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. Currently, a detection system to measure large-scale contamination of these radionuclides is not available and new remote detection techniques which overcome shortcomings of traditional detectors are needed. This project will therefore develop novel instrumentation and methods and a sustainable metrological infrastructure for outdoor-detection systems, which can detect remotely alpha-emitting radionuclides in the environment. 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 is currently 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 therefore need to be developed which overcome the shortcomings of traditional detectors and that will allow remote detection of alpha particles in the environment, reducing risk to personnel, detection costs and time. The techniques should 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.

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.

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

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

New method and instrumentation for the optical detection of alpha particle emitters in the environment by air radioluminescence

Optical detection of alpha particle emitters in the environment by air radioluminescence is a new technology that enables sensing of a radiological threat at safe distance, without putting first respond personnel and equipment in harm's way. This standoff detection technique gives advanced warning of potential threats, in contrast to conventional detectors for alpha contamination which are built in a hand-held format and must be positioned within a few centimetres of the source in order for the alpha radiation to be detected. At the heart of this novel technology developed in this project is an optical detection system, optimised to collect large numbers of radioluminescence photons. The instrument can be operated as a standalone device, for example, operated from an UAV vehicle or deployed on a vehicle and then operated from a tripod.

Development of a calibration system for the novel-type radioluminescence detector systems

To facilitate the deployment of novel-type alpha-radioluminescence detection systems developed in the objective 1, this project will establish SI traceable calibration procedures, new and specific calibration schemes and novel portable calibration standards. This state-of-the-art portable calibration standard will also allow the temporal stability of the radioluminescence detector, including potential wavelength shifts of the (interference) filters and changes in the spectral responsivity of the photomultiplier-based detectors to be monitored, hence contributing to quality assurance.

Extending the optical detection system to an imaging functionality for mapping of alpha contaminations in the environment

Mapping large-scale contaminations with traditional hand-held alpha detectors exposes operators to immense risks, on top of other difficulties such as contamination of the detector and poor areal resolution. Therefore, this project will develop an unmanned airborne monitoring system (UAMS) that enables automated scanning and imaging of large contaminated areas. The UAMS comprises a fit-to-purpose UAV, 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. Procedures to calibrate UAMS based radiation monitoring systems will be developed and validated using metrologically sound approaches, for the first time.

Feasibility study for a laser-induced fluorescence spectroscopic method for the detection of alpha emitters

The laser-induced fluorescence method is a novel technique for detection of alpha particles that complements the alpha-radioluminescence technique. Active laser probing of the contaminated area with high repetition rate (e.g. up to MHz) using lock-in amplification technique and short-time windowing could dramatically reduce the effect of ambient noise. Furthermore, this technique enables realisation of Fluorescence Light Detection and Ranging (Fluorescence LIDAR) which could provide a distance-dependent measure of radiation by recording the time the laser pulse is emitted and the laser-induced radioluminescence signal is measured.

Results

New method and instrumentation for the optical detection of alpha particle emitters in the environment by air radioluminescence

Four optical detection systems were built to test the light collection efficiency, background suppression, beam handling and filtering. One system is based in 240 mm diameter fused silica lens, two are based on Plexiglas Fresnel lenses with clear apertures of 257.6 mm and 452.9 mm, respectively, and the fourth system is based on a modular laser-line UV-enhanced Al-coated concave mirrors (6 mirrors, 75 mm diameter, 500 mm focal length). The Fresnel lens systems, being lightweight and having good light collection efficiency, have been targeted as the first systems to be installed on the UAV for contamination mapping. The modular mirror system is another candidate that has been considered to be scaled up for UAVs, as it combines large light collection efficiency with a flexibility due to its modularity. These optical systems were tested at the PTB Microbeam facility where accelerated alpha particles were injected in a quartz cuvette which was placed 2 m from the optical detection systems under test. The cuvette was filled with air, N_2 and N_2/NO mixture. Radioluminescence has been measured in two spectral regions, UV-C and UV-AB. Radioluminescence spectra were measured using a calibrated, high-precision, fast array spectroradiometer.

Three extended Am-241 sources with an active area of 20 mm x 100 mm and activities of 980 kBq, 9.6 MBq and 90 MBq were developed. To comply with radiation protection protocols in the EU, these sources were integrated into special housings with UV-transmissive quartz domes. These samples were used to test the developed optical detection systems and will be used in measurement campaigns.

The mapping capabilities of the fused silica lens-based detection system in UV-A and UV-C spectral region, both in air and N_2/NO atmospheres, were tested with the extended 980 kBq Am-241 source and with environmental pitchblende samples having activities less than 1.8 kBq. The radioluminescence image of the alpha sources was obtained by remotely scanning the narrow field of view of the optical detection system from a distance of 2 m over the user-defined region of interest, recording the photon count rate point by point.

Development of a calibration system for the novel-type radioluminescence detector systems

The spectral shape of alpha-particle induced radioluminescence in the air was measured by means of UV spectroscopy. An UV quartz cuvette containing an Am-241 surface source (990 kBq) positioned inside a spectrofluorometer was applied to study the radioluminescence emission spectrum of air. The results of spectral distribution measurements along with the literature values served as criteria to define the spectral calibration requirements.

The spectral distribution of a batch of 3 UV-A (1.3 mW) respectively UV-C (1 mW) LEDs to be used as optical radiation sources for the low photon flux UV radiance standard has been measured. The centre wavelength and the spectral bandwidth were found to be within the manufacturer specification, the wavelength offset to the corresponding alpha particle induced radioluminescence UV lines (259.1 nm / 337.1 nm) was less than 3 nm. Taking into consideration the LEDs spectral bandwidth (FWHM) of 10 nm, the selected LEDs have been considered suitable as radiation sources.

Pitchblende environmental samples for out- and indoor experiments were prepared. The analysis of these slices with a grid ionisation chamber, alpha-track and SEM is still in progress. First results show that the distribution of uranium is not homogeneous and different for each sample. In some samples the uranium is clearly separated from “dead” rock in other, it is located in small hotspots all over the sample. While the EDX of the SEM provides an element distribution of the sample in the range of microns the alpha-track gives a general distribution of all alpha emitters. By counting the alpha-tracks on the detector, the surface contamination (Bq/cm^2) can be calculated. With the grid ionisation chamber the energy of the alpha particles is measured and shown in a spectrum. Based on the alpha energy, the associated radionuclide can be identified. Additionally, the number of particles with a certain energy for a sample is known. First environmental surfaces like asphalt, metal, concrete brick and plant were also analysed with the SEM. Dependent on surface rough- or smoothness, the first material to be worked with was chosen: Concrete brick. The surface of the concrete brick is less rough than asphalt and can even be polished to decrease the seeping of tracer into the material. This is important because of the absorption of alphas in solids.

The required mechanical components for the low photon flux UV radiance standard according to the intended PTB design have been manufactured/machined. Together with commercially available opto-mechanical parts, two prototypes of fully functional low photon flux UV radiance standards were constructed. Due to a modular concept, different optical flux level and spectral (260 nm, 340 nm + broadband source) configurations are possible. The opto-electronic components, i.e. the LED source power supply and the monitor detector, were successfully tested on a breadboard level.

Two prototype low flux UV radiance standards have been radiometrically characterized 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.

A well-characterized Po-210 activity standard to establish a traceable relationship between the radioluminescence signal and alpha activity has been developed.

Extending the optical detection system to an imaging functionality for mapping of alpha contaminations in the environment

Preliminary calculations were performed to determine the capability of a UAV-mounted UV detection system to detect alpha-induced luminescence from air. These calculations include the spatial arrangement of the alpha sources. Two drones from the UPC UAV-fleet have been chosen, the DJI WIND 4 and the DJI Matrice 600 Pro. Decision on which drone will be used will depend on the final weight and size of the optical detection system to be mounted.

The optical detection system based on the PMMA Fresnel lenses with a lens diameter of 452.9 mm and a focal length of 424.5 mm was selected for use in the UAV. The performance of this system was tested at the PTB Microbeam facility. A carbon fiber reinforced polymer frame to reduce weight and ensure the required stability of the UAV has been built. Since the system weighs less than 5 kg, it can be mounted in UPC's UAV DJI Matrice 600 Pro.

A basic skeleton of the code for data acquisition, processing, transmission and analysis in the UAV has been developed. Data such as the tilt of the UAV (i.e, of the detector), altitude, position and speed will be compiled and send to the ground station in a format for further analysis.

Feasibility study for a laser-induced fluorescence spectroscopic method for the detection of alpha emitters

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. The model was validated with experimental data using different nitric oxide and oxygen concentrations. The model seems to work very well and reproduces experimentally measured data.

Impact

The project goals and results have been presented conferences such as: National Conference "SRRp_30" held in Romania by SRRp (Romanian Society for Radiological Protection); EURADOS Annual Meeting 2021 (WG 3.1 "Spectrometry Systems for Environmental Dosimetry"); Euramet TC-IR Annual Meeting 2021 (Session: "Highlights from EMPIR JRPs"); European Geosciences Union-General (EGU) Assembly 2021 (ERE 1.1 "Energy, Resources and The Environment"); Conference on Radiation Topics ConRad 2021 (24th Nuclear Medical Defense Conference, Bundeswehr Medical Academy); Ninth International Conference on Radiation in Various Fields of Research; 12th International Conference on Instrumental Methods of Analysis: Modern Trends and Applications (IMA-2021); Optics & Photonics Days 2021 (OPD2021); 15th International Symposium on Radiation Physics (ISRP-15), Kuala Lumpur – Malaysia (2021); Euramet TC-IR Annual Meeting 2022; MARC XII conference (2022).

Impact on industrial and other user communities

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. Novel instrumentation developed in this project is essential for a quick and adequate response by nuclear regulatory bodies and other decision makers (e.g. local authorities or aid organisations) both during and in the aftermath of a nuclear or radiological accident by providing reliable radiological data that will allow appropriate countermeasures and reduce the risk of exaggerated actions and preventable follow-up costs. The novel mobile detector system for the remote detection of alpha-emitting radionuclides in the environment has the opportunity to be licensed to instrumentation companies. Moreover, with its optimised optical system and filtering setup, this instrument has a huge potential as a remote alpha particle monitor in the nuclear industry sector.

Impact on the metrology and scientific communities

A calibration facility and a metrological basis to support the implementation of new technologies for radiological emergency management will be established. This includes the development of a fit-for purpose, state-of-the-art portable radiant standard and calibrated and well characterised alpha active environmental samples (mineral phase, organic, soil and plant). The characteristic limits (minimum detectable activities and largest standoff distances) for remote alpha detection will also be determined. Furthermore, models for the substrate dependent generation and optical path propagation of the alpha particle induced radioluminescence will be developed. The latter are of crucial importance to perform precise quantitative measurements and for applying accurate corrections to the deduced activities. These corrections include the influence of wind, which reflects changes in the nitrogen density that consequently affects the number of radioluminescence photons and, the influence of smoke and dust which change the refractive index of the medium where radioluminescence light is propagating.

Impact on relevant standards

The project will provide guidance for stakeholders and input to international standardisation bodies (ISO, IEC), as far as nuclear and radiological emergency preparedness is concerned. 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.

Members of the consortium are involved in the following committees: ISO/TC85 (Nuclear Energy), IEC/TC45 (Nuclear Instrumentation), EURAMET TCIR (Technical Committee for Ionising Radiation), ICRM (Gamma and Beta Spectrometry WG, Alpha Spectrometry WG and Low Level WG), BIPM CCRI I and II, and EURADOS WG 3. This will ensure that the harmonised procedures and methods developed in this project will provide input directly into European and international standardisation.

Longer-term economic, social and environmental impacts

Radiological accidents bring human activity in contaminated areas to a halt, thus disrupting both economic and social order to a large extent. Misinformation regarding geographic location of contamination sites and the associated dose rates will trigger an avalanche of losses that affect agriculture, tourism, export and domestic consumption. By providing traceable radiological data, overestimation of the resulting risks will be avoided and the effects on both economic and social life can be minimised.

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. Furthermore, it will contribute in guiding the emergency medical teams who provide health assistance to the affected/contaminated/exposed persons near to or in the contaminated areas and will also assist in defining safe evacuation routes and zones.

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 RADIOPROTECȚIE – „SRRp-30", ISBN: 978-973-1985-64-0 https://srrp.ro/wp-content/uploads/2020/12/Conf.Nat_._SRRp_-2020-A4-ver.12.pdf

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