

Topics in Applied Physics 140

Matthieu Hamel *Editor*

Plastic Scintillators

Chemistry and Applications

 Springer

Topics in Applied Physics

Volume 140

Series Editors

Young Pak Lee, Physics, Hanyang University, Seoul, Korea (Republic of)

David J. Lockwood, Metrology Research Center, National Research Council of Canada, Ottawa, ON, Canada

Paolo M. Ossi, NEMAS - WIBIDI Lab, Politecnico di Milano, Milano, Italy

Kaoru Yamanouchi, Department of Chemistry, The University of Tokyo, Tokyo, Japan

Topics in Applied Physics is a well-established series of review books, each of which presents a comprehensive survey of a selected topic within the domain of applied physics. Since 1973 it has served a broad readership across academia and industry, providing both newcomers and seasoned scholars easy but comprehensive access to the state of the art of a number of diverse research topics.

Edited and written by leading international scientists, each volume contains high-quality review contributions, extending from an introduction to the subject right up to the frontiers of contemporary research.

Topics in Applied Physics strives to provide its readership with a diverse and interdisciplinary collection of some of the most current topics across the full spectrum of applied physics research, including but not limited to:

- Quantum computation and information
- Photonics, optoelectronics and device physics
- Nanoscale science and technology
- Ultrafast physics
- Microscopy and advanced imaging
- Biomaterials and biophysics
- Liquids and soft matter
- Materials for energy
- Geophysics
- Computational physics and numerical methods
- Interdisciplinary physics and engineering

We welcome any suggestions for topics coming from the community of applied physicists, no matter what the field, and encourage prospective book editors to approach us with ideas. Potential authors who wish to submit a book proposal should contact Zach Evenson, Publishing Editor:

zachary.evenson@springer.com

Topics in Applied Physics is included in Web of Science (2019 Impact Factor: 0.633), and is indexed by Scopus.

More information about this series at <http://www.springer.com/series/560>

Matthieu Hamel
Editor

Plastic Scintillators

Chemistry and Applications

 Springer

Editor

Matthieu Hamel
Université Paris-Saclay, CEA, List
Laboratoire Capteurs et Architectures Electroniques
Palaiseau, France

ISSN 0303-4216

ISSN 1437-0859 (electronic)

Topics in Applied Physics

ISBN 978-3-030-73487-9

ISBN 978-3-030-73488-6 (eBook)

<https://doi.org/10.1007/978-3-030-73488-6>

© Springer Nature Switzerland AG 2021

This work is subject to copyright. All rights are reserved by the Publisher, whether the whole or part of the material is concerned, specifically the rights of translation, reprinting, reuse of illustrations, recitation, broadcasting, reproduction on microfilms or in any other physical way, and transmission or information storage and retrieval, electronic adaptation, computer software, or by similar or dissimilar methodology now known or hereafter developed.

The use of general descriptive names, registered names, trademarks, service marks, etc. in this publication does not imply, even in the absence of a specific statement, that such names are exempt from the relevant protective laws and regulations and therefore free for general use.

The publisher, the authors and the editors are safe to assume that the advice and information in this book are believed to be true and accurate at the date of publication. Neither the publisher nor the authors or the editors give a warranty, expressed or implied, with respect to the material contained herein or for any errors or omissions that may have been made. The publisher remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

This Springer imprint is published by the registered company Springer Nature Switzerland AG
The registered company address is: Gewerbestrasse 11, 6330 Cham, Switzerland

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/cm³, as compared to ~\$ 50–100/cm³ 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, LaBr₃, SrI₂, 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

References

1. C. Dujardin, E. Auffray, E. Bourret-Courchesne, P. Dorenbos, P. Lecoq, M. Nikl, A.N. Vasil'ev, A. Yoshikawa, R.-Y. Zhu, *IEEE Trans. Nucl. Sci.* **65**(8), 1977 (2018)
2. J.B. Birks, *The Theory and Practice of Scintillation Counting* (Pergamon Press, Oxford, 1964)
3. F.D. Brooks, *Nucl. Instr. Meth.* **162**(1–3), 477 (1979)
4. V.A. Li, T.M. Classen, S.A. Dazeley, M.J. Duvall, I. Jovanovic, A.N. Mabe, E.T.E. Reedy, F. Sutanto, *Nucl. Instr. Methods A* **942**, 162334 (2019)
5. N. Zaitseva, B.L. Rupert, I. Pawełczak, A. Glenn, H.P. Martinez, L. Carman, M. Faust, N. Cherepy, S. Payne, *Nucl. Instr. Methods A* **668**, 88 (2012)
6. G.H.V. Bertrand, J. Dumazert, F. Sguerra, R. Coulon, G. Corre, M. Hamel, *J. Mater. Chem. C* **3**(35), 6006 (2015)
7. C. Frangville, J. Dumazert, E. Montbarbon, R. Coulon, G.H.V. Bertrand, M. Hamel, *Mater. Chem. Front.* **3**(8), 1574 (2019)

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(ometallic) 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 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 opposes 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

Contents

Part I Materials

1	Introduction—Overview on Plastic and Inorganic Scintillators	3
	Christophe Dujardin and Matthieu Hamel	
1.1	History of Scintillators	4
1.2	Plastic Scintillator Chemists	8
1.3	The Scintillation Process in Plastics and Inorganic Materials/Crystals	8
1.4	Typical Preparation Process and Size Possibilities	11
1.5	Main Parameters and Tools for Modification or Improvement	13
1.5.1	Light Yield	14
1.5.2	Decay Time	16
1.5.3	Emission Wavelength	18
1.5.4	Behavior Against External Environment	19
1.5.5	Effective Atomic Number and Density	24
1.6	Summary	25
	References	26
2	Neutron/Gamma Pulse Shape Discrimination in Plastics Scintillators: From Development to Commercialization	35
	Natalia P. Zaitseva, M. Leslie Carman, Andrew M. Glenn, and Andrew N. Mabe	
2.1	Physical Basis for Neutron/Gamma Discrimination in Organic Scintillators	35
2.2	Plastic Scintillators with Efficient Fast Neutron/Gamma Discrimination	39
2.2.1	PPO-Based PSD Plastics	40
2.2.2	PSD Plastics Utilizing Alternative Dyes and Dye Mixtures	52
2.3	PSD Plastics for Combined Detection of Fast and Thermal Neutrons	68
2.3.1	<i>¹⁰B-loaded PSD Plastic Scintillators</i>	68

2.3.2	<i>⁶Li-loaded PSD Plastic Scintillators</i>	74
2.4	Commercialization and Further Directions of Studies	82
	References	86
3	The Detection of Slow Neutrons	91
	Jonathan Dumazert and Camille Frangville	
3.1	Slow Neutrons: Essential Features	91
3.1.1	The Definition of Slow Neutrons	92
3.1.2	The Origins of Slow Neutrons	93
3.2	Nuclear Reactions of Interest in Slow Neutron Detection	99
3.2.1	Natural Abundance, Reaction Cross Section, Q-Value, and Typology of Reaction Products	100
3.2.2	Main Nuclear Reactions of Interest	101
3.2.3	Size of the Scintillator: Slow Neutron Mean Free Path and the Interaction of Reaction Products	106
3.3	Detection of Reaction Products and n/γ Discrimination	109
3.3.1	Background Radiation	109
3.3.2	Pulse Height Discrimination	110
3.3.3	Pulse Shape Discrimination	112
3.3.4	Compensated Detectors	114
3.3.5	Multiplicity-Gated Detection	115
3.3.6	Capture-Gated Detection	115
3.4	Figures of Merit for Slow Neutron Detectors	117
3.4.1	Figures of Merit About the Response to Neutrons	117
3.4.2	Figures of Merit About the Response to Gamma Rays	119
3.4.3	Figures of Merit About the Response to Neutron Against the Response to Gamma Rays	120
3.5	Incorporation of Neutron Converters into Plastic Scintillator-Based Detectors	122
3.5.1	Homogeneous Incorporation	122
3.5.2	Heterogeneous Incorporation	125
3.6	Applications of Plastic Scintillators to the Detection of Slow Neutrons	130
3.6.1	Homeland Security	131
3.6.2	Neutron Flux Monitoring and Source Characterization	131
3.6.3	Reactor Antineutrino Experiments, Surveillance, and Monitoring	132
	References	133
4	Chemical Approach on Organometallic Loading in Plastic Scintillators and Its Applications	139
	Guillaume H. V. Bertrand and Matthieu Hamel	
4.1	Introduction/Context	139
4.1.1	Plastic Scintillation	139

4.1.2	Frame of This Chapter	140
4.1.3	Properties Optimization	140
4.1.4	Chemical Design and Material Science, What the Loading Implies	141
4.1.5	Organization of This Chapter: Application Driven	142
4.2	Scintillation Process Enhancement	143
4.2.1	Triplet Harvesting	143
4.2.2	Iridium Complexes	143
4.2.3	Europium Complexes	145
4.3	Photon Detection	146
4.3.1	Theory	146
4.3.2	X-ray Detection	147
4.3.3	Gamma Detection	149
4.4	Neutron Detection	156
4.4.1	Thermal Neutron	156
4.4.2	Lithium Loading	158
4.4.3	Boron Loading	159
4.4.4	Cadmium and Gadolinium Loading	160
4.5	Conclusion	162
4.6	Table by Elements	162
	References	164
5	Polysiloxane-Based Scintillators	169
	Sara Maria Carturan and Alberto Quaranta	
5.1	Foreword	169
5.1.1	Silicon-Based Polymer Properties: Chemistry	170
5.1.2	The Synthesis of Silicones	172
5.2	Optical Properties of Phenyl-Containing Polysiloxanes	174
5.3	Design of Polysiloxane-Based Scintillators	179
5.3.1	Energy Transfer in Organic Polymers	179
5.3.2	Polymeric Scintillators	181
5.3.3	Polysiloxane-Based Scintillators	183
5.4	Polysiloxane Scintillators for Neutron Detection	186
5.4.1	Neutron Detection in Organic Scintillators	186
5.4.2	B and Li Loaded Polysiloxanes for Detection of Thermal Neutrons	190
5.4.3	Design of Polysiloxane Scintillators for n/ γ Discrimination	194
5.5	Summary	196
	References	197
6	Composite Scintillators	201
	Masanori Koshimizu	
6.1	Introduction to Organic–Inorganic Composites	201
6.1.1	Overview on Fabrication Methods of Nanocomposites	203

6.1.2	Optical Properties Related to the Nanocomposite Structure	206
6.2	Plastic Scintillators Incorporating Non-emitting Inorganic Nanoparticles	208
6.2.1	Sol–gel-Derived Organic–Inorganic Composite Scintillators	209
6.2.2	Nanocomposite Scintillators Fabricated via Two-Step Synthesis	211
6.3	Nanocomposite Scintillators Comprising Luminescent Nanoparticles	214
6.3.1	Nanocomposite Scintillators Comprising Inorganic Phosphor Nanoparticles	214
6.3.2	Nanocomposite Scintillators Comprising Semiconductor Nanocrystals	217
6.4	Summary and Future Prospects	219
	References	220
7	Molecular Design Considerations for Different Classes of Organic Scintillators	223
	Patrick L. Feng	
7.1	Design Considerations for Crystalline, Plastic, and Liquid Scintillators	224
7.1.1	Background on Scintillation Mechanisms	224
7.1.2	Process (1): Direct Excitation into π -Electronic States	225
7.1.3	Process (2): Overview of Direct Ionization and Recombination of π -states	230
7.1.4	Physical and Mechanical Properties of Different Classes of Organic Scintillators	237
7.2	Future Opportunities	239
	References	240
8	Organic Glass Scintillators	243
	Patrick L. Feng, Nicholas R. Myllesbeck, and Joseph S. Carlson	
8.1	Introduction to Organic Glass Scintillators	243
8.2	Glassy State of Matter	244
8.3	Differentiating Characteristics of Organic Molecular Glasses	246
8.4	Design Strategies for Stable Organic Molecular Glasses	246
8.4.1	Nonplanar Structures	248
8.4.2	Increasing Molecular Size	249
8.4.3	Multiple Conformations	250
8.4.4	Physical Mixtures	252
8.5	Fluorescent Molecular Glasses as Organic Glass Scintillators (OGSs)	253
8.6	Organic Glass Scintillators: Case Studies	255

8.7	Organic Glass Thermal and Mechanical Properties	262
8.7.1	Mechanical Strength: Intermolecular Interactions	264
8.7.2	Mechanical Strength: Organic Glass/Polymer Blending	264
8.8	Properties of OGS/Polymer Blends	266
8.8.1	<i>Effect of Small-Molecule Additives on T_g</i>	266
8.8.2	Scintillation Properties of OGS/Polymer Blends	267
8.9	Organic Glass Scintillator Fabrication Methods	269
8.10	Long-Term Stability and Environmental Aging of Organic Glass Scintillators	270
8.10.1	Surface Versus Bulk Diffusion	270
8.10.2	Accelerated Aging of Organic Glasses and Mitigation Methods	271
8.11	Compatibility of OGS with Multi-functional Additives	275
8.11.1	Boron-Loaded OGS for Fast Neutron/Gamma PSD and Thermal Neutron Capture	275
8.11.2	Metal-Loaded OGS for Fast Neutron/Gamma PSD and Gamma-Ray Spectroscopy	277
8.12	Summary and Future Outlook	277
	References	278

Part II Applications

9	Optical Improvements of Plastic Scintillators by Nanophotonics	287
	Bo Liu and Xiaoping Ouyang	
9.1	Introduction	287
9.2	Enhancement of Light Extraction Efficiency of Plastic Scintillators by Photonic Crystals	289
9.2.1	Introduction of Photonic Crystals	289
9.2.2	Enhancement Mechanism of Light Extraction Efficiency by Photonic Crystals	290
9.2.3	Control of Directional Emission by Photonic Crystals	294
9.2.4	Consideration for the Structural Design of Photonic Crystals	296
9.3	Control of Directional Emission of Plastic Scintillators by Plasmonic Lattice Resonances	297
9.4	Patterning Techniques for Plastic Scintillators	298
9.4.1	Self-assembly Lithography	299
9.4.2	Nanoimprint Lithography (NIL)	301
9.4.3	X-Ray Interference Lithography (XIL)	301
9.5	Improved Scintillation Performance of Detectors by Photonic Crystals	302
9.6	Summary and Remark	304

References 305

10 Analog and Digital Signal Processing for Nuclear Instrumentation 309

Mathieu Thevenin and Yoann Moline

10.1 Introduction 309

10.2 The Light to Electric Signal Conversion 311

 10.2.1 Design of PMTs 311

 10.2.2 Solid-State Semiconductor Photodetectors 331

10.3 The Signal Acquisition Frontend 344

 10.3.1 Charge to Voltage Conversion 344

 10.3.2 Gain and Pulse Shaping Stage 346

 10.3.3 Voltage Limiters 347

 10.3.4 Impedance Matching and Other Effects 348

10.4 The Digitization Stage 349

 10.4.1 Signal Digitization Basics 349

 10.4.2 Digitizer Architectures 354

10.5 Signal Processing and Feature Extraction 356

 10.5.1 Low-Level Digital Stream Processing 357

 10.5.2 Digital Pulse Processing 365

10.6 Data and Information Processing 368

 10.6.1 Count Rate Analysis 369

 10.6.2 Discrimination of the Nature of the Interactions 369

 10.6.3 Spectral Unmixing and Radionuclide Identification 372

10.7 Conclusion 373

References 374

11 Radioactive Noble Gas Detection and Measurement with Plastic Scintillators 385

Krasimir Mitev and Philippe Cassette

11.1 Radioactive Noble Gas Isotopes 385

 11.1.1 Kr-85 388

 11.1.2 Xe-131m 389

 11.1.3 Xe-133 390

 11.1.4 Xe-133m 391

 11.1.5 Xe-135 392

 11.1.6 Ar-37 392

 11.1.7 Rn-222 and Progenies 393

 11.1.8 Rn-220 and Progenies 395

11.2 Application of Plastic Scintillators to the Detection of Noble Gas 396

 11.2.1 Xenon Detection Systems for the CTBT Network 396

 11.2.2 Kr-85 Monitors Using Plastic Scintillators 400

 11.2.3 Radon and Thoron Detection and Measurement with Plastic Scintillators 402

11.3 RNG-Related Properties of Plastic Scintillators 403

11.3.1	Noble Gas Absorption in Plastic Materials	403
11.3.2	<i>Application of Pulse Shape Discrimination to ^{222}Rn Measurements</i>	410
11.3.3	<i>Description of the Alpha-Particle Peak Shapes in ^{222}Rn Measurements with Plastic Scintillators</i>	414
11.4	Concluding Remarks	419
	References	420
12	Recent Advances and Clinical Applications of Plastic Scintillators in the Field of Radiation Therapy	425
	Sam Beddar, Irwin Tendler, François Therriault-Proulx, Louis Archambault, and Luc Beaulieu	
12.1	Introduction	426
12.2	Basic Dosimetry Properties of Plastic Scintillators	427
12.2.1	Basic Properties of Scintillators Used in Radiation Therapy	427
12.2.2	Relating Scintillation Signal to Absorbed Dose	428
12.2.3	Other Properties	429
12.3	Signal Processing in Plastic Scintillators Dosimeters	430
12.3.1	Cherenkov and Other Sources of Stem Effects	430
12.3.2	Multi-point Radiation Therapy Plastic Scintillator Dosimeters	432
12.4	Clinical Applications and Special Procedures	433
12.4.1	Photon and Electron External Beam Radiation Therapy	433
12.4.2	Proton External Beam Radiation Therapy	440
12.4.3	Brachytherapy	449
12.5	Conclusion	454
	References	454
13	Plastic Scintillators in Environmental Analysis	461
	Alex Tarancón, Héctor Bagán, and José Francisco García	
13.1	Environmental Analysis: Requirements and Characteristics. Contribution of Plastic Scintillators	462
13.2	Preparation of Plastic Scintillators for Environmental α/β Emitting Radionuclide Analysis	467
13.2.1	Fibers and Sheets	468
13.2.2	Plastic Scintillation Particles	468
13.2.3	Stability of Plastic Scintillation Microspheres	474
13.2.4	Plastic Scintillation Resins	476
13.3	Direct Surface and Soil Analysis	477
13.4	Sensors and Online Analysis of Alpha and Beta Emitting Radionuclides	481
13.4.1	Sensors Based on Plastic Scintillation Microspheres (PSm)	482
13.4.2	Sensors Based on Plastic Scintillation Resins	485

- 13.4.3 Sensors Based on Plastic Scintillation Fibers (PSF) 487
- 13.4.4 Sensors Based on Plastic Scintillation Sheets 488
- 13.5 Plastic Scintillators for Beta Emitting Radionuclide
Analysis 489
- 13.6 Plastic Scintillators for Alpha Emitting Radionuclide
Analysis 498
- 13.7 Conclusion 503
- References 504
- 14 Use of Scintillators to Study the Earth from Ground
to the Radiation Belts 509**
Philippe Laurent, Sebastien Celestin, Vincent Maget,
Pablo Caron, and François Trompier
- 14.1 Survey of Earth Natural Radioactivity and Potential
Nuclear Accidents, Citizen Science 509
 - 14.1.1 Source of Terrestrial Natural Radioactivity 509
 - 14.1.2 Sources of Radioactive Pollution
in the Environment 512
 - 14.1.3 Nuclear Activities Survey and Alert in Case
of Accident: Description of Official and NGO
Networks 514
 - 14.1.4 Measuring and Mapping the Radioactivity
in the Environment: The Citizen Science Approach 519
- 14.2 High-Energy Processes in the Atmosphere, Terrestrial
Gamma-Ray Flashes (TGFs), and Gamma-Ray Glows 521
 - 14.2.1 Gamma Ray Emissions During Thunderstorms 521
 - 14.2.2 Gamma-Ray Glows 522
 - 14.2.3 Terrestrial Gamma-Ray Flashes (TGFs) 523
 - 14.2.4 TARANIS: A Satellite to Study the Effects
of Thunderstorms and Lightning 524
 - 14.2.5 Measure of Gamma Rays and Particles in the Earth
Radiation Belts 526
 - 14.2.6 Measure of Gamma Rays/Particles
from the Radiation Belts 529
 - 14.2.7 IGOSAT: A Student Nanosat Project to Measure
Radiation Belts Gamma Ray/Particles 531
- 14.3 Conclusion 533
- References 534
- 15 Plastic Scintillator Detectors for Particle Physics 541**
Priscilla Brooks Cushman and David-Michael Poehlmann
- 15.1 Introduction 541
- 15.2 Principles of Calorimetry in High-Energy Physics 542
 - 15.2.1 Electromagnetic and Hadronic Showers 542
 - 15.2.2 Historical Review of Plastic Scintillating
Calorimetry 544

15.3	Modern Plastic Scintillator Calorimeter Design	559
15.3.1	Experiments at the Large Hadron Collider	559
15.3.2	High-Granularity Calorimeters for Particle Flow Strategies	564
15.3.3	Dual-Readout Solutions	570
15.4	Neutrino Experiments	571
15.4.1	Optimizing Calorimeters for Neutrino Physics	571
15.4.2	Tracking Calorimeters for Neutrinos	572
15.5	Large Plastic Scintillator Detectors in the Future	579
15.5.1	Neutron Sensitivity	579
15.6	Conclusion	583
	References	584
	Appendix: Molecules Cited in the Book	589
	Index	631

Contributors

Louis Archambault Département de physique, de génie physique et d'optique, et Centre de recherche sur le cancer, Université Laval, Québec, QC, Canada;
Service de physique médicale et radioprotection, Service de radio-oncologie et Axe oncologie du CRCHU de Québec, CHU de Québec - Université Laval, Québec, QC, Canada

Héctor Bagán Department of Chemical Engineering and Analytical Chemistry, University of Barcelona, Barcelona, Spain

Luc Beaulieu Département de physique, de génie physique et d'optique, et Centre de recherche sur le cancer, Université Laval, Québec, QC, Canada;
Service de physique médicale et radioprotection, Service de radio-oncologie et Axe oncologie du CRCHU de Québec, CHU de Québec - Université Laval, Québec, QC, Canada

Sam Beddar Department of Radiation Physics, The University of Texas MD Anderson Cancer Center, Houston, TX, USA;
The Graduate School of Biomedical Sciences, The University of Texas MD Anderson Cancer Center, Houston, TX, USA

Guillaume H. V. Bertrand Université Paris-Saclay, CEA, List, Palaiseau, France

Joseph S. Carlson Sandia National Laboratories, Livermore, CA, USA

M. Leslie Carman Lawrence Livermore National Laboratory, Livermore, CA, USA

Pablo Caron DPHY - Department of Physics, Instrumentation Environment and Space, ONERA, Toulouse, France

Sara Maria Carturan Dipartimento di Fisica e Astronomia, Università di Padova, Padua, Italy;
INFN – Laboratori Nazionali di Legnaro, Legnaro, PD, Italy

Philippe Cassette Sofia University “St. Kliment Ohridski”, Faculty of Physics, Sofia, Bulgaria

Sebastien Celestin Laboratory of Physics and Chemistry of the Environment and Space (LPC2E), University of Orleans, CNRS, CNES, Orleans, France

Priscilla Brooks Cushman School of Physics and Astronomy, University of Minnesota, Minneapolis, MN, USA

Christophe Dujardin Université de Lyon, Université Claude Bernard Lyon 1, CNRS, Institut Lumière Matière UMR 5306, Villeurbanne, France

Jonathan Dumazert Université Paris-Saclay, CEA, List, Palaiseau, France

Patrick L. Feng Sandia National Laboratories, Livermore, CA, USA

Camille Frangville Université Paris-Saclay, CEA, List, Palaiseau, France

José Francisco García Department of Chemical Engineering and Analytical Chemistry, University of Barcelona, Barcelona, Spain

Andrew M. Glenn Lawrence Livermore National Laboratory, Livermore, CA, USA

Matthieu Hamel Université Paris-Saclay, CEA, List, Palaiseau, France

Masanori Koshimizu Department of Applied Chemistry, Tohoku University, Miyagi, Japan

Philippe Laurent CEA, IRFU, CEA Saclay, Paris, France;
Laboratoire Astroparticule et Cosmologie, Paris, France

Bo Liu School of Physics Science and Engineering, Tongji University, Shanghai, People's Republic of China

Andrew N. Mabe Lawrence Livermore National Laboratory, Livermore, CA, USA

Vincent Maget DPHY - Department of Physics, Instrumentation Environment and Space, ONERA, Toulouse, France

Krasimir Mitev Sofia University "St. Kliment Ohridski", Faculty of Physics, Sofia, Bulgaria

Yoann Moline Université Paris-Saclay, CEA, List, Palaiseau, France

Nicholas R. Myltenbeck Sandia National Laboratories, Livermore, CA, USA

Xiaoping Ouyang Northwest Institute of Nuclear Technology, Xi'an, People's Republic of China

David-Michael Poehlmann Department of Physics and Astronomy, University of California, Davis, CA, USA

Alberto Quaranta Department of Industrial Engineering, University of Trento, Trento, Italy

Alex Tarancón Department of Chemical Engineering and Analytical Chemistry,
University of Barcelona, Barcelona, Spain;
Serra Hunter Fellow, Barcelona, Spain

Irwin Tendler Department of Radiation Physics, The University of Texas MD
Anderson Cancer Center, Houston, TX, USA

François Therriault-Proulx Co-Founder, CEO At Medscint Inc., Quebec City,
Canada

Mathieu Thevenin Université Paris-Saclay CEA SPEC CEA-CNRS UMR 3680,
Gif-sur-Yvette, France

François Trompier IRSN, Fontenay-aux-Roses, France

Natalia P. Zaitseva Lawrence Livermore National Laboratory, Livermore, CA,
USA

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.

C. Dujardin (✉)

Institut Lumière Matière, Université de Lyon, Université Claude Bernard Lyon 1, CNRS, 69622 Villeurbanne, France

e-mail: christophe.dujardin@univ-lyon1.fr

M. Hamel

Laboratoire Capteurs Et Architectures Électroniques, Université Paris-Saclay, CEA, 91120 Palaiseau, List, France

e-mail: matthieu.hamel@cea.fr

© Springer Nature Switzerland AG 2021

M. Hamel (ed.), *Plastic Scintillators*, Topics in Applied Physics 140,
https://doi.org/10.1007/978-3-030-73488-6_1

1.1 History of Scintillators

The first scintillator ever described was $\text{BaPt}(\text{CN})_4$ 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 Pestel [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 acetate 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,

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

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 $200 \times 50 \times 10$ 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-4xx	n.d.
Detec-Europe	France	n.d	Amcrys reseller
Eljen Technology	USA	EJ-2xx	21
Epic-Crystal	China	n.d	1
Institute for High-Energy Physics	Russia	SC-2xx (bulk) SC-3xx (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-4xx	21
Shandong Haiqiang Environmental Protection Technology Co., Ltd	China	n.d	n.d.
Win-Trust	China	SPxxx	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