RADIATION SAFETY ASSESSMENT OF MALAYSIAN CONSUMER PRODUCTS WITH POSSIBLE RADIOACTIVE ELEMENTS

HALMAT JALAL HASSAN

UNIVERSITI TEKNOLOGI MALAYSIA

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HALMAT JALAL HASSAN

A thesis submitted in fulfilment of the
requirements for the award of the degree of
Doctor of Philosophy

Faculty of Science
Universiti Teknologi Malaysia

AUGUST 2022

DEDICATION

I dedicate this thesis to my beloved parents, wife, sweet daughter and both sons, and closest friends with lots of love and gratitude. Each of you always makes me feel special, and thank you for believing in me.

ACKNOWLEDGEMENT

In the name of Allah, the Most Gracious and the Most Merciful. All praise to Allah, for His Mercy has given me patience and strength to complete this work.

First of all, I would like to express my most profound sincere appreciation to my supervisor, Prof. Dr. Suhairul Hashim, for his consultation, advice, and motivation to prepare this project, and also to my co-supervisor, Dr. Muhamad Syazwan Mohd Sanusi. Without their continued support and interest, this thesis would not have been the same as presented here.

I am also indebted to the librarian of UTM. Finally, thank you to all laboratories assistants and the Department of Physics for all of your help.

Finally, and most importantly, I am forever indebted to my loving parents, wife, daughter, sons, and family for their understanding, endless patience, and encouragement throughout the years. I had such a heart-warming experience, and such an experience will always be with me.

ABSTRACT

A wide range of consumer products containing radioactive materials are marketed for everyday use. Users are generally unaware of the presence of radioactivity in these products and the potential harm due to radiation exposures. Consumer products (CPs) containing naturally occurring radioactive material (NORM) continue to be sold in local and online markets in Malaysia, including lantern mantle, jewellery accessories, negative ion, healthcare, tourmaline products, tungsten welding rods, and gaseous tritium light sources (GTLS). A hyper-pure germanium detector was used to perform the gamma spectroscopy analysis and investigation of the spatial distribution of long-lived radionuclides (^{238}U, ^{232}Th, and ^{40}K) in these consumer products. The samples were prepared for elemental analyses which provide the composition of radioactive elements as well as rare earth elements (REEs) using X-Ray Fluorescence (XRF), inductively coupled plasma optical emission spectroscopy (ICP-OES), and X-Ray photoelectron spectroscopy (XPS). Monte Carlo (MC) simulations were performed to calculate the dose conversion factors (DCFs) that are used in estimating the equivalent organ doses and the annual effective dose (AED) from usage of these consumer products. Different scenarios in terms of use durations and consumer product placements on the human phantom were studied. The lantern mantle in sample M5 recorded the highest thorium activity at 12517 ± 1173 kBq, exceeding the exemption limit for thorium of 10 kBq adopted by the International Atomic Energy Agency (IAEA). Among the jewellery accessories products, glass pendant sample, GP11 recorded the highest activity at 6969 ± 483 and 687 ± 130 Bq of ^{232}Th and ^{40}K, respectively, while sample A14 (pendant pen shape) recorded the highest level of ^{238}U at 1168 ± 131 Bq. The annual effective dose for GP11 can reach up to 3 mSv y^{-1}, exceeding the public dose limit of 1 mSv y^{-1} recommended by the International Commission on Radiological Protection (ICRP). The glass disc coded A13 was estimated to give an AED of up to 2.5 mSv y^{-1} whereas the AED received from undergarment usage can be up to 2 mSv y^{-1}. However, the estimation of AED from the use of tourmaline products ranged between 0.8 µSv y^{-1} to 640 µSv y^{-1}. Currently, the investigated consumer products are available for purchase without information concerning their radioactivity content. Additionally, there is no impediment to the purchase of these products in Malaysia. There is a need to set specific criteria for the approval of these consumer products before their release for purchase and use by members of the public.

ABSTRAK

Pelbagai rangkaian produk pengguna yang mengandungi bahan radioaktif dipasarkan untuk kegunaan harian. Pengguna secara amnya tidak menyedari kehadiran keradioaktifan dalam produk ini dan potensi bahaya terhadap dedahan sinaran. Produk pengguna (PP) yang mengandungi bahan radioaktif semula jadi (NORM) terus dijual di pasaran tempatan dan dalam talian di Malaysia, termasuk mantel lantera, aksesori barang kemas, ion negatif, penjagaan kesihatan, produk turmalin, rod kimpalan tungsten dan sumber cahaya tritium bergas (GTLS). Pengesan germanium hiper-tulen digunakan untuk melakukan analisis spektroskopi gama dan penyiasatan taburan ruangan radionuklid hayat-panjang (^{238}U, ^{232}Th dan ^{40}K) dalam produk pengguna ini. Sampel telah disediakan untuk analisis unsur dan menyediakan komposisi unsur radioaktif serta unsur nadir bumi (REEs), menggunakan pendarflour sinar-X (XRF), spektroskopi pancaran optik plasma terganding beraruhan (ICP-OES) dan spektroskopi fotoelektron sinar-X (XPS). Simulasi Monte Carlo (MC) dilakukan untuk mengira faktor penukaran dos (DCFs) yang digunakan dalam anggaran dos organ setara dan dos berkesan tahunan (AED) semasa penggunaan produk pengguna ini. Senario berbeza dari segi tempoh penggunaan dan kedudukan produk pengguna pada fantom manusia telah dikaji. Mantel lantera dalam sampel M5 merekodkan aktiviti torium tertinggi pada 12517 ± 1173 kBq, melebihi had pengecualian untuk torium 10 kBq yang diterima pakai oleh Agensi Tenaga Atom Antarabangsa (IAEA). Antara produk-produk aksesori barang kemas, sampel loket kaca, GP11 mencatatkan aktiviti tertinggi masing-masing pada 6969 ± 483 dan 687 ± 130 Bq untuk ^{232}Th dan ^{40}K, manakala sampel A14 (bentuk pen loket) mencatatkan tahap tertinggi ^{238}U pada 1168 ± 131 Bq. Dos berkesan tahunan untuk GP11 boleh mencapai sehingga 3 mSv t^{-1}, melebihi had dos orang awam 1 mSv t^{-1} yang disyorkan oleh Suruhanjaya Antarabangsa Perlindungan Radiologikal (ICRP) untuk orang awam. Cakera kaca berkod A13 dianggarkan memberikan AED sehingga 2.5 mSv t^{-1}, manakala yang diterima daripada penggunaan pakaian dalam boleh sehingga 2 mSv t^{-1}. Walau bagaimanapun, anggaran AED daripada penggunaan produk turmalin adalah berjulat antara 0.8 µSv t^{-1} hingga 640 µSv t^{-1}. Pada masa ini, produk-produk pengguna yang disiasat tersedia untuk dibeli tanpa maklumat tentang kandungan keradioaktifannya. Selain itu, tiada halangan untuk membeli produk-produk ini di Malaysia. Terdapat keperluan untuk menetapkan kriteria khusus untuk meluluskan produk-produk pengguna ini sebelum dikeluarkan untuk pembelian dan digunakan oleh orang awam.

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LIST OF ABBREVIATIONS

AELB	-	Atomic energy licensing board
AERB	-	Atomic energy regulatory board
AED	-	Annual effective dose
ARPANSA	-	Australian radiation protection and nuclear safety agency
BE	-	Binding energy
CPs	-	Consumer products
DCFs	-	Dose conversion factors
ED-XRF	-	Energy dispersive x-ray fluorescence
EPA	-	Environmental protection agency
EURATOM	-	European atomic energy community
FWHM	-	Full width at half maximum
GTLS	-	Gaseous tritium light sources
HPGe	-	High purity germanium
IAEA	-	International atomic energy agency
ICP-OES	-	Inductively coupled plasma optical emission spectroscopy
ICRP	-	International Commission on Radiation Protection
ICSD	-	ionization chamber smoke detectors
MIRD	-	Medical internal radiation dose
NEA	-	Nuclear Energy Agency
NORM	-	Naturally occurring radioactive material
NRPB	-	National Radiological Protection Board
PHE	-	Public Health England
RCPs	-	Radioactive consumer products
TENORM	-	Technologically enhanced naturally occurring radioactive materials
TIG	-	Tungsten Inert Gas
XPS	-	X-ray photoelectron spectroscopy

LIST OF SYMBOLS

%	-	Percentage
Bq	-	Becquerel
Bq g^{-1}	-	Becquerel per gram
Bq Kg^{-1}	-	Becquerel per Kilogram
Cm	-	Centimeter
eV	-	Electron volt
g	-	Gram
Gy	-	Gray
h	-	Hour
hv	-	Energy of photon
^{40}K	-	Potassium-40
Min	-	Minute
MeV	-	Mega electron-volt
mL	-	Mili liter
mSv	-	Milisievert
mSv h^{-1}	-	Milisievert per hour
mSv y^{-1}	-	Milisievert per year
μSv	-	Microsievert
μSv h^{-1}	-	Microsievert per hour
μSv y^{-1}	-	Microsievert per year
°C	-	Degree celsius
mm	-	Millimeter
ppm	-	Part per million
^{232}Th	-	Thorium-232
^{238}U	-	Uranium-238
W	-	Watt
W_T	-	Tissue weighting factor
W_R	-	Radiation-weighting factor

CHAPTER 1

INTRODUCTION

1.1 Introduction

The International Atomic Energy Agency (IAEA) in GSR-Part 3, defined the term of “consumer products” (CPs) as “a device or manufactured item into which radionuclides have deliberately been incorporated or produced by activation, or which generates ionising radiation [hereinafter referred to as ‘radiation’], and which can be sold or made available to members of the public without special surveillance or regulatory control after the sale” [1].

Ionising radiation is a part of natural life and can be found naturally in the environment. Dominant in terms of radioactivity levels are the primordial radionuclides of thorium and uranium and their progenies (the so-called decay series), together with residual ^{40}K [2, 3]. From daily food intake to building materials, these radionuclides exist in traces or more substantial amounts [4, 5]. The extractive industries, including ore mining, and milling, discard radionuclides and alter their natural concentration and activity, which are also prevalent in the waste stream of the upstream oil and gas industry [6, 7]. This is referred to as Technologically Enhanced Naturally Occurring Radioactive Materials (TENORM), or in many cases, simply Naturally Occurring Radioactive Material (NORM) [8-10].

NORM as a term describes materials that contain radionuclides of natural origin and can be found almost everywhere, in soil, air, and consumer products [11]. The Canadian Nuclear Safety Commission (CNSC) defined NORM as the materials found in the environment that contain radioactive elements of natural origin and which contain uranium, thorium (elements that release radium and radon gas once they begin to decay) and potassium [12]. However, TENORM is used to describe natural radioactive materials in which the concentration of radionuclides is enhanced by man-made procedures [13]. Therefore, NORM has always been a part of our world. Exposure to ionising radiation emitted by ^{238}U, ^{232}Th, and ^{40}K, radionuclides of NORM, may cause risk to the environment and humans [14, 15]. NORM has a low radiation exposure dose in most environments. However, certain human activities can raise the radioactivity concentration level. Therefore, this may give rise to an above background radiation dose to consumers [16].

According to the IAEA guidelines for consumer products, three main distinct categories of consumer products are identified: (i) consumer products that have a small amount of NORM, such as an incandescent gas lantern mantle, radioluminous products containing gaseous tritium light sources (GTLS) or luminous paint; (ii) products that are able to generate radiation devices, for example, cathode ray tubes with the capability of producing x-rays; and (iii) products that contain activation products as a result of being intentionally exposed to radiation, for instance, irradiated gemstones, in which natural irradiation can occur naturally for an extended period of time. In contrast, artificial irradiation can be useful to increase the commercial value of the gemstones by enhancing the colour of the gemstones [17, 18].

Radioactive materials have been incorporated into a large variety of consumer products for many years, and this is the focus of present interest, in particular with regard to controls on safety. In the course of developing technologies in the modern age, the type and number of consumer products containing radioactive substances have increased markedly [19]. For the present research, radioactive materials include both natural and artificial radionuclides. Artificial radioactive materials, intentionally produced as radiation sources, are regulated by law in terms of their purchase, use, and disposal. However, in some instances, laws on radiation protection have excluded small amounts of naturally occured radionuclides, such as in the case of domestic smoke alarms [17, 18, 20].

The use of radioactive materials in medical treatments (radiotherapy and nuclear medicine) and in the nuclear fuel cycle, as in nuclear power plants, has established practices over many decades, tracing back to the latter part of the first half of the 20th century [21]. Conversely, in the 1920s, devoid of the knowledge of the risk of harm that we now possess, the word “atomic energy" gained particular popularity, promising an engendering of health-promoting effects, as for example, to "energise" the skin, rejuvenating it and providing a more youthful appearance. This was supported by the sale of cosmetics to be typically applied in the form of powder and creams containing uranium, radium, and other radioactive materials; kemolite, a volcanic mud included [16].

Apart from this, as a technological innovation, radium formed the basis of radioluminous paints, utilised from approximately 1914 through to as late as the 1970s, providing luminosity to watch and clock dials, allowing these to be read in the dark. Further to this has been the use of uranium to obtain sealed colour glazed ceramics and tiles, also providing iridescence in glassware [22]. Also, until relatively recent times, thorium was added to a number of products, prominently being used in lantern gas mantles, primarily because of its high melting point, yielding considerable white-light incandescence [23]. These are still in use in Malaysia's night markets [24].

Furthermore, consumer products containing NORM have been sold in online and local markets in Malaysia without information on the radioactivity contained within. For instance, scalar energy pendants, bracelets, and necklaces, cosmetic and healthcare products are obtainable by members of the public and maintain contact with the human body, especially with the skin [25, 26].

The manufacturers of cosmetic and healthcare products state that these products are composed of volcanic materials and/or other minerals. The purveyors and manufacturers link the supposed health benefits with negative ion technology and scalar energy, also using further jargon such as tourmaline products, negative ion products, and volcanic lava energy. They claimed health benefits include improved circulation and flexibility, with an ability to detoxify and enhance energy levels, also linked with the prevention of cancer [27]. Additional claims include that these products can maintain health, balance energy, and improve emotional well-being while also protecting against electromagnetic fields [28].

In many countries, the use of radioactive materials in consumer products is regulated. For instance, a Council Directive of the European Atomic Energy Community (EURATOM) prohibits the sale of foodstuffs, toys, personal jewellery, and cosmetics to which radioactive materials have been added. Unjustified exposures, as reported by the International Commission on Radiological Protection (ICRP-103), can be applied to a range of NORM consumer products [29]. If the benefits of the use of a consumer product to which NORM has been added cannot be shown to exceed the risk, then a ban would seem to be required [19]. Such products contain thorium and uranium, both concentrated in activity [26]. Given that over time, risks have been identified, NORM has become increasingly subject to monitoring and regulation [30]; the IAEA safety guide "Radiation Safety for Consumer Products" [31] recommends the use of such items to be justified. However, there appears no scientific proof that NORM products are good for the public [1, 31].

In order to limit radiation exposure from NORM-added consumer products, the Malaysian Atomic Energy Licensing Board (AELB) issued a directive entitled "Assessment and licensing of consumer products containing radioactive material" via its technical document (LEM/TEK/69), which established criteria for controlling consumer products [3]. This was preceded by the United States Environmental Protection Agency (EPA) guidelines for the regulation of NORM [32]. Despite such concerns, globally harmonized regulations for regulating and controlling radioactive content consumer products have yet to be established, [33].

Besides, there is still concern over increasing human exposure to radionuclides through the long-term use of particular consumer products that can cause significant health impacts on consumers, especially for children and senior citizens [34]. Although AELB established guidelines for consumer products, the regulation still does not provide a list of such consumer products or information on radioactivity levels, dose exposures, effective doses, and potential routes to exposure, external and internal. The guidelines also lack general information; for example, the annual number of manufactured products within the country or imported; and details of the point of sale that must be provided by suppliers, manufacturers, or distributors of such consumer products [22].

In terms of the exemption dose limit, the European Commission has introduced two-dose criteria for the gamma dose with an exemption criterion of 0.3 mSv y^{-1} and an upper limit of 1 mSv y^{-1}. Most of the European countries apply their control based on the upper limit, 1 mSv y^{-1} [22, 35], while the U.S., Canada, and Australia follow the IAEA exemption limit, in which 1 mSv y^{-1} dose limit for public exposure [36-38].

Similar to the EU states, there is a discrepancy in the exemption dose limit in Southeast Asian countries, which do not provide detailed lists of consumer products. For instance, in its LEM-TEK-69 regulation guideline, AELB dictates the effective dose for individual members of the public to be such that the practices are less than 10 µSv y^{-1} per each product [3]. The exemption dose limit for the general public in Indonesia is 0.5 mSv y^{-1}, 0.01 mSv per week, or 0.25 µSv h^{-1} [39]. However, Vietnam [40], Singapore [41], and the Philippines [42] reported the exemption limit to be 1 mSv y^{-1} for the public.

1.2 Problem statement

A variety of consumer products containing radioactive materials are widely marketed for everyday use. It is significant to ensure that consumer products contain as little radioactivity as practicable. Users generally are unaware of the radioactivity level of radionuclides contained in these products as well as the potential risk they may produce because of radiation exposure they may cause if used. Furthermore, the products should be assessed and approved before they are manufactured and supplied to the public. This is to ensure that even if these consumer products are handled in any manner and treated as domestic waste, the radiation doses to individual members of the public are small and hence constitute only a negligible radiation risk.

Under these circumstances, no special regulatory controls would be required to protect the public from unnecessary radiation exposure arising due to consumer products. The AELB should set the criteria for approving consumer products before they are released to the public. Furthermore, the standard specifications for consumer products should be developed. These standards are meant for compliance by the manufacturers of consumer products in order to meet the requirement of keeping the radiation dose to individual members of the public as low as reasonably achievable.

These standard specifications provide an important step towards the design and production of safer consumer products for public use.

The majority of countries have adopted the IAEA Specific Safety Guide SSG-36 as the standard for radiation safety guidelines on consumer products. In SSG-36, consumer products are additionally defined as products “which can be sold or made available to members of the public without special surveillance or regulatory control after-sale” [43]. In 2016, the AELB in Malaysia established technical advice (LEM-TEK-69) on consumer products to report the safety evaluation and implementation of consumer product handling [3]. The report defined consumer products as a “device or article or thing that contains the number of radioactive materials as determined by the appropriate authority” [3].

However, LEM-TEK-69 does not have detailed information on the radioactivity levels, effective doses, exposure doses, and potential routes. The LEM-TEK-69 report also lacks information (number of products manufactured or imported, and point of sale) provided by suppliers and manufacturers of consumer products (products that are subject to licensing, for example, ionisation chamber smoke detectors). For example, product availability, prohibited products, products that are subject to licensing, testing, and labelling, are crucial for guidelines and technical reports on consumer products containing radioactive material.

In order to obtain a comprehensive guideline and standard specifications on consumer products containing radioactive materials, it is proposed that a revision could be made to LEM-TEK-69 by including categories and types of consumer products used in Malaysia instead of mentioning ionisation chamber smoke detectors (ICSD) and pendants only. The focus may also be concentrated on healthcare products, gemstones, and accessories. A dose assessment is conducted on these products by assessing the dose of these items in households, offices, and the public domain. The radioactivity levels, dose assessment, and potential route figures of these products, and other aforementioned requirements previously discussed, are investigated.

1.3 Research Questions

Drawing from the problem statement, this research has three questions as follows:

1. How efficient and well-organized are the regulatory guidelines or national authorities in controlling consumer products being made or sold available in Malaysia to members of the public?
2. How much information or data about consumer products, such as a list of products is kept by the national authorities?
3. How efficient and structured is the process of licensing given to manufacturers?

1.4 Research objectives

This research aims to investigate the radioactivity contained and the concomitant radiation risk from long-term use of such consumer products. Hence, the objectives of this research are:

1. To identify and distinguish commercially available consumer products, including pseudo-products, by establishing baseline data on radioactivity level, equivalent, and effective dose received by the consumer.
2. To assess available quantitative data about the number of different types of consumer products obtainable in Malaysia.
3. To evaluate the worldwide requirements imposed by the competent authorities, such as, types of prohibited products and products that are subject to licensing, testing, labelling, and controlled disposal.
4. To propose particular guidelines for the assessment and licensing of Malaysian consumer products.

1.5 Scope of the study

This research focuses on broadening and reconditioning the current technical (LEM-TEK_69) by AELB. The research has two components: 1) inspection and tracking technique, and 2) empirical technique. The inspection and tracking technique is oriented towards identifying local and imported consumer products that fall under the following classification:

a) Products in which radionuclides (NORM or man-made sources) have deliberately been incorporated or added
b) The product contains naturally radioactive material (NORM)
c) The product that unintentionally contaminated with radioactive materials, specific isotopes, or irradiated.

The latter component (empirical) approach focuses on quantification of activity concentration as well as the dose received (absorbed, organ equivalent, and effective doses) and verification of the type of exempted products. The exemption and clearance levels utilised in this work are based on the AELB LEM-TEK-69 report. The research work excludes a few products that have been exempted by the competent official authorities in Malaysia according to the Atomic Energy Licensing Exemption Order (Atomic Energy Licensing Act 1984);

a) Smoke detector (Atomic Energy Licensing Exemption Order 1984)
b) Irradiating apparatus below 5 keV (Atomic Energy Licensing Exemption Order 2002)
c) Lightning arrester (Atomic Energy Licensing Exemption Order 1990)
d) Item- or product- associated isotopes or machines which produce radiation for medical diagnostic and therapy purposes.

This research describes consumer products in which radioactive substances are intentionally added, with members of the public having easy access to these without special control, including cosmetics and healthcare products, within the scope of consumer products.

1.6 Significance of research

Radioactive materials have been incorporated in a large variety of consumer products for many years, and this is the focus of present interest, in particular regard to controls on safety. In the course of developing technologies of the modern age, the type and number of consumer products containing radioactive substances have increased markedly. The situation is particularly concerning, with considerable numbers of consumer products purposefully incorporating radioactive materials in order to enhance the product function, all in the absence of consideration of health risks posed to the consumer. Indeed, via the Internet alone, it would appear that one can freely purchase such radioactive consumer products, devoid at the online purchasing level of any form of control. Internal and external exposure to humans as a consequence of the potential life-long utilisation of these consumer products could foreseeably cause significant health impacts to consumers, not least children and senior citizens.

It is significant for regulatory authorities of preparing proper guidelines and for public safety. These guidelines are meant for compliance by the manufacturers of consumer products in order to meet the requirement of keeping the radiation dose to individual members of the public as low as reasonably achievable. Such harmonization of practice must provide a significant step towards the design and production of safer consumer products for public use. For the first time this project is being conducted in Malaysia.

1.7 Thesis outline

The thesis is organised into five chapters. Chapter 1 discusses the research background, problem statement, research questions, objectives, scope, and significance of the research. Chapter 2 comprises the relevant literature reviews, which define the various types of radioactive consumer products as well as a description of the consumer products in terms of IAEA guidelines, and then review the data of available consumer products in worldwide countries. Furthermore, chapter 2 summarise the worldwide requirements and regulations imposed by the competent authorities, i.e., types of prohibited products and products that are subject to licensing, testing, labelling, and controlled disposal. The previous research data on consumer products and healthcare products were discussed. Chapter 3 describes the details of the sample preparation, experimental procedures, and the description of measurement techniques as well as estimation of effective dose by Monte Carlo (MC) simulation, that have been used in this research work. Chapters 4 discuss the results of various study investigations and analyses based on the experiments. Furthermore, chapter 4 discuss the radiation safety guidance of consumer products and gives recommendations to revise the LEM/TEK/69 documents, based on the finding in this research. Finally, chapter 5 presents conclusions drawn from the overall results and recommendations for future work.

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LIST OF PAPER CONFERENCE PROCEEDING

1. Hassan, H.J., S. Hashim, N.Z.H. Abu Hanifah, M.S.M. Sanusi, M.R.Fahmi, R.M. Tahar, and D.A. Bradley. Dose Assessment with NORM added Consumer Products Using Geant4 Monte Carlo Simulations. *Presented in conference ISRP-15 (2021).*
2. Hassan, H.J., S. Hashim, N.Z.H. Abu Hanifah, M.S.M. Sanusi, M.R.Fahmi, R.M. Tahar, and D.A. Bradley. (2022). Use of Tourmaline-based Healthcare Products and Associated Radiation Risks. *Presented in ISRP-15 conference (2021).*
3. Hassan, H.J., S. Hashim, N.Z.H. Abu Hanifah, M.S.M. Sanusi, and D.A. Bradley. Radioactivity in the Living Environment: A Need for Harmonization of Legislative Control. *International Conference on the Safety and Security of Radioactive Sources: Accomplishments and Future Endeavours (CN-295)-(IAEA).* (20-24 June 2022).