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100 Years of radionuclide metrology

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HIGHLIGHTS

- The driving forces for the development of radionuclide metrology.
- Radium standards to facilitate trade of this valuable commodity in the early years.
- After 1950, focus changes to healthcare and industrial applications.
- National Measurement Institutes develop new techniques, standards, and disseminate the best practice in measurement.
- Challenges in nuclear medicine, radioactive waste management and nuclear forensics.

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ABSTRACT

The discipline of radionuclide metrology at national standards institutes started in 1913 with the certification by Curie, Rutherford and Meyer of the first primary standards of radium. In early years, radium was a valuable commodity and the aim of the standards was largely to facilitate trade. The focus later changed to providing standards for the new wide range of radionuclides, so that radioactivity could be used for healthcare and industrial applications while minimising the risk to patients, workers and the environment. National measurement institutes responded to the changing demands by developing new techniques for realising primary standards of radioactivity. Looking ahead, there are likely to be demands for standards for new radionuclides used in nuclear medicine, an expansion of the scope of the field into quantitative imaging to facilitate accurate patient dosimetry for nuclear medicine, and an increasing need for accurate standards for radioactive waste management and nuclear forensics.

1. Introduction

An internationally-recognised infrastructure for radionuclide metrology can be traced back to certification of the primary radium standards at national measurement institutes in 1913. To mark the centenary of the field, this article aims to give a very brief overview of the developments since then and their impact, and look ahead to the future. This article is also to acknowledge the contributions of the scientists working in national measurement institutes worldwide; the authors cannot hope to name all those involved, but refer the reader to a special edition of Metrologia (Simpson and Judge, 2007) and references therein. The authors are also indebted to comprehensive reviews of the history of radioactivity, including work by Malley (2011), Collé (2007), Allisy (1994) and Smith (1975).

The first part of the article covers the period when primary standards were samples of radium. A concise review is then given of the methods developed to replace the artefacts by more fundamental techniques. The value of the work depends on the difference radionuclide metrology has made to healthcare and industry, which is summarised in the penultimate section. The article concludes with some reflections on the future of the discipline.

2. The early years of radionuclide metrology – the Curie (Ci)

Radioactivity was discovered in 1896 by Henri Becquerel; two years later Marie and Pierre Curie had separated radium from

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uranium ore. The discoveries were made at a time of public fascination with science, and the new phenomenon was put to use very quickly – the first documented use of gamma rays from radium was reported by Dr. Danlos in the St. Louis Hospital, Paris in 1901 (Danlos and Bloch, 1901).

The Buchler company in Braunschweig started purifying and selling radium for research purposes in 1902. With many valuable applications, the cost of radium rocketed, reaching $100,000/g in the early 1920s. Laboratories working in the field had their own methods to quantify radium, but wanted to compare research results and trade radium. It was against this backdrop that the pioneers in the field, including Marie Curie and Earnest Rutherford, identified the need for an international measurement system for the new material.

The first Radium Standards Committee held its inaugural meeting at the International Congress of Radiology and Electricity in Brussels in 1910, chaired by Lord Rutherford. The committee proposed to define a new quantity named the curie as 'the amount of radon in equilibrium with 10^{-8} g of radium'. Marie Curie refused to allow the use of the name curie for such a small quantity; the committee relented and 1 Ci was defined to be the amount of radon in equilibrium with 1 g of radium (Rutherford, 1910). It was reported that Marie Curie accepted the proposal 'for the honour rendered to the memory of Pierre Curie' (although Rutherford later said it was in honour of both M. and Mme Curie).

Marie Curie was charged with realising the international primary standard and, in 1911, prepared a standard of 21.99 mg of pure radium chloride in a thin glass tube. The standard was delivered to the Bureau International de Poids et Mesures (BIPM) in Paris and it was deposited 'in the lower division of the safe located in the first vault'. Marie Curie went on to found the Institut du Radium in Paris in 1914: the Laboratoire Curie was dedicated to the study and measurement of radioactivity, issuing thousands of certificates in the following decades.

Duplicate standards of the BIPM standard were prepared at the Institut für Radiumforschung in Vienna for governments that wanted them. Standard no. 3 was obtained by the UK's National Physical Laboratory (NPL) at a cost of £354; it was received in December 1912 and certificated by Rutherford, Meyer and Curie on 2nd June 1913 with a claimed accuracy of 0.3%. Standards were also supplied to other countries: Standard no. 2 was delivered to the laboratory led by Hans Geiger at the PTR (the Physikalisch-Technische Reichsanstalt, the forerunner of the Physikalisch-Technische Bundesanstalt (PTB)) in Germany in 1912. By 1925, primary standards had been established in Paris, Brussels, London, Washington, Vienna and Berlin.

Over the next few years, there was a rapid expansion in the medical applications of radium (needles, tubes and plaques for cancer therapy and external beam radiotherapy) plus industrial applications such as luminous paint. Such was the demand that there was a public appeal for funding in the UK to purchase radium for the government. National measurement institutes faced a high demand for measurement services – NPL issued 6000 certificates in 1930 alone, the National Bureau of Standards (the predecessor to the National Institute of Standards and Technology (NIST)) issued 38,000 certificates between 1914 and 1942 – and certification of ^{226}Ra content and the determination of the ^{228}Ra impurity dominated the work.

The main scientific issues that researchers faced were self-absorption corrections and the effect of wall thickness on the gamma spectrum emitted (it had been found that radium needles tended to buckle if the walls were too thin, but increasing the wall thickness affected the results). Fragility of the original primary standards was also an issue, and new standards were prepared in 1934 by Hönigschmid (the copies were found to agree with the original standard to better than 0.3%).

3. The need for a new unit (the becquerel (Bq))

Frédéric and Irène Joliot-Curie discovered artificial radioactivity in 1934 when observing the effect of alpha particles bombarding targets made of boron and aluminium; (alpha, n) reactions were observed which produced ^{13}N and ^{30}P, the first man-made radionuclides. This was the beginning of a new era for the study, production and use of radionuclides, leading to the construction of particle accelerators and nuclear reactors (the first reactor was constructed by Fermi and Szilard at the University of Chicago in 1942). Fields such as nuclear physics, radiochemistry and radiation detectors developed rapidly, mostly as a result of the Manhattan Project.

Medical applications of the new radionuclides soon followed: the first delivery of ^{14}C from the Oak Ridge National Laboratory (USA) to a hospital occurred in 1946. Industrial-scale production of artificial radionuclides for medical and industrial applications began in the 1950s: a wide range of radionuclides became available in the 1950s and 1960s.

There were also growing concerns over the safety of using radioactivity. Worries over the health risks were growing, and the environment worldwide was contaminated with low levels of radioactivity due to both the weapons testing programme and discharges (both planned and accidental) from the expanding nuclear power industry. In response to these concerns, governments introduced stringent regulations on the use and disposal of radioactive materials to protect the general public, the workforce and patients undergoing diagnostic scans or cancer therapy using radiopharmaceuticals.

The increasing variety of artificial radionuclides and the need for accurate measurement to underpin the new regulations made it necessary to change the definition of the curie based on the radium standard. In practice, this transition was made in several steps. First, in 1950, the curie was re-defined to be the quantity of any radioactive nuclide in which the number of disintegrations/s is 3.7×10^{10} (this value had been recommended by the International Radium Standards Commission in 1930 and was used for all the early work on radionuclides). Second, the ICRU decided to replace the term 'quantity of a radionuclide' by 'mean number of nuclear transformations per unit of time', called 'activity' and the name 'curie' was kept for the unit associated to this new quantity and defined as 3.7×10^{10} s^{-1}. This was endorsed at the 12th Conférence Générale des Poids et Mesures in 1964 which defined the unit of activity to be the second to the power of minus one. Finally, in 1975, the 15th Conférence Générale des Poids et Mesures decided 'by reason of the pressing requirement, expressed by the International Commission on Radiation Units and Measurements (ICRU), to extend the use of the Système International d'Unités to radiological research and applications, by reason of the need to make as easy as possible the use of the units for non-specialists, taking into consideration also the grave risks of errors in therapeutic work, to adopt the following special name for the unit of activity (of a radioactive source): becquerel, symbol Bq, equal to one reciprocal second.' The special name becquerel was specifically introduced because of the dangers to human health that might arise from mistakes involving the unit reciprocal second, in case the latter unit was incorrectly taken to identify the different quantities involved.

4. Realising the Bq

Very soon the wide range of primary standards needed proved to be a challenge to researchers at national measurement institutes. Perhaps the first general technique for realising primary standards can be traced back to Dunworth in Cambridge in 1940