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# Polysiloxane-based scintillators as radiation sensors: state of the art and future perspectives

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Polysiloxane scintillators are emerging as promising radiation sensors due to their high radiation resistance, mechanical flexibility, chemical inertness, and thermal stability compared to traditional plastics. This mini-review traces the evolution of these materials from early formulations to current high-performance scintillators. Their capabilities for particle discrimination, thermal neutron detection, and real-time dose monitoring in proton radiation therapy are highlighted. The chemical versatility of polysiloxanes allows the optimization of energy transfer and light yield, outperforming commercial plastics. Recent developments have led to photocurable, 3D-printable polysiloxane scintillators, opening new avenues. Despite remarkable progress, much room remains for innovations by leveraging the chemistry of siloxane precursors. The entrance of polysiloxanes into the landscape of radiation detectors represents an opportunity to further expand the applications of polymer-based scintillators.

## KEYWORDS

scintillators, polymers, silicones, particle discrimination, thermal neutron detection, flexible sensors

## Introduction

Polymer-based scintillators have found widespread use in several fields of physics as radiation sensors due to their peculiar chemical and physical properties. They are lightweight, easy to produce in different volumes and shapes, commonly offer a fast response in the order of ns to incoming radiation, and they are considered highly tissue-equivalent for density and elemental chemical composition. In high-energy physics, these features are harnessed to collect and reveal the extremely low energy deposited by incoming radiation by designing large-volume, highly segmented calorimeters based on numerous organic scintillator tiles or cubes and boarding lightguides to deliver scintillation light to faraway photoconverter devices (Fessler et al., 1985; Pari et al., 2019). In medical physics, their classical use as X-,  $\gamma$ -, and  $\beta$ -rays detectors, extensively adopted for real-time dose monitoring (Beddar, 2006), has been widened to the field of proton therapy dosimetry (Robertson et al., 2013). Moreover, organic scintillators also find practical and extensive use in nuclear safety control at borders to prevent the illicit trafficking of radioactive materials (Kouzes et al., 2008) and as neutron detectors in nuclear power plants, boron neutron capture therapy (BNCT) centers, and neutron-producing research facilities.

Although plastic scintillators based on carbon polymers appear to be favored for a wide variety of applications, they suffer from some drawbacks that eventually limit their use in specific fields; over the years, the class of polysiloxanes has proved an alternative.

In this mini-review, we will go through the most significant results achieved using polysiloxanes as radiation sensors in the broader sense of the classification. In particular, we will revise relevant outcomes related to radiation tolerance compared to traditional plastics as evidenced through different *in situ* and *ex situ* methodologies. This topic may be considered a boost for research into synthetic routes, production techniques, and different fluorophores to attain polysiloxanes scintillators, to not only reveal ionizing radiation—ions, X-,  $\beta$ -, and  $\gamma$ -rays—but also fast and thermal neutrons. This flourishing branch of research will be covered as well as the recently demonstrated capability of silicone scintillators to distinguish different radiations on the basis of time-resolved study of light pulses. Finally, the latest results on the synthesis of UV-curable, 3D-printable polysiloxane scintillators are discussed. Radiation resistance, sensing features, and versatility in manufacturing techniques make the advent of polysiloxanes in the field of radiation detectors an opportunity to further expand the application of polymer-based scintillators.

## Polysiloxanes as radiation-tolerant scintillators: from early stages to high-performance sensors

Traditional organic scintillators are made of carbon-based polymers, with the addition of fluorescent molecules in order to achieve optimal light yield under irradiation. Their practical use covers all the previously mentioned applications, so much so that plastic scintillators based on aromatic polymers now dominate the commercial offering. Notwithstanding their optimal features, a novel research field covering scintillators based on polysiloxanes emerged in the late 1980s and has since produced continuous new developments. The reason for this lies in the unique features of the Si–O–Si chemical bond, which might be exploited to overcome the known limitations of plastics. In particular, plastic scintillators suffer modest radiation resistance (Li et al., 2005), developing fogging and cracks when exposed to high humidity and temperature swings. With a consequent reduction in light response due to scattering effects, they are sensitive to organic solvents and temperature changes. In most cases, these drawbacks have been addressed through innovative formulations or synthetic pathways, but radiation-induced damage, lack of mechanical flexibility, and sensitivity to organics are inherently bound with the chemical nature of carbon-based polymers. In such a scenario, polysiloxanes offer great benefits because this macromolecule is composed of bridging Si–O bonds, the strength, rotational freedom, and partial ionic character of which account for a set of valuable physical and chemical properties. Among these, the most relevant properties to the field of radiation sensors—radiation and thermal resistance, high refractive index, low water and chemical permeability, and mechanical deformability—have been extensively explored and harnessed to meet the demanding requirements of each specific application.

Focusing on radiation tolerance, the quest for scintillators with enhanced radiation resistance compared to commercial, carbon-based plastics (i.e., BC-400, Bicron, and EJ-200, Eljen Technology) led several decades ago to the first experiments with polysiloxanes by the teams of Feygelman, Harmon, Zorn, and Bowen (Zorn et al., 1991; Harmon et al., 1991; Bowen et al., 1989). As a first step, copolymers containing dimethyl–diphenyl units along the chain and homopolymers based on methylphenyl siloxane repeating units were produced either by chemical industries or by research laboratories. Surprisingly, after cross-linking of the base and viscous resins added to selected fluorophores, the solid gums proved to be highly resistant to high doses of  $\gamma$ -rays, as demonstrated by the comparison of the transmittance spectra of the polysiloxane samples produced and of the plastic standard before and after irradiation. This optimal result related to radiation resistance was confirmed in subsequent years by different studies on several formulations of polysiloxane-based scintillators containing different dye combinations (as detailed below). But how were these new silicone-based scintillators designed? What are the basic properties to consider in order to obtain a polysiloxane with good light emission? As for light output, the design relied on previous, in-depth studies on the energy transfer mechanisms performed, in the case of carbon-based matrices by Birks (1964). Intrinsic fluorescence of the polymer itself or the use of non-fluorescent polymers, such as polymethylmethacrylate, added with a secondary fluorescent solvent, is a *conditio sine qua non* for the assembly of a well-performing scintillator. In the case of fluorescent base polymers such as polystyrene, phenyl pendant groups must be present in the macromolecule as the phenyl ring  $\pi$ -electrons' delocalization leads to emission in the UV range as a result of  $\pi \rightarrow \pi^*$  excitation transitions (Valeur, 2001). Relevant, in-depth studies on the correlation between the polymethylphenylsiloxane homopolymer or co-polymer structure related to the presence of alternating dimethyl–diphenyl units or dimethyl–methylphenyl units and their concentration and the photophysical properties of diluted solutions of polysiloxanes have been independently conducted by Salom et al. (1987), Horta et al. (1999), and Itoh (2001). They particularly focused on the presence of excited state dimers or excimers, their amount, and their emission maximum as well as the excimer-to-monomer intensity ratio— $I_{E/M}$ . This ratio is found to increase with phenyl concentration, as confirmed by the optical studies of Quaranta et al. (2010a) on solid-state polydimethyl-co-diphenylsiloxanes with varying contents of diphenyl units. Beyond the influence on optical emission features, the presence of a remarkable amount of phenyl substituents is crucial for another reason. This requirement is further strengthened by the need for fluorophore solubilization—primary dye and waveshifter—that usually belong to the class of aromatic compounds. This need is mandatory since the emission quantum efficiency of the bare polymer is very low, usually below 0.15, and the emission wavelength lies in a range (in general 250–350 nm) where most of the photoconverters, either silicon detectors or photomultiplier tubes, are almost insensitive. Moreover, the addition of the waveshifter reduces self-absorption phenomena, thus allowing scintillation light transmission at longer distance—higher attenuation length—and the production of large-volume scintillators with preserved performance. The primary dye, whose concentration is in the order of a few percent wt., receives

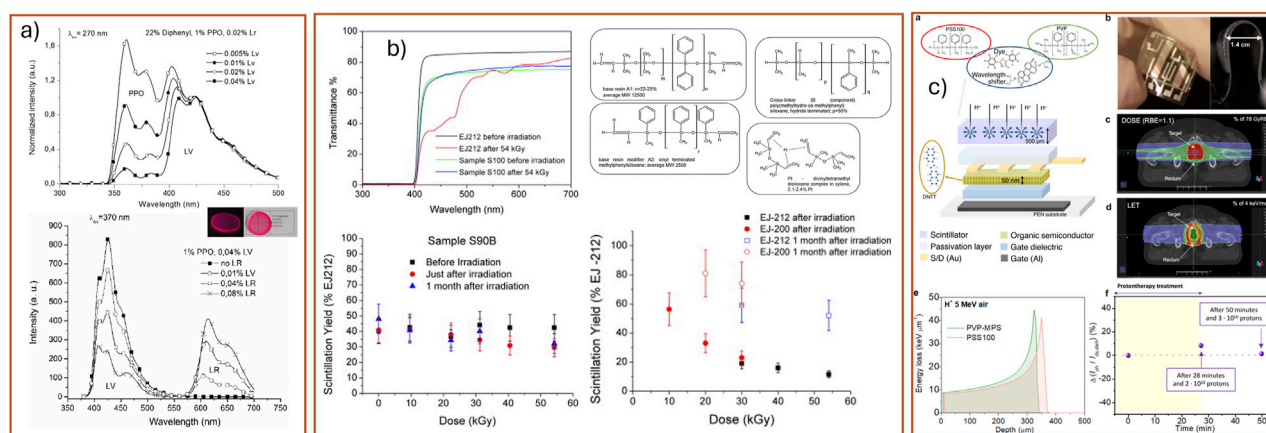


FIGURE 1

(a) Fluorescence spectra of polydimethylsiloxy doped with varying amounts of dye to optimize the energy transfer between the matrix and primary/secondary dye (Dalla Palma et al., 2014). (b) Radiation tolerance assessment of polysiloxanes either as homopolymers or blends as proved by transmittance and light yield changes following irradiation with  $\gamma$ -rays compared to standard plastic (Quaranta et al., 2013a). (c) Fully flexible device encompassing polysiloxane scintillator as sensor and organic thin-film transistor (OTFT) as photoconverter to be used for real-time dose monitor in proton therapy treatment: structure of the device, simulation of the proton dose delivery in the specific target corresponding to the cancer volume, and results of high-dose irradiation on the response of the device are shown (Calvi et al., 2023).

through efficient energy transfer enough excitation to emit light at longer wavelength at the limit of visible range. The waveshifter, loaded in the order of 0.01%–0.05% wt., plays a role in further shifting the emission toward the maximum sensitivity of photodetectors and in turn enhancing the attenuation length of the scintillator owing to reduced self-absorption. Energy transfer can take place through different processes, including radiative, non-radiative dipole–dipole exchange, known as “Förster transfer,” diffusion, or collisional exchange mechanisms. A complete and detailed description of energy transfer mechanisms and their kinetics has been comprehensively provided by Birks (1964), Birks and Conte (1968), and Hallam and Birks (1978); Steingraber and Berlmann (1963), Berlmann (1960), and Berlmann (1961); Avivi and Weinreb (1957); and Bengtson and Moszyński (1974) and Bengtson and Moszyński (1978) and has been extensively recently reviewed by Hamel (2021). The traditional dye composition for plastic, carbon-based scintillators—*p*-terphenyl and 1,4-bis(5-phenyl-2-oxazolyl) benzene (POPOP)—is not sufficiently soluble in phenyl-containing polysiloxanes. As explained in Birks (1964), a suitable concentration of primary dye and wavelength shifter is approximately 1% wt. and 0.1% wt., respectively. In polyphenylsiloxy, an evident lack of solubility is observed for *p*-terphenyl amounts as high as 0.1% wt., thus hampering the use of a “traditional” fluorophore mixture. This has led to the need to redesign the material composition in order to optimize the energy transfer between the various components. In fact, regardless of how different the energy transfer mechanisms may be, in all cases it starts from the necessity that there is at least a partial overlap between the excitation of the acceptor and the emission of the donor. In the case of phenyl-containing siloxanes, either homopolymers or copolymers, the photophysical properties of the base matrix have been extensively studied, starting from model compounds and ultimately arriving at polymers (Salom et al., 1987; Itoh, 2001; Quaranta et al., 2010a; Quaranta et al., 2010b). Fluorescence

emission from excited states in the form of monomer and excimer, which results from the interaction of phenyl chromophore in excited states with adjacent ground states, has been observed and correlated to the polymer structure. On these bases, several polysiloxane scintillator compositions with different dye combinations have been produced and analyzed in terms of optical properties and light output under irradiation with ionizing radiation (Bowen et al., 1989; Zorn et al., 1989; Feygelman et al., 1990), and their response has been compared to the standard plastic scintillator. Over the years, the response to alpha particles,  $\gamma$ -rays, and ion beams has been enhanced through extensive experimental work on both matrices and dyes, with the aim of not only optimizing energy transfer but also shifting the emission wavelength to ranges more suitable for the sensitivity of the photoconverter; red-emitting siloxane-based scintillators have thus been prepared (Dalla Palma et al., 2014).

In Figure 1a, fluorescence spectra of poly dimethyl-co-diphenylsiloxy are doped with different amounts of 2,5-diphenyloxazole (PPO) as primary dye and Lumogen Violet (LV) and Lumogen Red (LR) as waveshifters (Dalla Palma et al., 2014). Extensive work on the synthesis of new fluorophores with intrinsically high quantum efficiency in photoluminescence, optimal solubility in commercial phenyl siloxanes, and good matching in spectral features of acceptor and donor species led to scintillators that actually exceed the capabilities of commercial scintillators, as demonstrated by Lim et al. (2020). Concerning radiation resistance, several years after the pioneering work cited above, Quaranta’s and Carturan’s in-depth study on homopolymers, co-polymers, and blends based on siloxane under proton beams and intense  $\gamma$ -rays irradiation highlighted the persistence of performance using both post-irradiation characterizations and ion-beam-induced luminescence (IBIL) for *in situ* analysis of radiation damage (Quaranta et al., 2010b; Quaranta et al., 2013b; Quaranta et al., 2010c). In Figure 1b, the comparison of transmittance spectra and light yield measurements prior to and following irradiation of the

standard plastic scintillator with polysiloxanes provides clear evidence of the superior performance achievable with the latter. More recently, the radiation damage, limit of detection, and dose monitoring sensitivity of a flexible proton sensor encompassing a thin layer of polysiloxane scintillators coupled with a flexible photoconverter device have been tested under low-energy proton beams (Figure 1c) (Calvi et al., 2023). The same device has been tested at the proton therapy center APSS (Trento Hospital) on an anthropomorphic phantom and proved to be a wearable, human tissue-equivalent, real-time dosimeter for use during patient cancer treatment (Fratelli et al., 2025).

## Particle discrimination and thermal neutron detection with polysiloxane scintillators

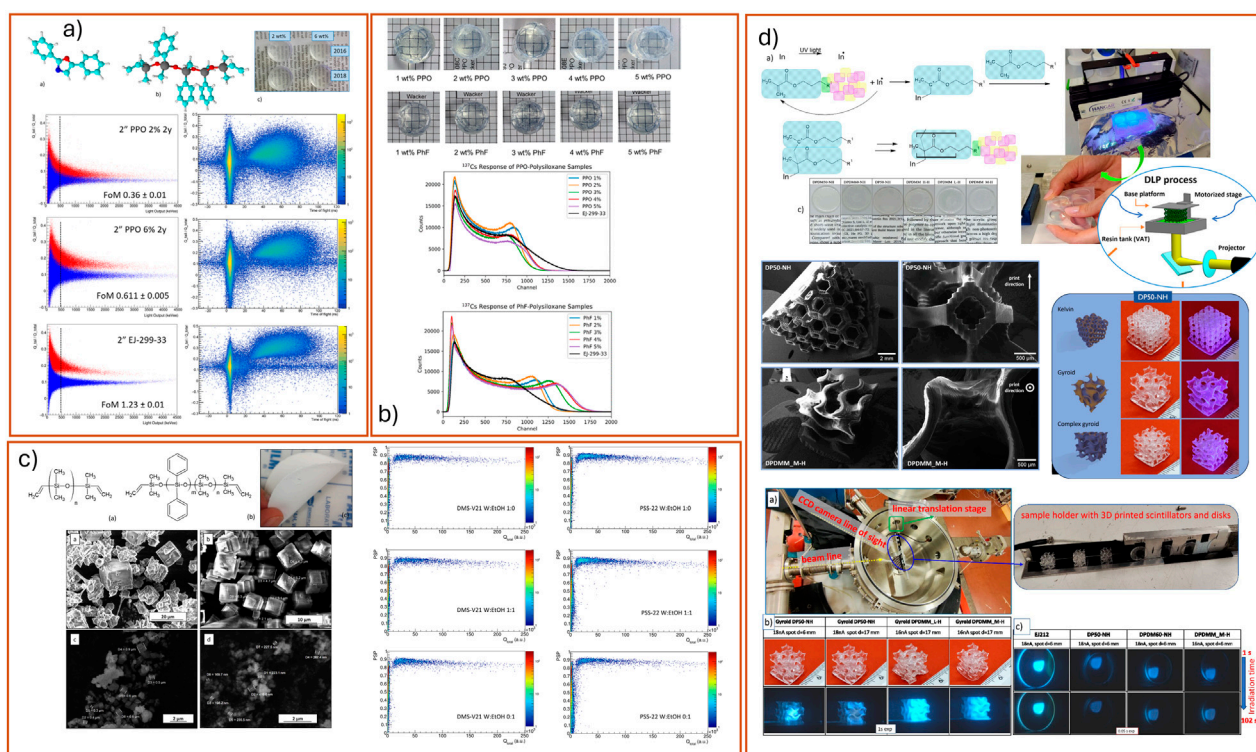
The ability to discern various particles using polymer-based scintillators was discovered several decades ago by Brooks (1979), who observed that the addition of naphthalene or mono-isopropyl-biphenyl as a secondary solvent to an aromatic polymer base of polyvinyltoluene or polystyrene doped with fluorophores surprisingly enabled neutron–gamma discrimination based on the different shape of the light pulse. Several decades later, Zaitseva et al. (2012) rediscovered this research topic and made great progress in the production of plastic-based scintillators with particle discrimination ability, also at large scale and with long-lasting performance (Zaitseva et al., 2024). By leveraging the ability to boost triplet-state concentration via primary dye concentration adjustments, the Zaitseva group demonstrated that neutron–gamma discrimination was achievable using plastics. The process that accounts for discrimination on the basis of different time profiles of the scintillation pulse has been known for many years and was clarified by Berlmann and Steingraber (1973). The technique based on over-doping the base polymer with primary dye—Zaitseva used 2,5-diphenyloxazole—leads to a close proximity of primary dye molecules and, in turn, of ground and excited states. Light pulse shape is affected by the different linear energy transfer (LET) of impinging particles, in that high-LET ionizing particles, such as alpha or protons, induce a high ionization density in the material, thus increasing the population of triplet states  $T_1$ . Annihilation of vicinal  $T_1$  states leads to *delayed fluorescence* through the process  $T_1 + T_1 \rightarrow S_0 + S_1 \rightarrow S_0 + S_0 + h\nu$ , giving rise to a time profile characteristic for these particles. As a result, the possibility of disentangling the contribution to light pulse led to high-LET ionizing particles, like protons, generated by the elastic scattering interaction of neutrons with matter, and low-LET  $\gamma$ -rays are exploited by using suitable data processing methods (Ranucci, 1995). Siloxane-based scintillators are available with different formulations for phenyl groups and conformation of the macromolecule. This versatility in designing chemical structures and, in turn, the polarity of the medium might be harnessed to allow the addition of primary dye in high concentration, preserving optimal solubility and avoiding dye aggregation. In fact, primary dye over-doping, aimed at promoting excimer formation, can extend well beyond the necessary concentration to achieve optimal light output—hence, maximum solubility limit must be considered. Marchi et al. (2019) first demonstrated that n- $\gamma$  discrimination capability in polymethylphenylsiloxane is pursued using a much lower

concentration of 2,5-diphenyloxazole (PPO) as primary dye than in case of carbon-based matrices. This fact has been related to the intrinsic features of siloxanes: torsional freedom of the Si–O bridges, bond length, and bond angles lead to enhanced mobility of the dissolved dye molecules within the matrix, while in carbon-based polymers, their diffusion is hindered by the rigidity of the structure. Therefore, stacks of phenyl substituents of the macromolecule act as harvesting sites for PPO molecules due to local higher chemical affinity in terms of polarity, ultimately promoting the formation of excimers. The optimal figure of merit in n- $\gamma$  discrimination, 0.611 evaluated at 480 keVee, has been reached using 6% wt. PPO only and the persistence of discrimination capability over 2 years have also been demonstrated (Figure 2a). More recently, Lim et al. (2020) exploited commercial phenyl-containing polysiloxanes produced either by Wacker or Shin-Etsu as base matrices for scintillators with n- $\gamma$  discrimination features, adding either traditional PPO dye or home-synthesized new primary dopants based on fluorene derivatives (Figure 2b) (Arrue et al., 2023; Lim et al., 2020; Han et al., 2022). Outstanding results have been demonstrated related to light yield, PSD capabilities, aging, and thermal resistance, outperforming the very same features of commercial plastic scintillators. As for thermal neutron detection, the pioneering work of Bell et al. (2004) inspired several research groups in the design of both functionalized siloxanes (Quaranta et al., 2011; Carturan et al., 2011) and boron-based elemental-organic compounds suitable for dissolution in silicones (Chandler et al., 2024) to indirectly reveal thermal neutrons through the nuclear reaction capture on  $^{10}\text{B}$ -producing ionizing  $\alpha$  particles. Analogously, indirect revelation through  $^6\text{Li}$  addition, thus exploiting the capture and generation of back-to-back  $\alpha$  and triton, has been also extensively explored and led to remarkable results for detection efficiency (Figure 2c) (Carturan et al., 2019; Pino et al., 2023).

## Polysiloxane-based nanocomposites as stretchable and tough radiation sensors

Beyond the nanocomposite thermal neutron detector described in the previous section, much effort has been dedicated to the fabrication of flexible, real-time ion beam and X-ray monitoring. Radiation-resistant scintillators have adopted the approach of nanoparticle inclusion in the elastomeric silicone matrix. In the field of oncological surgery, polydimethylsiloxane resin added with gadolinium oxysulfide:terbium (GOS:Tb) powder (Dietsch et al., 2025) allowed the production of scintillating soft and stretchable films for intraoperative X-ray imaging of tumors, thus enhancing the precision of surgical removal. Real-time proton beam visualization on the patient's skin during a proton therapy run is highly beneficial for reducing the impact of patient movements during irradiation. This challenging task can be pursued by the patient wearing garments with fabrics impregnated with polysiloxane loaded with Ag-doped ZnS nanoparticles (Yamamoto et al., 2025) so that the proton beam image can be collected in real time using a CCD/CMOS camera. While in these described scenarios, and other similar approaches cited in Dietsch et al. (2025) and Yamamoto et al. (2025), the silicone matrix just acts as a suitable host for the inorganic scintillating particles, and a much more relevant role in determining scintillation efficiency, environmental stability, and radiation tolerance is played by functionalized polysiloxanes





**FIGURE 2**  
**(a)** Main outcomes related to PSD achieved with polysiloxanes adopting the primary dye over-doping approach; transparency preservation and long-lasting performance are highlighted (Marchi et al., 2019). **(b)** Polysiloxanes from commercial resins and in-house fluorene-based synthesized primary dye outperform standard plastic scintillators in light yield (Lim et al., 2020; Arrue et al., 2023). **(c)** Nanostructured polysiloxane-based scintillators containing 6Li as thermal neutron converter display optimal detection efficiency and high  $\gamma$ -ray rejection (Carturan et al., 2019). **(d)** Additive manufacturing by digital light processing (DLP) led to polysiloxane scintillators with complex geometries showing optimal proton beam detection capabilities (Carturan et al., 2024).

described by Zheng et al. (2024a) and Zheng et al. (2024b), where metal-halide perovskite nanocrystals (PVKs) demonstrated superior stability in trifluoromethyl-substituted polysiloxanes. The exciting optoelectronic properties of PVKs have attracted great interest from many different research groups in the last decade owing to the high photoluminescence quantum yield resulting from the efficient passivation of surface shallow defects of the nanocrystals pursued by capping ligands, such as oleylamine and oleic acid. However, the ligands proved to have limited stability in harsh environments, thereby hampering the PVKs and their composites' applicability. In Zheng et al. (2024b), a mutual reinforcement of both PVKs and the polytrifluoromethylsiloxane-embedding matrix was demonstrated; while superior luminescent stability of PVKs under UV and X-ray irradiation was achieved owing to strong ion-dipole interactions, the polysiloxane matrix showed enhanced mechanical strength under high-dose irradiation thanks to the free-radical-scavenging effect of metal-halide nanocrystals.

### 3D-printed siloxane scintillators with complex geometries

Aiming at radiation sensing using carved or complex scintillators—as in the case of high-energy physics calorimeters (Acerbi et al., 2020) or range prediction and mapping of dose

distribution in clinical proton beams (Beddar, 2019)—the production of photocurable, 3D-printable polysiloxane scintillators has been pursued, and very recently, additive manufacturing through digital light processing (DLP) of sol-gel-derived precursors has been demonstrated. The scintillators display light output as high as 44% of the standard EJ-212 and proved to reveal low-energy proton beams through CCD camera image collection (Carturan et al., 2024). A summary of results is reported in Figure 2d. Further experiments are currently ongoing to provide proof of concept of their implementation on the ground both in high-energy physics and proton therapy facilities.

## Conclusion and outlook

Polymer-based scintillators have been historically applied not only in nuclear physics experiments but also in whatever scientific and technological fields to reveal and discriminate particles using lightweight, low-cost, high-volume sensors, offering dose monitoring in real time and particle discrimination capability. In the last three decades, scintillators based on polysiloxane have been produced and extensively studied from many different perspectives depending on the application for which they have been designed. Polysiloxanes are unique as scintillators in that they offer unprecedented radiation tolerance, mechanical deformability,

chemical inertness, and thermal resistance. In this mini-review, we have retraced the various stages that led from the first silicone-based scintillators to the most high-performing materials currently available, encompassing scintillators with features that even outperform those of commercial plastics regarding light output, radiation tolerance, mechanical flexibility, and wearability. In this scenario, the latest exciting evidence of the applicability of polysiloxanes has been demonstrated, with a focus on mixed-radiation field sensors with PSD capability, thermal neutron detectors, and real-time, *in situ* dose monitors in proton therapy treatments. In spite of these remarkable achievements, there is still considerable room for improvement and innovation in these materials: the chemistry of polysiloxanes is extremely versatile, being able to draw from polymer synthesis through various methods, as well as sol-gel synthesis, starting from silicon-based precursors with countless possible functionalities. A striking example of this versatility is the cutting-edge development of scintillators using photocurable polysiloxanes, enabling sensor fabrication through additive manufacturing techniques.

Nevertheless, some issues currently limit the spread of using polysiloxanes in different areas of interest, from nuclear physics to high-energy physics and medical physics, on a commercial scale that need to be addressed. For  $\gamma$ -ray detection, and more generally for the detection of low-LET ionizing particles, the interaction volume is critical, and therefore large-scale polysiloxane scintillators are needed. Good performance has been observed in the case of laboratory-made, large-size cylinders with a diameter and thickness of 50 mm (2 inches) (Marchi et al., 2019). However, production methods specifically designed for large-scale scintillators should be proposed and validated, starting from commercially available resins, and a comprehensive evaluation of production costs also should be made.

## Author contributions

SC: Validation, Data curation, Supervision, Methodology, Conceptualization, Resources, Investigation, Funding acquisition, Writing – original draft, Visualization. SM: Funding acquisition, Resources, Formal analysis, Methodology, Writing – review and editing, Investigation, Software. AQ: Writing – review and editing, Investigation, Conceptualization, Funding acquisition, Supervision, Resources, Methodology.

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## Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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