DTU Library

Separation of ^{99m}Tc from ^{99}Mo0$_3$. A high performance sublimation generator

Tomicic, M.

Publication date:
1977

Document Version
Publisher's PDF, also known as Version of record

[Link back to DTU Orbit](#)

Citation (APA):
Tomicic, M. (1977). *Separation of ^{99m}Tc from ^{99}Mo0$_3$. A high performance sublimation generator*. Risø National Laboratory. Risø-M No. 1943

Risø-M-1943

Title and author(s)	Date 26th July 1977
Separation of ^{99m}Tc from $^{99}MoO_3$. A high performance sublimation generator by M. Tomicic	**Department or group** Isotope Division
	Group's own registration number(s)

20 pages + 7 tables + 3 illustrations

Abstract

At the present time ^{99m}Tc is widely used in nuclear medicine and its uses are increasing. It can be produced by various methods, and of those most frequently used today two have special features making them particularly applicable to the large-scale production of instant ^{99m}Tc - these are solvent extraction with methyl-ethyl--ketone and the sublimation methods.

This report presents a bibliographic review of these methods, their main performance parameters, and experience obtained from the development and operation of a sublimation generator.

Separation of ^{99m}Tc from irradiated MoO_3 was carried out with high yields (75-95%) after multiple repetition of the separation process, with molybdenum trioxide heated for half an hour at a maximum temperature of 850-890°C in an air flow. The activity ratio of molybdenum in the separated ^{99m}Tc was of the order of 4×10^{-5}.

Copies to

Library 100
Author 50
Isotop-Apoteket 25

Summary

At the present time ^{99m}Tc is widely used in nuclear medicine and its uses are increasing. It can be produced by various methods, and of those most frequently used today two have special features making them particularly applicable to the large-scale production of instant ^{99m}Tc - these are solvent extraction with methyl-ethyl-ketone and the sublimation methods.

This report presents a bibliographic review of these methods, their main performance parameters, and experience obtained from the development and operation of a sublimation generator.

Separation of ^{99m}Tc from irradiated MoO_3 was carried out with high yields (75-95%) after multiple repetition of the separation process, with molybdenum trioxide heated for half an hour at a maximum temperature of 850-890°C in an air flow. The activity ratio of molybdenum in the separated ^{99m}Tc was of the order of 4×10^{-5}.

Contents

INTRODUCTION

Technetium-99m is one of the most versatile radionuclides used in nuclear medicine at the present time. It possesses a combination of excellent physical characteristics that make feasible the administration of millicurie amounts, resulting in improved information and more rapid procedures, with a relatively low radiation dosage. Furthermore, when it is in an appropriate chemical state, it can easily be incorporated into a great variety of pharmaceuticals of biological interest.

Technetium-99m is a decay product of ^{99}Mo, which is usually obtained either by the nuclear reaction $^{98}Mo(n,\gamma)^{99}Mo$ with thermal neutrons, or as a fission product of ^{235}U.

Three general methods of separating ^{99m}Tc from ^{99}Mo are in current use:

a)   Elution from ^{99}Mo adsorbed on a column filled with suitable material. There are several materials reported in the literature as suitable for such separation (1). The most commonly used adsorbent is alumina, on to which the ^{99}Mo is loaded, whereafter the ^{99m}Tc is eluted by an appropriate solution, generally physiological saline. The low capacity of alumina for Mo (2, 3) makes it necessary to use ^{99}Mo of high specific activity, which is only obtainable by irradiation enriched ^{98}Mo in a high neutron flux, or as a fission product.

b)   Extraction of ^{99m}Tc in an appropriate organic solvent. Different solvents have been studied (4, 5); those most frequently used are methyl-ethyl-ketone, acetone (6), methyl-isobutyl-ketone (7) and tri-isooctylamine (8). After the ^{99m}Tc extraction in an organic solvent, it is necessary to evaporate the solvent to dryness and redissolve the residue in the appropriate solution.

c)   Sublimation of ^{99m}Tc from $^{99}Mo-MoO_3$. This method is based on the work of C. Perrier and E. Segré (9), who found that a difference in the volatilities of the oxides of technetium and molybdenum could be used to separate them. The method has been developed commercially at Lucas Heights, Australia, to obtain ^{99m}Tc in multicurie amounts (10).

These three methods produce ^{99m}Tc of the quality required for medical applications, but the last two have additional ad-

vantages that make them very attractive from the point of view of a centralized production of "instant ^{99m}Tc". The main performance parameters of these two methods are briefly summarised.

The extraction of 99^{m}Tc by methyl-ethyl-ketone (M.E.K.)

Based on studies reported by Gerlit (5) on the chemistry of technetium, a method has been developed for separating ^{99m}Tc from ^{99}Mo. The distribution coefficients, reported by Gerlit, from a 5M NaOH solution in M.E.K., are 49 and 0.2, respectively.

Richards (11) investigated a number of variables involved in the extraction of ^{99m}Tc by M.E.K. He found that the extraction of ^{99m}TcO$_4$ is excellent in the range of NaOH molarities from 1 to 7. However, at low molarities the extraction of molybdenum is increased. He also found no appreciable drop in yield until the volume of the organic solvent is less than one half that of the aqueous phase. The major problem in the separation was the low yield from the first extraction when the solution is more radioactive. Richards states that the pertechnetate appears to be readily reduced to a lower, non-extractable state either directly by radiolysis or by a radiolysis product. The addition of H$_2$O$_2$ greatly improves but does not completely correct the situation. Frequently, however, the yield from the first extraction is low even with the addition of peroxide. This author stresses the use of high quality, freshly destilled methyl-ethyl-ketone, and a low evaporation temperature because decomposition products present in the ketone, or produced by overheating, can reduce pertechnetate.

Procedures

Several systems have been reported for carrying out a good separation of ^{99m}Tc from ^{99}Mo with methyl-ethyl-ketone (10, 11-18). All are basically the same and only differ in the sophistication of the apparatus.

The method consists of mixing a solution of molybdate in 5 or 6 M NaOH with freshly destilled methyl-ethyl-ketone. The use of KOH instead of NaOH has been suggested (10, 12) because the solubility of potassium molybdate is very much higher than the corresponding sodium salt, and the use of KOH permits an increase in the capacity of the apparatus and allows the use of a more concentrated solution without the risk of blockages through

crystallisation. Recently, Constant et al. (15) reported the use of a K_2CO_3 2,5 M solution.

After mixing, the less dense methyl-ethyl-ketone layer containing the pertechnetate is passed through a dry alumina column to remove traces of base, and then evaporated to dryness at a relatively low temperature (45-55°C). The carrier-free ^{99m}Tc is taken up in a sterile physiological saline solution. The methyl-ethyl-ketone distillates are re-destilled and re-used. The time required for the process is about 30 min (12, 17).

Most of the apparatus illustrated in the literature was designed for small-scale operation, except a stainless steel apparatus, scaled-up for amounts of 200 Ci, that was developed by the Atomic Energy Commission in Australia (10).

Separation efficiency

All systems reported in the literature (10, 12-20) for the production of ^{99m}Tc by extraction with methyl-ethyl-ketone have high yields. The values are between 75-95%, although some authors have pointed out certain anomalies regarding the separation efficiency. Richards (11) found a low yield particularly from the first extraction, as mentioned earlier, and Baker (12) found an average yield of 63% when using low specific activity molybdate ($\sim$ 0.4-0.6 g Mo/ml). However, Belkas and Perricos (14) studied the influence of the amount of inactive molybdenum in the extraction of technetium and concluded that it is of no significance in the range of concentrations studied (0.13-1.3 g Mo/ml). The time of mixing is not considered critical and good yields may be obtained in 3-5 min (11, 14).

Radionuclidic purity

The radioactive impurities found in a ^{99m}Tc sample depend, of course, on the method used to obtain it. In any case, the principal radionuclidic impurity that can be expected is ^{99}Mo. The acceptable limit for ^{99}Mo in radiopharmaceutical quality ^{99m}Tc preparations is 1 µCi of ^{99}Mo per 1 mCi of ^{99m}Tc, with a maximum of 5 µCi of ^{99}Mo per patient dose (21). The ^{99}Mo content in samples of ^{99m}Tc extracted by methyl-ethyl-ketone is more than 100 times lower than this recomended level (10, 12, 16).

Some authors (22, 23) also found ^{186}Re and ^{124}Sb in the

extracts, but in very low concentrations, and the extra radiation dose to the patient from these impurities is minimal at the time of extraction, being of the order of 0.1 per cent of the total body radiation dose from ^{99m}Tc(24).

Chemical purity

The concentration of aluminium (III) in the eluates when alumina is used to dry the methyl-ethyl-ketone is less than 1 µg/ml (17). Colorimetric analysis (10) of ^{99m}Tc solutions obtained by extraction with methyl-ethyl-ketone in a stainless steel apparatus gave Mo < 50 ppm, heavy metals < 10 ppm, and Fe < 5 ppm. Slight traces of residual ketone have been observed, < 10^{-3} mM/ml (15).

Sublimation of ^{99m}Tc

The method developed at Lucas Heights consists of passing a stream of oxygen through a tube furnace packed with MoO_3 powder (150 g, 200 Ci). The oven is heated to a suitable temperature for the ^{99m}Tc release and the vapour passed through a filter that removes entrained molybdenum trioxide. The vapour then condenses in the cold parts of the outlet tube; the carrier gas leaves the apparatus through a small vial containing a sterile saline solution. After the furnace has cooled down, the outlet tube is flushed with the solution using a suck/blow syringe and the washings are finally returned to the vial. The whole operation takes approximately 60 minutes (10).

Another sublimation generator is described in the literature (25). Here Na_2MoO_4 is used as initial product and the generator consists of a tubular element with a sintered glass disk for the reception of the sample and a heating device surrounding it. A stream of air is passed through the tube and the sublimation product is collected in the cooler part of the apparatus, or in a special collection container from where it can be dissolved under sterile conditions.

Separation efficiency

The separation efficiency of the sublimation generator is low, around 25-30 per cent, and careful control of the temperature is necessary to maintain adequate yields (10).

Recently, V. Machàu, J. Vlcek, V. Rusek, et al. (27) studied the possibilities of separating ^{99m}Tc from ^{99}Mo with high yield from repeated separations without using any carrier gas.

Separation of ^{99m}Tc from MoO_3 powder has been performed with a yield higher than 80% upon heating for 30 min at $800^{\circ}C$ in air (27). The authors in question studied the release of ^{99m}Tc from neutron-irradiated MoO_3 powder in the range of 480 ∿ $700^{\circ}C$.

Radionuclidic purity

Boyd (10) reported that radionuclidic purity of sublimated ^{99m}Tc varies both with the age and with the temperature of sublimation. He found very low contamination by ^{99}Mo and ^{188}Re, the latter being the major impurity in a fresh sample at the first separation temperature. His experimental results indicate that there is a marked increase in the vapour pressure of molybdenum trioxide as the temperature is increased. The greatest part of the sublimated ^{99}Mo does not dissolve in saline solution and can be removed by filtration up to a value < 10^{-4}%. The level of ^{188}Re contamination is not influenced by the temperature of sublimation and decreases with the age of the generator approximately parallel to its decay line.

Recently, Colombetti et al. (26) measured the principal radionuclidic impurities in ^{99m}Tc sublimated from fission product molybdenum. They found ^{131}I as the principal impurity with ^{99}Mo, ^{103}Ru, ^{106}Ru and ^{132}Te at very low levels, and these impurities were reduced in any successive sublimation. The authors point out that the contribution from these impurities to the total dose is negligible.

Chemical purity

The chemical purity of the filtered ^{99m}Tc solution obtained by sublimation of analytical grade reagent molybdenum trioxide, containing a very low proportion of volatile material, is very high. It is rare to observe any positive result and in all cases the levels do not exceed 1 µg Mo per mCi ^{99m}Tc, or 0.1 µg heavy metal per mCi ^{99m}Tc (10).

Both methods are good enough to produce ^{99m}Tc on a routine basis and of the quality required for medical application. The simplicity of the equipment and the fewer operations involved in the sublimation method make it very attractive for routine pro-
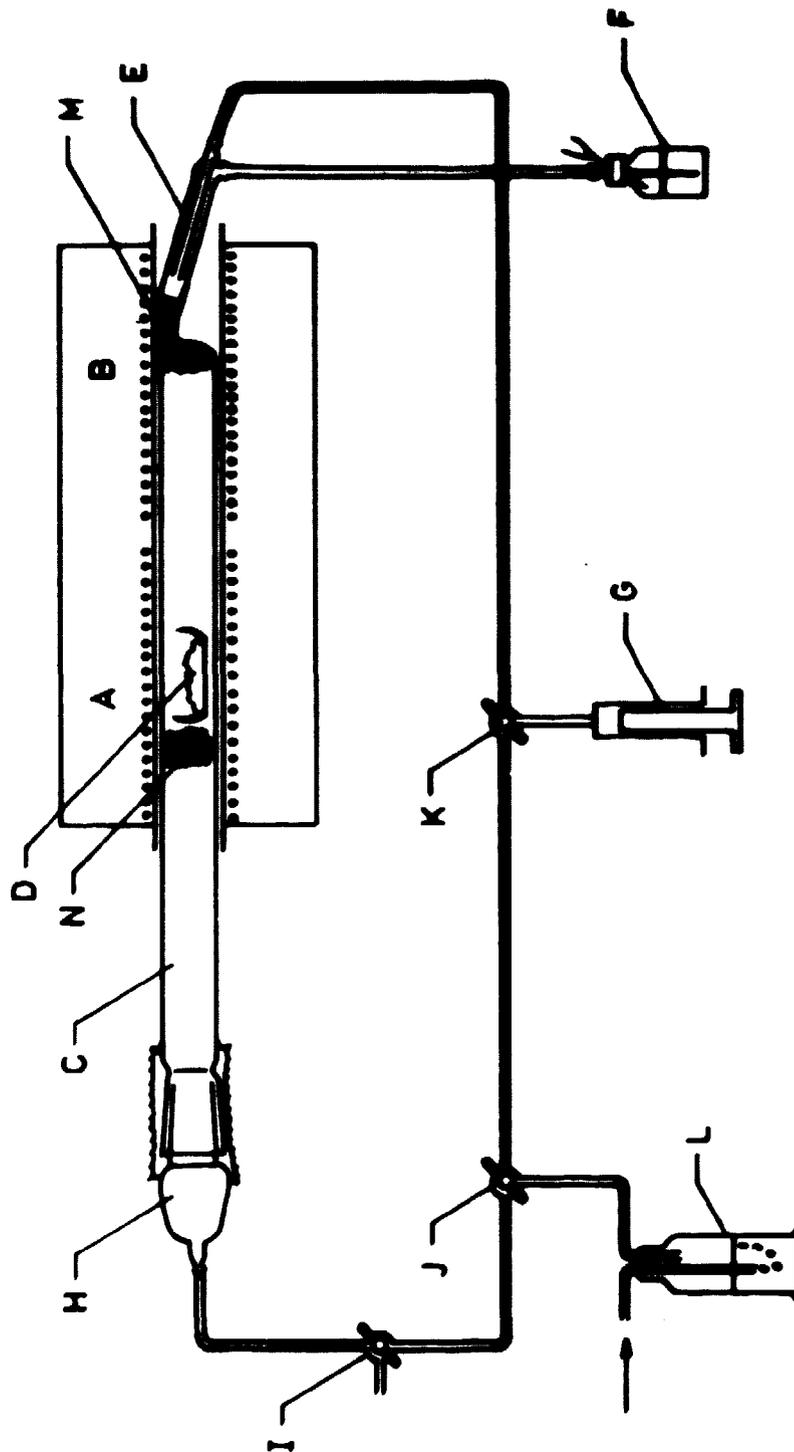
Fig. 1. Equipment used for the experimental preparation of
^{99m}Tc by sublimation from irradiated MoO_3 (D) in the electric
furnace (A,B).

duction. Furthermore, any breakdown of equipment during operation will not result in a significant loss of the parent material, as in the case of the wet method. Also in the dry method the ^{99m}Tc is dissolved directly in the saline solution thus avoiding possible problems arising from use of methyl-ethyl-ketone.

This report describes experience obtained from separation of ^{99m}Tc from irradiated MoO_3 by sublimation, with special emphasis on obtaining high and constant yields with each successive separation.

EXPERIMENTAL WORK

Molybdenum trioxide (Merck, p.a.) was irradiated in the R 1 facility of the Danish Reactor DR 2. A typical irradiation consisted of five grams of molybdenum trioxide dried at 120^oC, irradiated in a 60 x 16 mm^o polyethylene vial for two hours in a thermal neutron flux of 8 x 10^{12} n/cm^2 s.

The sublimation equipment is shown in fig. (1). It is a modified version of that patented and used at Lucas Heights, Australia. The main differences lie in the kind of furnace used and the method of sublimation, which results in different yields obtained. The home-made furnace consists of an alumina tube (31 cm long, 5 cm diameter) on which two independent heating zones (A, B) each of about 14 cm were made by winding with heating coils (Kanthal DSD, 0.50 mm^o, 7.18 Ω/m). The core is covered by a layer of refractory cement, insulated with stone fiber (Rockwool), and finally wrapped in asbestos cloth. The temperature in each zone is controlled with a variable transformer; the temperature gradient on the central axis of the furnace, with only one zone heated (A or B), is shown in figures (2) and (3).

The quartz tube (C) is 50 cm long, 3.5 cm in diameter, and has a ground quartz joint (H) at one end to which the carrier gas supply is connected. The opposite end finishes in a narrower part (E) (10 cm long, 1 cm diameter) inclined at an oblique angle to the main part of the tube and containing an inner silica tube (0.5 cm diameter, 9 cm long). The inner tube is connected by PVC tubing to a syringe (G), the outer part is connected through PVC tubing and a hypodermic needle to a small rubber--capped vial (F) that is vented through a similar needle.
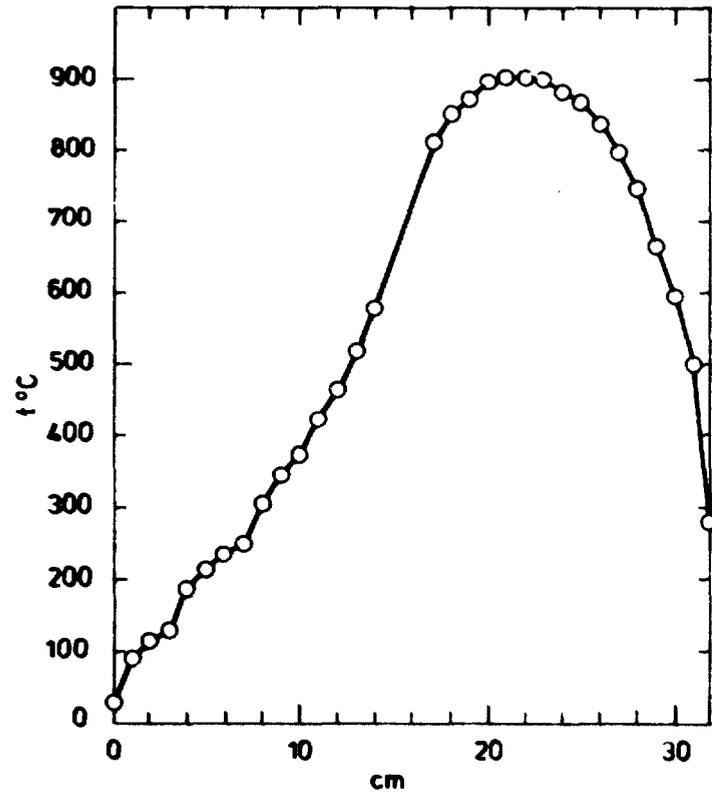
Fig. 3. Heating zone B. Temperature variation along the central axis of the furnace.
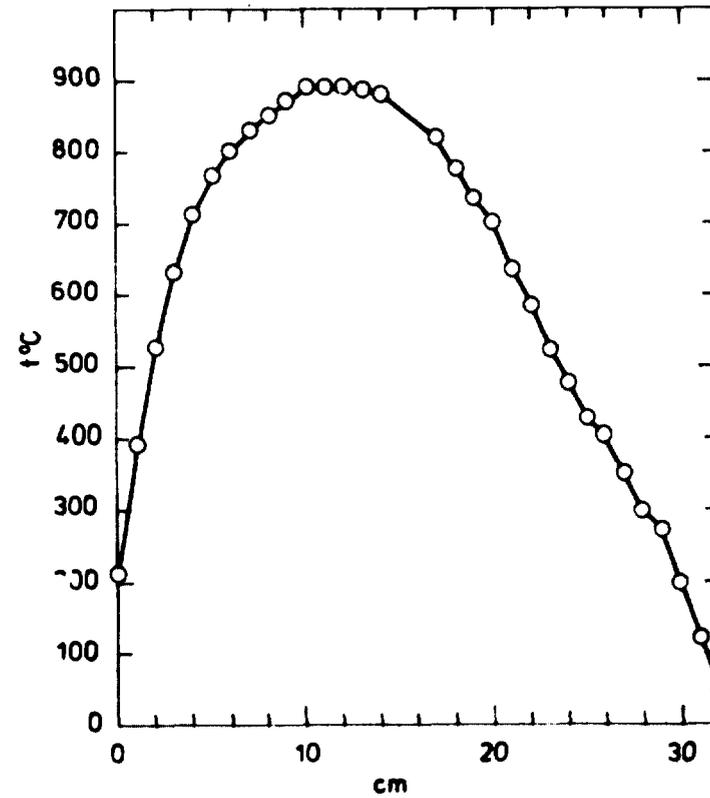

Fig. 2. Heating zone A. Temperature variation along the central axis of the furnace.

The carrier gas is introduced into the system through a concentrated sulphuric acid trap (L) and enters at one end or the other by means of three-way stopcocks (I), (J), (K).

After the molybdenum trioxide was irradiated, it was left for at least 48 hours to achieve equilibrium with ^{99m}Tc, then it was measured in an ionization chamber (NPL Type 1383A) and transferred to a small quartz boat. The quartz boat (D) was placed in the quartz tube in the position of maximum temperature (heating zone A). The quartz tube had previously been charged with a quartz wool filter (M) and placed in such a position that two-thirds of the condenser part was held inside heating zone B. After the boat was placed in the correct position, a quartz wool plug (N) was inserted next to it.

A capped vial (F) filled with 8 ml saline solution was connected and stopcock K closed. By opening J and I, a slow stream of air was allowed to flow through the quartz tube, leaving it through the hypodermic needle in the vial.

Heating zone A was switched on with the appropriate voltage and when the temperature reached $800^{\circ}C$ (25-30 minutes) heating zone B was switched on; heating continued for a further half an hour until the maximum temperature, $850-890^{\circ}C$, was reached in zone A. The filter was maintained at about $600^{\circ}C$. The temperatures were measured with two Pt-Pt/Rh thermocouples placed between the quartz tube and the furnace wall. On completion of the process, the apparatus was allowed to cool for 5-10 min. and then partly removed from the furnace to allow an increase in the rate of cooling of the condenser. Once it was cold, the air supply was stopped and stopcock I was closed and K opened. The condenser was flushed two or three times with the saline solution and then with 8 ml of NaOH 0.1 M solution, by means of the syringe (G).

After all ^{99m}Tc had been removed from the condenser, this was washed with distilled water and the quartz tube moved to the original position. An air flow was introduced by means of stopcocks (J) and (K), passed through the condenser in the opposite direction, and vented through stopcock (I). The heating coil of zone B was swithched on and zone B heated at a maximum temperature of $860-890^{\circ}C$ for half an hour; the heating of zone A was maintained so that the temperature in the region where the boat was placed did not exceed $600^{\circ}C$. After heating, the furnace

was switched off and left to cool. After 20-25 minutes, the air flow was stopped and the equipment was then ready for a new sublimation once the maximum activity of ^{99m}Tc had been reached (24 hours).

The activity of ^{99m}Tc was measured in an ionization chamber. From the known initial amount of ^{99}Mo and the quantitative relationships between parent and daughter nuclides, the separation yields were calculated at the time when heating ceased. According to the decay scheme of ^{99}Mo, and by means of classical equations, it is possible to calculate the activity ratios at any given moment; when the system has reached transient equilibrium the ^{99m}Tc activity is 96.4% of the ^{99}Mo activity.

The ^{99}Mo content was checked using a 512-channel pulse height analyser provided with a 3" x 3" NaI(Tl) scintillation detector. The bottle sample, which had been measured initially in the ionization chamber, was placed in a lead container (0.66 cm thickness) and the molybdenum content assayed by detection of the 740 KeV gamma ray. This was made as a rough check to confirm that the activity measured with the ionization chamber was only ^{99m}Tc. Another check was made after the ^{99m}Tc activity had completely decayed.

RESULTS

Table I shows the yields obtained in the sublimations of ^{99m}Tc from irradiated MoO_3 repeated every twenty-four hours under the described experimental conditions. In this case, the MoO_3 sublimated from the boat and deposited in the filter region was not back-sublimated after the initial sublimation was completed.

TABLE I

Sublimation No.	^{99m}Tc yield (%).
1	85
2	12
3	19
4	24

As can be seen, the yields are very low and only the first sublimation gives a high yield.

Table II shows the yields of the theoretically available amount of ^{99m}Tc obtained when, after sublimation, the MoO_3 that had been condensed in the filter region was sublimated back to the zone in which the boat was situated. These results originate from five generators, on each of which five sublimations were made. As can be seen, the yields of ^{99m}Tc are drastically improved in comparison with the former results.

TABLE II

Sublimation No.	Yield of ^{99m}Tc in different sublimations (%)				
	A	B	C	D	E
1	93	70	78	71	100
2	87	81	90	78*	90
3	97	74	79	80	73*
4	94	86	90	85	78
5	82	75	85	72	70

* sublimation after 48 h, ‡ sublimation after 72 h,

⁞ sublimation after 96 h, ⁞ sublimation after 120 h.

A rather unexpected and surprising result was the small amount taken up by the saline solution after the first sublimation. Table III shows the per cent of the total amount of ^{99m}Tc obtained in the washing solution that was found in the NaCl 0.9% solution. A subsequent washing with NaOH 0.1 M was necessary to remove practically all the activity in the condenser. In the first sublimation of the generator, C, D, and E, after washing with the NaCl 0.9% solution, a second wash was made with 8 ml NaCl 0.9% solution containing 1 ml H_2O_2 30%. An additional amount of ^{99m}Tc was removed from the condenser and the per cent of the total amount obtained from these two washes is indicated in brackets in Table III.

TABLE III

Sublimation No.	% ^{99m}Tc removed by NaCl 0.9%				
	A	B	C	D	E*
1	41	43	51 (78)	47 (82)	24 (74)
2	99	85	98	91	78
3	92	91	83	96	92
4	93	88	99	99	95
5	93	92	98	92	94

*irradiated in a quartz ampoule

It is clearly seen that only in the first sublimation is the difference significant. It was considered that this behaviour could be due to impurities on the surface of the quartz tube, and to exclude this possibility and any others related to the equipment itself, the D run was made using the same equipment as the C run without any change other than the introduction of a new batch of irradiated ^{99}MoO$_3$. The results, as can be seen in Table III, were the same.

Another reason could have been impurities present in the MoO$_3$ used. To take account of this, several tests were performed; a different commercial type of MoO$_3$ (BDH Analar), a molybdenum trioxide purified by sublimation and an earlier irradiated molybdenum trioxide were used. In the case of the formerly irradiated MoO$_3$, the ^{99m}Tc sublimated in the last four sublimations was completely removed by the saline solution.

As shown in Table IV, the results obtained were similar to the former ones and the different conditions did not improve the amounts of ^{99m}Tc taken up in the saline solution.

TABLE IV

	^{99m}Tc dissolved in 0.9% NaCl (First sublimation)	^{99m}Tc Total yield (%)
MoO_3 BDH Analar	39	75
MoO_3 sublimated	47	71
MoO_3 re-used	44	90

Table V shows the per cent of ^{99m}Tc dissolved in the NaCl 0.9% solution when oxygen was used as a carrier gas in the first three sublimations and nitrogen in the fourth. In this experiment the results were similar to that obtained when air was used as a carrier gas, except for a significant difference when nitrogen was used. Only 6% more could be washed out with the NaOH 0.1 M solution, and it was necessary to wash repeatedly with nitric acid to remove all the activity from the condenser.

TABLE V

Sublimation No.	^{99m}Tc in the saline solution (%)	^{99m}Tc % Total yield
1 with O_2	38	92
2 - -	88	70
3 - -	96	89
4 with N_2	26	70

Table VI shows the results obtained in three sublimations with a batch irradiated in one of the FST-rigs of the Danish Reactor DR 3, with a thermal neutron flux of $1.5 \times 10^{14} n/cm^2$ for four hours. The initial activity of the molybdenum trioxide charged on the generator was 250 mCi.

TABLE VI

Sublimation No.	^{99m}Tc in the saline solution (%)	^{99m}Tc % Total yield
1	63	87
2	96	73
3	99	71

The same general behaviour can be seen, but in this case the amount dissolved by the NaCl 0.9% solution in the first sublimation was clearly higher than in the former results.

The existence of two technetium oxides, the volatile Tc_2O_7 and the relatively involatile TcO_2, has been firmly established (28). Technetium dioxide is stable in air at room temperature but is readily oxidized by oxygen or by heating in air to Tc_2O_7 (28, 29). Tetravalent technetium, as TcO_2, is oxidized by atmospheric oxygen in an alkaline medium to produce pertechnetate without the use of hydrogen peroxide (30). From observations of the deposition temperature on a quartz tube, Tachimori et al. (18) concluded that ^{99m}Tc was present in the form of ^{99m}Tc (IV) in the MoO_3 crystal lattice and that by heating it was released in the form of ^{99m}TcO_2$, which in air further oxidized to ^{99m}Tc_2O_7$. The sublimation effectuated with nitrogen seems to confirm that ^{99m}Tc is released as TcO_2. The amount dissolved in the saline solution, probably being Tc_2O_7, could be explained as ^{99m}TcO_2$ oxidized by the residual oxygen inside the equipment before the sublimation with nitrogen was carried out.

According to the results, the ^{99m}Tc in the condenser seemed, except in the first sublimation to be in the form of Tc_2O_7, which is completely dissolved by the saline solution. In the first sublimation, the ^{99m}Tc was probably in the form of Tc_2O_7 and TcO_2; the reason for this behaviour cannot be unambiguously established with the results obtained. We aimed to obtain high yields rather than to determine the chemical state of the technetium in the irradiated molybdenum trioxide, or the form that is released and deposited in the quartz tube.

A paper chromatographic analysis of the ^{99m}Tc dissolved in

the saline solution gave more than 98% perthechnetate, and reduced technetium was found in the o.1 M NaOH solution.

The ^{99}Mo content was assayed in almost every sublimation, and the ratio of the activities at separation time in one of the generators is shown in Table VII.

<u>Table VII</u>

Sublimation No.	Activity ratio ^{99}Mo/^{99m}Tc
1	4.2×10^{-5}
2	7.2×10^{-5}
3	1.4×10^{-5}
4	3.2×10^{-5}
5	5.5×10^{-5}

CONCLUSIONS

An attempt was made to use the sublimation method for separation of ^{99m}Tc from molybdenum trioxide with high yields on repeated separation. For this purpose, equipment was designed and built that permitted the irradiated molybdenum trioxide to be sublimated forward and backward in each separation. Between 70 and 99% of the theoretically available amount of ^{99m}Tc was separated when molybdenum trioxide was heated for one hour, in an air stream, from room temperature up to a maximum of 890°C.

The ^{99}Mo content of the separated ^{99m}Tc was well below the requirements for medical applications and of the order of 4 x 10^{-5}.

A peculiar phenomenon was noted in the first sublimation of ^{99m}Tc. The ^{99m}Tc deposited on the condenser seemed to be in two chemical states, presumably Tc_2O_7 and TcO_2, and less than 50% of the activity on the condenser was removed by the saline solution. No explanation can be offered for this behaviour at the present time. Any effect resulting from the molybdenum trioxide or from the quartz equipment was eliminated. A better result was achieved when a 250 mCi sublimation generator was

used; in this case, 63% of the activity on the condenser was washed out by the saline solution. More appropriate experiments must be designed and carried out in order to determine the cause of this behaviour.

According to experience from these sublimations, a better tube furnace should be designed that can be opened horizontally in two halves. This would allow work with tongs and also allow easier positioning of the quartz tube.

ACKNOWLEDGEMENTS

REFERENCES

1. Radioisotopes Production and Quality Control. Technical Report Series No. 128, 698-706, IAEA, 1971.

2. V. Rarnik, B. Gorenc and J. Novak, Atomproxis 13 (6), 258-9 (1967).

3. Mikheev, N.B., El-Garhy, M., Moustafa, Z., Atomproxis 10, 263 (1964).

4. Boyd, G.E., Larson Q.V., J. Phys. Chem. 64, 988 (1960).

5. Gerlit, J.B., Proc. Internat. Corp. Peaceful Uses of Atomic Energy 7, 145 (1956).

6. Kuzina, A.F., Tagil T.S., Zamoshnikora, N.N., Spytsyn, V.I., Dokl. Akad. Nawk. SSSR 145, 166 (1962).

7. Reichold, P., Wolf, P., Rep. HMI-B52,15 (1967).

8. Klofutar, C., Stular, V:, Krasoved, F., Z. Anal. Chem. 214, 27 (1967).

9. Perrier, C., Segré, E., J. Chem. Phys. 5, 712 (1937).

10. R.E. Boyd, Symposium on new developments in Radiopharmaceuticals and labelled compounds. IAEA Vol I, 3, (1973); Also English patent 1, 378, 557, 27. Dec. 1974.

11. Richards, P., Report BNL-12564 (1969).

12. Baker, R.J., Int. J. Appl. Radiat. Isotopes Vol. 22, 483 (1971).

13. Lathrop, K.A., Radiopharmaceuticals from generator produced radionuclides, Proceeding of a Symp. Vienna 11-15 May 1970.

14. Belkas, E.P., and Perricos, D.C., Radiochem. Acta 11, 56 (1969).

15. Charlier, R., Fallais, Z., Constant, R., Rapport I.R.E. 3/1973.

16. MEKTEC 99 Catalogue, California Radiochemicals, Inc.

17. Robinson, G.D., J. Nucl. Med. 12, 459 (1971).

18. Tachimori, S., Amano, H., Nakamura, H., J. Nucl. Sci. Technol. (Tokyo) Vol. 8, No. 7 357 (1971).

19. Mani, R.S. and Narasimhar D.V.S. Symposium on new develop-
    ments in Radiopharmaceuticals and labelled compounds.
    IAEA Vol I, 3 (1973).

20. Archundia, C. and Butement, F.D.S., Rev. Soc. Quim. Mex. 12
    (3) 99A (1968).

21. Claig Harris, C., Radiologic Clinics of North America Vol.
    VII No. 2, 1969.

22. Billinghurst, M.W., Groothedde, F., and Palser, R., J. Nucl.
    Med. 15, No. 4, 266 (1974).

23. Robinson, G.D., J. Nucl. Med. 13, No. 5, 318 (1972).

24. Colombetti, L., Barnes, W.E., Int. J. Appl. Radiat. Isotopes
    25, 455 (1974).

25. Smith, E.M., and Hupf, H.B., German Patent 2, 152, 794, 4.
    May 1972.

26. Colombetti, L.O., Husak, V. and Dvorak, V., Int. J. Appl.
    Radiat. Isotopes 25, 35 (1974).

27. Machan, V., Vlcek, J., Kokta, L., Rusek, V., Smejkal, Z.,
    Rokoiek, J., and Vitkova, J., : Radioch. Radional. Letters
    20, 1, 33 (1974).

28. Kokgov, K., Pavlov, O., and Shvedov, V., Advances in
    Inorganic Chemistry and Radiochemistry Vol. II, 1 (1968).

29. Smith, W.T. et al.: J. Amen. Chem. Soc., 75, 5773, 5777
    (1953).

30. Spytsyn, V., Kuzina, A., Zamashnikora N., and Oblova A.,
    Isotopes and Radiation Technology 2, No. 3, 229 (1965).