



Scintillation for the future in a changing world

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Abstract

Liquid Scintillation has two main characteristics: the lack of resolution and the capability to determine alpha and beta emitters. Society evolves toward a situation where information is key for management and where sustainability is also of paramount importance for the future of the planet. In this scenario, this article suggests to continue implementing the use of Plastic/Extractive Scintillating resins and to rescue some capabilities of the scintillation to design specific and miniaturize detectors that include alpha/beta discrimination based on particle attenuation or scintillators emission wavelength. These approaches will complement laboratory analysis and are aligned with the demands of the society.

Keywords Sustainable development goals · Plastic/Extractive scintillating resins · alpha/beta discrimination · alpha/beta penetrating power · Wavelength discrimination · Detector miniaturization.

Introduction

The **world is changing** and so are the demands of society, steering towards the wellbeing and sustainability of both the human population and the planet. This change had been narrowed down in recent years through the Sustainable Development Goals (SDG) [1]. The 17 SDG point at different aspects that must guide the goals of almost every discipline. Radionuclide determination using scintillation is an activity that provides necessary information for the accomplishment of several among these SDGs.

We can summarize the actions related to sustainability and the scintillation techniques in: pollution reduction, waste management and safe use of chemicals [2]. On the other hand, the use of scintillation has enabled gathering information of social interest through routine analysis in specific scenarios while minimizing the resources needed to achieve them.

Liquid Scintillation (LS) is a well-established technique whose road began during the first “Liquid Scintillation” conference in Chicago in 1957 [3]. Soon followed landmark

developments in the field like the fundamental text by J.B. Birks, “The Theory and Practice of Scintillation Counting” (1964) [4] and the editions of the “Handbook of Radiochemical analysis” by M. Annunziata [5]. The current advances in the field have been the subject of many LSC conferences (the most recent happening in 2021 in Shenzhen, China), specialized journals (JRNC, Applied Radiation & Isotopes and Nuclear Instruments & Methods) and newsletters and technical documents from private parties commercializing Scintillation equipment [6, 7] & or consumables for radionuclide measurement [8, 9].

This combined scientific research has contributed to **advances in LS** over time specially in the areas of: Background reduction (active shielding, pulse shape analysis and mathematical methods), Alpha/Beta Discrimination based on pulse shape temporal distribution, Detector design (Triple to Double Coincidence Ratio TDCR), Calibration, CIEMAT-Nist, Deconvolution methods, Cocktail formulation, NPE-free cocktails, plastic scintillators, sample processing and automation [5].

Nonetheless, some other aspects described in the initial studies have not progressed to the same extent. The development of the scintillation technique has left some options for alpha and beta radionuclide emitter discrimination that have ever been incorporated as part of protocols of analysis. This could be a good moment to review some of them and, if convenient, recover them for certain applications. The new capacities could be of interest for the improvement

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of determinations in the laboratory but also for the development of specific devices for measurements in specific scenarios.

However, the future of LS cannot be considered without considering the characteristics of the **context associated with the field of radioactivity**. The first is the specific regulation and supervision of the nuclear activities that conditions the measurements that must be done and how they must be performed. In recent years, it is also relevant for the implementation of innovation, the concentration of companies that produce detectors and consumables and the appearance of some new ones based in Asia (China, Korea and Japan).

Another key consideration concerning the development process of LS equipment and in its various applications is the convergence between professionals from different fields using different approaches to solve the same problems.

Furthermore, the expected outcomes related to radionuclide detection using scintillation are increasingly diverse, varying both in relationship to the discipline using them (environmental monitoring, radiopharmacy, decommissioning metrology) and the circumstances in which they are used (on-site, continuously, emergencies etc.)

This complex scenario is a challenge with advantages and limitations for the development of new LS equipment and methods. As a consequence, the **objective** of this article is to suggest to the LS community a discussion about the potential implementation in the scintillation detection process of some capabilities based on: chemical selectivity based on solid scintillators; alpha/ beta discrimination based on their different attenuation interacting with materials; alpha/beta discrimination based on the wavelength of emission of the fluors and the instrumental miniaturization. Most of these capabilities have been proposed and essayed in the past. But they had not finally been implemented in routine analysis.

This article collects a personal perspective of the aspects that could be interesting to investigate in the future evolution of the scintillation technique.

Discussion

This section is organized in four separated parts according to the above-mentioned objectives: Chemical selectivity, Penetration power, Wavelength of emission and Instrument miniaturization.

Chemical Selectivity

One of the main limitations of scintillation techniques is the lack of spectrum resolution, either due to the continuous distribution of energies of the Beta emissions or due to the widening of the bands because of different energy losses

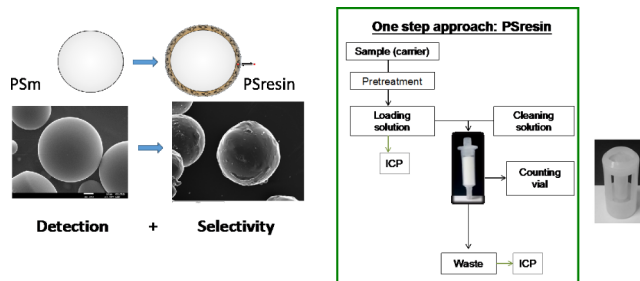


Fig. 1 Images of PSm and PSresin. Separation scheme using a PSresin [11]

of monoenergetic alpha emissions during the detection process. In any case, the determination of the activity of a given alpha or beta emitting radionuclide generally implies a previous step of chemical separation aimed at isolating it totally or partially.

These chemical separation processes require a large amount of resources (manpower and reagents); they generate a significant amount of waste and require time to be completed.

In recent years, proposals have appeared based on a solid scintillator support coated with a selective extractant “Plastic/Extractive Scintillating Resins”. In these cases, the selective separation and detection steps continue as main parts of the determination process, but both are integrated into a single element [10, 11].

The following Fig. 1 shows the configuration of the Plastic Scintillating Resins (PSresins). These PSresin are formed by a core of Plastic Scintillating (PSm) coated with the selective extractant for the radionuclide of interest. These PSresins are packaged in a cartridge and the separation process is performed following the same procedure as when using Extractive Resins.

The main difference when using PSresins is that after separation and cleaning steps, the radionuclide has not to be eluted and mixed with LS cocktail because the cartridge, that already contains the isolated radionuclide and the scintillator, can be measured directly. PSresins have been developed for the determination of ^{99}Tc , ^{14}C , $^{89}\text{Sr}/^{90}\text{Sr}$, ^{210}Pb , U in environmental, decommissioning, emergencies samples.

The incorporation of PSresins to routine work will allow the reduction of reagents, treatment time and waste generated, especially avoiding the production of mixed waste generated using LS cocktails. In fact, after measurement, the radionuclides could be eluted from the PS resins by changing the medium, which would allow the recovery of the inactive plastic residue and the radioactive aqueous solution. Last can be treated like any other aqueous radioactive waste. These characteristics can contribute to sustainability while maintaining the radionuclide monitoring capacity.

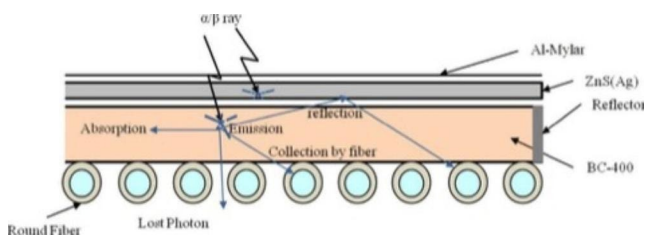


Fig. 2 Scheme of the detection system using two successive layers of scintillators [12]

The main limitation of PSresin is its limited detection efficiency for low energy beta radionuclides.

α and β attenuation

It is well known the different attenuation of different types of ionizing radiation into materials. When the material is made up of low atomic number elements, as is the case with plastic scintillators, the penetration of alpha particles between 4 and 6 Mev is 35–55 μm , while beta particles with energy between 0.2 and 2 Mev would reach between 0.4 and 9.5 mm [12]. The different attenuation for both radiations is sufficient for penetration to be considered a variable with the ability to discriminate between alpha and beta particles.

This approach has been tested with an overlapping sheets geometry of different scintillators by Y. Ifergan et al. [13]. In Fig. 2 the structure of the scintillator system used in detection can be observed.

The system consists of a first ZnS scintillator layer with a silver foil; followed by a thicker layer of plastic scintillator BC-400. This is separated from the outside by reflective foils except in its lower part where there is a set of optical fibers that collect the scintillator emission and transfer it to the detector.

In this device, the alpha particles only produce a signal in the surface scintillator while sufficiently energetic beta emissions would be able to pass through this first sheet and produce a signal also in the second.

The detection efficiencies recorded in the ZnS for an alpha emitter such as ^{238}Pu are between 62 and 65%, depending on the configuration of the optical fibers; while for beta emitter $^{90}\text{Sr}/^{90}\text{Y}$, the efficiency of BC-400 plastic scintillator detector is between 87 and 90%.

A configuration similar to the one described above is the one marketed by Eljen Technologies in its product EJ-444 [14]. As in the previous case, this system consists of a silver activate sulfide sheet followed by another of EJ-212 plastic scintillator. EJ-444 is presented in sheets and discs of different diameter and thickness.

This same proposal could be transferred to a spherical geometry as shown in Fig. 3. In this case, the plastic scintillator microspheres (PSm) could be formed by a single

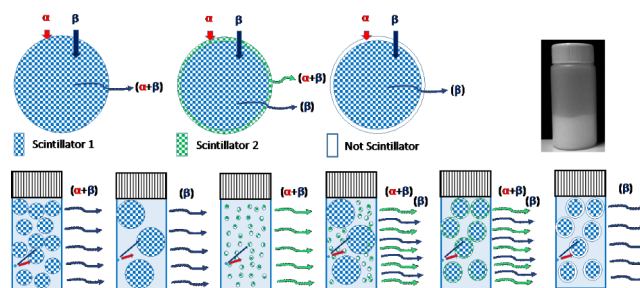


Fig. 3 Different structures of Plastic Scintillating microspheres (PSm) and detection configurations

scintillator material or by more than one organized in concentric layers. As in the foils, the outer scintillator would detect the alpha and beta emissions, while the interior, only the betas. The use of a non-scintillating outer layer could allow even the unique detection of beta emissions.

This type of configuration would allow the preparation and measurement of aqueous solutions in a similar way to that developed in liquid scintillation: a vial filled with PSm whose interstices are occupied by the solution to be analyzed. The detection efficiency is greater the smaller the diameter of the microsphere (145–17 μm) [11]. In fact, this geometry would be an enormous magnification of the LS with scintillation cocktails where scintillator and aqueous phase form a micellar system. In the case of PSm, the distance between the disintegration points and the solvent or fluor is of few tens of microns, depending on the size of the microsphere. This path of the emitted particle through the aqueous solution before reaching the microsphere acts as a “shield” against alpha emissions and, consequently, could also be used as a variable to limit the type of particles detected.

Regarding other variables such as the duration of the pulse, one aspect in favor of the discrimination of alpha and beta particles based on penetration is its simplicity. However, its implementation also shows weak points since its ability for alpha / beta discrimination depends on different variables such as the energy of the particles and the design of the scintillator system. The difficulty of producing the designed systems can also be considered a weak point. Thus, this approach based on penetration cannot be a general solution, but it could be the basis for devices oriented to inform about activity on specific situations or parameters.

For example, a very important screening parameter is gross alpha / beta activity in drinking water. The results of a European interlaboratory comparison [15] involving 71 laboratories using different measurement techniques, including LSC, and analyzing waters of different composition showed that, for alpha activity, results reported by between 36 and 63 (depending on the complexity of the analyzed water) presented a deviation of more than 30% with respect to the

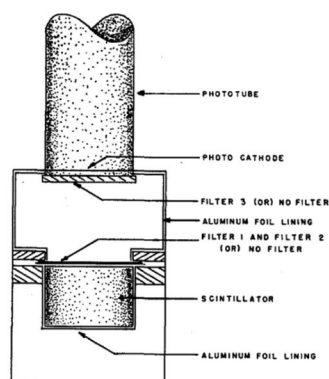


Fig. 3. Sketch of detector model used to test the performance of the various scintillator-filter combinations.

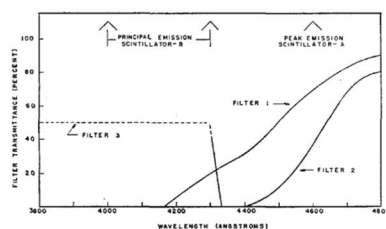


Fig. 2. Filter transmittance as a function of the wavelength of normally incident light. The dotted portion of the curve for filter 3 does not exhibit the detailed structure of the function.

TABLE II. Summary of measurements.^a

Scintillator	Light transmitting media	Relative phototube pulse height
A	clear	1 (by definition)
A	Filters: 1+2	0.4
A	Filters: 1+2+3	<0.001
B	clear	0.5
B	Filter: 3	0.2

Fig. 4 Scheme of the detector, filter transmittance behavior for filters and relative signal detection for scintillator A and B [16]

reference values. For beta activity, this deviation was found in the values determined by between 27 and 61 laboratories, depending also on the composition of the water studied. This would be a parameter for whose determination the different alpha/beta attenuation could be the basis of an alternative analytical procedure.

The implementation of approaches of this type would reduce waste, could simplify procedures and favor sustainability and the implementation of routine measures.

Wavelength-based discrimination

The implementation of the systems based on different scintillators for alpha / beta discrimination according to their different attenuation would be greatly favored if the emission of these scintillators could be differentiated by spectroscopic methods. This approach could take advantage of all the progress made in the field of ultraviolet / visible spectroscopy.

This wavelength-based discrimination was early studied by E. Boldt et al. [16] in 1961 combining different plastic scintillators (Scintillator A: Polyvinyltoluene with p-terphenyl, tetraphenylbutadiene and POPOP - emission peak at 400 nm; Scintillator B: Polyvinyltoluene with p-terphenyl - emission 390–460 nm), and filters (Filter 1: Kodak Wratten gelatin filter type 2D; Filter 2: Kodak Wratten gelatin filter type 3; Filter 3: sharp cutoff interference filter type B-5-3). The scheme of the device together with the filters transmittance distributions and the results achieved can be observed in Fig. 4.

The results show that combining the different filters it was possible to detect with reduced intensity but without interference the signals emitted by both scintillators.

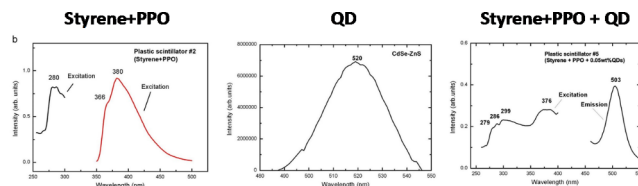


Fig. 4. Photo-induced excitation and emission spectra of (a) plastic scintillator #1 (Styrene + PPO + POPOP) and (b) plastic scintillator #2 (Styrene + PPO).

Fig. 5. Excitation spectra of QD (CdSe-ZnS) under an excitation peak at 315 nm.

Fig. 5. Photo-induced excitation spectra (under 303 nm) and emission spectra (under 303 nm) of plastic scintillator #1 (Styrene + PPO + POPOP) and #2 (Styrene + PPO + QD).

For wavelength discrimination, the use of scintillators capable of shifting the emission frequency towards ranges of lower energy may be useful. These scintillators are marketed by different companies such as Eljen Technologies that have wavelength shifting plastic such as EJ-280, EJ-282, EJ-284 and EJ-286 based on polyvinyltoluene and with maximum of emission and absorption at 490 and 427, 481 and 390, 608 and 574, and 425 and 355 respectively [17].

In addition to this shift, another aspect that can facilitate wavelength discrimination is the narrowing of the emission bands. In recent years, different plastic scintillators doped with quantum dots have been prepared and studied. J.M. Park et al. prepared a series of these scintillators based on styrene and PPO and quantum-dots with a core of CdSe and shell (ZnS) [18]. In Fig. 5 it can be seen how the scintillator emission shifts its maximum from 380 to 503 with the addition of QD.

Another option interesting to explore the capability of particle type discrimination based on the wavelength of scintillator emission is the use of plastic scintillators doped with organometallic compounds. P.E. Feng et al. showed how the inclusion of an organometallic compound as the

$\text{Ir}(\text{ppy})_2(\text{acac})[(2\text{-phenylpyridine}), (2\text{-3-pentanedionate})]$ in a plastic scintillator as the PVK (poly (9-vinylcarbazole)) may induce triplet harvesting by the metal-to-ligand charge transfer (MLCT) [19]. But what is interesting is that the extension of this process depends on the type of ionizing particle. Triplet harvesting produces a shift on the wavelength emission that will allow to differentiate different particles based on the spectral distribution (Spectral-Shape Discrimination SSD). The main limitation of this approach could be related to the time required for this process that could be too long to be applied for counting purposes [20].

Despite the possibilities pointed out by these examples, the implementation of alpha/beta discrimination based on the emission frequencies of the scintillators has different difficulties to overcome. The first is the synthesis of scintillators and their combination in structures that generate sufficient and separate signal intensities for the two types of excitation. The second, the design and construction of the detectors that allow the efficient collection of the emitted signals in addition to their spectroscopic discrimination and their segregated detection with high sensitivity and a signal/background ratio that is also adequate.

Wavelength discrimination complements the penetration discrimination proposal and, as indicated in the previous section, opens the options for future the development of specific detectors.

Instruments miniaturization

Most commercial liquid scintillation counters are designed to work in the laboratory. Today, many analytical determinations are moving from the laboratory to the place where the problem (and the sample) is located, allowing information to be obtained in real time and in a continuous way. This availability of information can be especially important in the field of radioactivity both in routine analysis and in specific scenarios as emergencies.

In recent years, many technologies have been adapted to provide services in the specific conditions generated by new social demands. The best-known case of all is moving from the public phone box to the private mobile phone.

The author does not have the technical knowledge to propose how the miniaturization of liquid scintillation detectors could be guided but is confident that the same evolution of electronic components, communication capabilities and data processing that has allowed the design of specific devices for needs/specific applications, will also allow this evolution of the LS counters. Instruments designed for specific applications may allow the reduction of elements and the incorporation of the new miniaturized electronic components. This can be a way to reduce costs and expand the market for this type of specific equipment.

Presentations at LSC2020 [21] such as those by P. Casette (New Developments in miniature TDCR counters), B. Sabo (Performance of a portable TDCR system), Q. Zhou (Development of a portable TDCR system), J. Janda (The influence of plastic scintillator dimensions, PMT size and photocatode geometry on the detection efficiency) contribute to this development.

The evolution of scintillation detectors is a key point to implement future innovation in the field of radioactivity detection based or not on what is suggested in this article.

The miniaturization and adaptation of LS detectors to specific scenarios and problems will contribute to reducing resources and waste and to improve information to support decisions related to radioactivity management. This development is aligned with sustainability in this society where information is increasingly important.

Conclusions

Scintillation detectors should evolve in the future by developing approaches that complement the currently existing capabilities for the determination of alpha and beta emitting radionuclide activity in the laboratory.

The evolution of the equipment must be adapted to the specific needs of each problem, facilitating “in situ” measures and increasing the information provided.

In these evolutions, the miniaturization of the equipment is of great importance, maintaining only the necessary components and incorporating alpha/beta discrimination and selectivity options such as those associated with Plastic / Extractive Scintillating Resins, the differential attenuation of these particles or the use of detection devices capable of emitting, discriminating and detecting different wavelengths.

These evolutions can contribute to Sustainability and the information and security needs of society.

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