Current Landscape of Alternative Technologies for Calibration Applications

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Abstract

Radioisotopic sources are necessary for instrument calibration in numerous applications worldwide. According to a recent National Academy of Sciences report (NAS) (2021), there are over 19,000 radionuclide specific licenses in the United States alone at research facilities, industrial facilities, universities, hospitals, cancer treatment facilities, and nuclear medicine clinics. The Office of Radiological Security (ORS) encourages and facilitates the adoption of non-radioisotopic alternative technologies, including X-ray generators, electron beam (eBeam), and medical linear accelerators (LINACs), when viable options are available in an application space. Successful adoption of non-radioisotopic alternatives has been demonstrated in technologies used for blood irradiation, radiation for scientific research, sterile insect technique (SIT), and medical device sterilization. However, radioisotopic instrument calibration is an application that has not made this transition, in part due to a current lack of viable non-radioisotopic alternatives. Calibration systems with precise and traceable decay rates and energies are critical for periodic calibration and verification of proper device function of radiation technologies. End users and organizations that provide calibration services rely on calibration systems to ensure they are obtaining accurate measurements. Calibration systems primarily consist of IAEA Category 2 cesium-137 (Cs-137) and cobalt-60 (Co-60) sources, which are used to comply with health and safety licensing requirements.

The focus of this paper is the current use of calibration systems, their applications, and market availability of non-radioisotopic calibration alternatives.

What Are Alternative Technologies?

The mission of the National Nuclear Security Administration's (NNSA) ORS is to "enhance global security by preventing high-activity radioactive materials from being used in acts of terrorism" (Norris et al. 2022). ORS accomplishes this mission by implementing three strategies: Protect, Remove, and Reduce. Through the Reduce program, ORS encourages the development and adoption of non-radioisotopic alternative technologies to permanently reduce the risk of terrorism by eliminating the availability of high-priority radioactive materials.

ORS defines alternative technologies as technologies that do not contain radioactive materials and are able to perform an equivalent (or better) function compared to corresponding radioactive source-based devices. The function these devices provide is equally as important to the security of the radioactive sources used. Calibration systems is one application where viable alternatives have yet to be fully developed. Calibration systems produce radiation fields of known energy and intensity for calibration of radiation monitoring equipment, dosimeters to ensure their accurate operation (NAS 2021). Alternative technologies for radioisotopic calibration systems are desirable due to the potential risk of exposure to radioactive material, either through acts of terrorism, radiological accidents, or through accidental loss of radioactive sources

Study Background

According to the NAS, there are over 19,000 radionuclide-specific licenses in the United States (U.S.) alone, many of which are required to ensure compliance with government health and safety regulations (NAS 2021). Agencies and programs rely on instruments that require recurring calibration to provide and certify accurate measurements. Millions of calibrations are performed globally each year on radiation detection equipment in nuclear power plants, oil exploration, industrial and medical facilities, and for emergency response incidents. Finding 15 in the NAS 2021 report indicated that "No progress has been made domestically and internationally with adopting alternative technologies for calibration systems to replace cesium-137 and cobalt-60 sources. No obvious non-radioisotope alternatives exist for replacing the cesium chloride sources used in these applications, and there is currently no research and development dedicated to exploring alternatives. This poses an obstacle in global efforts to eliminate cesium-137 in the form of cesium chloride." As a result of this finding, the NAS issued the following recommendation: "The National Institute of Standards and Technology (NIST) should engage with the research community as well as federal, industry, and international partners to initiate research on alternatives to cesium chloride for calibration applications. This engagement should start immediately to prepare for the possible future elimination of the use of cesium-137 in the form of cesium chloride."

Since this recommendation, the Bureau International des Poids et Mesures (BIPM) has actively engaged the international metrology community, including all the primary metrology labs in the world to discuss the future of calibration metrology using ionizing radiation. After several conversations, the BIPM created a task group to engage the international community. To date, several meetings have been held on this topic and each time members have arrived at the same conclusion: there are currently no alternative technologies available to replace calibration gauges that use radioisotopes. The metrology community is mindful that they are a small user community and decisions made for more numerous Cs-137 applications will likely drive supply decisions. The community acknowledged the need for research and development (R&D) collaboration across metrology laboratories as well as the requirement to interface with experts who develop alternative technologies for other applications and learn from their experiences. The example cited was the highly successful transfer from self-shielded Cs-137 irradiators to X-ray irradiators for blood transfusion and research applications. The community expressed the need to identify where success has been achieved and build on these examples.

Radioactive Isotopes of Concern

There are many devices worldwide that contain risk-significant quantities of radioactive material. While these devices provide a variety of benefits, their use must be balanced against being adequately secured to prevent inappropriate use of radioactive materials. The NAS indicated that not only do radioactive sources continue to be broadly used, but there are more high-risk (Category 1 and Category 2) sources and very likely more Category 3 radioactive sources in the U.S. today than when the initial 2008 NAS report was released (80,000 Category 1 and 2 sources in 2021 vs. 54,000 in 2008) (NAS 2021).

There are currently thousands of different radioactive isotopes in use daily throughout the world, but not all pose the same risks if used in a radiation dispersal device (RDD) or "dirty bomb." Out of the thousands of manufactured and natural radionuclides, Cs-137, Co-60, iridium-192 (Ir-192), and americium-241 (Am-241) account for nearly all (over 99 percent) of the radioactive sources that pose the highest security risk (NAS 2008). Of these four isotopes, Cs-137 in the form of cesium chloride (Cs-137-Cl) presents the greatest concern based on its dispersibility and its presence in population centers throughout the world (NAS 2008 and 2021). As a result, the focus of this landscape study is on the use of Cs-137 in calibration systems and replacement challenges to the metrology community.

Cs-137 in Calibration Systems

U.S. and international systems of radiation measurements are based on the energy spectrum of Cs-137 (NRC 2008a). Cs-137 has been the basis for radioisotopic device calibration internationally for over fifty years due to its single energy spectrum (662 keV) and long half-life (30.2 years). Calibration systems generate radiation fields of known intensity which are used to calibrate radiation monitoring equipment and dosimeters to accepted current standards (NAS 2008). National and international protocols, recommendations, and standards all rely on the use of cesium irradiators for instrument calibration. The activity in calibration systems typically ranges from millicuries (mCi) (NRC 2010) up to 2,200 curies (Ci) (NAS 2008). Some of these calibrators are located at nuclear power facilities with stringent security requirements, but in 2008 over 100 of these systems were in locations other than nuclear power plants in the U.S. (NAS 2008). These systems commonly consist of radioactive sources, radiation shielding, a mechanism for positioning the source, and a track or internal chamber for positioning the items to be calibrated (NAS 2008). Calibration systems are disseminated to secondary laboratories, who in turn calibrate instruments for tertiary users. This network ensures that every radiation detection instrument used in the U.S. provides accurate measurements and is traceable back to NIST standards.

Stakeholders at the 2008 Nuclear Regulatory Commission (NRC) "Stakeholder Workshop on the Security and Continued Use of Cesium-137 Chloride Sources" discussed the potential impact on instrument calibration if there were changes to Cs-137 regulations or if Cs-137 calibration systems were eliminated. In summary, these changes would potentially be "catastrophic" for the calibration metrology community (Minniti 2008). First responder and emergency response equipment such as personnel self-reading dosimeters, portal monitors, and portable survey instruments used by the U.S. Department of Homeland Security all require the use of Cs-137 for calibrations (NRC 2008b) and eliminating Cs-137-Cl from use in calibration instrumentation could be detrimental to the nation's emergency response capabilities (Minniti 2008; NAS 2021). The combined challenge of finding an alternative to cesium and rewriting national and international standards and procedures is a daunting task. It is not surprising that given the long half-life of Cs-137, there is little enthusiasm for change within the metrology community. Lack of Cs-137 as a calibration standard would place regulators, radiation detection equipment manufacturers, users of radioactive material, and first responders worldwide out of regulatory compliance within a year as they would no longer be able to perform calibrations as legally required.

Replacing current NIST standards with less soluble/dispersible forms of Cs-137 could be acceptable for calibration applications because the Cs-137 energy spectrum would be maintained (NRC 2008a). However, there are currently no available replacements for Cs-137-Cl as a reference standard in calibration systems (NAS 2021). Until a new chemical or physical form of Cs-137 becomes available, calibration facilities will continue to rely on Cs-137-Cl. Any alternatives the metrology community has considered are associated with measurement uncertainties that essentially make calibrations less accurate.

There is concern that if cesium replacement projects are successful worldwide, manufacturers may decide that the market is unsustainable and stop offering Cs-137 calibration sources which would result in applications with no available alternatives being orphaned or abandoned. The only location where Cs-137 is currently separated and produced for commercial purposes is at the Production Association (PA) Mayak in Chelyabinsk, Russia (Pomper et al. 2014). NAS (2021) recommends R&D into alternatives to address this need effective immediately, including possible interim use of vitrified cesium. Within this same report, NAS indicated that evolution of a creative idea to a commercial product can take years to more than a decade (if successful) and requires substantial investments (NAS 2021). NAS recommends the NNSA prioritize funding of projects aimed at developing non-radioisotopic alternatives for applications where there are currently no acceptable alternative technologies (NAS 2021). NNSA has an opportunity to advance progress by identifying promising alternatives for these applications. Calibration systems were identified as one area where NNSA should prioritize funding (NAS 2021). In response, NNSA NA-22 included this topic in their FY24 Department of Energy (DOE) Small Business Innovation Research solicitation (SBIR) funding opportunities listing.

Assessment of Alternative Technologies for Calibration Systems

While no acceptable alternatives currently exist for calibration systems, it is not entirely accurate to state that there have been no R&D efforts dedicated to exploring alternatives. The French company ATRON, in collaboration with the French National Centre for Scientific Research (CNRS) and the Alternative Energies and Atomic Energy Commission (CEA) has developed an innovative method for calibration and verification of radiation survey meters which fulfills the needs of the French nuclear industry (Chapon et al. 2016). ATRON's calibration method uses braking radiation generated by a rapid deceleration or deflection of charged particles (particularly electrons) near strong electric fields produced by atomic nuclei. An electrostatic electron accelerator with a tantalum target is used to generate the braking radiation (Chapon et al. 2016) and the accelerator is tuned to produce X-rays between Cs-137 and Co-60 gamma energies (662 to 1332.5 keV) (Bordy et al. 2019). On-site air kerma measurements and recurrent calibration and verification provides traceability to French national references. According to Chapon et al. (2016), this method is closer to the actual use conditions of radiation survey meters while eliminating exposure risks for operators and the environment. In addition, the proportionality between the delivered dose rate and the intensity of the primary electron beam allows the dose rate response of the radiation survey meter to be measured over its full range by acting only on the accelerator parameters (Chapon et al. 2016). According to the NAS (2021), drift of the ATRON device is 0.3% over 11 months and the uncertainty in the calibration of radiation survey meters is +/- 2.1~~less than 7~~% (A. Chapon, personal communication, May 3, 2023), both which are considerably larger than the accuracy of Cs-137 calibrators (0.1% drift for months to years and 0.5% uncertainty for Cs-137) (NAS 2021). The NAS also notes that the working life of the tube is indicated at about 4,000 hours which is significantly less than the service life of a Cs-137 calibration system, potentially resulting in higher maintenance costs. Furthermore, calibration of the ATRON systems is still referenced back to the French national metrological facility which still uses Cs-137 (Chapon et al. 2016).

X-rays are currently used to calibrate the low energy range of detectors between 60 and 300 kV. X-ray tubes are presently available up to 450 KeV, which inches the metrology community closer to replacing

Cs-137 with X-rays. LINAC type sources start at 1 MeV which are currently available as alternatives to Co-60 calibration systems. For X-rays to serve as an alternative to cesium, development of X-ray tubes that span the gap in radiation standards between the upper limit of X-rays (300 kV) and the lower limit of LINACs (1 MeV) will be essential. Furthermore, the metrology community is hesitant to move to LINACs due to infrastructure issues in Low- and Middle-Income Countries (LMICs). This primary reason they continue to support Cs-137 for use in LMICs.

Researchers at the National Institute of Metrology (NIM) in Beijing, China built a graphite cavity ionization chamber to measure X-ray air kerma in the 250 to 450 kV range (Zhao et al. 2020). Five reference radiation qualities falling within this range were established between 250 to 450 kV using two different ionization chambers, a spherical graphite cavity ionization chamber and a plate free-air ionization chamber. The X-ray radiation qualities established were N-250, N-300, N-350, N-400, and N-450. Correction factors were determined for each of the radiation qualities tested. The relative standard uncertainty of the method proposed was 0.45%, and air kerma measurements taken from two different types of ionization chambers were in good agreement (0.09%) (Zhao et al, 2020). Researchers indicate that this study will help fill the gap in existing radiation standards.

Similar studies were performed at the National Metrology Institute of Japan (NMIJ) in Japan. As in the Chinese study, two ionization chamber designs were compared: a free-air ionization chamber and a graphite-walled cavity ionization chamber. The energy range of X-ray reference field measurements was expanded up to 450 kV and calibration coefficients obtained by both methods were in concurrence (Ishii et al. 2022). Authors noted that one potential disadvantage to the graphite-walled cavity ionization chamber was that measurements below 100 keV had high deviation. X-ray standards have been used in dosimeter calibration up to 210 KeV (free-air ionization chamber method), but gamma-ray emitters (commonly Cs-137 or Co-60) have been used for calibration at higher energies. Improving the energy range of X-ray standards is challenging because air kerma measurements made using free-air ionization chambers requires a prohibitively large air chamber volume at higher energies. Switching to a graphite cavity ionization chamber avoids the chamber volume issue (Ishii et al. 2022).

Research by NIMs in China and Japan indicate that the energy range of X-rays can be expanded up to 450 kV; while this remains well below the 662 keV energy of Cs-137, recent progress has been encouraging and is an area that deserves further research. Ronaldo Minniti (NIST) (R. Minniti, personal communication, January 26, 2023) indicated that it could be possible for an X-ray manufacturer to boost the energy of an X-ray tube to 700 kV, resulting in an X-ray energy approaching 600 keV. This would require a prototype or proof of principle, followed by fabrication and manufacturing before it could be made available to primary metrology laboratories. Once available, extensive testing would be required to transition from cesium-137 sources to higher kV X-ray calibration gauges, including revising national and international standards which are currently based on cesium. Documentary standards would need to be amended to ensure that the transition from cesium to X-ray calibration gauges is smooth and the current structure is not jeopardized. This is the impetus behind the NAS 2021 recommendation that alternatives be pursued effective immediately to help address the transition to alternatives for calibration gauges. The immediacy of this recommendation is an effort to address the “valley of death” which is the gap between evolution of a creative idea to a commercial product (Islam 2017).

Alternative Forms of Cs-137

In their “Status of Interagency Research to Identify a Lead Agency to Champion Development of Alternative Chemical Forms of Cesium-137” report, the NRC (2014) indicated that alternative forms of cesium have only been deployed in lower activity Cs-137 sources (< 10 Ci) in medical and well-logging applications. The report specified that the most promising alternative forms of cesium were phosphate ceramics and cesium aluminophosphate glass, but significant R&D was needed to confirm if high activity cesium sources (i.e., 400-2000 Ci) could be produced. Communications with private industry stipulated that without market demand, development of alternative forms of cesium is not financially viable (NRC 2014). One manufacturer specified that development of larger activity sources (up to 1,000 Ci) using alternative chemical forms of Cs-137 could be attainable in 5 to 10 years if market demand existed.

The NAS (2021) recommendation of possible interim use of vitrified cesium is in direct conflict with a 2014 NRC memorandum stating that “development of alternate forms of Cs-137 would not significantly reduce dispersibility and clean-up costs associated with the malevolent use of such sources. Therefore… staff is not recommending further Federal efforts to identify a lead agency or agencies to conduct research to facilitate development of alternative chemical forms for Cs-137” (NRC 2014). The most convincing argument for the pursuit of alternative forms of cesium is maintenance of the Cs-137 energy spectrum, which is critical to current calibration metrology needs both domestically and internationally. Less dispersible forms of cesium have been available for over 40 years, with solid Cs-137 in the form of fused glass being used in gauging and well logging sources. One disadvantage of using alternative forms of cesium is the increased source size required due to the lower specific activity of alternative forms. Another issue is that neither the technology has been proven nor are fabrication facilities available in the U.S. to scale up the activity level for phosphate ceramics and cesium aluminophosphate glass to levels necessary for calibration systems sources (i.e., 400-2000 Ci). Significant investment in R&D would be needed to determine if this capability could be developed for the manufacture of high activity sources.

However, examples of alternative forms of Cs-137 do exist. An innovative and cost-effective technique was developed, demonstrated, and deployed for the large-scale preparation of Cs-137 brachytherapy glass sources with a matrix activity of 40 mCi. Development of a novel immobilization technique enabled fabrication of sources in stainless steel capsules having uniform activity with a negligible leach rate (Dash et al. 2009). Another example where alternative forms of cesium are currently being deployed is in blood irradiators manufactured in India (Diwan 2022). Sources composed of vitrified Cs-137 poured into stainless steel pencils were developed at the Bhabha Atomic Research Center. This effort resulted from India’s pursuit of an alternative to Co-60 sources and involves recovery of cesium from nuclear waste. More than 1,200,000 Ci of Cs-137 was recovered within a period of one year (Kaushik et al. 2017). India now has a facility designed and commissioned for production of vitrified Cs-137 sources for use in blood irradiators. Ten pencil sources are used in each irradiator with single sources having an activity of approximately 300 Ci (Patil et al. 2015). Ronaldo Minniti (R. Minniti, personal communication, January 26, 2023) indicated that “curie levels between 100 and 500 Ci would fit most metrology community needs”, which is possible through efforts currently being deployed in India. Future applications being targeted for use of vitrified Cs-137 sources in India includes food irradiation, brachytherapy, and sterilization of medical equipment.

Conclusions

While the use of radioactive materials remains critical in calibration systems worldwide, it is encouraging to learn that the metrology community has acknowledged the need to collaborate across laboratories as well as the need to look where success has happened and learn from those experiences. Metrology laboratories currently rely on long-term stability of delivery systems and radionuclides for calibrations and any change to alternative technologies will be disruptive and may lead to lower performance. Cs-137 sources used in calibration devices are fundamental to calibration metrology laboratories as the gold standard for the calibration of radiation personal protection instrumentation of all types. To date, there has been little effort to develop alternatives to Cs-137-Cl sources. This is not to say that there is no appetite for alternatives, but alternatives need to be fit for purpose and at present no viable alternative technologies exist.

According to one stakeholder, "It is that simple… [impacts to] medical, research establishments, universities, industry, nuclear power, the armed forces, first responders and the public will be adverse if personal radiation protection instrumentation is not calibrated to show that it is responding correctly. The major concern would be health and safety could be jeopardized if new Cs-137 sources are not available and existing Cs-137 sources go to burial rather than being made available for recycling for applications such as the metrology community. One of the greatest concerns is that calibration laboratories will simply be viewed as acceptable collateral damage by government organizations focused solely on nuclear security" (M. F. Shepherd (JL Shepherd & Associates), personal communication, December 4, 2022). The reproducibility the cesium-137 spectrum provides cannot be replicated with X-rays at this time. Any alternatives the metrology community has considered are associated with measurement uncertainties that essentially make calibrations less accurate. According to Ronaldo Minniti (R. Minniti, personal communication, January 26, 2023), "when you talk to a scientist, we want to make things better, not worse. At the end of the day, at the metrology level we start with a low uncertainty because down the chain, when we transfer this measurement to the end user this uncertainty increases. So that's the reason we need to find an alternative that at least meets the specifications of cesium. As of today, we're not there yet.".

It is the opinion of the authors that the intent of the NAS 2021 recommendation is to challenge the metrology community into considering their future reliance on Cs-137. Currently, there is no looming deadline for the elimination of radioisotopic sources, but rather there should be recognition that if an incident involving the illicit use of radioactive sources were to occur the landscape for calibration gauges which rely on radioisotopic sources could change rapidly. R&D is being advocated to consider possible alternatives so stakeholders might prepare for a future transition to alternative technologies. Strategic influence from a national level is critical to the advancement alternative technology efforts. Without this leverage, organizations will continue to progress without meaningful direction, leadership, or the necessary authority to ensure accountability and an alignment of functions.

While development and adoption of alternative technologies continues to rise, governments throughout the world remain vulnerable to the financial exposure associated with the theft or accidental release of high-risk radioactive materials due to the lack of a national strategies for permanent risk reduction. This is the impetus behind the NAS recommendation to immediately engage federal, industry, research, and international partners to prepare for the possible future elimination Cs-137-Cl by pursuing alternatives for Cs-137 in calibration applications (NAS 2021).

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