. The main trace elements in Johnson and Matthey Mg, 99.99%, determined by neutron-activation analysis, were found to be Cu Mn and Zn, (4.5, 23.5, 61.6 ppm respectively) which originate the radionuclides 12.8-h Cu^{64}, 2.56-h Mn^{56}, 13.8-h Zn^{69m} and 245-d Zn^{65}. Activation analyses were carried out by 10-min irradiations in the pneumatic system for Mn and Cu, and 10-h-irradiations in pool for Zn traces. About 0.1-g samples were treated; Zn was determined by non destructive γ-spectrometry on long-lived Zn^{65} after a 3-d decay period, while for Mn and Cu radiochemical separations were performed.

During the electrolysis process, the contaminating activity is partially recovered into the final Na^{24} solution, as tracer tests demonstrated.

The contamination can be reduced when Cd-lined matrices are irradiated, due to the rather high Cd-ratios for Cu, Mn an Zn (41, 45, 23-28 respectively, as experimentally evaluated in the irradiation position used for Na^{24} production). Since no appreciable fast flux attenuation was found when using a Cd shield, Na^{24} activity can be produced at high radiochemical purity levels. When the experimental conditions as outlined in Table III (see footnote) are employed, the radiochemical contamination can be kept within a 0.1% level.

As Hg is used for transporting and discarding the solutions of the electrolytic cell, when remote-handling procedures are carried out, the contamination of the cathode and the diffusion in the Hg bulk could originate more serious contaminations for successive Na^{24} preparations: anyway, the decay of Mn^{56}, Cu^{64}

and Zn^{69m} prevents a substancial activity build-up in Hg, and, moreover, decontamination of Hg in the reservoir outside the shielded Pb cell can be easily carried out whenever necessary.

4. PRODUCTION EXPERIMENTS AT mC LEVEL

0.5 samples of pure Mg are irradiated in high fast-neutron fluxes, using Cd shielding. The irradiated sample transferred in a Pb-shielded manipulation cell is quickly dissolved in about 3.5 ml of concentrated HCl, and the solution, diluted to 30 ml with 13% citric acid, is pipetted in the electrolytic cell E (see Fig. 1), and electrolysed for 4 hours. The aqueous Na^{24} solution from the outer compartment is recovered through the stopcock 12, after opening the connection 14 with the Hg reservoir A; the inner anodic compartment 3 is then removed and the exhausted solution is released to waste through the "ad hoc" holes 11 and the stop cock 11, by raising the Hg level.

By using the arrangement shown in Fig. 1, and the above described procedure, some Na^{24} production experiments were performed (see the results reported in Table IV and V); it must be noted that higher Na^{24} activities could be easily prepared, by simply prolonging the irradiation time and reducing the decay down to a 4.5-h time, sufficient for the whole operation sequence (extraction from the reactor, dissolution and radiochemical separation); a proper choice of irradiation position can be, moreover, made.

5. DISCUSSION AND CONCLUSIONS

The problem set by the radiochemical separation of the Na^{24} activity from the bulk of the Mg matrix is somewhat complicated by the conditions required in the specific case.

Due to the plenty of potential contamination sources, some simple single-step procedures are not useful. Thus ion-exchange techniques fail, as the possibility of selective Na^{24} retention on small resin columns, washing out the bulk of the matrix, is excluded (owing to the need of pH adjusting on the acid solution); the alternative procedure (selective adsorption of the matrix) requires too large resin amount, so that a large contamination by inactive sodium may be expected. In effect, it was found in preliminar experiments that a resin column for the absorption of 0.5g of Mg, releases more than 70 μg of Na.

The above reported method for Na^{24} production seems to be very useful for the preparation of mC of Na^{24} at high specific activity. The main limitation is probably the need of treating rather small matrices (~0.5 g Mg), so that the maximum activity which can be reached is controlled by the irradiation conditions such as flux level and irradiation time.

The chemical recoveries (93 ± 5%), the decontamination factor from the matrix (1.5 to 5×10^3) and the overall operation time (4 h) are quite satisfactory. The specific activity is also good, as a 6-fold factor relatively to Na^{24}.production from Na thermal irradiation was found. Since more than 40% Na found to dilute the final Na^{24} activity is originated from the matrix, better results can be reached when using higher-purity Mg, and moreover a further advantage can be expected if a quartz cell is adopted.

The main advantage of the method is probably the simple arrangement for remote handling operations which the use of Hg as cathode and for solution transport in the cell makes possible.

BIBLIOGRAPHY

[1] R. Beaugé, (CEA Report), DEP-SEPP 148, (1962).

[2] M. Chemla, J. Pauly, Bull. Soc. Chim. France, 432, (1953).

[3] R. Malvano, "Neutron activation analysis of Na traces in magnesium", (in press).

List of Figures

Fig. 1 - Electrolytic cell and arrangement for remote-handling procedure

A - Hg reservoir

B - Remote handlings rods for stopcoks

C - Pb shielding wall

D - E.M. stirrer

E - Electrolytic cell

Fig. 2 - Influence of citric acid concentration on Na^{24} recovery rate 30 ml solution;0.5g Mg; citric acid (1) 1.5g, (2) 2.5g, (3) 3.5g, (4) 6g, turbidity or precipitation.

Fig. 3 - Influence of Mg concentration on Na^{24} recovery rate 30 ml solution; 3.5g citric acid; Mg (1) 0.2g, (2) 0.5g, (3) 1g.

Fig. 4 - Influence of the matrix type on Na^{24} recovery 30 ml solution; (1) Mg 0.5g, citric acid 3.5g; (2) Al 0.2g, citric acid 1.5g; (3) Al 0.2g, citric acid 3g; (4) Al 0.5g, citric acid 4g precipitation.

TABLE I.

Values of $\bar{\sigma}_f$ for the production reactions for Na^{24}

Reaction	$\bar{\sigma}_f$ (mb)	
	Found	Literature [2]
$Mg^{24}(n,p)Na^{24}$	1.29 ± 0.09	1.2
$Al^{27}(n,\alpha)Na^{24}$	0.62 ± 0.01	0.61 ± 0.01

TABLE II

Na determinations in reagents and materials, by neutron activation analysis

Material	Na found			Independent determinations
H_2O, deionized	0.021	± 0.000	µg/ml	2
HCl, Merck reagent-grade	5.1	± 0.5	µg/ml	3
HCl, R. Pont reagent-grade	1.3	± 0.4	µg/ml	4
HCl, C. Erba reagent-grade	0.59	± 0.04	µg/ml	4
Citric acid, Merck reagent-grade	0.89	± 0.08	µg/g	4
Citric acid, C. Erba reagent-grade	4.0	± 0.8	µg/g	3
Silicone grease, Edwards (high vacuum)	330	± 15	µg/g	2
Idem, purified (a)	133	± 16	µg/g	2
Silicone fluid, Edwards (optical contact)	0.15			1
Contribution from the electrolysis cell: glass	7.7	± 1.3	µg	5
polymethylmethacrylate	8.2	± 2.2	µg	2
Total contribution for standard conditions (b)	137	± 1.3	µg	

a) Purification performed by dissolving silicone in $CHCl_3$ and washing with diluted HNO_3

b) 3.5 g citric acid Merck, 3.5 ml HCl Erba, glass cell.

TABLE III.

Contaminating activities in the final Na^{24} solution (a)

Activity	mC
Na^{24}	3.2
Cu^{64}	8×10^{-4}
Mn^{56}	3×10^{-3}
Zn^{69m}	1×10^{-4}
Zn^{65}	2×10^{-5}

a) The following conditions are supposed : 0.5 g Mg, 5 ppm Cu, 25 ppm Mn, 60 ppm Zn ; 4-d irradiation, fast flux 2×10^{13} n/cm^2 sec, thermal flux 8×10^{13} n/cm^2 sec, Cd- shielding used ; 15-h waiting time ; 93 Na^{24} yield ; 50 % Cu Mn Zn recovery.

TABLE V

Characteristic of the final Na^{24} solution (a)

Total Na^{24} activity	3.2 mC
Na^{24} radioactive concen	160 /uC/ml
Specific activity	92 C/g
pH of the solution	6
Mg present	0.15 mg
Contamination of foreign activities	0.1 %

a) The data refer to the conditions of 4-d irradiation at a fast flux of 2 x 10^{13} n/cm^2 sec, and 15-h decay.

TABLE IV

Na^{24} production experiments

Run	Irradiation time (h)	Fast flux (n/cm^2 sec x 10^{13})	Decay time (h)	Matrix weight (g)	Chemical yield (%)	Separated Na^{24} activity (mC)
A_1	10	0.75	6	0.51	97	0.71
A_2	10	0.75	11	0.50	96	0.56
B_1	10	1.25	6	0.52	93	1.16
B_2	10	1.25	12	0.50	95	0.88
C_1	10	1.9	5.5	0.49	88	1.65
C_2	10	1.9	11	0.50	86	1.39

Fig. 1 - Electrolytic cell and arrangement for remote-handling procedure

A - Hg reservoir

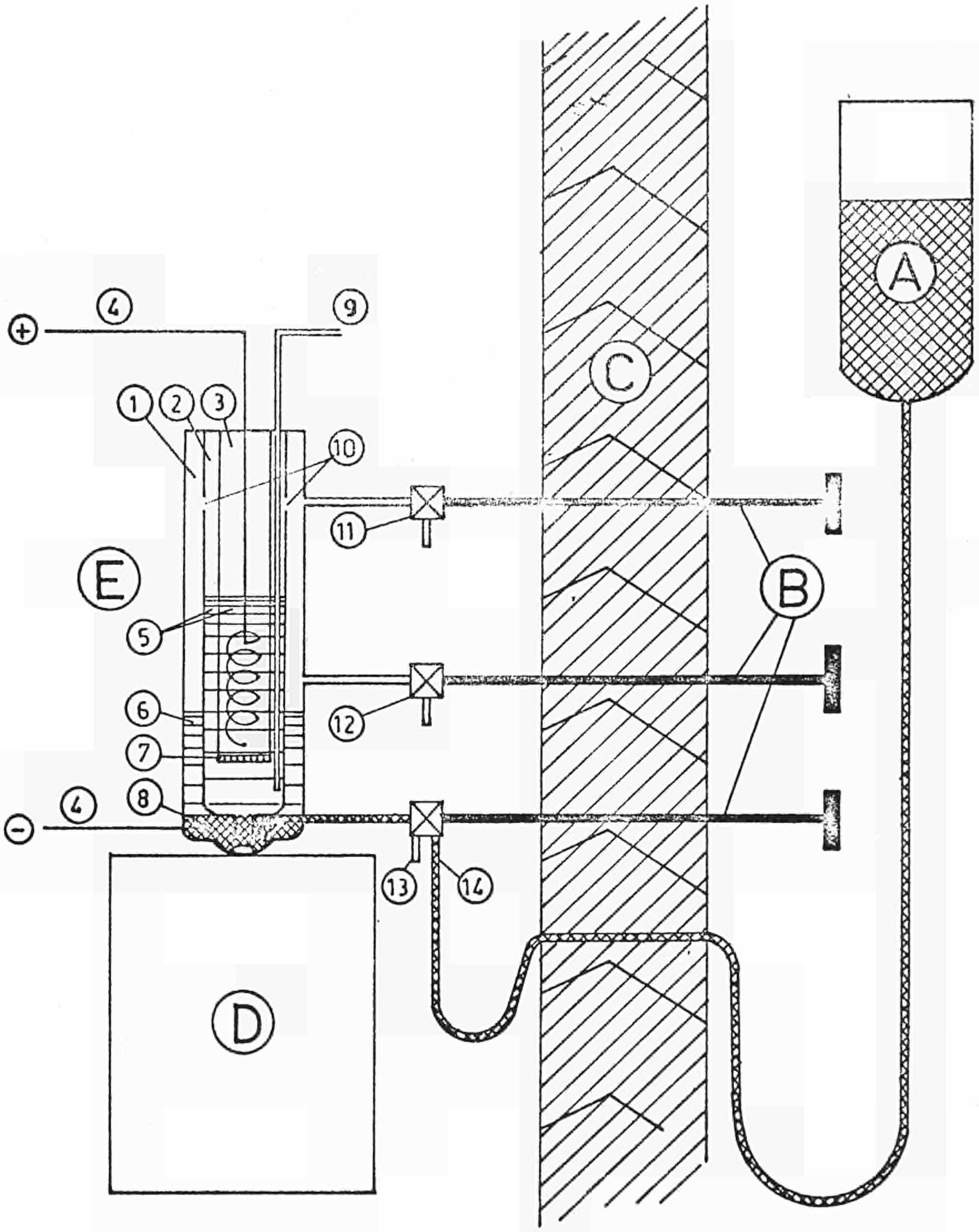
B - Remote handlings rods for stopcocks

C - Pb shielding wall

D - E.M. stirrer

E - Electrolytic cell

1 - Outer compartment for Na^{24} recovery from amalgam
2 - Cathodic compartment
3 - Anodic compartment
4 - Pt wire
5 - Mg solution
6 - H_2O
7 - Sintered glass diaphragm
8 - Hg cathode
9 - N_2 inlet
10 - Electrolyzed solution outlet from the inner compartment
11 - Electrolyzed solution outlet, to the waste
12 - Na^{24} solution outlet
13 - Hg outlet, for emptying the cell and the reservoir
14 - Hg reservoir connection



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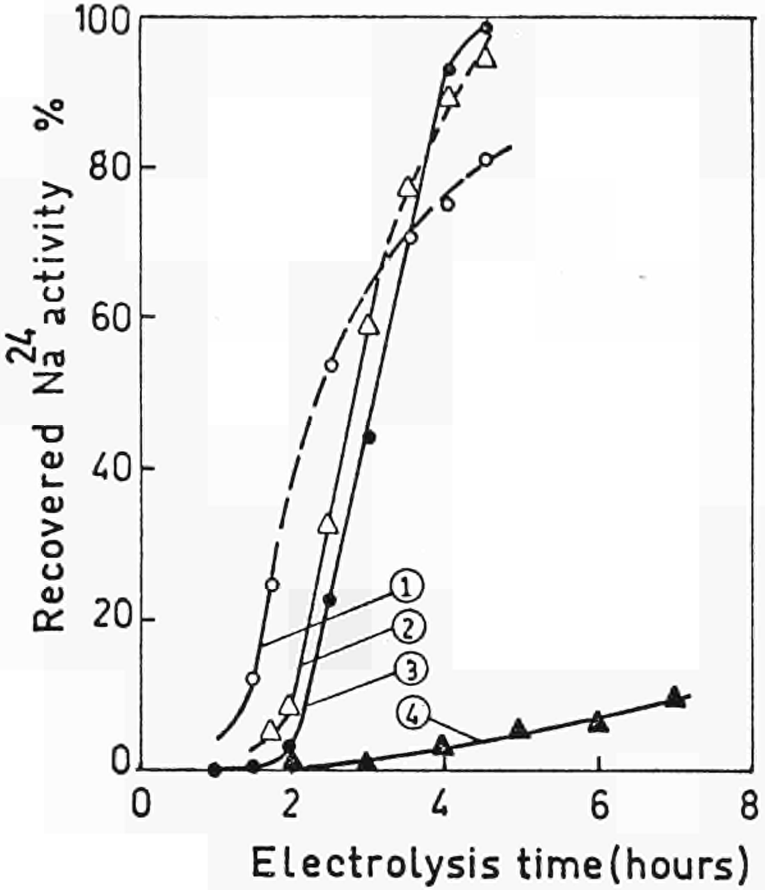

Fig. 2 – Influence of citric acid concentration on Na^{24} recovery rate 30 ml solution; 0.5 g Mg; citric acid (1) 1.5 g, (2) 2.5 g (3) 3.5 g, (4) 6 g, turbidity or precipitation

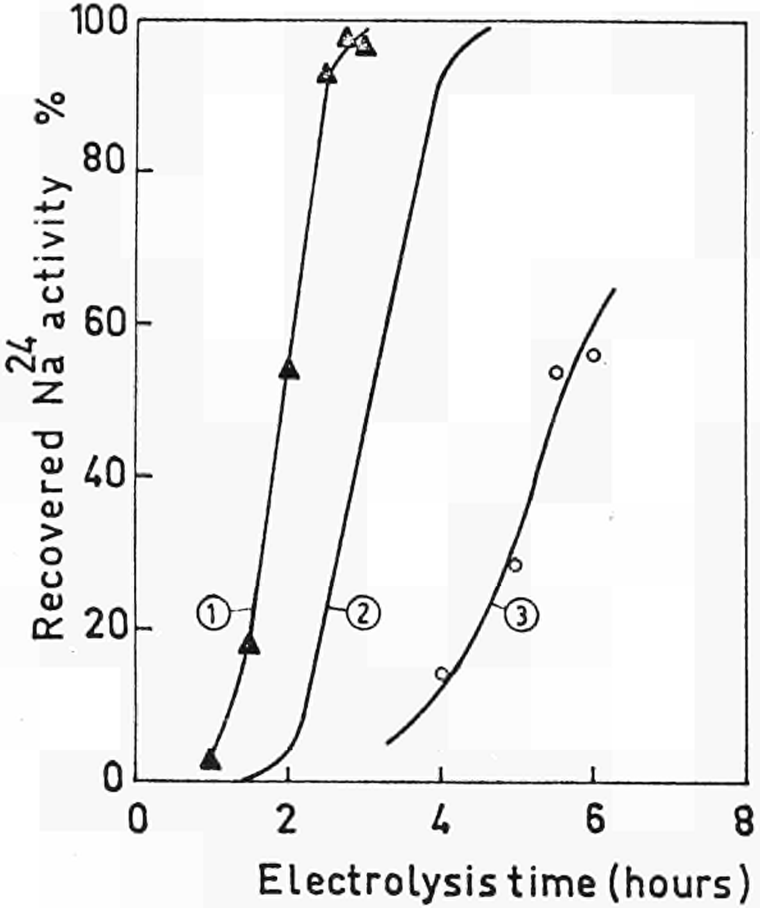

Fig. 3 – Influence of Mg concentration on Na^{24} recovery rate 30 ml solution; 3.5 g citric acid; Mg (1) 0.2 g, (2) 0.5 g, (3) 1 g.

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