Publishable Summary for 19ENV02 RemoteALPHA
Remote and real-time optical detection of alpha-emitting radionuclides in the environment

Overview

Radiological emergencies involving an accidental or deliberate dispersion of alpha emitting radionuclides in the environment can cause significant damage to humans and societies in general. A detection system to measure large-scale contamination of these radionuclides was not available and new remote detection techniques which overcome shortcomings of traditional detectors were needed. This project has developed novel instrumentation and methods and a sustainable metrological infrastructure for outdoor-detection systems, which can detect remotely alpha-emitting radionuclides in the environment. This includes two lens-based radioluminescence detection systems, a novel calibration methodology based on radiance standards, environmental standards, and an unmanned aerial monitoring system. This will lead to real-time collection of traceable radiological data and faster, more reliable information for the decision-making authorities.

Need

Alpha particles represent the biggest risk to soft biological tissues compared to all nuclear decay products due to their high energy, large mass and high linear energy transfer. The amount of deposited energy is about 2 000 000 to 6 000 000 times higher than that of an ordinary chemical reaction (ordinary chemical energy used by the cells in the body), thus implying that a single alpha particle has the ability to severely damage or kill all cells within its range (typically, two to four cells). Therefore, the release of alpha emitting radionuclides in the environment, such as by nuclear terroristic attacks or transportation accidents, as well as by severe emergencies in nuclear installations, represents the greatest radiological threat for human beings if they enter the human body.

A detection system to measure large-scale contamination of these radionuclides was not available. In case of an emergency, the only option is to evacuate the population from the affected areas and then run diagnostics by hand, thus exposing the emergency teams to considerable risk. Even then, the results of emergency field applications are notoriously ambiguous, time consuming and tedious due to the centimetre range of the alpha particles in air.

Instrumentation and methods that overcome the disadvantages of existing detectors and allow remote detection of alpha particles in the environment were thus required to be developed, lowering personnel risk, detection costs, and time. Two radioluminescence detection systems were developed to establish the metrological basis for the optical detection of alpha-emitting radionuclides. One detection system is based on a high-quality UV fused silica lens (UVFS), while the other uses a PMMA Fresnel lens. Both systems operate as mapping systems by generating the radioluminescence image of alpha radiation sources by remotely scanning a narrow field of view over the user-defined region of interest while recording the photon count rate. While the UVFS system was optimized for use as a scanning telescope on a tripod due to limitations in its weight and mechanical stability, the Fresnel lens-based lens system was optimized for use as an unmanned airborne monitoring system (UAMS) for mapping alpha contamination in the environment. The UAMS is based on the unmanned aerial vehicle (UAV) DJI Matrice 600 Pro and uses the RIMASpec software architecture which allows the real-time viewing of alpha contaminations. The techniques aim at ensuring an adequate level of preparedness and response, and assist on-site incident management, creation of evacuation plans as well as in developing strategies for protecting public from harm. These measures are required by the European Union (EU) legislations defined in the Council Directive 2013/59/EURATOM and are compulsory for all EU Member States.

The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Objectives

The overall objective of this project is to develop novel optical systems for the remote detection and quantification of large-scale contamination with alpha emitters in the outdoor environment for the first time, allowing sound and quick countermeasures in the case of a radiological emergency.

The specific objectives of the project are:

1. To develop a new method and instrumentation for the optical detection of alpha particle emitters in the environment by air radioluminescence over a detection range of more than two metres. This includes the development of the first prototype of a mobile-outdoor optical detection system for real-time radioluminescence mapping of alpha sources in the environment.
2. To develop and establish a calibration system for the novel-type radioluminescence detector systems. This includes a new metrological infrastructure with a dedicated UV radiance standard, well characterised alpha-active environmental sample (mineral-phase, soil, organic and plant specimen spiked with alpha emitters) and a validated calibration scheme for the remote detection of optical system.
3. To extend the optical detection system to an imaging functionality for mapping of alpha contaminations in the environment. This includes the development of an unmanned airborne monitoring system (UAMS) that will integrate the unmanned aerial vehicle (UAV) and the novel alpha-radioluminescence detection system developed in the objective 1 to scan and obtain an image of the contaminated area.
4. To prepare and run a feasibility study for a laser-induced fluorescence spectroscopic method for the detection of alpha emitters. This method complements alpha-radioluminescence and, depending on laser parameters such as pulse power, photon wavelength and pulse duration, can enhance the detectable activity limit to below 1 kBq/cm^2.
5. To facilitate the take up of the results by stakeholders and provide input to relevant standardisation bodies and radiation protection authorities. Information on the project research results will be disseminated by the partners to standards committees, technical committees and working groups such as EURADOS, ISO, IEC, IAEA, BIPM CCRI (I)-(II), ICRM and EURAMET TC-IR. In addition, knowledge will be transferred to the nuclear industry sector.

Progress beyond the state of the art

Objective 1: Two optical detection system optimized to collect large numbers of radioluminescence photons were developed. The radioluminescence mapping capability was demonstrated with (a) accelerated alpha particles and dedicated ^{241}Am sample designed to simulate an extended alpha source [5], (b) environmental samples such as pitchblende minerals [4] and ^{241}Am-spiked sand, soil and leaf samples [8], (c) ^{241}Am-spiked concrete samples, (d) a ^{210}Po activity standard and radiance standards [8], (e) depleted uranium nuclear waste materials [8], and with an UAMS from a 5 meter height [7]. Monte Carlo simulation routines based on Geant 4 and FLUKA were also developed to simulate radioluminescence [1-2,9].

Objective 2: To facilitate the deployment of novel-type alpha-radioluminescence detection systems developed in the objective 1, this project has established for the first time two SI traceable calibration procedures [4,7-8]: (a) application of well-characterised activity standards such as ^{210}Po activity standard and environmental samples to establish a traceable relationship between radioluminescence intensity and alpha activity, and (b) the use of all-optical radiation-based devices (UV-A and UV-C radiant standards) that, when calibrated against an alpha activity standard, simulate the radioluminescence induced in nitrogen (N_2) and nitric oxide (NO) gases by alpha particles in specific spectral regions.

Objective 3: An unmanned airborne monitoring system (UAMS) for automated scanning and imaging of large, contaminated areas has been developed. The UAMS comprises a fit-to-purpose DJI Matrice 600 Pro, the optical detection system developed in objective 1 and calibrated in objective 2, real-time data transmission and analysis module, and the ground control station [6]. The UAMS hardware uses the RIMASpec software architecture with a web application developed to generate flight plans and visualise the count rate on a map in real time [6].

Objective 4: A rate-equation model to determine the dynamics of fluorescent transitions in different gas environments has been developed.

Results

Objective 1

Two radioluminescence detection systems have been developed and optimized to establish the metrological basis for the optical detection of alpha-emitting radionuclides. One detection system is based on a high-quality UV fused silica lens (UVFS), while the other uses a PMMA Fresnel lens. Both systems operate as mapping systems by generating the radioluminescence image of alpha radiation sources by remotely scanning a narrow field of view over the user-defined region of interest while recording the photon count rate. While the UVFS system was optimized for use as a scanning telescope on a tripod due to limitations in its weight and mechanical stability, the Fresnel lens-based lens system was optimized for use as an unmanned airborne monitoring system for mapping alpha contamination in the environment [5].

The UVFS lens system has been used to quantify total surface activities of pitchblende minerals [4], ^{241}Am-doped environmental samples (leaves, sand, and soil) and nuclear materials involving a standard ^{239}Pu source, a set of depleted uranium sources with complex geometry, and a UO_2 pellet [7]. The ^{241}Am-doped environmental samples and nuclear materials were selected to test the feasibility of radioluminescence instrumentation and methods for scenarios encountered (a) when fallout containing alpha emitters (in the form of radioactive dust or washout during rain) is deposited in common environmental materials such as soil, sand, concrete, leaves, etc., and (b) during decommissioning of nuclear facilities where alpha emitters are produced, handled, used, and stored. Activities of up to 290(90) Bq could be determined in air. With N_2+NO flushing, activities as low as 51(7) Bq were detected in contaminated leaves. The activities deduced from the radioluminescence measurement in air compare well with those measured using the triple-to-double coincidence ratio (TDCR) technique [7]. For higher activity (> 164 Bq) leaf and soil samples, the results agree to within 7%. In the N_2+NO atmosphere, the disagreement is larger (about 25%) predominantly due to an uneven gas flow through the chamber. Very low activity (< 164 Bq) environmental samples have been quantified only in UV-C under an N_2+NO atmosphere with an agreement better than 13%. Concrete samples doped with ^{241}Am to surface activities ranging from 0.161 MBq to 18.44 MBq have been also successfully mapped with the fused silica detection system from a reference distance of 2 m. Since concrete is a widespread material in urban areas, these sample would simulate a scenario of deliberate or accidental dispersion of alpha emitting radionuclides in urban areas.

A dedicated ^{210}Po alpha activity standard with a sharp peak of less than 32 keV FWHM at 5.3 MeV and a surface activity of 648 kBq has been used to characterise the fused silica lens system in terms of its sensitivity to alpha-induced radioluminescence in different atmospheres (air, N_2, N_2 + NO mixture) in the UV-A and UV-C (solar blind) spectral regions. The UV-A sensitivity measured in air, 662(44) $s^{-1}MBq^{-1}$, compares well with the value measured with reference sources of ^{239}Pu, 760(100) $s^{-1}MBq^{-1}$, under similar experimental conditions. In the N_2 atmosphere, the sensitivity is increased by factor of 8, which is due to the removal of quenchers such as oxygen and humidity from the chamber. The use of N_2 + NO mixture led to about 25% increase of the sensitivity relative to N_2. The effect of N_2+NO purging is especially apparent in case of UV-C spectral region. Here, an increase of sensitivity by more than three orders of magnitude relative to the air measurements is observed. The UV-C sensitivity in air is 34.1(24) $s^{-1}MBq^{-1}$.

The instrumentation and methodology for radioluminescence mapping of the contaminated area and the derivation of activity was developed. In addition, the prerequisites (purging conditions and gas composition) under which the radioluminescence signal is amplified by more than three orders of magnitude were determined. With these developments, objective 1 was fully achieved.

Objective 2

Sixteen environmental samples have been prepared and characterised to facilitate the deployment of novel-type alpha-radioluminescence detection systems in the environment. These include pitchblende minerals (10 samples) [4] and, sand, soil, and leaves (2 samples each) spiked with standard solution of ^{241}Am1[7].

Two low flux UV radiance standards operating in UV-A and UV-C spectral regions, respectively, have been radiometrically characterised in terms of lateral homogeneity, angular distribution and absolute photon radiance. The absolute photon radiance was calibrated by comparison with the PTB radiance primary standard, the High Temperature Blackbody HTBB3200pg. They simulate radioluminescence induced in nitrogen (N_2) and nitric oxide (NO) gases by alpha-emitting sample of a circular shape with a diameter of 25 mm. The UVFS lens-based reference instrument has been used to cross-calibrate both radiance standards in terms of alpha activity. Both radiance standards can be used as transfer standards simulating up to 510 MBq when used in the UV-A and up to 8.7GBq when used in the UV-C spectral regions.

A well-characterised ^{210}Po activity standard to establish a traceable relationship between the radioluminescence signal and alpha activity has been developed. The ^{210}Po source was characterized in a Defined Solid Angle α-Spectrometer where the alpha spectrum has been measured. The source has a peak at about 5.3 MeV with energy spread (FWHM) of only 26.8(7) keV at 69° to 31.9(16) keV measured at 0° relative to the surface normal.

A new calibration methodology based on two complementary approaches has been developed: (a) application of a well-characterised ^{210}Po standard and environmental samples [7,8] and (b) use of low flux UV radiance standards [8]. With these developments, objective 2 was fully achieved.

Objective 3

An Unmanned Aerial Monitoring System (UAMS) integrating the DJI Matrice 600 Pro unmanned aerial system (UAS) and the PMMA Fresnel lens-based radioluminescence detection system was developed to scan and map the contaminated area [6]. The Fresnel lens system was built with carbon fiber reinforced polymer frame to reduce weight and to ensure the required mechanical stability. The UAMS hardware uses the RIMASpec software architecture already successfully used in the previous gamma detection projects (e.g., EMPIR project 16ENV04 Preparedness). For the RemoteALPHA project, the software was upgraded to visualize a map of alpha-emitting radionuclides in the environment in real time [6].

Investigation of the detector response was first performed under light-controlled conditions with UV LEDs to determine the field of view (FOV) and the best focusing configuration for the flights. In addition, the change in efficiency as a function of the angle of incidence was also investigated. Following the choice of the optimum height of 5 m, the flight speed is primarily determined by the FOV of the detector. With the FOV of about 10 cm, the measurement time of 100 ms necessitates flying at a speed of 1 m s^{-1}. The flight planner software module of the RIMASpec ground control station can generate flight plans for the UAV that cover an area with a back-and-forth pattern which allowed detection of one 100 MBq extended ^{241}Am source with 20 mm x 100 mm active area, and 5 UV-C LEDs [6]. The probability of detecting a point source constitutes approximately 60%. This probability can be boosted by increasing the field of view of the detection system. However, increasing the FOV also has some other consequences such as the reduction of the detector efficiency and background light leakage. Extended sources with larger lateral dimensions (e.g., on the order of GPS uncertainty), on the other hand, would not be susceptible to difficulties encountered in the case of point sources.

In summary, the lightweight Fresnel lens-based optical detection system developed in objective 1 has been extended to an imaging functionality for mapping of alpha contaminations in the environment. The UAMS hardware uses the RIMASpec software architecture and was used to detect 100 MBq ^{241}Am source and 5 UV LEDs that simulate distribution of point alpha sources from a height of 5 m. With these developments, objective 3 was fully achieved.

Objective 4

A rate equation numerical model to simulate potential re-excitation yields in nitrogen molecules in air has been developed. Oxygen is found to quench the most of the potential radioluminescence states in air. A model has been developed to study how oxygen quenching works for nitric oxide. Laser re-excitation measurement with a wavelength tunable Optical-Parametric-Oscillator (OPO) -laser (Expla NT342) were performed. The laser with a pulse width of 4 ns and energy of 1 mJ was directed 4 mm over americium alpha source that has total activity of 32 MBq. The source was a 60 mm long and 3 mm wide stripe, and the laser beam was aligned along the stripe. The laser-induced emission of nitrogen ions was not detected at the nitrogen ion concentration of 10^8 ions/cm^3 which could be achieved with the used laser. The re-excitation and detection efficiency would be increased by one to two orders of magnitude by choosing a laser with a narrower linewidth and higher pulse energy. Narrower linewidth laser is absorbed more efficiently by the narrow spectral lines of nitrogen ions, and more pulse energy directly increases the amount of radioluminescence signal.

The objective of this feasibility study was only partially achieved, as the detection limit below 1 kBq/cm^2 was not reached. Nevertheless, the models for predicting the dynamics of the excited states related to radioluminescence were developed and the experimental procedure for carrying out laser-induced radioluminescence measurements was established.

Impact

The objectives and results of the project have been presented at more than 40 conferences and workshops including national conferences on radiation protection and radiation fields, global conferences on radiation topics and standards and regulatory meetings.

The project generated two exploitable products: (a) UV fused silica lens-based radioluminescence detection system for mapping contamination with alpha emitting radionuclides, and (b) low photon flux UV radiant standards for calibrating radioluminescence detection systems. The sectors of applications include nuclear industry, radiation protection, emergency preparedness and response, and UV radiometry.

Impact on industrial and other user communities

The feasibility of the developed UVFS radioluminescence system to detect and map low activity environmental and uranium samples has illustrated the potential application of the system towards item (source, container) integrity checks during the transfer and storage of nuclear materials. With the optical detection method, the hotspot localisation and quantification can be done on a sample of arbitrary shape and size without limiting the inspection to flat external surfaces. The Safeguards System at IFIN-HH, Romania is planning early uptake of the alpha imaging technology developed in RemoteALPHA project. It is foreseen that the optical method will be first implemented for detecting contamination on objects that are brought under safeguards to IFIN-HH or on objects already stored there. Wider use of the developed technology for nuclear safeguards and nuclear forensics applications is also considered.

Impact on the metrology and scientific communities

A novel calibration methodology has been developed to provide valuable information about, and confidence in, the performance of radioluminescence detection systems. The proposed calibration methodology is based on two complementary approaches: (a) application of well-characterised activity standards (^{210}Po source) to establish a traceable relationship between radioluminescence intensity and alpha activity, and (b) use of all-optical radiation-based devices (radiance standard) that, when calibrated against an alpha activity standard, simulate the radioluminescence induced in nitrogen (N_2) and nitric oxide (NO) gases by alpha particles in specific spectral regions. These radiance standards simplify substantially routine quality control of radioluminescence detection systems by eliminating the need for open alpha sources, which are always associated with strict radiation safety precautions. Furthermore, since the intensity of radiance standards is adjustable over a very wide range, linearity and detection limits of radioluminescence detectors can be readily determined.

Impact on relevant standards

The project has provided guidance for stakeholders and input to international standardisation bodies such as ISO as far as nuclear and radiological emergency preparedness is concerned. The methodology developed in this project will set the basis for new standards for the remote measurement of alpha emitters. Current standards such as ISO 8769:2020 (Measurement of Radioactivity) do not include neither the radioluminescence method nor radiance standards as a means of detecting and calibrating, respectively, alpha emitters. The project will help to fulfil the IAEA requirements listed in the Convention on Early Notification of a Nuclear Accident and in the Convention on Assistance in the Case of a Nuclear Accident or Radiological Emergency of the European Commission.

Longer-term economic, social and environmental impacts

The wider long-term impact of this project is to allow for a rapid, coordinated and effective response in emergency situations involving dispersion of alpha emitting radionuclides in the environment. The development of new calibration procedures for radiometric traceability of radioluminescence detection systems, will enable appropriate accident and post-accident radiation measurements that will lead to more effective countermeasures and better protection of people, wildlife, and the environment. The instrumentation and methodology developed in this project will assist response teams to assess the breakdown phase (i.e., the initial location of the accident and whether the cause of the accident is moving or fixed). It will help authorities take immediate targeted action for the public protection, including measures to reduce panic and prevent unnecessary chaos by providing the public with reliable data on the spread of radioactive particles.

More accurate determination of the extent of land contamination will help to reduce the area designated for exclusion and evacuation zones, thereby minimising associated follow-up costs.

List of publications

[1] I. Lalau and M.-R. Ioan, Simulation of radioluminescence induced by alpha particles in the air by Monte Carlo method, Ninth International Conference on Radiation in Various Fields of Research, June 2021. https://doi.org/10.21175/RadProc.2021.07

[2] I. Lalau and M.-R. Ioan, Modelling the radioluminescence of alpha particles in the air by the Monte Carlo method, Ninth International Conference on Radiation in Various Fileds of Research, June 2021. https://doi.org/10.21175/rad.abstr.book.2021.30.6

[3] M. -R. Ioan, I. Radulescu, M. Zadehrafi, L. Tugulan and C. Barna, Proiecte naționale și europene de metrologia radiațiilor, suport pentru implementarea Directivei 2013/59, în sănătate și protecția mediului, SOCIETATEA ROMÂNĂ DE RADIOPROTECŢIE, CONFERINȚA NAȚIONALĂ ANIVERSARĂ A SOCIETĂȚII ROMÂNE DE RADIOPROTECŢIE – „SRRp-30", ISBN: 978-973-1985-64-0 https://srrp.ro/wp-content/uploads/2020/12/Conf.Nat_._SRRp_-2020-A4-ver.12.pdf

[4] A. Klose, M. Luchkov, V. Dangendorf, F. Krasniqi, A. Lehnert, C. Walther, On the way to remote sensing of alpha radiation: radioluminescence of pitchblende samples, Journal of Radioanalytical and Nuclear Chemistry 2022, https://doi.org/10.1007/s10967-022-08540-6

[5] M. Luchkov, V. Dangendorf , U. Giesen, F. Langner, C. Olaru, M. Zadehrafi, A. Klose, K. Kalmankoski, J. Sand, S. Ihantola, H. Toivonen, C. Walther, S. Röttger, M.-R. Ioan, J. Toivonen, F. S. Krasniqi, Novel optical technologies for emergency preparedness and response: Mapping contaminations with alpha-emitting radionuclides, Nuclear Inst. and Methods in Physics Research, A 1047 (2023) 167895, https://doi.org/10.1016/j.nima.2022.167895

[6] Royo, P.; Vargas, A.; Colls, T.G.; Saiz, D.; Pichel Carrera, J.; Rabago, D.; Duch, M.A.; Grossi, C.; Luchkov, M.; Dangendorf, V.; Krasniqi, F. Mapping of Alpha-Emitting Radionuclides in the Environment Using an Unmanned Aircraft System. Remote Sensing (2023). Available at https://doi.org/10.20944/preprints202310.1698.v1

[7] M. Luchkov, C. Olaru, I. Lalau, M. Zadehrafi, I. R. Nikolényi, Z. Gémesi, M.-R. Ioan, F. Krasniqi, Radioluminescence mapping of 241Am-doped environmental samples and nuclear materials, Journal of Radioanalytical and Nuclear Chemistry (2023), https://doi.org/10.1007/s10967-023-09235-2

[8] F. S. Krasniqi, R. D. Taubert, S. Röttger, M. Luchkov, F. Mertes, A. Honig, C. Baltruschat, V. Dangendorf, U. Giesen, A calibration methodology for the novel radioluminescence detector systems, , https://doi.org/10.7795/EMPIR.19ENV02.PA.20231027

[9] C. Olaru, M.-R. Ioan, M. Zadehrafi, Modeling alpha particle-induced radioluminescence using GEANT4, Modeling alpha particle-induced radioluminescence using GEANT4, Romanian Journal of Physics (2023), Accepted; https://doi.org/10.48550/arXiv.2310.14601

This list is also available here: https://www.euramet.org/repository/research-publications-repository-link/

Project start date and duration:	01 September 2020, 36 months	
Coordinator: Faton KRASNIQI, PTB Tel: +495315926223 E-mail: faton.krasniqi@ptb.de Project website address: http://remotealpha.drmr.nipne.ro/index.php		
Internal Funded Partners: 1. PTB, Germany 2. BFKH, Hungary 3. IFIN-HH, Romania	External Funded Partners: 4. ALFA RIFT, Finland 5. LUH, Germany 6. TAU, Finland 7. UPC, Spain	Unfunded Partners: 8. MATE, Hungary
RMG1: IFINN-HH, Romania (Employing organisation); PTB, Germany (Guestworking organisation)		