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New technologies for radioactive waste monitoring: Results and perspectives from recent experience

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Summary. — With the initial support of the INFN-Energy committee throughout the last ten years a few activities have been pursued aimed at the improvement of radioactive waste management. Nowadays new technologies make possible the development of low-cost systems capable of monitoring the radioactivity coming out of (spent) nuclear fuel casks or radwaste drums. Simple, compact and effective radiation counters were developed at INFN for the detection of gamma rays and neutrons, suitable to be deployed as a distributed network for the real time monitoring of radioactive waste from the very low up to the highest level, and possibly preluding to a mass deployment benefitting both safety and security. Indeed, in such a framework any change in counting rates, or even a sensor blackout, could be interpreted as a possible safety or security breach. Techniques for sorting and segregating the radioactive waste have also been studied and a dedicated prototype for the gamma/visible imaging with spectroscopic features is currently being developed. On top of the new interesting technical features accessible by means of distributed networks of devices, one should also focus on the psychological impact they could have on the general public acceptability of a finely monitored repository. Tests performed on radioactive waste of all categories, including spent fuel, showed quite promising results.

1. – Introduction

Throughout the last ten years the Italian National Institute of Nuclear Physics (INFN) has kept fostering initiatives aimed at the improvement of radioactive waste management by means of its INFN-Energy committee. Several activities have been carried out in collaboration with national and international companies/agencies, including the participation to currently ongoing EU funded projects, focused on radioactive waste monitoring, nuclear decommissioning, accident remediation, sorting and segregation. Nowadays new technologies have made possible to develop low-cost systems capable of monitoring the radioactivity coming out of (spent) nuclear fuel casks or radwaste drums, by means of simple, compact and effective counters for the detection of gamma rays and neutrons. However, the main issue is not technological but rather sociological: can we convince the general public to accept a radwaste repository if we do not first convince ourselves? The question is not trivial at all, as it can be easily verified by looking at the radwaste data available on the IAEA public website. The database of ref. [1] is updated up to year 2013, whereas the new one in ref. [2] typically starts from 2019. Not all data are directly provided in numerical form, as for several countries only PDF documents are available. By comparing, for instance, the 2013 and the 2019 data from Italy one can observe an almost total discrepancy, as if the data would refer to different countries. Moreover, as Italy so far has no national repository, one does not expect any big difference, as all of the radwaste is still in storage and none has been disposed of. The 2013 data by overall categories include the HLW (High Level Waste), ILW (Intermediate Level Waste) and LLW (Low Level Waste), whereas the corresponding data for 2019 lack the HLW category and include the VLLW (Very Low Level Waste) that was missing in the previous data. Moreover, for 2013 radwaste data with detailed origin are provided, whereas for 2019 only the amounts of currently stored spent fuel in nuclear power plants and research reactors are reported, and in tons instead of cubic meters. Of course it might well be that by carefully studying the released documents one can understand the reasons of this apparently schizophrenic behavior, but this is something typically done by the hands-on experts in the field. And what about the general population, or even scientists? The feeling one gets when trying to understand these data is that numbers are unclear and do not add up correctly, and the PDF reports are written in a rather obscure legalese-technical language. This could easily create or increase mistrust and foster conspiracy theories, and the lesson to be learned should be that what is clear to experts is NOT clear to the general population. A step in the direction of the improvement of this situation could come from the implementation of real time monitoring systems based on a mesh of sensors distributed all over the storage sites around the radwaste containers, which also offers several safety and security advantages that are going to be discussed in the following.

2. – Monitoring

In most radwaste storage sites the drums are periodically inspected by operators making use of more or less sophisticated tools and detectors, and a few specific radiation detectors and dosimeters installed in several locations keep under control the local environment. This method has at least two safety disadvantages: (i) the local ambient monitoring is limited to few positions; (ii) it requires people moving around in a potentially dangerous environment, and perhaps this should be strongly reduced if not deprecated. As for security, on the one hand the usual two-people rule of surveillance is and will still be necessary along with cameras. On the other hand this is also one of the main concerns when considering the risk of insider threats [3]. And this is where new technologies can come into play and provide a considerable improvement. A relevant example from our everyday life illustrates how a low-cost and distributed technology can provide also benefits for initially unforeseen applications: the widely used Google Maps application provides information on the road traffic based on the number of mobile phones simultaneously present in each location.

In the case of radwaste the radioactivity has been so far only considered as a danger, and for this reason periodically monitored/inspected. But the penetrating outcoming radiation, basically only gamma and neutron, is also conveying information from the inside of each container. Why not using it to have a continuous real-time monitor? Listening to this information can produce initially unexpected benefits, provided that arrays of low-cost radiation sensors can be built and installed among the radwaste containers. As the radiation keeps flowing continuously the required sensors do not need to have high detection efficiency, and this could make them suitable for the full range of activity levels, from VLLW up to HLW and spent fuel.

Within the framework of radioactive waste monitoring INFN has taken part in three Euratom H2020 projects, namely MICADO [4], PREDIS [5], CLEANDEM [6], where it contributes with its long-standing expertise in the development of radiation detection systems. These developments aim at getting rid of paper (or paper-like, *i.e.*, PDF) monitoring notes replacing them with numerical data automatically collected and stored into files and databases. In order to monitor the penetrating radiation two detector technologies were developed in the MICADO Work Package 7 (WP7) and applied to the detection of gamma rays and neutrons, named SciFi and SiLiF respectively. A set of detectors of the two types is shown in fig. 1. As a task of the MICADO project demonstration 16 SciFi and 16 SiLiF radiation counters for gamma rays and neutrons were installed around four very low activity radwaste drums which were monitored during forty days (fig. 2). The same detection technologies are also being exploited in the PREDIS WP7, along with a newly developed wireless front-end and data acquisition electronics, for the specific case of cemented radwaste monitoring. For the CLEANDEM WP3 a miniaturized system was developed which incorporates two miniature counters for neutrons and gamma rays and the related front-end and data acquisition electronics. This system will be installed on an unmanned autonomous vehicle (UAV) for radiological inspections during decommissioning of nuclear installations or accident remediation.

3. – The SciFi gamma counter

The wide range of gamma detectors and techniques present on the market are typically from moderately to highly expensive, therefore they do not look well suited for a

Fig. 1. – A set of SiLiF neutron (top) and SciFi gamma (bottom) counters developed in the MICADO project.

Fig. 2. – Four very low activity radwaste drums surrounded by 16 SciFi and 16 SiLiF radiation counters for gamma rays and neutrons respectively.

finely distributed monitoring. In order to obtain small, inexpensive, robust, easy-to-use and reliable detectors one should use well known materials and properties, avoiding too sophisticated technologies which would lead to expensive and delicate components. These guidelines led to the development of the SciFi gamma radiation counter, based on a 3 mm diameter 80 cm long plastic scintillating fiber [7,8] and two Silicon Photomultipliers (SiPM) [9-11].

A scintillating fiber is an optical fiber whose core is made from a plastic scintillator material. Whenever a gamma ray interacts with the fiber, mainly by means of Compton electrons, it produces a tiny flash of light (scintillation photons), that propagates along the fiber in both directions. The fraction of photons reaching each fiber end is about 6% of those initially produced, provided that the fiber is much shorter than its 3.5 m attenuation length, and these photons can be detected by the SiPM. The recommended length is thus $\leq$ 1m.

By requiring that (i) the fast signals from the two SiPMs overcome a predefined amplitude threshold, and (ii) the two signals occur in coincidence within several tens of nanoseconds, one can make sure to reject any spurious background event and to be sensitive to real radiation, that is gamma and cosmic rays. A more detailed description of the SciFi gamma counter can be found in refs. [12] and [13].

One might argue that a 3 mm thick plastic scintillator is very poorly sensitive to the gamma radiation. However, even though a gamma ray deposits a small amount of energy in the fiber, the small number of scintillation photons thus produced can still be detected by the single-photon-sensitive SiPM detector. In the MICADO project the SciFi detectors front-end and readout electronics was provided by a single module capable to handle up to 32 detectors [14]. The counting rates of 36 SciFi detectors, measured with a 1.5 MBq ^{137}Cs source in 200 s at 50 cm distance from the fiber midpoint, along with the background rate, were uniform and stable [12].

In fig. 3 left the signal amplitude spectrum for a SciFi detector installed on a very

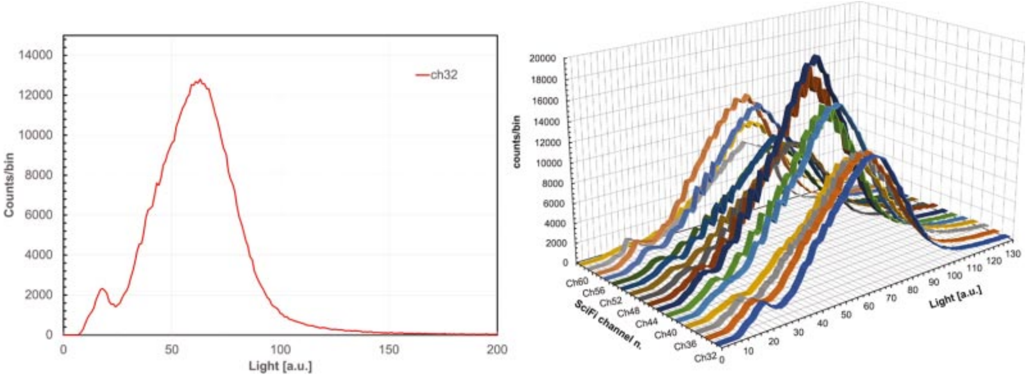

Fig. 3. – Left: Signal amplitude spectrum for a SciFi detector installed on a radwaste drum mainly containing ^{137}Cs. The small peak to the left is due to cosmic rays, the bigger one to the radiation from the drum. Right: Signal amplitude spectra for 16 SciFi detectors installed around the four radwaste drums of fig. 2 for the MICADO project.

low activity drum (fig. 2) mainly containing ^{137}Cs is shown. The small peak to the left is due to cosmic rays, the bigger one to the radiation from the drum. Figure 3 right is an overall 3D view of the signal amplitude spectra for 16 SciFi detectors installed around four radwaste drums for the MICADO project. The different height of each peak reflects the different activity and emission (an)isotropy of the four drums.

4. – The SiLiF neutron counter

The detector required for neutron monitoring purposes should be simple, small, robust, easily handled, possibly operated at low voltage, reasonably inexpensive, and have thermal neutron detection efficiency of at least a few percent. The effort to fulfil all of these requirements at once led to the development of the SiLiF detector which represents a viable alternative to the gold standard for neutron detection, namely the ^{3}He tube. It consists of a semiconductor detector coupled with a neutron converter, which is a material containing nuclei having a high cross section for neutron capture with charged particles in the final state [15].

The two isotopes commonly considered as suitable neutron converters are ^{6}Li (thermal neutron capture cross section 940 b) and ^{10}B (thermal neutron capture cross section 3840 b). ^{6}Li was chosen because after capturing a neutron it has a unique decay channel into a triton and an alpha with no emission of gamma rays. Moreover, the Q-value is higher and the charged particles produced are lighter, therefore easier to detect than in the case of ^{10}B. Used in form of ^{6}LiF it is a very stable and inexpensive salt which can be evaporated uniformly in thin films onto several substrates. At variance with other authors of previous papers who deposited the converter directly onto the semiconductor, the SiLiF detector described here features two converters onto two independent substrates which are then sandwiched almost in contact with a double-sided silicon diode, then incapsulated into an aluminum box as illustrated in fig. 4. The choice of using independent detector and converter allows a much better modularity and easy reconfigurability, reducing the cost of the technology and increasing the production yield [16-22].

A typical deposited energy spectrum obtained exposing a SiLiF detector with a single

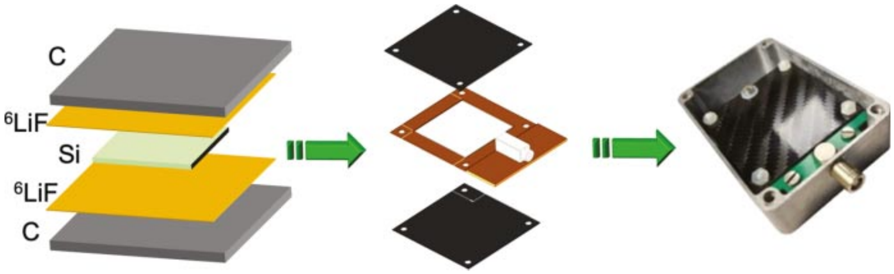

Fig. 4. – Construction of a SiLiF detector. A ^{6}LiF layer is evaporated onto carbon fiber plates, then a sandwich is assembled with the silicon diode in between and installed into an aluminum box.

thin ^{6}LiF layer (1.6 μm, 0.5% intrinsic detection efficiency) to an AmBe neutron source is shown in fig. 5 left. By employing thicker layers one can increase the detection efficiency worsening the spectrum shape because of the degradation of the kinetic energy of the produced particles while crossing the converter. A tradeoff solution was found in a thickness around 16 μm. A typical deposited energy spectrum with a sandwich of such a converter thickness is shown in fig. 5 right. A thin SiLiF has been fully characterized at the PTB metrology institute [23] and the area under the triton peak can be used as a reference to determine the detection efficiency of other detectors. Enforcing a threshold at 1.5 MeV for the thicker version has shown a gamma/neutron discrimination of the order of 10^{-10} [24], with a thermal neutron intrinsic detection efficiency of 5%. In order to make it efficient also to higher energy neutrons a suitable $10\times10\times10$ cm^{3} polyethylene moderator was designed, simulated and built to contain the detector box assembly. The neutron detection efficiency in such a configuration is basically constant from thermal energy up to 1-2 MeV and then slightly decreasing. The response of silicon diodes along with the employed charge sensitive preamplifier [25] is rather uniform and reproducible. This behavior is shown in fig. 6, where the deposited energy spectra of 34 SiLiF detectors

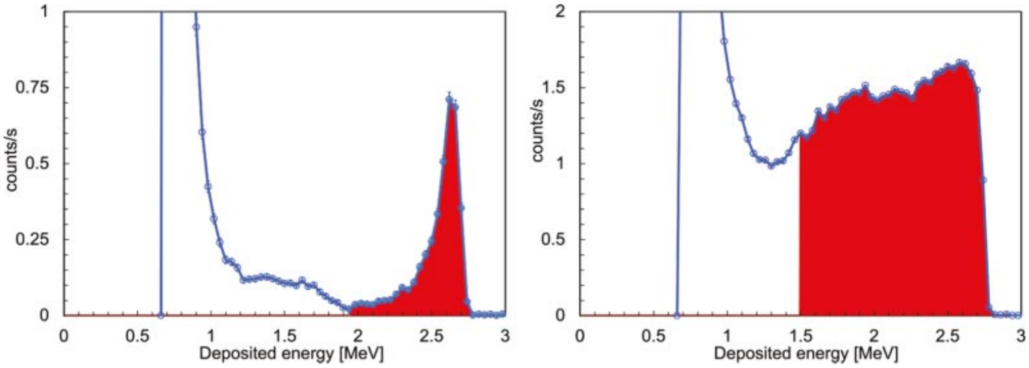

Fig. 5. – Typical deposited energy spectra obtained with SiLiF detectors exposed to an AmBe neutron source. Left: single-sided detector with converter thickness 1.6 μm, the colored region indicates the main contribution of tritons. Right: double-sided detector with converter thickness 16 μm, the colored region, above the selected threshold at 1.5 MeV, indicates the contribution of tritons+alphas safe from gamma contamination.

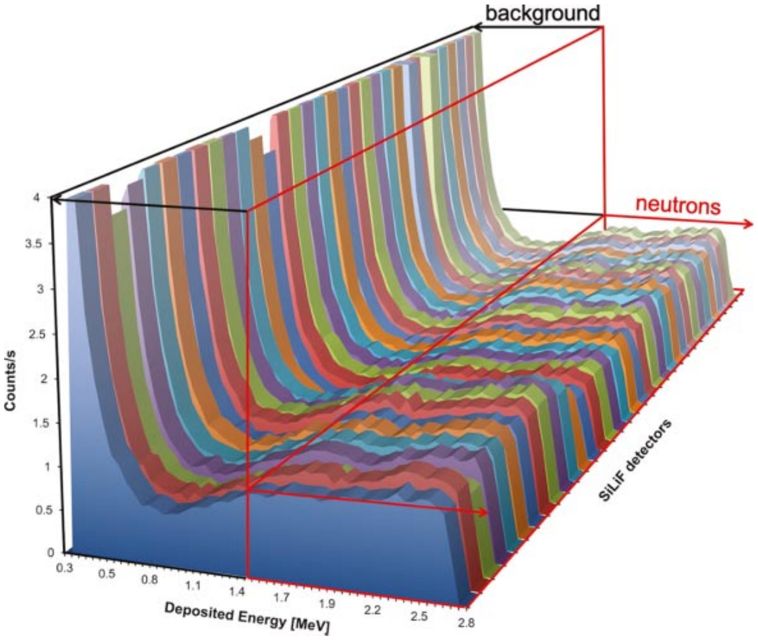

Fig. 6. – 3D arrangement of the deposited energy spectra from 34 SiLiF detectors collected in a predefined position in front of an AmBe neutron source. The gamma background discriminating threshold at 1.5 MeV is highlighted.

collected in a predefined position in front of an AmBe neutron source are arranged in a 3D fashion that also highlights the gamma background discriminating threshold at 1.5 MeV.

5. – Discussion

SciFi and SiLiF detectors have thoroughly been tested and characterized on the bench with several gamma and neutron laboratory sources, from low to high activity and with mixed radiation fields. Monitoring tests have also been performed throughout ten years with real radwaste containers in several storage sites: VLLW at Nucleco (Rome, Italy), LLW and ILW at Sogin (Sessa Aurunca, Italy), HLW at Orano (La Hague, France), Spent Fuel at Zwilag (Würenlingen, Switzerland). The measured SciFi gamma counting rates for the different waste types range from a few counts per second (cps) to few hundred cps. As for neutrons the measured SiLiF counting rates range from 10^{-6} to 200 cps, thus proving to be gamma blind. Indeed, the very low counting rates occurred when monitoring drums containing quite small quantities of actinides with activity of the order of few 10^3 Bq, whereas the background neutron counting rate far from any source was basically null. Remarkably, even though neutron monitoring of VLLW on a longer term produced very few counts their spectral shape is in agreement with the expectation. The 16+16 detectors of fig. 2 were kept in operation throughout 40 days, proving to be quite stable. The measured counting rates are shown in fig. 7.

The availability of SciFi and SiLiF detectors can open quite interesting perspectives not only concerning safety and security. Indeed, due to their low cost, good reliability

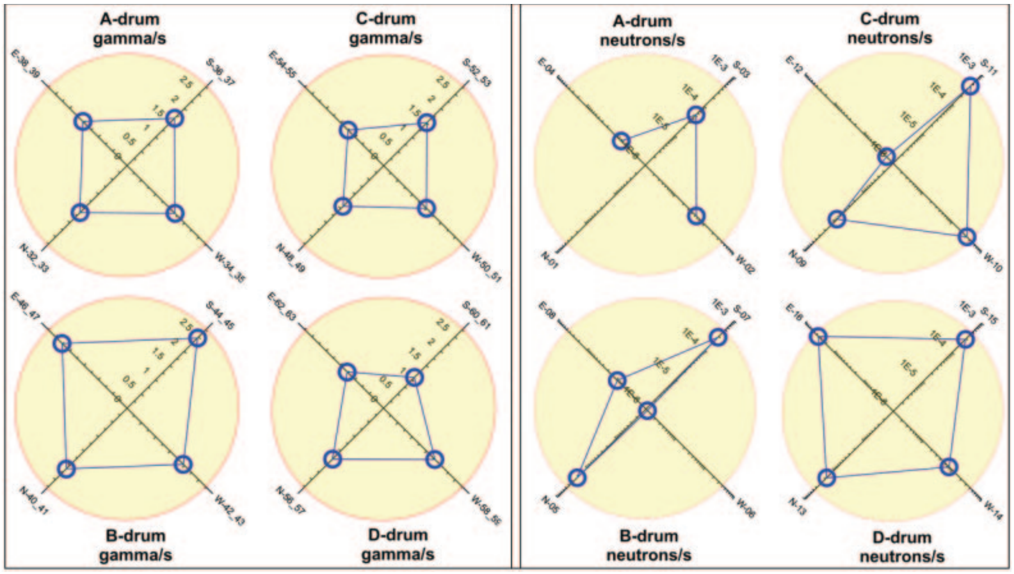

Fig. 7. – The measured counting rates of the 16+16 detectors installed around the VLLW drums of fig. 2 during 40 days.

and stability they could usefully be exploited to pre-screen radwaste drums in view of their final characterization. In a storage site where many drums are queued while waiting to be inspected and characterized, a few SciFi and SiLiF could be hung on each drum and kept measuring throughout a sufficient amount of time. The collected data would provide useful preliminary information about the gamma dose rate, the possible presence of actinides, the anisotropy of the radiation emission indicating a corresponding internal asymmetry. This could take advantage of the development of WiFi front-end and data acquisition electronics currently under way within the PREDIS project, where the detectors could be operated on batteries. Moreover, an interesting topic in several current projects is the so-called digital twin, consisting in a computerized replica of a physical system or installation enriched with field data. Such an exercise was done in a previous project [26], and a visual 3D example of how a storage site equipped with SciFi and SiLiF sensors could appear is shown in fig. 8. The virtual storage site could be navigated, the color of each drum reflecting the measured activity, and all of the available information about each item could be accessed interactively.

6. – Conclusions

In the coming years the nuclear decommissioning will become a more and more relevant activity, as most of the 422 nuclear power plants all over the world are 40-50 years old. This implies that new radwaste storage sites will have to be put in operation and one should to be prepared to the increasing needs of safety and security. The implementation

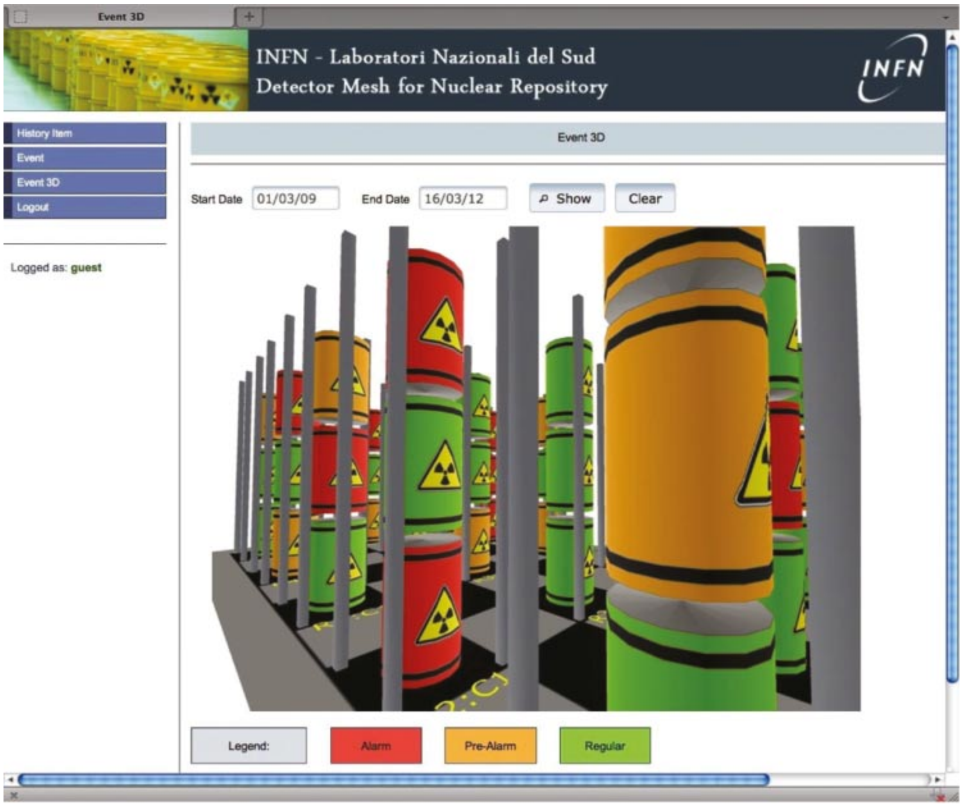

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Fig. 8. – Visual example of the possible digital twin of a storage site equipped with SciFi and SiLiF sensors. The virtual storage site can be navigated, the color of each drum reflecting the measured activity, and all of the available information about each item can be accessed interactively.

of a modern monitoring architecture could represent a relevant step in the direction of a better acceptability of the repositories for the population, as well as an additional and transparent tool both for the control authorities and for the general public. The radiation sensors here described, and others to be likely derived from them, could be a tool with quite appealing features in terms of cost, modularity, flexibility, safety, security. A realistic mass deployment looks feasible and compatible with advanced information technology tools capable of efficiently handling and using in real time big data coming from nuclear material. This would make possible to collect longer term behavior info and possible early hints of anomalies while at the same time reducing the need of human presence in radioactive environment, that become more and more important as moving up along the scale of the radwaste activity, from VLLW to HLW and spent fuel.

In view of the forthcoming construction of the Italian nuclear repository research institutions and universities can play a fostering role, as they have taken part in many pertinent research projects, and this represents a big chance for a step forward as it will be a hub where different disciplines and technologies can mix and progress. However, a solid and effective strategy will be needed in order to convince the general public and the local population to accept the repository.

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