Conference of Radiological Health Specialists

Problems in Radium Control

JOHN C. VILLFORTH, M.S.

ON DECEMBER 28, 1898, the Curies announced to the world the discovery of radium. It was not, however, until 45 months later that they were able to prepare the first 100 mg. of pure radium salts. This was the beginning of man's use of one of the most versatile radioactive materials that, in spite of the present widespread use of artificially produced radionuclides, is still commonly employed in medicine, industry, research, and various consumer items.

The biological effects of this new material were observed soon after its discovery. Two German chemical manufacturers reported in 1900 that radium had destructive action on the epidermis (*1*). Becquerel received a chest burn from carrying radium in his vest pocket. Pierre Curie produced a similar burn on his arm by placing it near a radium source for 10 hours (*2*). These and many more early observations led to the use of radium in medicine. In 1901 radium was used for the treatment of lupus at the St. Louis Hospital in Paris. The first radium used in medicine in the United States (150 mg.) was obtained by Dr. Robert Abbe from the Curie Laboratory in 1903. During that year experiments were conducted on its effect on certain nasopharyngeal conditions and in intrauterine applications. Its use on superficial conditions seemed to be well established. Not until 1910 were interstitial needles first used, although Alexander Graham Bell suggested in 1903 that radium in glass tubes be "inserted into the very heart of cancer" (*3*).

The decades that followed saw many improvements and innovations in the use of radium and its daughter product, radon, as a therapeutic tool in medicine. Kelly, Burnham, Stevenson, Joly, Janeway, and others helped pioneer this usage (*4*).

Industrial applications of radium lagged behind the medical uses. Although radium and mesothorium were used in luminous compounds during World War I, the first commercial use of radium in industrial radiography did not take place until 1930. Gradually, radiographic procedures using radium became an accepted quality control method. During World War II, the U.S. Navy purchased more than 17 grams of radium for industrial radiography (*5*). These were used to inspect highly stressed structural parts of ships—castings, weldments, and valves and fittings in the high-pressure steam and feed lines. In addition to radiography, radium is now used for other industrial and research purposes, such as neutron sources, instrument calibration sources, thickness and density gauges, static eliminators, and fire alarm devices.

Along with the fascinating history of the early applications of radium were also references suggesting that radium be used with caution. It is perhaps unfortunate that we have not profited more from the early mistakes and observations of others. For example, in 1906 Dr. Robert Abbe reported on the "explosion" of a 50-mg. glass radium tube while he was removing it from a container. Upon searching the literature, he noted that similar experiences

---

Mr. Villforth is chief of the Assistance to State Radioactive Materials Program, Division of Radiological Health, Public Health Service.

722-915—64——5

had occurred in Europe. This was thought—and not too incorrectly—to be caused by "either emanation or helium," which increased the pressure in the sealed tube (*6*).

Dr. George E. Pfahler became increasingly concerned over the report of the deaths of five radiologists from aplastic anemia caused directly or indirectly by radiations from radium or X-ray. Therefore, in 1921, he investigated the general health of all radiologists in the United States through a questionnaire. He also requested that each radiologist have a complete blood examination and carry a dental film in his pocket for 2 weeks, an early attempt at film badge dosimetry. Pfahler concluded from this survey that "increased protection is needed by those who are working with the gamma rays, or with the higher voltage X-rays," and "complete protection can undoubtedly be obtained. It requires not only the means, but the continual caution on the part of the individual" (*7*).

Soon after Pfahler's study, Dr. R. C. Williams evaluated the physical condition of persons employed in the radium section of the National Bureau of Standards. He observed certain blood changes and positive effects upon dental film worn by employees, and recommended, among other things, that all employees "utilize to the greatest possible extent all practical protective devices, such as screens, lead-lined carrier boxes, and handling forceps" (*8*).

Although the hazards of external radiation from improper handling of radium were slowly becoming recognized at this time, the effects of the ingestion of radium were not fully appreciated until the middle twenties when Dr. Harrison S. Martland published his classic findings on "radium poisoning" of the New Jersey dial painters (*9–11*). His studies have been the basis for present knowledge on the behavior of the so-called bone seekers in the body. These studies also had an impact on the then common use of radium "tonics" as a panacea for rheumatism, gout, and various other afflictions. As a result of these abuses, radium for internal administration was removed from listing in "New and Nonofficial Remedies" of the American Medical Association in 1932.

Uses of radium in medicine and industry continued to expand as new and better methods were developed for its application and as more efficient processing techniques made it more readily available at lower cost. Before World War I, radium was valued at $180 per mg.; during the 1920's the price dropped to $70 per mg., and in the 1930's, to $40 per mg. (*12*). The 1962 price is quoted at $16 to $21.50 per mg. of radium content, depending on quantity (*13*).

Unfortunately, application of radiological health principles and precautions, identified by earlier investigations, did not always keep pace with the use of radium. One reason for this may have been that many users were unaware of the biological damage that might manifest itself later, and therefore they disregarded precautions in handling radium. Also, government agencies have exerted little or no control over radium users to enforce adoption of proper precautions. Radium has not been subject to control by the Atomic Energy Commission although it is a greater potential hazard than virtually any AEC-licensed material.

Inherent Disadvantages of Radium

Radium 226, a naturally occurring radionuclide found as a daughter product in the uranium decay series, has a half-life of 1,622 years. As a member of the alkaline earth series, radium is chemically similar to calcium, strontium, and barium. Therefore, if it enters the body, it will probably be deposited in the bone where it may cause significant biological damage because of its long effective half-life and alpha decay. These properties make radium the most radiotoxic of the commonly used radionuclides.

Radium is a brilliant white metal that quickly turns black (nitride) in contact with the atmosphere. It behaves like the other alkaline metals, attacking glass or quartz and decomposing water to form an hydroxide (*14*). Therefore, the radium used in medicine and industry is combined in a more stable form as a chloride, bromide, or sulfate. Most of the radium processed in recent years is in the form of the insoluble sulfate. However, in 1933 Sayer reported that in a survey of radium sources in medical institutions, 18 percent of the radium was in the form of a chloride, 26 percent in a bromide form, and 54 percent was in a sulfate form. The remaining 2 percent was radium carbonate or some other compound (*15*). These salts are hermetically sealed in tubes, needles, cells, or capsules where they can be manipulated to produce the desired exposure. Since these salts are in the form of a fine powder, rupture of the sealed container will result in the dispersal of the radium, making recovery and decontamination operations difficult. Because radium 226 has a 1,622-year half-life, decontamination is necessary rather than restricting the area until the activity has decayed to acceptable levels, as can be done in similar incidents with some of the short-lived radionuclides.

It should also be pointed out that the dosage calculations in radiation therapy are based on the assumption that the radium is uniformly distributed throughout the active length of the source. Barium salts are usually added to the radium salts to fill the container completely. However, Freed and co-authors (*16*) have observed by autoradiography of radium needles that these salts may become unevenly packed, resulting in a considerable discrepancy in the expected tissue dose as calculated from a uniformly packed needle. They recommend a periodic inspection and autoradiograph of sources used in therapy.

Another characteristic of radium is its complex series decay. Radium 226 is primarily an alpha emitter, as are its daughters, radon 222, polonium 218, and polonium 214. The gamma activity that makes radium applicable in medicine and industry comes from the beta-gamma decay of two other daughters, lead 214 and bismuth 214. To be an effective gamma emitter, a radium source must be sealed for approximately 30 days to allow its daughter products to reach equilibrium with the radium. Unfortunately, internal pressure builds up inside the sealed source. This pressure is partly caused by the radon gas (3.28×10^{-3} atmospheres per year for a 1-mg. source), the helium buildup from the alpha decay (1.09 atmospheres per year for a 1-mg. source), but primarily by the generation of hydrogen and oxygen in the dissociation of any water present as a result of inadequate drying of the salts before they are sealed in the source. The pressure from this dissociation has been estimated to reach several hundred atmospheres (*17*). Extreme care must be exercised in handling sealed radium sources, for there is no way of determining when a source may have reached the point of failure. Rough handling and heat sterilization should be avoided as they may add sufficient external stress to initiate rupture or leakage. Older sources encapsulated in glass or fitted with a friction plug without threads are particularly susceptible to failure from internal pressure (*18*).

Leaking sources are not only a potentially severe health hazard because of the spread of finely divided radium salt if the hermetic seal fails completely, but, in addition, the sources will not deliver the expected gamma dose because the radon daughter products are not in equilibrium with radium (*19*).

The decrease in the cost of radium over the years has also contributed to unnecessary exposures. Many users who purchased radium when the price was inflated are reluctant to part with the sources at the present low price. Therefore, some owners who no longer have a use for radium have retained it as a kind of radioactive "white elephant." Sources have been stored in such unlikely places as safe deposit boxes (*20*).

The characteristics of radium just described merely suggest its potential hazards. In order to evaluate public health aspects, it is necessary to know the frequency and extent of the uses of radium.

Problems of Usage

Unfortunately, there is little information on the extent radium is used in the United States. Most current information has been obtained from the few State health departments that have established radium control programs and extrapolated to a nationwide basis.

A review of this limited information indicates that at least 4,500 facilities use radium in the United States, a group equivalent to approximately 45 percent of the number of Atomic Energy Commission licenses.

Of the 4,500 facilities, 1,800, or 40 percent, use radium for industrial or research purposes, and the remaining 2,700, or 60 percent, for therapeutic purposes at 1,700 private offices and 1,000 hospitals and clinics. It has been estimated that radium is used to treat 80,000 patients per year in hospitals throughout the United States (*21*).

Between 300 and 700 grams of radium are in use in medicine and industry as identifiable sources. This amount is much less than the 2,000 grams imported or processed in the United States since the discovery of radium (*22*). The radium that is unaccounted for has probably been used in luminous compounds, static eliminators, and electron tubes, disposed of as radioactive waste, lost as the result of improper handling, or stored in attics or safe deposit boxes.

Losses and thefts of radium are not uncommon (*23–26*). Almost every State health department has participated in a search for lost radium. Taft records 187 incidents through 1946 (*27*). The Public Health Service has collected information on approximately 300 incidents and is informed of others at the rate of approximately 2 per month.

In addition to the thefts and losses resulting from the failure to follow proper radiological health procedures, numerous facilities have been contaminated by the rupture of radium sources or by careless processing techniques (*28–33*). Perhaps the most serious contamination incident reported resulted from the rupture of a single 50-mg. radium sulfate capsule used for instrument calibration. Contamination was so extensive that it resulted in damages of more than $250,000 (*34*).

The State health departments that conduct radium inspection programs have found contamination from leaking sources to be prevalent. The number of facilities possessing leaking sources varies from 13 to 58 percent, depending on the methods of leak testing. Gallaghar and co-workers report 27 of 664 sources tested (5.6 percent) showed evidence of radon leakage; however, 3 of 4 hospitals surveyed had leaking sources in their inventory (*19*).

Radium is frequently stored improperly. Information from State health departments indicates that 20 to 90 percent of the facilities have failed to provide adequate shielding or proper security for the radium sources. Twenty-three years ago when Cowie and Scheele surveyed 45 hospitals, it is partly understandable that they found that storage facilities in 16 would allow "definite overexposures" to personnel, and 13 other hospitals had questionable storage areas (*35*). However, in light of the many excellent articles that have since been published on radium handling and storage (*36–41*), and the increased emphasis placed on radiation protection by professional societies, standards groups, and government agencies, there are no apparent reasons why radium cannot be stored and handled so as to reduce the gamma exposure to personnel within acceptable levels.

Radium Management Programs

There is no Federal control over possession and use of radium, but some State and local agencies have assumed responsibility for control, paralleling the development of other State and local radiological health programs. At present only 16 States license or register and inspect radium facilities, and not all of these include all users (medical, industrial, institutional, and research). In order to assist some States in starting a radium management program and to help others in planning and operating one, the Division of Radiological Health, Public Health Service, has established the Assistance to State Radioactive Materials Program. This project is designed to stimulate States to plan a program based on an analysis of sources and uses in each State and to assist them with consultation on legislation and regulations, evaluations of survey methodology and instrumentation, and training for State personnel.

There is a need to investigate the substitution of less hazardous, artificially produced radionuclides for radium in medicine and industry. For example, metallic cobalt 60 has been used for interstitial and intracavitary therapy. Gold 198 "seeds" have been substituted for radon "seeds." Tritium is rapidly replacing radium in luminous compounds, and cesium 137 is accepted in industrial radiography. It is hoped that more effort will be directed at evaluating the efficacy of these and other radionuclides as radium substitutes.

Summary

Of all the more commonly used radionuclides, radium has the longest and most dramatic history. Certain of its inherent characteristics

make it a potential radiological health problem—its radiotoxicity, its series decay through alpha, beta, and gamma emitting daughters including its first daughter product, radon, which is a "noble" gas, its chemical and physical form, its ability to build up pressure in a sealed source, and its depreciation. These qualities and the fact that radium has not been systematically and generally under regulatory control contribute to its potential hazard.

Radium continues to be used at approximately 4,500 facilities throughout the country, although cheaper, more adaptable AEC-licensed radionuclides are more readily available. Incidents and accidents are not uncommon, and contamination from leaking and ruptured sources has resulted in costly damage and required expensive decontamination. Most of these incidents could have been avoided by following accepted radiological health procedures.

States are developing programs for the licensing, registration, and inspection of radium users, and the Public Health Service has established a program to assist the States in these activities. But the assistance of other agencies and professional groups is required to educate users to the potential hazards of improper handling and the use of antiquated sources and to investigate less hazardous substitutes for radium.

REFERENCES

(*1*) Hevesey, G. de: Marie Curie and her contemporaries. J Nucl Med 2: 169–182, July 1961.

(*2*) Becquerel, H., and Curie, P.: Comptes rend (Paris) 132: 1289–1291 (1901).

(*3*) Zowers, Z. T.: The uses of radium. Correspondence between Z. T. Zowers and Alexander Graham Bell. Amer Med 6: 261 (1903).

(*4*) Case, J. T.: The early history of radium therapy and the American Radium Society. Amer J Roentgen 82: 574–585, October 1959.

(*5*) Briggs, C. W., and Gezelius, R. A.: Instructions for radium radiography. Radium Chemical Co., Inc., New York, 1951.

(*6*) Abbe, R.: Explosion of a radium tube. Med Rec 69: 615–616, April 1906.

(*7*) Pfahler, G. E.: The effects of the X-ray and radium on the blood and general health of radiologists. Amer J Roentgen 9: 647–656, October 1922.

(*8*) Williams, R. C.: Preliminary note on observations made on physical condition of persons engaged in measuring radium preparations. Public Health Rep 38: 3007–3028, Dec. 21, 1923.

(*9*) Martland, H. S., Conlon, P., and Knef, J. P.: Some unrecognized dangers in the use of and the handling of radioactive substances, with especial reference to the storage of insoluble products of radium and mesothorium in the reticulo-endothelial system. JAMA 85: 1769–1776 (1925).

(*10*) Martland, H. S.: Microscopic changes of certain anemias due to radioactivity. Arch Path Lab Med 2: 465–472 (1926).

(*11*) Martland, H. S.: Occupational poisoning in manufacture of luminous watch dials. JAMA 92: 466–473, 552–559 (1929).

(*12*) Tyler, P. M.: Radium. Department of Commerce, U.S. Bureau of Mines Information Circular 6312, U.S. Government Printing Office, Washington, D.C., August 1930.

(*13*) Bureau of Mines, Division of Minerals, U.S. Department of Interior: Minor metals and minerals. Minerals Yearbook 1, U.S. Government Printing Office, Washington, D.C., 1962.

(*14*) Sanderson, L.: Radium. Canad Mining J 82: 57–58, December 1961.

(*15*) Sayers, R. R.: Radium in medical use in the United States. Radiology 20: 305–310, April 1933.

(*16*) Freed, J. H., Pendergrass, E. P., and Raufer, H.: Use of autoradiographs to detect defects in radium needles and tubes and inequalities in the distribution of the radium. Radiology 56: 99–103, January 1951.

(*17*) Morgan, J. E.: Gas pressure in sealed radium containers. Amer J Roentgen 85: 949–954, May 1961.

(*18*) Lind, S. C.: Care of radium salts. Nucleonics 11: 56–58, April 1953.

(*19*) Gallaghar, R. G., Evans, R. D., and McAllister, R. G.: Testing radium capsules for radon leakage. Amer J Roentgen 90: 396–402, August 1963.

(*20*) Peterson, D. R.: Radioactive material in bank vaults. Public Health Rep 75: 1190, December 1960.

(*21*) Terrill, J. G., Jr., Ingraham, S. C., II, and Moeller, D. W.: Radium in the healing arts and in industry. Public Health Rep 69: 255–262, March 1954.

(*22*) Bureau of Mines, Division of Minerals, U.S. Department of Interior: Metals and minerals. Minerals Yearbook, annual issues, U.S. Government Printing Office, Washington, D.C., 1918–61.

(*23*) Miller, A. L.: Searching ash wagon for a clinker. Hospital Management 15: 45–47, May 1923.

(*24*) Taft, R. B.: Lost and found radium. Amer J Roentgen 37: 87–92, January 1937.

(*25*) Williams, M. M. D.: Recovery of radium tubes from sewer. Radiology 41: 478–482, November 1943.

(26) Anonymous: Three-day radiation scare grips downtown Toronto. Canad Nucl Technology, Summer 1961, p. 36.

(27) Taft, R. B.: Radium: Lost and found. Ed. 2. Walker, Evans & Cogswell Co., Charleston, S.C., 1946.

(28) Gallaghar, R. G., Zavon, M. R., and Doyle, H. N.: Radioactive contamination in a radium therapy clinic. Public Health Rep 70: 617–624, July 1955.

(29) Saenger, E. L., Gallaghar, R. G., Anthony, D. S., and Valaer, P. J.: Emergency measures and precautions in radium accidents. JAMA 149: 813–815 (1952).

(30) Krabbenhoft, K. L.: Radium accidents [Editorial]. Amer J Roentgen 83: 584–586, March 1960.

(31) Cross, F. H., and Hunnings, G.: An unusual radium incident in Nottingham. Amer J Roentgen 72: 989–994, December 1954.

(32) Skow, R. K., Vandivert, V. V., and Holden, F. R.: Hazard evaluation and control after a spill of 40 mg. of radium. Nucleonics 11: 45–47, August 1953.

(33) Hoover, R. L.: Industrial hygiene techniques in the decontamination of a building contaminated with radium. Amer Industr Hyg Assoc J 22: 83–85, April 1961.

(34) Gallaghar, R. G., and Saenger, E. L.: Radium capsules and their associated hazards. Amer J Roentgen 77: 511–523, March 1957.

(35) Cowie, D. B., and Scheele, L. A.: A survey of radiation protection in hospitals. J Nat Cancer Inst 1: 767–787, June 1941.

(36) Kaye, G. W. C., Bell, G. E., and Binks, W.: The protection of radium workers from gamma radiation. Brit J Radiol 8: 6–26, January 1935.

(37) Quimby, E. H.: Radium protection. J Appl Physics 10: 604–608, September 1939.

(38) DeAmicis, E., and Cowing, R. F.: Personnel exposures associated with the therapeutic use of radium, radon and cobalt-60. New Eng J Med 251: 1–4, July 1954.

(39) Satterfield, R. W.: Caution signs for radium handling. Hospitals 33: 75–78, March 1959.

(40) Baily, N. A.: Facilities for handling and storage of medical radiation sources in a large hospital unit. Amer J Roentgen 83: 462–464, March 1960.

(41) Webb, H. P.: An improved radium safe. Brit J Radiol 33: 654–655, October 1960.

Education Notes

Graduate Course in Accident Control. The University of North Carolina School of Public Health invites applicants to enroll September 1964 in an 11-month graduate program of study in public health accident control leading to the master of public health degree. Designed primarily for students without prior preparation in public health, the program provides basic preparation in public health with emphasis on aspects of public health practice relating to accident control activities. Students with prior preparation in medicine, nursing, education, and the social and physical sciences will be considered.

Traineeships providing tuition and fees as well as a stipend for support of the student while enrolled are available. Write Dr. Charles M. Cameron, Jr., Department of Public Health Administration, University of North Carolina School of Public Health, Chapel Hill, N.C., 27515.

Short Course in Accident Control. A course in program development in public health accident control will be given for the second time at the University of North Carolina School of Public Health, May 24–29, 1964. It will deal with the fundamentals of program planning and development as related to initiating community and regional accident control activities. The course is designed for local and State public health personnel of all disciplines (physicians, nurses, health educators, environmental health personnel, administrative specialists), who are not engaged full time in accident control programs.

A limited number of traineeships are available. Address inquiries to Dr. Charles M. Cameron, Jr., Department of Public Health Administration, University of North Carolina School of Public Health, Chapel Hill, N.C., 27515.