# Inradoptics

## Scintinel<sup>™</sup> Stilbene for Fast Neutron Detection

Crystalline stilbene is an organic scintillator used for radiation detection and is well-suited for fast neutron identification in environments with high gamma-ray flux. A fast neutron is one with kinetic energy above approximately 1 MeV. Fast neutron counting, spectroscopy and imaging have applications in research, defense, nuclear nonproliferation and homeland security. Stilbene has a high sensitivity to fast neutrons, nanosecond timing and permits excellent discrimination between neutron and gamma radiation. The 380 nm scintillation emission is easily detected by photomultiplier tubes or silicon photomultipliers and self-absorption is low. It is solid, not flammable and non-hazardous. Stilbene can be cut and polished and is not hygroscopic.

New methods for stilbene production<sup>i</sup> have facilitated the commercial availability of high-quality material. Fabrication methods developed at Inrad Optics yield stilbene crystals in shapes and sizes ranging from 5 inch diameter cylinders to 3 mm cubes and at levels of packaging and integration to suit customers from university researchers to OEMs.

#### FAST NEUTRON DETECTION ADVANTAGES

The detection of neutrons is a strong indicator of a fission source due to the absence of background sources of fast neutrons, whereas there are many benign sources of gamma radiation. Additionally, detection of radiation from shielded nuclear material is best conducted using neutrons since gamma rays are absorbed by materials which are transparent to neutrons.

The majority of neutron detectors, including He-3, are sensitive only to thermal neutrons, and use moderators to reduce the speed of fast neutrons prior to detection. By directly detecting fast neutrons, such as with stilbene, the direction, timing and energy of the source is retained. Directional history can permit imaging with neutrons and identify the location of the neutron source. Neutron energy and timing can facilitate determination of the type of source and mass.

Crystalline stilbene is recognized as the preferred material for detection of fast neutrons in environments where there is a gamma-ray background, due to stilbene's superior ability to allow neutron-gamma discrimination.

#### **STILBENE PROPERTIES**

Melting Point	124 °C
Density	1.15 g/cm³
Refractive Indices (at 589 nm)	1.703, 1.724, 1.844
Maximum Emission Wavelength <sup>ii</sup>	380 nm
Decay Time (Prompt)"	4.3 ns
Rise Time <sup>iii</sup>	0.1 ns

#### **NEUTRON DETECTION PERFORMANCE**

The scintillation light pulse emitted from stilbene consists of both a prompt and a delayed fluorescence. The fraction of light resulting from the slow component often depends on the type of particle interacting with the crystal. Pulse shape discrimination (PSD) methods exploit this effect to separate events arising from neutrons and gamma rays. The pulse shape discrimination pattern of Inrad Optics stilbene shows exceptional neutron-gamma separation, with a figure of merit of 4.7 measured near 500 keVee. The figure of merit is the separation between gamma ray and neutron peaks divided by the sum of the full-widths at half maximum of the relevant peaks. The Compton edge of <sup>137</sup>Cs is used to calibrate the energy scale, which is presented in units of keVee (keV electron equivalent) to account for the particle-dependent variation in light output per keV deposited in the scintillator. Details regarding experimental methods for pulse shape discrimination with stilbene are provided by Zaitseva.<sup>ii</sup> The figure of merit is strongly dependent on the energy range and the crystal size.



Pulse shape discrimination data from Inrad Optics stilbene (<sup>252</sup>Cf source). Data courtesy of N. Zaitseva, LLNL.

Organic scintillators such as stilbene have a fast time response. The prompt decay time for stilbene is 4.3 ns.<sup>ii</sup>

Separation of neutron and gamma signals using pulseshape-discrimination has been successful down to energies of 22 keVee (0.5 MeV neutron energy). Gamma misclassification rates of 10<sup>-6</sup> have been reported.<sup>ix</sup>

The intrinsic neutron efficiency of 2 inch Inrad Optics stilbene is 15.1% for  $^{252}Cf$  increasing to 38.6% for 4 inch stilbene. $^{i \varkappa}$ 

The resolution of stilbene was found to be 8.5% at 470 keVee.  $^{\mbox{\tiny iv}}$ 

A method for determining the light yield of stilbene is given by Galunov.<sup>v</sup> Temperature dependence of the light yield of crystalline stilbene has been reported by Baker.<sup>vi</sup>

Position resolution of 4.9 mm was achieved using 50 mmlong stilbene bars connected to silicon photomultipliers on both ends.<sup>vii</sup>

### COMPARISON TO LIQUID AND PLASTIC ORGANIC SCINTILLATORS

Crystalline stilbene is an alternative to liquid organic scintillators and plastic scintillators, and is appealing as a replacement for liquid organic scintillators both due to its physical properties (solid, not flammable) and response to neutrons. Recent publications comparing performance of Inrad Optics stilbene to liquid and plastic organic scintillators consistently report superior pulse shape discrimination figure of merit for stilbene.<sup>viii, ix</sup> Relative to EJ-309 liquid scintillation detectors, stilbene has a factor-of-5 better gamma misclassification rate and 10% greater intrinsic neutron efficiency.<sup>x</sup> Additionally, stilbene has superior PSD and energy resolution to EJ-309, with an approximately 17% improvement in neutron detection efficiency.<sup>xi</sup>

#### **APPLICATIONS**

Stilbene has applications in homeland security, nuclear nonproliferation and physics research. Inrad Optics supplies stilbene for customers in academia, industry and at national laboratories.

Stilbene can be used in fixed or handheld fast-neutron counting applications.<sup>x</sup> Additionally, the anisotropic response of stilbene may allow the direction of a source to be located.<sup>xii</sup>

Stilbene has been applied to the characterization and mass determination of fissile materials such as uranium oxide and plutonium.<sup>xiii</sup> This has applications in material accounting for nuclear safeguards.<sup>xiv</sup>

Stilbene crystals have been fabricated into pixels for neutron and gamma-ray imaging.<sup>xv, xvi, xvii</sup> Inrad Optics offers both bare pixels and custom stilbene arrays.

Stilbene has shown promise for use in applications which employ active interrogation to detect fissile material, such as vehicle and cargo scanning and assay of uranium in nuclear fuel assemblies.<sup>xviii</sup>

Stilbene has applications in neutron spectroscopy for materials identification and physics research.<sup>xix</sup>

The use of stilbene in radioxenon detection for nucleartest-ban-treaty monitoring has shown promise due to substantially lower memory effect relative to the conventional material.<sup>xx</sup>

#### **CRYSTAL STRUCTURE AND ORIENTATION**

Trans-stilbene crystallizes in the monoclinic system, with a structure first described by Robertson and Woodward.<sup>xxi</sup> We use the space group and lattice constants from the Bernstein refinement<sup>xxii</sup> as they are consistent with earlier reports of anisotropic behavior in organic scintillators: space group P2,/a, a = 12.382 Å, b = 5.720 Å, c = 15.936 Å and  $\beta = 114.15^{\circ}$ .

Solution-grown stilbene boules exhibit two sets of broad natural faces which have been identified as  $(\overline{2}03)$ and (001) by x-ray diffraction; these planes can be quickly differentiated visually due to the much larger birefringence through  $(\overline{2}03)$  relative to (001). (010) is orthogonal to the broad natural faces. When stilbene is fabricated with square cross-sections, faces are cut parallel to (010) and (001) (Figure 1). We denote the third set of cube faces as a\* and the direction normal to (001)is identified as c'. The (001) is the strongest cleavage plane in stilbene and the c' axis corresponds to the largest thermal expansion axis.<sup>xxiii</sup>

Stilbene is optically biaxial, with three indices of refraction and two optic axes which together define an optic plane; cube faces can be quickly verified by examination between crossed polarizers. Extinctions for (010) faces occur at a small angle from the cube edges, since the optic plane is parallel to *b* and rotated away from (001) by approximately 5°. Looking down *b* reveals strong colors on a\* faces. The final set of faces (001) yield extinctions symmetric with the cube edges.

The standard orientation for Inrad Optics cylindrical stilbene is with the (010) plane parallel to the end face coupled to the photodetector (or stated differently, the b-axis is normal to the end face). Crystallographic orientation may be marked on stilbene products: a black dot indicates a (010) face and a red dot indicates a (001) face.

#### ANISOTROPY

The scintillation behavior of stilbene exhibits anisotropy: there is a dependence on the direction of proton and carbon recoils with respect to the crystallographic axes. Early descriptions of the anisotropy were provided by Heckmann,<sup>xxiv</sup> followed later by Birks<sup>xxv</sup> and Brooks.<sup>xxvi</sup> This behavior has been exploited in the search for dark matter.<sup>xxvii</sup> In studies of stilbene anisotropy it has been found that the scintillation efficiency and light yield of stilbene varies as a function of the angle around the *c*' axis.



A detailed investigation of the anisotropic response of stilbene and other organic scintillators to 14 MeV neutrons has recently been conducted by Shuster.<sup>xxviii</sup> Weldon has presented a detailed study of the scintillation response of stilbene crystals for protons recoiling along *a*, *b* and *c'* axes and has notably shown that the direction of maximum scintillation response for proton recoils is along the a-axis, in disagreement with previous literature.<sup>xii</sup>

#### FABRICATION, PACKAGING AND INTEGRATION

Inrad Optics manufactures stilbene crystals in standard and custom dimensions. Sizes from 3 mm to 150 mm are available. Common items are right cylinders and cubes or bars. Circular cross-sections are appropriate for use with photomultiplier tubes and square cross-sections are used with silicon photomultipliers.

	MINIMUM DIMENSION	MAXIMUM DIMENSION
Cube or bar	3 mm x 3 mm	150 mm
Cylinder	10 mm diameter	5 x 3"; 4 x 4"
Plates, disks	1 mm thick (freestanding);	
	40 microns thick (on backing)	

We have also manufactured tapered cones, hollow cylinders and multi-part assemblies of bonded stilbene crystals. Compared with conical stilbene, a tapered cone geometry has 22.6% higher figure of merit in the 25-100 keVee region and 10.1% improvement in energy resolution at 478 nm.<sup>xxix</sup>

Stilbene is not hygroscopic; however, exposed surfaces can degrade over time. Inrad Optics offers stilbene bonded to a thin protective window to preserve the polished surface and facilitate mounting to a photodetector without damage. Alternatively, the stilbene crystal can be mounted in a protective aluminum housing to reduce the risk of thermal or mechanical shock to the crystal. Stilbene is also offered bare or wrapped in white PTFE tape or in a custom-designed array.

When choosing the length of the crystal, keep in mind that although the neutron detection efficiency increases with scintillator length,<sup>iii</sup> the pulse shape discrimination figure of merit decreases with increasing length, due to self-absorption in the crystal. Lengths below 4 inches are generally recommended.



Bare stilbene cylinders and cube; cylinders with protective window and PTFE wrapping; protective housing; stilbene array.

#### **CHOICE OF PHOTODETECTOR**

Stilbene scintillates with a peak wavelength near 380 nm which red-shifts with increasing crystal size due to self-absorption.<sup>ii</sup> The scintillation light from stilbene is commonly detected with a photomultiplier tube (PMT) and more recently, with silicon photomultipliers (SiPMs). PMTs with fast timing and peak sensitivity near 400 nm have been used successfully with stilbene; many are identified in the references.

Silicon photomultipliers offer the benefit of lower cost, lower power requirements and compact size with comparable performance for detection of scintillation light from stilbene.<sup>xxx, xxxii</sup>, xxxii In a recent comparison of organic scintillators and silicon photomultipliers, the best time resolution was obtained using Inrad Optics stilbene coupled to a SensL MicroFJ-SMA-60035 SiPM.<sup>xxxiii</sup> When stilbene is provided in a bare state, silicone greases such as BC-630 and EJ-550 may be used for optical coupling to the photodetector. These materials may be left in contact with stilbene for extended periods.

Inrad Optics Scintinel<sup>™</sup> stilbene product line includes assemblies with integrated photomultiplier tubes.

#### **SAFETY AND HANDLING**

Stilbene can be handled in a similar manner as other crystalline materials. Avoid thermal shock and temperature gradients within the crystal. Consult the Inrad Optics Stilbene Handling Instructions for more detailed information.

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