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Matthieu Hamel *Editor*

Plastic Scintillators

Chemistry and Applications

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Foreword

As one looks back over the history of radiation measurements, one finds distinct eras of intense development and progress, sometimes driven by emergent applications or by innovations in other fields. The initial radiation detectors were the gas-filled detectors (ionization chamber, proportional counter, and Geiger–Mueller tube) which stood as the dominant form of radiation detector until ~1950. The inorganic scintillator, introduced by Hofstadter, then revolutionized radiation detection for gamma rays since it offered high efficiency and reasonable energy resolution. The introduction of new inorganic scintillators was slow but steady, driven to meet particular application needs. The focus of detector development was changed by the introduction of the silicon semiconductor (~1970). Even with its small size, the superb energy resolution for charged particles opened advanced measurement possibilities. Larger detector volumes, better energy resolution, and higher efficiency became possible with germanium, which evolved to become today's gold standard for gamma-ray energy resolution. Such was the general state of our field at the beginning of this millennium.

The September 11, 2001 terrorist attack in New York City heralded a shift in the direction of radiation detector R&D. Coupled with the activity inspired by CERN's Crystal Clear Collaboration, an intense push was developed to deploy radiation sensors on a large scale for national security. This has led to two decades of development focused almost entirely on **inorganic** scintillators. The goal has been to develop large area, highly efficient, robust, inexpensive, gamma-ray detectors with much better energy resolution and particle discrimination than was previously available. Figure 1 from [1] depicts the productive outcome of this impressive effort.

The point of recalling this history is to show that inorganic scintillators have been the focus of the R&D limelight for decades, garnering the attention for both analytical method and material development. In the meantime, their organic counterparts have quietly evolved and continue to produce some very attractive detector materials.

While the push to improve detector performance has led to a slew of new inorganic scintillators, only a few of these have proven to provide practical solutions to real problems, particularly for the market of security applications. These applications

require large area and volume to subtend and stop incident neutrons and gamma rays coming from afar, have a low cost for widespread deployment, high environmental robustness, and simplicity in execution. Since inorganic scintillators and semiconductors have difficulty in meeting these constraints, attention has now fully swung to explore and exploit the organic scintillators.

Any history of organic scintillators will inevitably include the fiducial works of J. B. Birks [2] and F. D. Brooks [3] (the latter reference provides a nice review of the field up to ~1980). But since then, a small group of dedicated researchers have pushed to improve light yield, understand molecular dynamics and quenching phenomena, improve environmental acceptability, and leverage the ability to discriminate the fast neutrons from the gamma-ray background. What has been lacking is a comprehensive treatise that elucidates the science governing these processes, the existing and new materials and their properties, and the future pathway. This much-needed volume, written by these dedicated researchers, admirably fills this gap.

Whereas inorganic scintillators derive their scintillation capability from their crystalline properties, organic scintillators can function in liquid, plastic, or crystalline form. This obviates the necessity of growing crystals atom-by-atom, which enables larger, cost-effective volumes to be achieved. Since organic scintillators can be acquired for ~$ 1/cm^3, as compared to ~$ 50–100/cm^3 for their inorganic counterparts, large volumes strongly favor organics. This property (in addition to their prompt scintillation decay times) has led to their widespread adoption, e.g., for security applications and rare event physics searches [4], despite their inherent disadvantages of low density and efficiency, limited spectral capability, and age-related degradation. But over the last decade, material advances have made these disadvantages less operationally restrictive. This can be seen in the commercially available, larger stilbene detectors with reasonable light yield and particle discrimination capability, and newer plastic scintillator properties that rival the traditional liquid scintillator [5].

By loading scintillators with dopants of heavy metals or neutron absorbers, the effective stopping power of any organic scintillator to gamma rays or neutrons can be dramatically improved, and recent work has led to larger volumes and higher scintillation light yields (likely due to reduced quenching) [6]. If the current light yields of ~10 photons/keV can be improved by factors of ~3–5, the spectroscopic capability can become competitive with some inorganics without requiring heavy spectral unfolding. Exploiting new work in nanotechnology, nanoparticles have been homogeneously introduced into a matrix and have led to new detector materials that are individually sensitive to slow and fast neutrons, as well as gamma rays. This triple particle discrimination ability could foreshadow a significant advance for homeland security applications [7].

While the past few decades have brought remarkable developments in inorganic scintillators (e.g., LYSO, $LaBr_3$, SrI_2, CLYC, …), we are now on the verge of similar breakthroughs in organic scintillators. With a deeper understanding of the underlying

physical and chemical mechanisms in play, coupled with rapid and inexpensive fabrication, one can foresee more affordable, larger volume, and capable fast detectors that can fill critical roles in our most important detection applications. This volume is particularly useful not only for elucidating the current state of the art, but also for highlighting those research areas that will lead to these breakthroughs by our detection community.

Ann Arbor, MI, USA David K. Wehe

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Preface

Cross-disciplinary research is extremely pleasant to open-minded scientists who love to exit from their comfort background, and learn new subjects. The scintillation domain—and more particularly plastic scintillator, which is the subject of this book—is one of those disciplines that require skills in many fields such as organ(ometall)ic chemistry, materials chemistry, photophysics of condensed matter, electronics, signal processing, and nuclear physics. All these are linked such as a puzzle, a defective piece leading undoubtedly to scintillating systems of low detection performances. The extensive research that has been performed this these last two decades on fast neutron/gamma discriminating plastic scintillators is a living example: chemistry to stabilize the material or to introduce triplet-harvesting fluorophores, high-level photophysical understanding, new digitizers and solutions offering real-time pulse shape discrimination. Since this thematic is scientifically rich, it finds applications in many fields such as fundamental physics, homeland security, radioprotection, and environmental assays.

This book consists of 15 chapters written by leading experts in all the above-mentioned areas. Besides, I am very indebted to all the Authors for the time they have considered writing this wonderful book. It is divided in two main sections: Part I—*materials* is the chemical modifications of plastic scintillator and is composed of eight chapters. Chapter 1 introduces the concept of plastic scintillator and opposites it against another variety of state-of-the-art materials: inorganics scintillators. I am glad to be associated with C. Dujardin (expert scientist in inorganic scintillator) in this chapter. Chapter 2 is a success story from N. Zaitseva and collaborators from Lawrence Livermore National Laboratory about a fast neutron/gamma discriminating plastic scintillator that is now commercially available. Chapters 3 and 4 present the latest application-driven developments in elemental loading for two main applications: first is written by J. Dumazert and C. Frangville on thermal neutron detection, second on gamma spectrometry (and other applications) by G. Bertrand and M. Hamel. In Chap. 5, M. Koshimizu shatters the limits of plastic scintillation to their inorganic counterparts: composite scintillators are materials that merge all advantages of the two worlds. Chapter 6 focuses on a special polymer for scintillation mostly studied and developed by S. Carturan and A. Quaranta. The last two chapters of this first part are proposed by P. Feng and coworkers. The first chapter by Patrick

presents an overview on molecular design considerations for the three main organic scintillators: single crystal, liquid and plastic. The second chapter reports organic glass scintillators as a brand new class of materials that look particularly efficient in terms of scintillation properties.

In Part II—*applications*, standard, or chemically modified plastics are used in particular fields or combined with new possibilities in signal processing. In Chap. 9, B. Liu and X. Ouyang present the use of photonic crystals to mitigate light confinement in these materials. Chapter 10 by M. Thevenin and Y. Moline introduces plastic scintillators to smart algorithms and powerful analog stages to enhance their detection properties, putting at a higher scale new considerations in nuclear instrumentation. Chapter 11 is the application of plastic scintillators to gas detection by two field specialists: P. Cassette and K. Mitev. Then, the leading collaboration around S. Beddar presents the recent advances and clinical applications of plastic scintillators in the field of radiation therapy in Chap. 12. Other experts from the University of Barcelona (A. Tarancón and coworkers) present the use of plastic scintillators for environmental analysis in Chap. 13. This team is one of the few that gathers skills equally in material synthesis and their application to radiation detection. Chapter 14 by P. Laurent and coworkers initiates the readership to a less known application of plastic (and inorganic) scintillators: their use to study the Earth from ground to the magnetosphere. Then large experiments that use these materials are presented by P. B. Cushman and D. M. Poehlmann in Chap. 15.

This book is intended to become a screenshot of what are plastic scintillators in early 2021 (the date of completion of this book) and how they can be used as a simple, yet versatile sensor for nuclear instrumentation. Beginners will find all the definitions of the important parameters (light output, figure of merit, radiation/matter interaction), and experts will have access as well to the most comprehensive state of the art of the chemical modifications of these materials and revisited instruments. I hope the book will deserve the community to a better knowledge in this field.

Last, it is important to mention the "special" conditions the Authors have faced while preparing their chapters. This year 2020 has seen the spread of the COVID-19 disease. Most of our work has been upset due to lockdown(s) or subsequent delays on the project. I am very grateful the Authors succeeded, however, in preparing their manuscript in due time.

"l'important, c'est pas la victoire, mais le combat" Daïtro.

Saclay, France Matthieu Hamel

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Part I
Materials

Chapter 1
Introduction—Overview on Plastic and Inorganic Scintillators

Christophe Dujardin and Matthieu Hamel

Abstract Scintillators are materials that are able to emit photons when impinged with ionizing radiations. This family of materials is covered by both organic and inorganic structures, with some similarities but also with different photophysical processes occurring underneath. The scientific fields and communities are in fact rather separated, while it would benefit from more interactions and collaborations. This book is mostly focused on plastic scintillators, which are polymer-based materials, and the purpose of this chapter is to introduce and link them to well-known inorganic scintillators. In addition, hybrids materials are new developments based on inorganic nanocrystals in organic host. In such hybrid materials, a complex interplay occurs along the energy relaxation leading to the emission of light. Thanks to their chemical versatility, plastic scintillators can easily be modified. Whereas the first decades have seen their use as "all-purpose" detectors, the most recent developments afford specialization of the materials toward a given application. Thus, various modification stages are possible: the simplest is to tune its chemical properties. In addition, this material is an optical device, and complicated photophysical phenomena occur in the radiation/matter interaction volume. Finally yet importantly, current developments in artificial intelligence, as well as highly sophisticated algorithms, are used to overcome intrinsic limitations of plastics properties. This chapter thus gives a historical perspective on the development of plastic scintillators with a mention of past and current main actors. Then, a discussion follows on the basic principles in plastic scintillation design. Their main properties are finally presented and compared with inorganic scintillators. Some of these properties will be partially discussed herein since they will be fully explained in dedicated chapters.

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1.1 History of Scintillators

The first scintillator ever described was $BaPt(CN)_4$ for X-ray detection in 1895. It was under the form as a powder, now called X-ray phosphor and played a major role in medical x-ray radiography. With the emergence of photomultipliers, the research on scintillating materials has been intensified. Thus, plastic scintillators (PSs) were first depicted in the literature by Marvin G. Schorr and Franklin L. Torney as early as 1950 [1], a few months only after their liquid equivalents were discovered [2, 3]. Regarding inorganic scintillators as single crystals, the main known compositions were discovered after the 1950s as depicted in [4].

This first plastic was composed of *m*-terphenyl dissolved in polystyrene.[1] Rapidly various formulations appeared which highlighted the necessity to add a wavelength-shifter (i.e., a molecule that is able to absorb UV light from the primary fluorophore and emits at longer wavelengths) to the abovementioned binary mixture so as to afford better scintillation external efficiency, including the light extraction as mentioned by Pichat and Pesteil [5]. These discoveries lead to the commercial plastic formulations we currently know. In 1953, the first loading attempts with organometallics in the form of bismuth hexahydrobenzoate or triphenylmethyllead were published by the same French group, already with the aim of increasing the effective atomic number for gamma spectroscopy [6]. This scintillator modification is one of the hottest topics in the context of homeland security [7, 8], the another one is the pulse shape discrimination between fast neutrons and gamma rays [9, 10] which was first described by Frank D. Brooks in 1960 [11]. Concerning thermal neutrons (i.e., with kinetic energy in the range of 25 meV) detection, loading with dedicated elements allowing their capture appeared in the early sixties. Thus, lithium was described in a Russian patent [12] and boron in a British report, later on popularized in a paper from Anisimova [13], thanks to isopropenyl boron carbohydride. Gadolinium appeared a few years later in the form of gadolinium benzoyl acetonate at low concentration (typically $\leq$0.2% of Gd) [14]. Gamma spectroscopy and neutron detection with plastics will be fully described in dedicated chapters of this book. Some other application-driven but peculiar developments were also performed: fluorine loading for high-energy neutron detection [15–17], cadmium [18], hafnium [19], various lanthanides and actinides [20], etc. Various elements can thus be loaded into plastics. Selected developments for both inorganic and organic scintillators are summarized in Fig. 1.1.

Later, John B. Birks published the first book on the principle of scintillation counting in 1964, covering notably the latest developments and use of plastics and inorganics [21]. During these first fifteen years, plastics were extensively studied with the aim to find the best composition in terms of scintillation efficiency, decay time, chemical stability, etc. It is noteworthy that the well-known combinations of fluorophores, e.g., *p*-terphenyl/POPOP or PPO/POPOP were discovered at this time,

[1] Topological representation and key information of these molecules is given in the Appendix section at the end of the book.

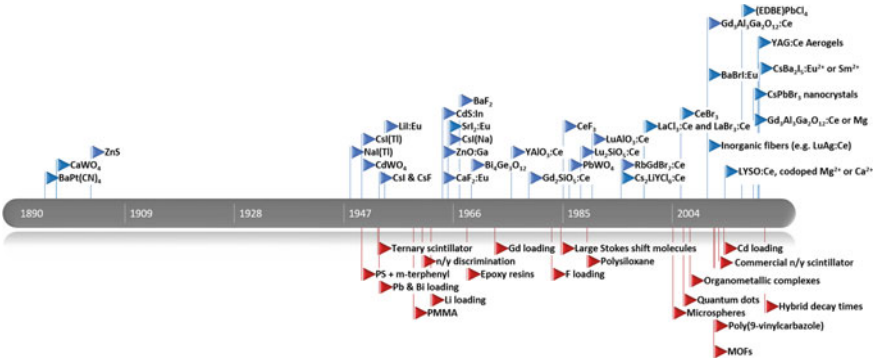

Fig. 1.1 Timeline of the main scintillator developments from 1890 to 2020 (illustrative only; for inorganics rather fast scintillators are considered)

and more details will be given in the following sections. Nowadays important developments are still performed as can be seen in the increasing number of publications talking about plastic scintillator modifications (Fig. 1.2).

Soon after the discovery of plastic scintillators, physics and chemical researches were followed by their industrial production. The Cold War probably took a key role as it involved important needs in the context of nuclear and radiological detection and radioprotection of the workers involved in the preparation of nuclear weapons. The two Blocs thus independently created their own facilities to cover them. Several companies emerged for the Western Bloc: *Pilot Chemicals* (USA, est. 1951), *National Radiac INC.* (Sintilon material, USA, before 1956 [22]), *Bicron* (USA, est. 1969), *Koch Light* (UK and USA, before 1969), and *Nuclear Enterprises Ltd.* (Scotland). Plastic scintillator NE 102 (NE standing for *Nuclear Enterprises*) was cited as early as 1959 [23]. Naton scintillators (*Nash & Thompson*, UK) appeared in the first part

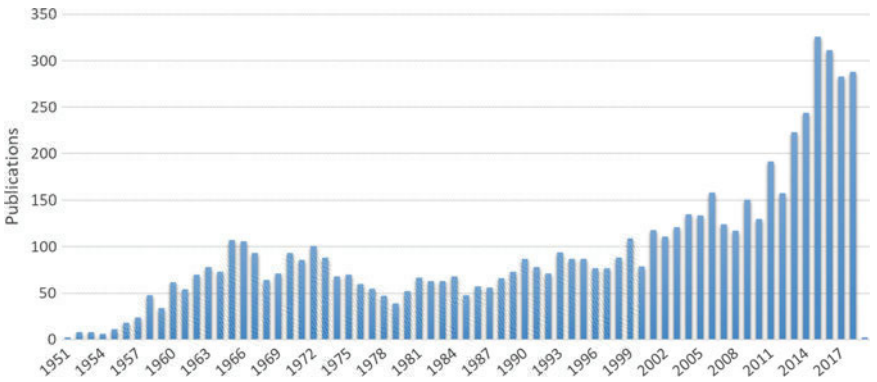

Fig. 1.2 Number of publications referring chemical modifications of plastic scintillators and sorted yearly (*source* SciFinder®, as of November 2019)

of the 1960s. Later on, the Pilot-*x* references appear in the *Nuclear Enterprises* catalog. *Altulor* (France, 1957) and *Polivar* (Italy) were involved in the preparation of PMMA-based PSs, mostly for CERN experiments during the years 1985–1990. Some old catalog front covers can be seen in Fig. 1.3.

From the Eastern Bloc, three main institutes were created. First is the *Institute for High-Energy Physics* (IHEP, USSR, 1963). The research and development program of plastic scintillators was started in the 1970s from the initiative of V. Rykalin. In the beginning, the technology of manufacturing PSs by the method of large-block thermal polymerization (SC-2*xx* series) followed by mechanical treatment and polishing was settled. Then extruded plastics (SC-3*xx* series) were extensively studied. Other trade codes from IHEP exist under the form PSM-*xxx* series. For example, PSM-115 (an injection-molded plastic) was used in HERA-B (a particle physics experiment at the HERA accelerator at DESY, see Chap. 15). These PSM-*xxx* codes have disappeared from IHEP catalog. The maximum dimensions of the bulk scintillators are 200 × 50 × 10 cm, with a production of up to 100 tons per year. Reference [24] presents all the key developments of IHEP in the field. In Ukraine, the *Institute of Scintillating Materials* (ISMA) was established in 2002 as a part of *Institute for Single Crystals* who found birth as early as 1955. ISMA has extended its inorganic research expertise to several other materials, including thus plastic scintillators. Former Czechoslovakia also has a long history in scintillators, whose research and development work started as early as 1952 in the Research Institute of Electronic Physics. In 1959, research and production were associated with the *TESLA National Corporation.* In the early 1990s, part of this institute was transformed into a private-owned company called *SM&D* (*Scintillation Materials and Detectors*). This company was then acquired by *Envinet a.s.* in 2008, then *Nuvia CZ* in 2016.

In 1990, *Bicron* claimed a production capacity of 12 tons of plastic scintillator per month. It seems that *Saint-Gobain Crystals and Detectors* acquired both *Nuclear Enterprises* after 1987 and *Bicron* after April 1993. In 1997, *Eljen Technology*, a subsidiary of *Ludlum Measurements* was created by C. Hurlbut (formerly from *Bicron*) in Texas.

It is also worth mentioning *Kuraray* from Japan, who started the production of scintillating fibers but also some plastics as early as 1982. In 2013, the discovery of the Higgs Boson was achieved in part with the use of these scintillating fibers. Currently, *Kuraray* is a global leader together with *Saint-Gobain Crystals and Detectors* in fibers technology. Table 1.1 inventories current manufacturers as well as some trade characteristics. It seems also that other companies may provide scintillators on-demand, such as *FUJITOK* [25] or *Plaken co., Ltd* [26] in Japan; such companies are not listed in Table 1.1.

Through history, it may be difficult to find reliable data on all commercial scintillators. Table 1.2 lists the various scintillator codes their main property. This table could be useful for further cross-comparison since several materials are now out of commercialization. Some other commercial PSs might incidentally be omitted. Despite this large choice (especially in the case of standard formulation, which is obviously both the most available and the most sold as well), there are still exciting chemical challenges to overcome and new possibilities to be discovered. They will now be described.

Fig. 1.3 Catalog front covers of Nuclear Enterprises (1980), Bicron (1990) and TESLA (late 1970s), and advertisement of National Radiac INC. (this advertisement is reproduced from [22] with permission from the American Chemical Society)

Table 1.1 Main providers of plastic scintillators (in alphabetic order). Some companies may be resellers of other manufacturers

Name	Country	Trade codes	Estimated number of products
Advatech UK Limited	UK	n.d	Amcrys reseller
Amcrys	Ukraine	UPS-*xxx*	> 10
Beijing Nuclear Instrument Factory	China	ST-4*xx*	n.d.
Detec-Europe	France	n.d	Amcrys reseller
Eljen Technology	USA	EJ-2*xx*	21
Epic-Crystal	China	n.d	1
Institute for High-Energy Physics	Russia	SC-2*xx* (bulk) SC-3*xx* (molded)	24
Kuraray	Japan	SCSN-*xx*	3
Nuvia CZ	Czech Republic	NuDET Plastic	2
Perkin-Elmer	UK	Meltilex®	1
Rexon	USA	RP-*xxx*	3
Saint-Gobain Crystals and Detectors	USA	BC-4*xx*	21
Shandong Haiqiang Environmental Protection Technology Co., Ltd	China	n.d	n.d.
Win-Trust	China	SP*xxx*	3

n.d. not determined

1.2 Plastic Scintillator Chemists

Figure 1.4 tries to overview the main teams, laboratories, institutes or companies that are currently or were previously involved in plastic scintillators modification, as well as their period of activity when the information is available. It may be sometimes difficult to judge whether active research is still ongoing in some groups. Only the key laboratories (with two or more publications written in this field) are shown, and collaborations cannot be added, so the publications are granted to the corresponding author.

1.3 The Scintillation Process in Plastics and Inorganic Materials/Crystals

Whatever the liquid or plastic nature, an organic scintillator can be resumed as a matrix that contains one or several organic fluorophores and potentially some dopants for giving special application features. These fluorophores are usually called primary

Table 1.2 Classification of past and current commercial scintillators. Scintillating fibers and prototypes are omitted

Main application	Scintillator code (alphabetical order)
General purpose	BC-400, BC-404, BC-408, BC-412, BC-416, EJ-200, EJ-204, EJ-208, EJ-212, NE 102A, NE 110, NE 114, Pilot F, RP-200, RP-400, RP-408, SC-201, SC-202, SC-205, SC-301, SC-302, SC-304, SC-306, SC-307, SC-308, SC-309, SCSN-38, SCSN-61, SCSN-81, SP32, SP101, ST-401, UPS-89, UPS-90, UPS-96, UPS-923A
Long decay time	BC-444(G), EJ-240, NE 115, UPS-92S
Ultrafast timing	BC-418, BC-420, BC-422(Q), EJ-228, EJ-230, EJ-232(Q), KL 236, Naton 136, NE 104, NE 104B, NE 111A(ZIP), Pilot B, Pilot M, Pilot U, Pilot U2, SC-206, SC-207, SC-305, UPS-91F
Green emitting	BC-428, EJ-260, EJ-262, SC-203, SC-204, SC-303, SP33, UPS-974
Red emitting	BC-430, NE 108*
Lead loading	BC-452, EJ-256, NE 142, SC-223, SC-322
Tin loading	NE 140, SC-221, SC-222, SC-321
Fast neutron/gamma discrimination	EJ-276(G), NE 150, UPS-113NG
Deuterated polymer	BC-436, NE 125
Boron loading	BC-454, EJ-254, SC-231, SC-331
High temperature applications	BC-434, BC-438, BC-440(M), BC-448(M), EJ-244, EJ-248, NE 160
Low energy gamma rays or X-rays	BC-450, NE 105
Radiation hard	PSM-115, SCSN-81 T, UPS-92RH, UPS-98RH

*in Nuclear Enterprises catalog this scintillator is mentioned to emit at $\lambda_{max} = 538$ nm, despite being classified as "red" plastic. However and according to the CIE rules, 538 nm is "yellowish green"

and secondary fluorophores, this name contracting to their respective role within the organic scintillator. In standard liquids or plastics, the matrix accounts for $\geq$95% of the material, so radiation/matter interactions occur here. What happens afterwards has been extensively described elsewhere [21] and will be introduced in several chapters of this book. In a few words, excitons are created and are transferred from the matrix to the primary fluorophore, usually (but not exclusively) by Förster Resonance Energy Transfer (FRET), which is a non-radiative transfer. The second process is radiative and undergoes emission/absorption between a donor and an acceptor; this is the roles of the primary and the secondary fluorophores, respectively. Ultimately, the emission wavelength and potentially the scintillation decay time are usually governed by the secondary fluorophores. But various exceptions exist.

Similarly, an inorganic scintillator can be seen as a host containing emitting centers. The latter can be a doping ion or a point defect (extrinsic), but can be as