

## RADIOCHEMICAL SEPARATION OF RADIOTHALLIUM FROM PROTON-IRRADIATED LEAD

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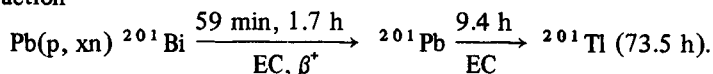
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Absorption of carrier-free Tl from nitric acid solutions of lead by ammonium 12-molybdophosphate fixed in the matrix of porous Teflon (AMP-sorbent) has been investigated. Effective separation of Tl and Pb is shown to take place. Elution processes of Tl from AMP-sorbent have been investigated. It is found that complete extraction of Tl is achieved upon dissolving AMP in concentrated ammonia. Further purification and concentration of Tl are performed by means of cation exchange chromatography on Dowex-50 or KU-2 resins. Investigation of high temperature behaviour of Tl ultramicroamounts in Pb melt showed that Tl is quantitatively separated out into the gas phase when fluorinating additions of  $PbF_2$  or NaF solid salts covering the melt surface are used. The volatile compounds of radiothallium formed were transported by a stream of inert gas (He or  $N_2$ ) from the evaporation zone to the thermochromatographic column, where they were sorbed on the surface in a limited zone with the maximum at  $\sim 240^\circ C$ . Liquid and gas thermochromatographic methods for separation of carrier free radiothallium from proton-irradiated lead material have been developed. The radiochemical, chemical and radionuclidic purity of Tl samples complies with requirements of nuclear medicine for  $^{201}Tl$  product. Both methods ensure  $\sim 95\%$  chemical yield of Tl and take about two hours each.

### Introduction

Methods of  $^{201}Tl$  radiochemical separation including processes of coprecipitation, ion exchange, extraction have been considered in Reference 1. With targets of enriched thallium<sup>2,3</sup> a two-stage procedure is performed: first radioactive Pb fraction (thoroughly purified from stable Tl admixtures) is separated, then, after a decay time of 30-40 hours during which an optimum growth of  $^{201}Tl$  from  $^{201}Pb$ , radiothallium is separated from lead.

Lately some studies<sup>4-7</sup> have shown advantages of a recently proposed method for production of  $^{201}Tl$  from Pb target irradiated with protons of energy  $\leq 100$  MeV in reaction



In this case Tl ultramicroamounts were separated from the Pb target material through a one-stage procedure by means of some selective methods, e.g. by lead bromide precipitation and adsorption of anion halide complexes Tl(III) on cation-exchange resins,<sup>4,6</sup> or by sublimation of volatile thallium fluorides and oxides.<sup>8,9</sup>

In view of further progress in work on production of  $^{201}\text{Tl}$  from proton-irradiated Pb material the development of new selective methods for separation of radiothallium from massive lead targets (tens of grams) becomes of current importance. Investigations in two promising directions were proposed. Firstly, to extract Tl from relatively large volumes of lead-saturated nitric acid solutions, produced by dissolving irradiated Pb, one can employ cation exchange on salts of heteropolyacids. Due to their selectivity to singly charged ions of heavy metals,<sup>10</sup> one could expect effective separation of Pb grams and Tl ultramicroamounts with minimal losses of Tl ( $\leq 5\%$ ), which was impossible in the system  $[\text{TlBr}_4]^- - \text{PbBr}_2$ .<sup>4</sup> Secondly, it is possible to employ high-temperature separation of volatile Tl compounds from Pb, followed by gas thermochromatography of volatile products.<sup>8,9</sup> The advantage of this method, is that it allows eliminating target material destruction; due to this, the amount of work with highly active materials is reduced, e.g. no Pb regeneration is required for enriched targets.<sup>6,7</sup>

Hence the present investigation is aimed at: (1) studying sorption and desorption of Tl ultramicroquantities in nitric acid solutions when ammonium 12-molybdophosphate is used as a sorbent, and at (2) studying high temperature sublimation of Tl from molten Pb followed by gas thermochromatography of Tl in an open quartz column. The results obtained became a basis of methods developed for radiothallium separation from proton-irradiated lead targets.

## Experimental

### *Separation of radiothallium from lead with ammonium 12-molybdophosphate*

**Sorbent.** The use of selective inorganic cation exchangers, containing insoluble and fine particles of heteropolyacid salts, is somewhat restricted in radiochemical processes because of difficulties related mainly to poor flowing of solutions through sorbents, except those based on porous Teflon with inorganic ion-exchangers firmly fixed on them.<sup>11</sup> One can pass solutions as fast as several tens of  $\text{ml} \cdot \text{cm}^{-2} \cdot \text{min}^{-1}$  through columns with a sorbent like this. We used a sorbent with 9.5% of ammonium 12-molybdophosphate (AMP) in the Teflon matrix.

**Thallium solutions.** Solutions of pure carrier free Tl(I) in 0.1 mol/l  $\text{HNO}_3$  and nitric acid solutions of irradiated Pb containing a complex mixture of radionuclides,

along with Tl radionuclides, were taken as primary solutions. Working solutions were prepared with different concentrations of  $\text{HNO}_3$  and  $\text{Pb}(\text{NO}_3)_2$ . A carrier Tl solution ( $\text{TlNO}_3$  in 0.1 mol/l  $\text{HNO}_3$ ) with a known concentration determined as  $\text{Tl}_2\text{CrO}_4$  was employed<sup>1,2</sup> to find the exchange capacity of AMP sorbent.

*Determination of AMP-Tl(I) exchange capacity.* A solution of  $\text{TlNO}_3$  of known concentration (0.4619 mg/ml; 4 mol/l  $\text{HNO}_3$ ) enriched in  $^{201},^{202}\text{Tl}$  was passed through columns with known amounts of the sorbent at elution rate of 0.5 ml/min. The activity and concentration of Tl in 1 ml eluent fractions were determined.

*Determination of chemical admixtures in thallium samples.* The contents of inactive Mo, Pb and P admixtures in Tl samples obtained at the final stage of Tl separation from lead were determined by colourimetric methods.<sup>1,3,14,15</sup>

*Measurements of samples activity.* The gamma-spectrometric method was used to determine the activity of samples from intensity of gamma-quanta of the energy: 167 keV ( $^{201}\text{Tl}$ ), 267 and 1205 keV ( $^{200}\text{Tl}$ ), 439 keV ( $^{202}\text{Tl}$ ), 278 keV ( $^{203}\text{Pb}$ ), 183 keV ( $^{206}\text{Bi}$ ).<sup>6</sup> Besides, in some experiments the relative activity of Tl samples was measured by a single-channel analyser type 20626 (Messelektronik, GDR) with a NaI detector.

*Sorption of thallium on the AMP-sorbent.* Sorption of carrier free Tl was investigated under static and dynamic conditions. In the first case 8 ml of the solution and 0.1 g of the sorbent was shaken in a 10 ml testing tube for 2 minutes. It was determined in advance that equilibrium of Tl sorption was achieved in 45 seconds. Under dynamic conditions, columns of 3 mm in diameter were used, changing the column length from 30 to 120 mm, which corresponded to variations in sorbent amount from 0.2 to 0.75 g. The column with the sorbent was pre-washed with 10 ml 1 mol/l  $\text{HNO}_3$ . After flowing of the Tl solution ended, the column was thoroughly washed with 1 mol/l  $\text{HNO}_3$  solution. Distribution of Tl between the solution and the sorbent was determined by its activity in the solution before and after elution.

*Results and discussion.* The capacity of the AMP-sorbent was calculated from the experimental results shown in Fig. 1. The mean value, 5.78 mg of Tl per 1 g of the sorbent, corresponds to 0.31 meq of Tl(I) per 1 g of  $(\text{NH}_4)_3\text{PMO}_{12}\text{O}_{10} \cdot 5\text{H}_2\text{O}$ , which was 20% of the theoretical value of 1.53 meq/g.

As can be seen in Fig. 2 sorption of carrier-free Tl on the AMP sorbent from nitric acid solutions, including those containing Pb, had a quantitative character with a distribution coefficient  $> 10^4$  in the  $\text{HNO}_3$  concentration range from 0.2 to 6 mol/l. Then it decreased as  $\text{HNO}_3$  concentration grew to 12 mol/l, which can be accounted for by competing sorption of Tl and hydrogen ions and production of Tl complexes with nitrate ions. Various concentrations of  $\text{Pb}(\text{NO}_3)_2$  in the solution did not affect Tl sorption (Table 1). The results of Table 2 show that completeness

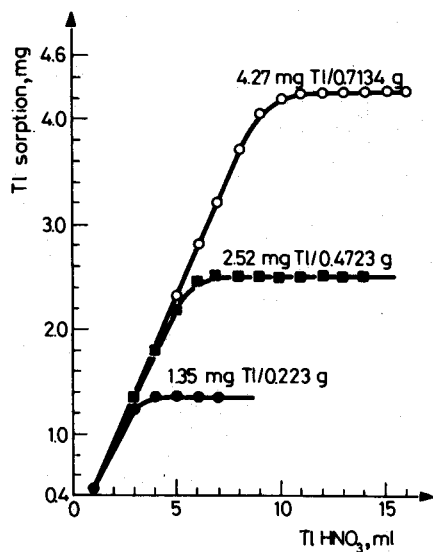


Fig. 1. Sorption of Tl on AMP sorbent:  $TlNO_3$  in 4 mol/l  $HNO_3$  (0.4619 mg/ml)

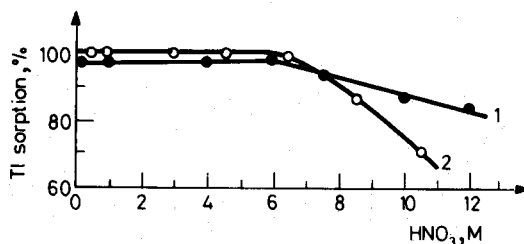


Fig. 2. Sorption of carrier free Tl by AMP sorbent; curve 1 –  $HNO_3$ , curve 2 –  $HNO_3$  + 28.9 g/l  $Pb(NO_3)_2$  (static conditions)

Table 1  
Influence of  $Pb(NO_3)_2$  amount on Tl sorption; AMP 9.5 mg, 4 mol/l  $HNO_3$  8 ml, mixing time 2 min, 20 °C

$Pb(NO_3)_2$ , g/l	0	1.1	5.4	10.8	21.7	43.4	65.1	86.7
Tl sorption, %	97	98	99	99	97	97	98	98

Table 2  
Influence of AMP sorbent quantity and of solution  
flow rate in the column ( $\phi$  3 mm) on Tl sorption;  
10 ml of 4 mol/l  $\text{HNO}_3$ , 80 g/l of  $\text{Pb}(\text{NO}_3)_2$

Weight of sorbent, g	Height of column, cm	Flow rate, ml/min	Sorption, %
0.75	12	1.7	99
0.40	6	2.1	99
0.20	3	3.7	100

of Tl(I) extraction from nitric acid solutions by the AMP sorbent did not depend on the sorbent column length and on the elution rate. Quantitative sorption of Tl on the AMP sorbent is probably due to fast kinetics of the exchange processes and large distribution coefficients. In this case Pb was not extracted from nitric acid solutions by the AMP sorbent and the passing of active solutions through AMP columns thus ensured quantitative separation of radiothallium and Pb target material. The filtrate, which is a solution of  $\text{Pb}(\text{NO}_3)_2$  in  $\text{HNO}_3$ , is suited both for regeneration of enriched Pb targets, and for processing of its removal.

As is known,<sup>1,6</sup> sorbed Tl can be eluted from AMP with a solution of 0.1 mol/l  $\text{Br}_2$ —4 mol/l  $\text{HBr}$ , which oxidizes  $\text{Tl}^+$  to  $\text{Tl}^{3+}$ , producing a complex anion  $[\text{TlBr}_4]^-$ . However, use of both eluents and a neutral solution 0.1 mol/l  $\text{Br}_2$ —0.4 mol/l  $\text{KBr}$  for washing Tl out of the AMP column gave no satisfactory results (Table 3). The hydrophobic Teflon matrix probably hampers in some way the heterogeneous oxidation of Tl by bromine. This made us use a concentrated  $\text{NH}_4\text{OH}$  solution to dissolve AMP and wash it out with Tl from the column. Experiments showed that after washing 0.3 g of AMP-sorbent by 3—5 ml of 25%  $\text{NH}_4\text{OH}$  or by diluted  $\text{NaOH}$  solutions, the Teflon matrix remaining in the column was completely deactivated (Table 3).

It is well known that Tl(I) has strong alkaline properties, therefore, it is present in an ammonia solution of  $\text{pH} \sim 12$  in a cationic form. This makes it quite easy to extract Tl from the solution and purify it from molybdate and phosphate ions by means of cation-exchange resins like Dowex-50 or KU-2. According to our determination, the coefficient of Tl distribution between the resin and the ammonia solution is  $\sim 10^3$ . Thallium was easily eluted from the cation exchange column by  $\text{NaCl}$  solutions (Fig. 3).

The procedure of Tl separation, purification and concentration in this way allowed obtaining  $^{201}\text{TlCl}$  samples for nuclear medical applications in isotonic solution.

The results were used for developing a radiothallium production technique based

Table 3  
Influence of solution composition and flow rate through the column  
with AMP sorbent ( $\phi$  3 mm, AMP 0.3 g) on Tl yield from the column (%)

Solution	Flow rate, ml/min	Yield of Tl, %				
		No of solution fraction (2 ml each)				
		1	2	3	4	5
0.1 mol/l Br <sub>2</sub> in 4 mol/l HBr	0.2	34	14	4	2	2
	0.9	24	4	2	2	2
0.1 mol/l Br <sub>2</sub> in 5% KBr	0.2	35	3	2	2	2
	0.3	27	3	2	2	1
25% NH <sub>4</sub> OH*	0.7	86	1	1	0	0
	1.5	85	1	0	0	0
	3.3	70	11	1	1	0
NaOH:* 0.1 mol/l 0.5 mol/l 1.0 mol/l	2.0	0	3	76	5	1
	1.7	87	2	0	0	0
	2.0	90	4	1	0	0

\*According to the residual activity of the sorbent after washing it by solutions, the yield of Tl was  $\geq 99\%$ .

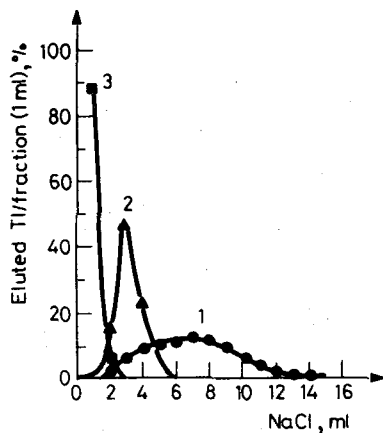


Fig. 3. Elution of Tl from the Dowex 50 x 4 column by NaCl solutions (%); curves: 1 - 0.9; 2 - 1.8; 3 - 6.6; rate of elution 1 ml/min.

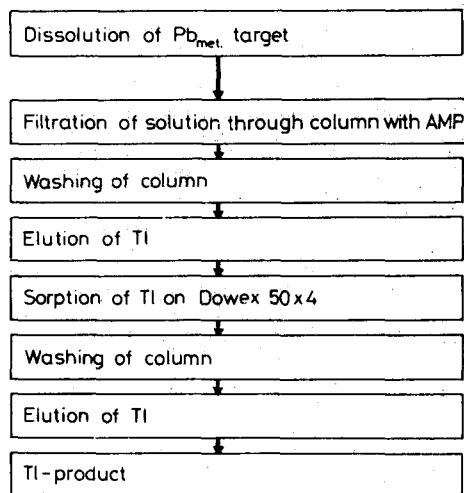


Fig. 4. Flow-sheet of radiothallium separation from irradiated Pb target by ion exchange processes

on separation of Tl from Pb on the AMP sorbent, followed by purification and concentration on cation-exchange resin (Fig. 4). Irradiated Pb target was dissolved in 4 mol/l  $\text{HNO}_3$  at 70–90 °C (25 ml/1 g Pb). The solution was cooled to room temperature and passed through a column 3 mm in diameter, packed with 0.3 g of AMP sorbent, at an elution rate of 3–4 ml/min. Traces of Pb and  $\text{HNO}_3$  were removed from the AMP column by washing with 10 ml of 1 mol/l  $\text{HNO}_3$  and then with 10 ml of  $\text{H}_2\text{O}$ . AMP (Tl\*) was dissolved in 10 ml of 25%  $\text{NH}_4\text{OH}$  solution while passing through the column at an elution rate 1–1.5 ml/min. Then radiothallium was sorbed from the ammonia solution in a column (3 mm in diameter and 50 mm in height) packed with Dowex 50 X 4 in the  $\text{NH}_4^+$ -form. The elution rate of ammonia solution through the cation exchange column was 1–2 ml/min. Then the resin was washed with 10 ml of  $\text{H}_2\text{O}$  and 5 ml of 0.09% NaCl at the same elution rate. Radiothallium was eluted with 5 ml of 1.8% NaCl. For a 10 g Pb target, the chemical yield of Tl was  $95 \pm 2\%$ , duration of the procedure did not exceed 2 hours.

Testing the technique with lead material irradiated by 680 MeV protons and containing, besides radiothallium, many other spallogenic radionuclides showed that the final TlCl product in 1.8% NaCl was of high radionuclide purity (Fig. 5). Thallium was present in the solution in the form of Tl(I), which was proved by its total adsorption on AMP sorbent. Radiochemical purity of the sample did not change for 2 weeks. This period is much longer than the application time set for pharmaceutical  $^{201}\text{TlCl}$  product (8 days). The content of inactive admixtures of Mo, Pb and P did not exceed 1  $\mu\text{g/ml}$ , which is close to the sensitivity level of the analytical





les of  $\text{PbF}_2$  in a quartz vessel (1) installed in a quartz tube (2), already heated to the necessary temperature. An inert carrier gas, purified from moisture and oxygen by traps (6–9) flowed through the tube at a rate of  $\sim 100 \text{ cm}^3/\text{min}$ . Gaseous products emanated from molten Pb into a quartz tube (3) 3 mm in diameter and 300 mm in length, which had a negative temperature gradient and served as a thermo-chromatographic column (TC). Distribution of radioactive products, deposited in various parts of TC, was determined by the gamma-spectrometric method.<sup>6</sup> A series of experiments was carried out in order to investigate the influence of volatilization time, temperature, Pb and  $\text{PbF}_2$  amounts and geometry of liquid lead upon escape of volatile trace Tl compounds from molten Pb. Several experiments dealt with investigations of radiothallium separation when  $\text{PbCl}_2$  and NaF are used as chlorinating and fluorinating agents and substitutes nitrogen and air for helium.

*Results and discussion.* Paper<sup>1,7</sup> reported on the separation of radiothallium from lead in dynamic vacuum with HF vapour as reactive gas. This study is aimed at solving another problem: to find conditions for radiothallium extraction from molten Pb with an inert gas (helium or nitrogen) flowing over the surface at a normal pressure. It is obvious that these conditions would simplify processing of a highly active target. The experiments showed that if there are many grams of irradiated Pb, the problem cannot be solved due to evaporation of Tl in the elemental form. Earlier there was a successful high-temperature separation of radiothallium as TlF from several hundred grams of  $\text{PbF}_2$ , irradiated with high-energy protons, in a nitrogen atmosphere.<sup>8</sup> However, the  $\text{PbF}_2$  target is not quite suitable for being irradiated by intensive proton beams. Therefore it was decided to use  $\text{PbF}_2$  only as a fluorinating agent with which radiothallium must react to produce TlF on the boundary between molten metal and solid salt. Halogenation of microadmixture by adding salts is used for analysis,<sup>1,8</sup> but never employed in preparative radiochemistry. Experiments with Pb samples irradiated with 100 and 680 MeV protons proved the assumption of effective influence of fluorinating additives upon behaviour of Tl ultramicroamounts at high temperature.

The results obtained by investigation of volatile Tl escape under different experimental conditions are shown in Fig. 7. Points on the curves show mean values for 3–6 determinations with maximum deviations. It is seen that Tl yield from molten Pb increases with temperature, time of volatilization, amount of  $\text{PbF}_2$  added, and decreases with increasing amounts of lead. Figure 8 shows that evaporation of Tl depends not only on the total amount of molten metal, but also on its geometrical dimensions: the higher the layer of the same surface area, the lower Tl yield in the gas phase within the same time. This agrees with our assumption on Tl fluorination at the phase boundary. Comparison of results in Fig. 7 reveals an identical behaviour of Tl in an atmosphere of inert gases ( $\text{He}$ ,  $\text{N}_2$ ) and its noticeably slower

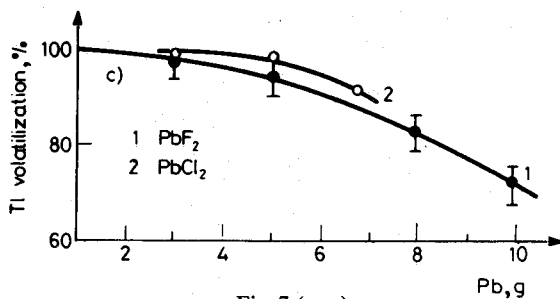
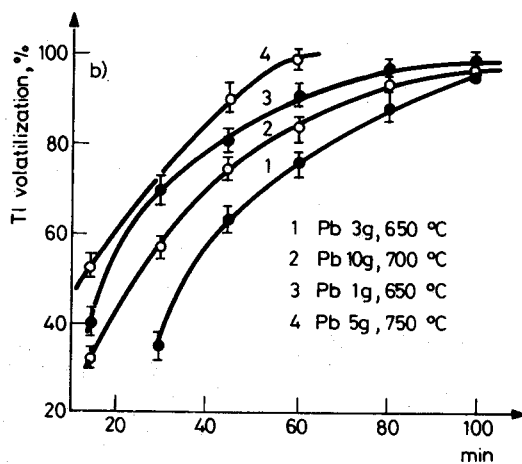
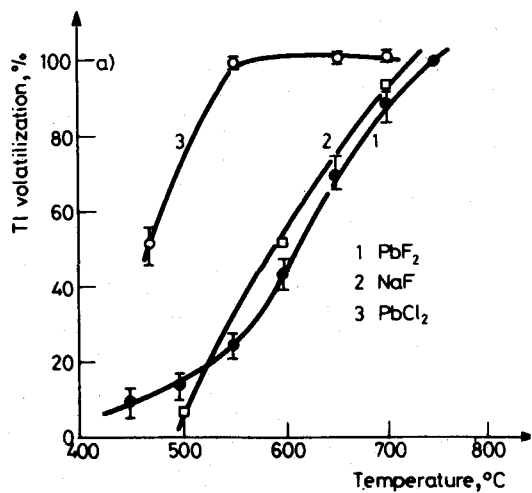


Fig. 7 (a-c)

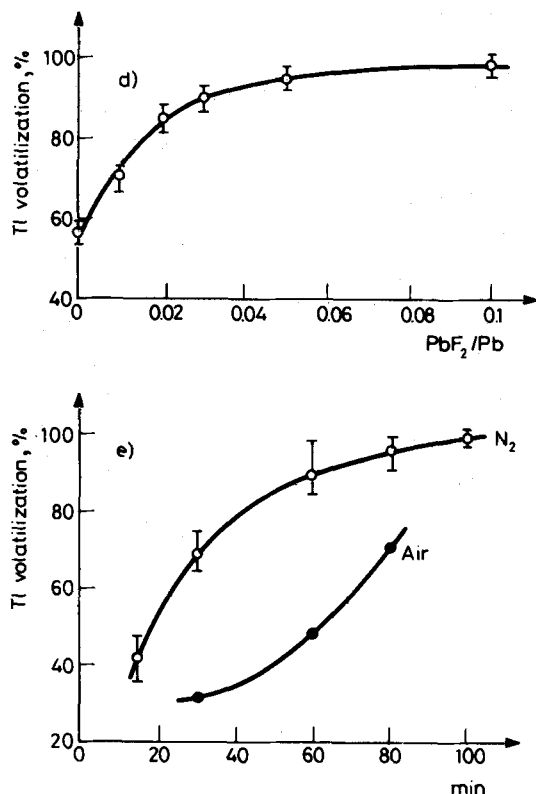


Fig. 7. Dependence of Tl volatilization from molten Pb on temperature (a), time (b), amounts of Pb (c) and  $PbF_2$  (d), carrier gas (e). a) 5 g Pb +  $PbF_2$  (curve 1), 3 g Pb + NaF (curve 2), 3 g Pb +  $PbCl_2$  (curve 3);  $PbF_2$  (NaF,  $PbCl_2$ ): Pb = 0.1; time 60 min, helium carrier gas; b)  $PbF_2$  : Pb = 0.1; c) Pb +  $PbF_2$ , 700 °C (curve 1); Pb +  $PbCl_2$ , 650 °C (curve 2);  $PbF_2$  ( $PbCl_2$ ) : Pb = 0.1, sublimation time 60 min; d) 3 g Pb, 700 °C, sublimation time 60 min; e) 3 g Pb, 700 °C,  $PbF_2$  : Pb = 0.1

escape into the air, which can be caused by production of PbO or the PbO –  $PbF_2$  system.<sup>8</sup>

The results show that effective evaporation of Tl from molten Pb takes place at 700–750 °C with both  $PbF_2$  ( $t_{melt} = 822$  °C) and NaF ( $t_{melt} = 996$  °C) as fluorinating agents. Quantitative evaporation of Tl was also obtained in the Pb- $PbCl_2$  system (Fig. 7a, b). Due to the higher pressure of  $PbCl_2$  vapours (than that of  $PbF_2$ ) some part of  $PbCl_2$  is carried from the high-temperature zone by a gas flow and contaminates TC, including the Tl adsorption zone.

On the basis of known general ideas, one can assume that the process leading to escape of volatile Tl ultramicroquantities from molten Pb unites several individual

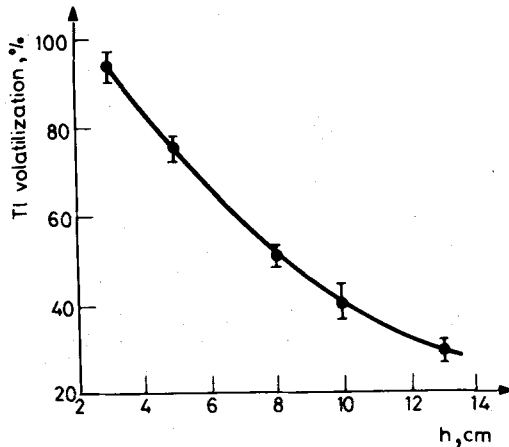


Fig. 8. Volatilization of Tl from molten Pb as a function of the layer height, 700 °C, sublimation time 60 min, diameter of the quartz crucible 8 mm

processes, including diffusion of Tl atoms in liquid Pb, surface reaction and evaporation of products from the phase boundary surface. Results, obtained for the escape of volatile Tl into different carrier gases (He, N<sub>2</sub>, air), and for mass and height of molten Pb layer allow regarding them as decisive factors. As far as diffusion of Tl atoms as admixture to Pb is concerned, their abnormally high diffusion mobility is known,<sup>19</sup> and one can assume that carrying Tl to the surface of molten metal will be a fast process.

Deposition of Tl vapours on TC walls occurred in a relatively narrow temperature zone with a half-width of ~ 1 cm and a maximum at ~ 240 °C (Fig. 9). The gamma-spectrometric analysis showed that there were no admixtures of other radio-nuclides in this zone.

The results show the possibility of effective use of PbF<sub>2</sub> or NaF additions for fluorinating carrier-free Tl and separating its ultramicroamounts from irradiated Pb at 700–750 °C, followed by gas-thermochromatographic purification and concentration. Based on these data, Figure 10 schematically illustrates the method of Tl separation from Pb target. Noteworthy is that the target mass will depend on irradiation conditions: it will not exceed 10 g of lead at  $E_p \leq 100$  MeV,<sup>6</sup> and it can be an order of magnitude larger at  $E_p = 680$  MeV. Amount of lead does not change the Tl separation technique in principle; it only affects geometric dimensions of the high-temperature part of the apparatus and of the crucible for melting Pb.

When Pb targets (about 10 g) with 1 g of PbF<sub>2</sub> were processed, practically all of the thallium produced was carried out from the high-temperature zone in the TC by

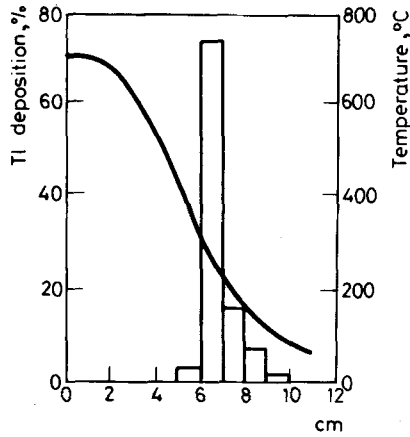


Fig. 9. Distribution of Tl along the column; 6 g of Pb, irradiated with protons of  $E_p = 680$  MeV;  $PbF_2 : Pb = 0.1$ ;  $700^\circ C$ , sublimation time 90 min

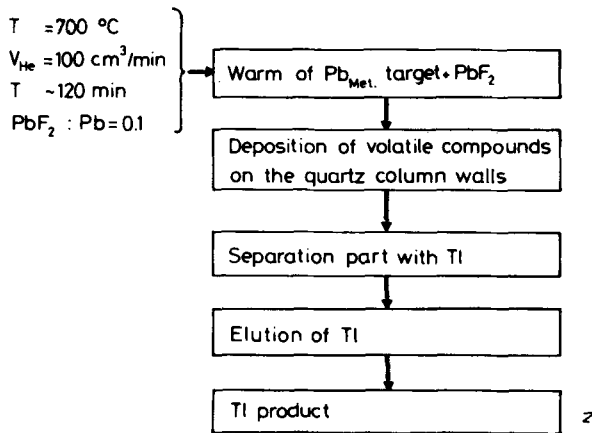


Fig. 10. Flow-sheet of radiothallium separation from irradiated Pb target by the gas-thermo-chromatography method

the inert gas flow ( $\sim 100 \text{ cm}^3/\text{min}$ ) within 1.5–2 hours. In the quartz TC, Tl was sorbed in the  $240 \pm 20^\circ C$  temperature zone and then was completely washed off the tube walls by a 1.8% solution of NaCl. Contamination of this solution by Pb was  $\leq 1 \mu\text{g}/\text{ml}$ . Radiothallium had high radionuclidic and radiochemical purity [ $\geq 99\%$  Tl(I)].

The gas-thermochromatographic method for radiothallium production from irra-

diated lead with solid fluorides as fluorinating agents and inert gases used for carrying TlF to TC is relatively more complicated than the liquid method with AMP sorbent, if equipment behind the biological protection is considered. With a proper design, however, it seems to be more suitable, especially for targets of large masses. First of all, it is due to localization of the greater part of radioactive products in Pb and PbF<sub>2</sub> (during the whole process) and, consequently, due to better prevention of accidental contamination in the laboratory.

### Conclusion

The investigations yielded new information about adsorption of Tl ultramicroquantities from lead nitrate solutions by ammonium 12-molybdophosphate fixed in the matrix of porous Teflon (AMP sorbent). Effective separation of Tl and Pb is shown to take place during adsorption. Processes of Tl washing out of AMP sorbent have been studied; it is found that the most effective way is to dissolve AMP in a concentrated aqueous solution of ammonia. Then thallium is easily purified and concentrated from the ammonia solution via cation-exchange chromatography on Dowex-50 or KU-2 resins.

Investigation of carrier free Tl in molten Pb has shown that radiothallium can be quantitatively separated as gaseous TlF when solid salts of PbF<sub>2</sub> or NaF, covering the surface of the molten metal, are used as fluorinating agents. Radiothallium vapour is carried by the He or N<sub>2</sub> flow from the evaporation zone to the thermochromatographic column, where it forms a limited adsorption zone with a maximum at ~ 240 °C.

It is shown that radiothallium, separated, purified and concentrated by the liquid and gas methods, is characterized by radionuclide, radiochemical and chemical purity which complies with requirements for <sup>201</sup>TlCl samples. Both methods ensure chemical yields of ~ 95%, and each takes about two hours.

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