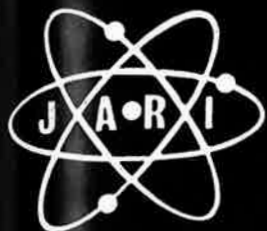


Applied Radiation and Isotopes

Including Data, Instrumentation and Methods for Use in
Agriculture, Industry and Medicine



Volume 45 Number 10
October 1994

Editors-in-Chief

W. L. McLaughlin, NIST, Gaithersburg

D. M. Taylor, UWCC, Cardiff

Editors

P. L. AIREY
Menai

D. A. BRADLEY
Kuala Lumpur

G. A. BRINKMAN
Amsterdam

C. G. CLAYTON
March

B. M. COURSEY
Gaithersburg

K. DEBERTIN
Braunschweig

W. C. ECKELMAN
Bethesda

E. A. EVANS
High Wycombe

T. FLORKOWSKI
Krakow

M. GUILLAUME
Liège

D. M. LYSTER
Vancouver

T. TOMINAGA
Tokyo



PERGAMON

HEALTH SCIENCES LIBRARY
University of Wisconsin

NOV 11 1994

1305 Linden Drive
Madison, WI 53706

AdvanCell - Ex. 1003 - Page 1

APPLIED RADIATION AND ISOTOPES

EDITORS-IN-CHIEF

W. L. McLAUGHLIN
(Editor-in-Chief for North America)
*Radiation Physics Division,
National Institute of Standards
and Technology, Gaithersburg,
MD 20899, U.S.A.*

D. M. TAYLOR
(Editor-in-Chief)
*University of Wales, College of Cardiff
Department of Chemistry,
P.O. Box 912, Cardiff CF1 3TB,
Wales*

CONSULTING EDITOR EMERITUS

W. B. MANN, 5710 Warwick Place, Chevy Chase, MD 20815, U.S.A.

EDITORS

- P. L. AIREY, *ANSTO, Lucas Heights Research Laboratories, Private Mail Bag 1, Menai, N.S.W. 5097, Australia*
D. A. BRADLEY, *Asia Lab (Malaysia) Sdn. Bhd., No. 6 Jalan 4/91, Taman Shamelin Perkasa, Cheras, 56100 Kuala Lumpur, Malaysia*
G. A. BRINKMAN, *NIKHEF-K, P.O. Box 41882, 1009 DB Amsterdam, The Netherlands*
C. G. CLAYTON, *Sprole House, 125 Knights End Road, March, Cambridgeshire PE15 9QD, England*
B. M. COURSEY, *Center for Radiation Research, National Institute of Standards and Technology, Gaithersburg, MD 20899, U.S.A.*
K. DEBERTIN, *Physikalisch-Technische Bundesanstalt, Postfach 3345, D 38023 Braunschweig, Germany*
W. C. ECKELMAN, *NIH/Clinical Center, Building 10, 1C495, 9000 Rockville Pike, Bethesda, MD 20892, U.S.A.*
E. A. EVANS, *5 Willow Chase, Hazlemere, High Wycombe, Buckinghamshire HP15 7QP, England*
T. FLORKOWSKI, *Institute of Physics & Nuclear Techniques, Academy of Mining & Metallurgy, Al. Mickiewicza 30, 30-059 Krakow, Poland*
M. GUILLAUME, *Centre de Recherches du Cyclotron, Université de Liège, Bat. B30 Sart Tilman, B-4000 Liège, Belgium*
F. F. KNAPP, *Health Sciences Research Division, Mail Stop 6229, Building 4501, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831, U.S.A.*
D. M. LYSTER, *Division of Nuclear Medicine, Vancouver General Hospital, 855 West 12th Avenue, Vancouver, B.C., Canada V5Z 1M9*
T. TOMINAGA, *Department of Chemistry, Faculty of Science, University of Tokyo, Bunkyo-ku, Tokyo, Japan*

Published Monthly

Copyright © 1994 Elsevier Science Ltd

Publishing Office

Elsevier Science Ltd, Bampfylde Street, Exeter EX1 2AH, England
(Tel. Exeter (01392) 51558; Fax 425370)
Production Editor: Alison J. Snell

Subscription and Advertising Offices

North America: Elsevier Science Inc., 660 White Plains Road, Tarrytown, NY 10591-5153, U.S.A.

Rest of the World: Elsevier Science Ltd, The Boulevard, Langford Lane, Kidlington,
Oxford OX5 1GB, England
(Tel. Oxford (01865) 843000; Fax 843010)

Subscription Rates

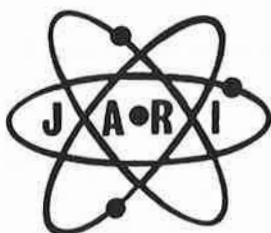
Annual Institutional Subscription Rates 1995: North, Central and South America, U.S.\$1021.00, Rest of the World £685.00. Associated Personal Subscription Rates are available on request for those whose institutions are library subscribers. Sterling prices exclude VAT. Non-VAT registered customers in the European Community will be charged the appropriate VAT in addition to the price listed. Prices include postage and insurance and are subject to change without notice.

Back Issues

Back issues of all previously published volumes, in both hard copy and on microform, are available direct from Elsevier Science offices.

Second class postage paid at RAHWAY NJ. Postmaster send address corrections to *Applied Radiation and Isotopes*, c/o Elsevier Science Inc., 660 White Plains Road, Tarrytown, NY 10591-5153, U.S.A.

Applied Radiation and Isotopes



Editors-in-Chief

W. L. McLaughlin, NIST, Gaithersburg

D. M. Taylor, UWCC, Cardiff

HEALTH SCIENCES LIBRARY
University of Wisconsin

NOV 11 1994

1305 Linden Drive
Madison, WI 53706



PERGAMON

AIMS AND SCOPE

Applied Radiation and Isotopes aims to provide a high quality medium for the publication of substantial original papers and short articles describing all aspects of the use of radiation and radionuclides in science and technology, including chemistry, physics, biochemistry, biology, medicine, environmental science and engineering. Papers describing fundamental data and of methods of data acquisition as well as theoretical papers in applied radiation science are encouraged.

Relevant topics for *Applied Radiation and Isotopes* include the following, however, authors are encouraged to suggest other topics which might also be published in the journal:

Radiation sources: The design, construction, production and characteristics of radiation or radionuclide sources.

Radionuclides: The production and processing of radionuclides for use in radiation sources or for the preparation of labelled compounds. Including measurements of activation cross-sections, target design and quality control procedures.

Synthesis of labelled compounds: Methods for the synthesis, purification and quality control and *in vitro* testing of radionuclide-labelled compounds for use as radiopharmaceuticals or scientific and medical research.

Measurement of radiation and radioactivity: Theoretical and experimental studies of the measurement of x-rays, γ -rays, α - and β -particles and other forms of radiation; nuclear instrumentation, including radiation spectrometry, radiation and radionuclide imaging techniques, dosimetry, novel counting systems and whole-body counters.

Radioanalytical methods: Activation analysis, isotope dilution analysis, radioimmunoassay, radionuclide tomography including radiation spectrometry, etc., as well as the development of *in vivo* techniques such as positron emission (PET) and single-photon emission spectrometry (SPECT).

Nuclear physics and chemistry topics, including data compilations, directly relevant to practical applications.

Radiochemistry including chemical behaviour and speciation of radionuclides and labelled compounds in geological, environmental and biological systems, and of the factors which modify this behaviour.

Manuscripts, which will be subject to peer review, should take one of the following forms:

Original papers, which should be definitive and describe a reasonably complete investigation.

Technical Notes which may describe new, unpublished information, including *preliminary communications* and *work in progress*; or may take the form of **Letters to the Editor** containing comments related to articles previously published in the journal. This type of communication should not exceed two printed pages in order to expedite their publication.

Review articles, *conference proceedings* and *complete conference abstracts* may also be accepted for publication following discussion with an editor of the journal.

Copyright © 1994 Elsevier Science Ltd

It is a condition of publication that manuscripts submitted to this journal have not been published and will not be simultaneously submitted or published elsewhere. By submitting a manuscript, the authors agree that the copyright for their article is transferred to the publisher if and when the article is accepted for publication. However, assignment of copyright is not required from authors who work for organizations which do not permit such assignment. The copyright covers the exclusive rights to reproduce and distribute the article, including reprints, photographic reproductions, microform or any other reproductions of similar nature and translations. No part of this publication may be reproduced, stored in a retrieval system or transmitted in any form or by any means, electronic, electrostatic, magnetic tape, mechanical, photocopying, recording or otherwise, without permission in writing from the copyright holder.

Photocopying Information for users in the U.S.A. The Item-fee Code for this publication indicates that authorization to photocopy items for internal or personal use is granted by the copyright holder for libraries and other users registered with the Copyright Clearance Center (CCC) Transactional Reporting Service provided the stated fee for copying, beyond that permitted by Section 107 or 108 of the U.S. Copyright Law, is paid. The appropriate remittance of \$7.00 per copy per article is paid directly to the Copyright Clearance Center, 222 Rosewood Drive, Danvers, MA 01923, U.S.A. [Tel. (508) 750 8400; Fax (508) 750 4744].

Permission for other use. The copyright owner's consent does not extend to copying for general distribution, for promotion, for creating new works or for resale. Specific written permission must be obtained from the publisher for such copying. The Item-fee Code for this publication is: 0969-8043/94 \$7.00 + 0.00

DISCLAIMER

Whilst every effort is made by the publishers and editorial board to see that no inaccurate or misleading data, opinion or statement appears in this journal, they wish to make it clear that the data and opinions appearing in the articles and advertisements herein are the sole responsibility of the contributor or advertiser concerned. Accordingly, the publishers, the editorial board and editors and their respective employees, officers and agents accept no responsibility or liability whatsoever for the consequences of any such inaccurate or misleading data, opinion or statement.

©TMThe text paper used in this publication meets the minimum requirements of American National Standard for Information Sciences—Permanence of Paper for Printed Library Materials, ANSI Z39.48-1984.



b89043307727a

- Chun-Nan Hsu, Dai-Chin Liu and Chi-Lung Chuang 981 Equilibrium and kinetic sorption behaviors of cesium and strontium in soils
- M. T. Teli, L. M. Chaudhari and S. S. Malode 987 Attenuation coefficients of 123 keV γ -radiation by dilute solutions of sodium chloride
- J. A. Novotny, D. R. Lacaze, M. L. Moore and M. Forsbacka 991 Production of millicurie quantities of ^{85m}Kr in an aluminum vessel in a 1-megawatt nuclear reactor
- Amon R. Wafelman, Monique C. P. Konings, Cornelis A. Hoefnagel, Robert A. A. Maes and Jos H. Beijnen 997 Synthesis, radiolabelling and stability of radioiodinated *m*-iodobenzylguanidine, a review
- S. N. Muddukrishna, P. Jorge, W. Machner, T. R. Sykes and A. A. Noujaim 1009 An improved method for analysis of technetium radiolabeled antibody by size exclusion HPLC
- Sindre P. Hassfjell and Per Hoff 1021 A generator for production of ^{212}Pb and ^{212}Bi
- S. A. Gerasimov 1027 Simple and fast method for separating peak doublet
- K. N. Yu and S. Y. Mao 1031 Application of high resolution gamma ray spectrometry in measuring radioactivities in drinks in Hong Kong
- Carol Gamblin, Christopher L. Gordon, David C. F. Muir, David R. Chettle and Colin E. Webber 1035 *In vivo* measurements of bone lead content in residents of Southern Ontario
- C. M. Bartle 1039 Measurement of the composition of dairy products using the neutron/gamma transmission (NEUGAT) method
- Technical Notes*
- M. R. Zaman 1047 4-Bromoacetanilide-methanol vapour attacks the paraffin cylinder surrounding the (Ra-Be) neutron source in charge plate collection experiments
- Ron Hulst, Ben L. Feringa, Hilde K. Sierstema, Erik J. F. Franssen, Gerben M. Visser and Wim Vaalburg 1049 Quality control of purified [^{18}O] target water using ^{31}P NMR
- Letters to the Editors*
- F. M. Nortier, S. J. Mills and G. F. Steyn 1051 Excitation function of the $^{111}\text{Cd}(\text{p}, \text{n})^{111\text{m}}\text{In}$ reaction
- F. Tárkányi 1052 Reply by Dr F. Tárkányi to Letter by Dr Nortier *et al.*
- Book Reviews*
- 1053
- Events*
- 1055



0969-8043(1994)45:10;1-2

Appl. Radiat. Isot. is Indexed/Abstracted in: Anal. Abstr.; App. Health Phy. Abstr.; Biosis Data.; CAB Inter.; Cam. Sci. Abstr.; Chem. Abstr. Serv.; Curr. Cont./Life Sci.; Curr. Cont./Phy. Chem. & Earth Sci.; CABS; Eng. Ind.; Excerpt. Med.; Health & Saf. Sci. Abstr.; Ind. Med.; INSPEC Data.; Curr. Cont. ISI/BIOMED Database; MEDLINE; PASCAL-CNRS Data.; Res. Alert; Curr. Cont. SCISEARCH Data.; Soc. Sci. Cit. Indx; SSSA/CISA/ECA/ISMEC; Tox. Abstr.



PERGAMON

ISSN 0969-8043

ARISEF 45(10) 981-1056 (1994)



A Generator for Production of ^{212}Pb and ^{212}Bi

SINDRE P. HASSFJELL* and PER HOFF

Department of Chemistry, Section for Nuclear Chemistry, University of Oslo, P.O. Box 1033, Blindern, N-0315 Oslo, Norway

(Received 3 May 1994)

A generator has been developed for the production of the α -emitting radionuclide ^{212}Bi and its parent nuclide ^{212}Pb . The generator is based on the emanation of ^{220}Rn from ^{228}Th barium stearate. The decay product of ^{220}Rn , ^{212}Pb deposits on the walls of a polyethylene bottle, and can be washed off with distilled water. The generator shows no leakage of any long-lived parent nuclides, is easy to operate and has a high degree of radiation safety.

Introduction

The potential for α -emitters in the radiation-therapy of tumors is well documented (Huneke *et al.*, 1992; Vergote *et al.*, 1992; Ford and Terzaghi-Howe, 1993; Humm and Chin, 1993). The α -emitters of choice, ^{211}At and ^{212}Bi , have been investigated in several *in vitro* and *in vivo* experiments (Macklis *et al.*, 1989; Huneke *et al.*, 1992; Vergote *et al.*, 1992). As ^{211}At must be produced in an accelerator, its availability is limited and the production costs relatively high; ^{212}Bi occurs in the natural decay series from ^{228}Th (Fig. 1), and can be produced by an isotope generator. Several generators have been made, based on different principles, for the separation of ^{212}Bi from its parent nuclides.

One type of generator utilizes the different cation and anion exchange properties of the individual decay products of ^{228}Th (Zucchini and Friedman, 1982; Atcher *et al.*, 1988). Generators based on ^{228}Th ($t_{1/2} = 1.9$ yr) experienced problems with radiolytic damage in the resin, with consequent diminished yield and serious radiation safety problems. To avoid these problems, another generator based on ^{224}Ra ($t_{1/2} = 3.7$ days), was made. However, because of the short half-life it must be regenerated after 1–2 wk. This generator has given good yields of ^{212}Bi and its parent nuclide ^{212}Pb , but demands regular maintenance and skilled chemists to operate it.

Another system is based on separation of ^{220}Rn from ^{228}Th by emanation of Rn gas from Th salts. It has been shown (Gregory and Moorbath, 1951; Wahl and Daniels, 1958) that nearly quantitative separation of ^{220}Rn from ^{228}Th of ^{219}Rn from ^{223}Ra , can be

achieved if the $^{228}\text{Th}/^{223}\text{Ra}$ is incorporated into solid Ba stearate $[\text{Ba}(\text{C}_{18}\text{H}_{35}\text{O}_2)_2]$. One type of generator collects ^{211}Pb by distillation of ^{219}Rn from ^{223}Ra barium stearate under reduced pressure. Negligible contamination from parent nuclides is reported, but the yield is low, approx. 10% (Atcher *et al.*, 1989).

Because of its short half-life, ^{220}Rn has a relatively short diffusion distance at room temperature and atmospheric pressure. It therefore appeared to be a simpler and better approach to collect ^{212}Pb and ^{212}Bi by enclosing the ^{228}Th barium stearate in a gas-tight collection chamber. The Rn gas will then fill the volume of the collection chamber, and after decay of ^{220}Rn and ^{216}Po , ^{212}Pb will deposit on the walls. Patent rights have been claimed on a generator based on this principle (Norman *et al.*, 1991), including claims for Rn emanation from ^{228}Th barium stearate already described in 1951 (Gregory and Moorbath, 1951) as well as different kinds of collecting media for the ^{212}Pb and ^{212}Bi .

The generator described in this paper is also based on the well-known Rn emanation, but with a different collection principle.

Materials and Methods

Production of ^{228}Th barium stearate

^{228}Th was purchased commercially as dry nitrate (Harwell, U.K.). The ^{228}Th barium stearate was prepared by a method outlined by Hursh and Lovaas (1967). A 5 ml 0.01% HCl solution containing 0.025 g $\text{BaCl}_2 \cdot 2\text{H}_2\text{O}$ and 20–30 MBq of $^{228}\text{Th}(\text{NO}_3)_4$ was added to 25 ml of 100% methanol and stirred at low speed for 5 min. Then 0.084 g of sodium stearate dissolved in 25 ml of 100% methanol was added

*To whom all correspondence should be addressed.

slowly. The desired [^{228}Th]barium stearate precipitated from the solution was filtered onto a $0.8\ \mu\text{m}$ filter membrane (Gelman, U.S.), and washed with 25 ml of 100% methanol. The filter membrane with the precipitate was folded and wrapped up in two additional filters to avoid any loss of [^{228}Th]barium stearate. The packet was secured with plastic strips. Several packets were made. The whole chemical procedure was performed in a sealed plastic glove-box which was discarded afterwards. For security reasons, the use of electrical equipment inside the glove-box was avoided.

The generator

The generator is shown in Figs 2 and 3. All generator parts were made of stainless steel, and connected to standard high vacuum equipment to avoid leakage of radioactivity to the surroundings. The generator was made with two compartments, a lower compartment which is a collection chamber and an upper compartment which is a housing chamber for the source when the collection chamber is removed. These two compartments are connected via a gate valve (VAT). The packets of [^{228}Th]barium stearate are placed in a filering basket mounted by threads to the end of a long tube closed in both ends. The filering basket has a capacity of 3–4 [^{228}Th]barium stearate packets. The tube can slide on two O-rings, thus facilitating the filering basket to move between the two chambers, through the gate valve. To neutralize pressure differences when the tube is moved, an on-off valve (VAT) with connection to a filtered air outlet is placed in the upper chamber. To

collect the ^{212}Pb and ^{212}Bi , a polyethylene bottle is placed inside the collection chamber.

When the tube is lowered, the filering basket with the emanating [^{228}Th]barium stearate becomes positioned inside the bottle. The decay products from the emanated ^{220}Rn , ^{212}Pb and ^{212}Bi , deposit on the walls of the bottle. After an appropriate collection time, the tube is pulled up, placing the ^{228}Th source in the upper compartment, and the gate valve is closed. The polyethylene bottle is then removed and the radioactivity easily washed off with a suitable solution.

Results

Filter membrane packets with [^{228}Th]barium stearate with ^{228}Th at the kBq level have shown unchanged emanation ability for ^{220}Rn during 1 yr. At higher radioactivity levels, however, a loss in emanation ability was observed during this period. The generator has been operated with 40–50 MBq (1.1–1.4 mCi) of ^{228}Th for 1 yr, and the total amount of ^{212}Pb that can be collected inside the polyethylene bottle at saturation has decreased from 50 to 11% of the ^{228}Th radioactivity.

About 70% of the radioactivity inside the polyethylene bottle can be easily washed off with approx. 30 ml of distilled water. By increasing the ionic strength of the washing solution or by adding a chelating agent, the yield increases. With a 1.0 M NaCl solution, 99% of the ^{212}Pb can be washed of the walls of the bottle. No contamination from ^{228}Th or ^{224}Ra outside the filter membrane packets has been detected.

Despite the relatively strong ^{228}Th source, no detectable leakage of any kind of radioactivity out of the generator has been observed.

Discussion

The obvious advantage of this generator compared to those based on ionic exchange is the ease of operation. It does not require experienced chemists for safe operation and handling. The two compartments of the generator, one for collection and one for storage, combined with the possibility of decreasing the inside pressure, gives the generator a high degree of radiation safety. Moreover, the system is static with the long tube as the only moving part, and therefore the only regular maintenance required is lubrication of the tube and the O-rings by a silicon based grease. This operating simplicity diminishes the probability of accidents and improves the radiation safety.

Since the ^{212}Pb and ^{212}Bi can be removed nearly quantitatively from the walls of the polyethylene bottle after a few minutes washing, it may be concluded that implantation of ^{212}Pb into the plastic due to recoil in the ^{216}Po decay is of no importance.

The dismounting of the filering basket from the tube is simple, and the [^{228}Th]barium stearate packets

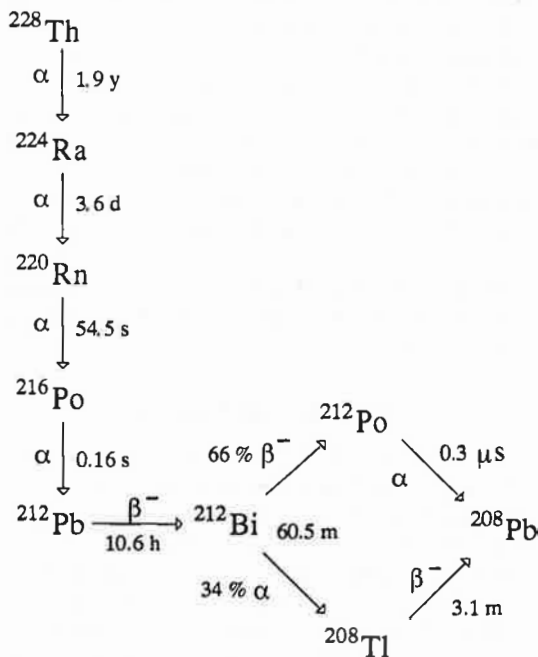


Fig. 1. The decay mode of ^{228}Th to stable ^{208}Pb .

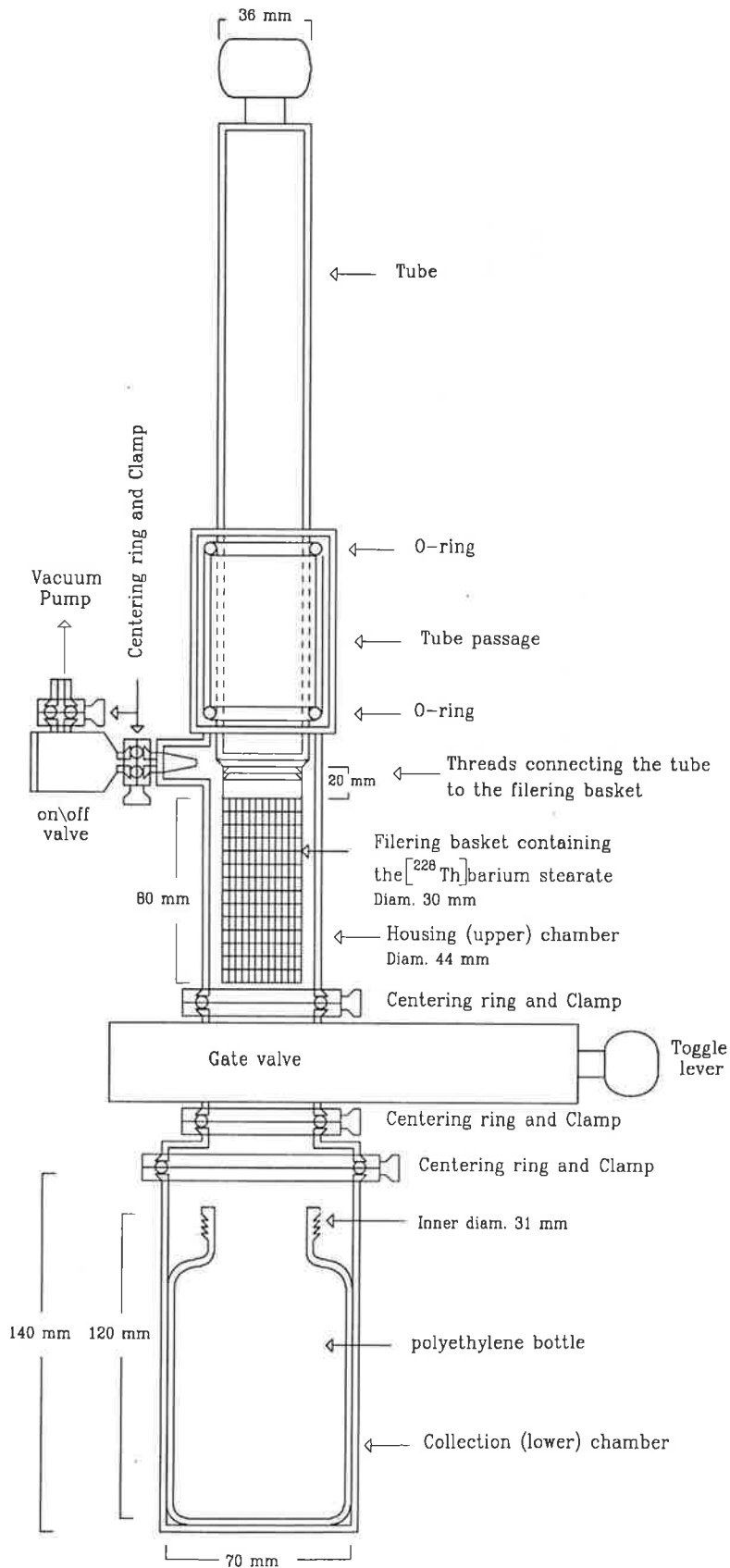


Fig. 2. The design of the generator showing functional details and operating mechanism.

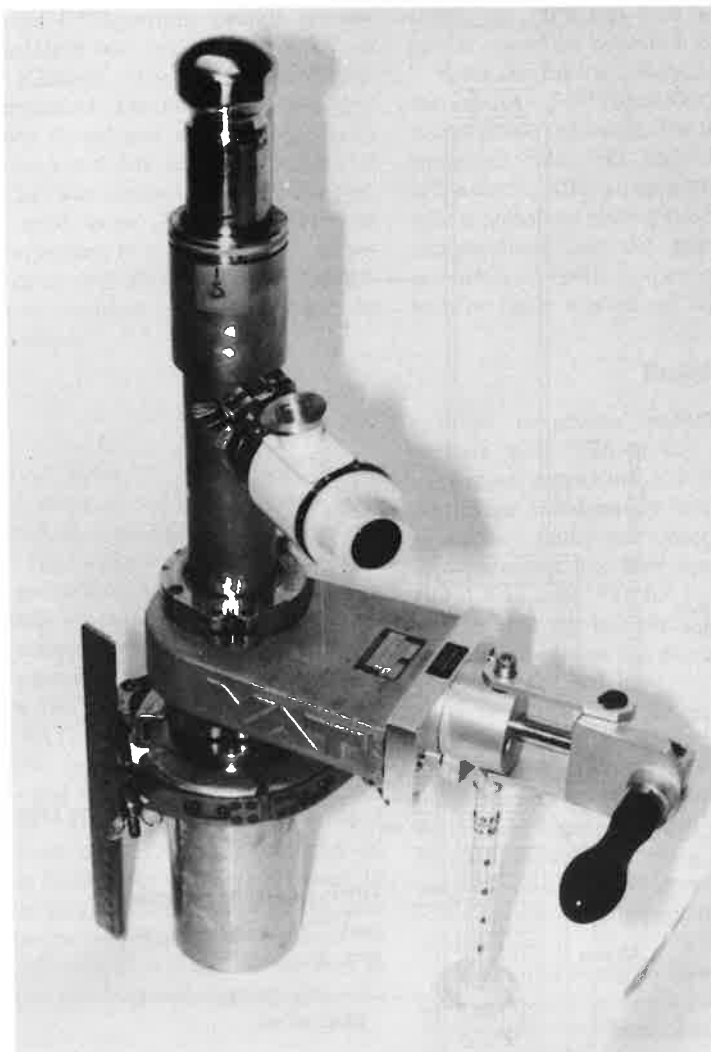


Fig. 3. A picture of the generator.

are easily replaced. If desired, it is easy to introduce safety obstacles to prevent unintentional dismounting of the ^{228}Th source or free exposure to the surroundings.

In conclusion, this generator produces ^{212}Pb and ^{212}Bi without any contamination from the parent nuclides. It has a high degree of radiation safety, needs very little maintenance and is simple to operate. The decreasing yield of ^{212}Pb and ^{212}Bi is due most likely to radiation damage of the stearate or the filter membranes. Methods for regaining the emanation ability will be investigated.

Acknowledgements—The authors thank Professor Gösta Rudstam, Studsvik, for valuable discussions in the initial phase of the work. The skilled technical assistance from chief engineer Roald Østbu and engineer Yngve Kristiansen of the mechanical workshop at our institute is highly appreciated.

References

- Atcher R. W., Friedman A. M. and Hines J. J. (1988) An improved generator for the production of ^{212}Pb and ^{212}Bi from ^{224}Ra . *Appl. Radiat. Isot.* **39**, 283.
- Atcher R. W., Friedman A. M., Huizenga J. R. and Spencer R. P. (1989) A radionuclide generator for the production of ^{211}Pb and its daughters. *J. Radioanal. Nucl. Chem. Lett.* **135**, 215.
- Ford J. R. and Terzaghi-Howe M. (1993) Effects of ^{210}Po alpha particles on survival and preneoplastic transformation of primary rat tracheal epithelial cells irradiated while in suspension or in the intact tissue. *Radiat. Res.* **136**, 89.
- Gregory J. N. and Moorbath S. (1951) The diffusion of thoron in solids. Part II—The emanating power of barium salts of the fatty acids. *Trans. Faraday Soc.* **47**, 1064.
- Humm J. L. and Chin L. M. (1993) A model of cell inactivation by alpha-particle internal emitters. *Radiat. Res.* **134**, 143.
- Huneke R. B., Pippin C. G., Squire R. A., Brechbiel M. W., Gansow O. A. and Strand M. (1992) Effective

- α -particle-mediated radioimmunotherapy of murine leukemia. *Cancer Res.* **52**, 5818.
- Hursh J. B. and Lovaas A. I. (1967) Preparation of a dry ^{228}Th source of thoron. *J. Inorg. Nucl. Chem.* **29**, 599.
- Macklis R. M., Kaplan W. D., Ferrara J. L. M., Atcher R. W., Hines J. J., Burakoff S. J. and Coleman C. N. (1989) Resident's essay award: alpha particle radioimmunotherapy: animal models and clinical prospects. *Int. J. Radiat. Oncol. Biol. Phys.* **16**, 1377.
- Norman J. H., Wrasidlo W. A. and Mysels K. J. (1991) Method and generator for producing radioactive lead-212. U.S. Patent, Patent number 5,038,046.
- Vergote I., Larsen R. H., De Vos L., Nesland J. M., Bruland Ø., Bjørgum J., Alstad J., Tropé C. and Nustad K. (1992) Therapeutic efficacy of the α -emitter ^{211}At bound on microspheres compared with ^{90}Y and ^{32}P colloids in a murine intraperitoneal tumor model. *Gynecol. Oncol.* **47**, 366.
- Wahl A. C. and Daniels W. R. (1958) Emanating power of barium stearate for 3.9 second actinon (^{219}Rn). *J. Inorg. Nucl. Chem.* **6**, 278.
- Zucchini G. L. and Friedman A. M. (1982) Isotopic generator for ^{212}Pb and ^{212}Bi . *Int. J. Nucl. Med. Biol.* **9**, 83.