Radiation Detection and Measurement

Lecture 17

Chapter 7: Scintillation Detection Principles

Inorganic scintillators

Scintillation mechanism in inorganic crystals with activators

- Determined by the lattice structure of available energy states
- Valence band: electrons bound to lattice sites
- Conduction band: electrons with sufficient energy to migrate through the material
- Forbidden band: the band between the valence and conduction band where in a pure crystal no electrons can reside, the band gap region

Inorganic scintillators

- Activator energy levels: energy states produced by the introduction of impurities into the lattice. The energy levels are in the forbidden band and allow de excitation from the conduction band to the valence band through scintillation. The levels are also called luminescence centers or recombination centers. Fig 8.6 shows the relationship between these levels.

Inorganic scintillators

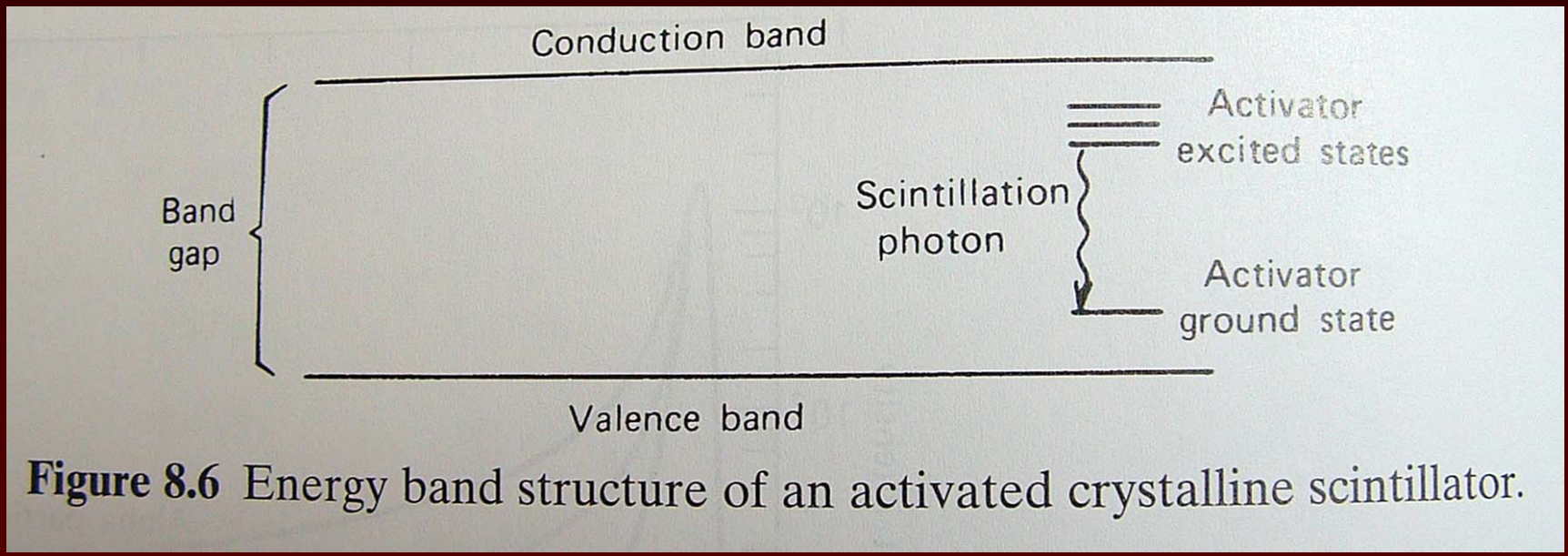

Figure 8.6 Energy band structure of an activated crystalline scintillator.

Inorganic scintillators

The process of scintillation:

1. traveling charged particle forms a large number of electron –hole pairs by exciting electrons from the valence to the conduction band
2. the positive hole then quickly travels to an activator site where it ionizes the site

Inorganic scintillators

3. the electron in the conduction band travels until it is captured by an ionized activator site
4. The neutralized activator site may be in an excited state, upon de excitation produces a photon in the visible range.
5. Half lives of these excited states are ~ 50-500 ns, which is the characteristic time of scintillation light.

Inorganic scintillators

Competing Processes:

- An electron may be captured by the activator site at a level with a forbidden transition to the ground state. At this point more energy must be absorbed to raise to an energy level with a possible de excitation pathway
- the energy absorbed is usually thermal in nature and the time scale corresponds to a phosphorescence event and contributes to background light or “after glow”

Inorganic scintillators

- Quenching may also compete where an electron is captured at an activator site and undergoes a radiation less transition from excited states to the ground state via electron capture.

Inorganic scintillators

Example of an efficiency calculation for NaI:

- On average it takes ~ 3X the band gap energy to create electron hole pair, for NaI this is ~ 20 eV, for 1MeV absorption, 5 X 10^4 electron hole pairs are formed.
- With an absolute scintillation efficiency of 12%, 1 MeV would yield about 1.2 x 10^5 total light energy, with average photon energy of 4 eV, there is the production of ~ 4x10^4 photons. So there is ~ 1 photon for every electron-hole pair formed.

Inorganic scintillators

- In general one should choose a scintillator material that produces light in the most sensitive energy of the detector device (Fig 8.7 shows several common scintillation materials as well as the response curve for two common photocathode).

Inorganic scintillators

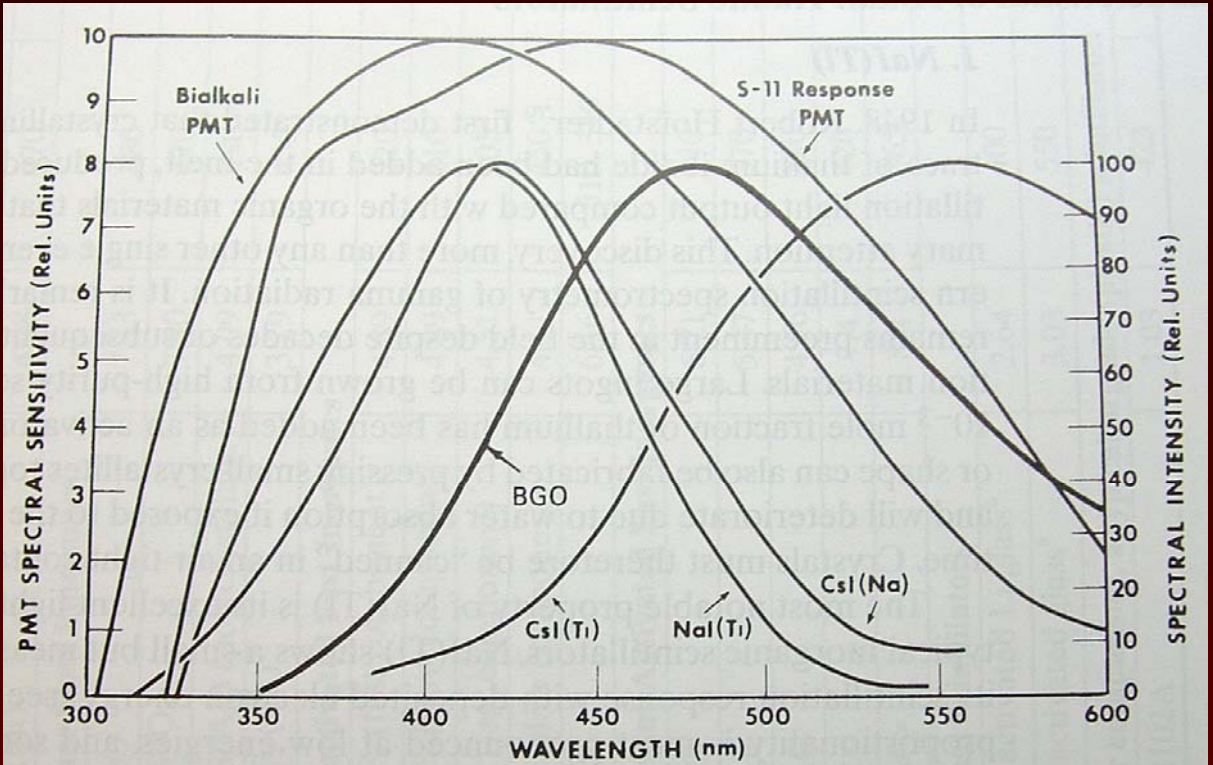

Figure 8.7 The emission spectra of several common inorganic scintillators. Also shown are the response curves for two widely used photocathodes. (Primarily from *Scintillation Phosphor Catalog*, The Harshaw Chemical Company. The emission spectrum for BGO is from Ref. 72.)

Inorganic scintillators

- Inorganic scintillator tend to be more linear than their organic counterpart (although non linearities are present from quenching)
- As in organics, heavy charged particles produce less light per unit energy, providing alpha – to –beta ratios of 0.66 – 0.67 for NaI (Tl) and CsI(Tl) or 0.2 for BGO and GSO.

Characteristics of alkali halide scintillators

NaI(Tl):

- Hygroscopic with excellent light yield
- A small but measurable non –linearity od scintillation response with deposited electron energy (shown in Fig 8.8, linearity would be a line (horizontal)).
- Dominant decay time of a scintillation pulse is 240 ms.
- Phosphorescence with a characteristic decay time of 0.15 s

Characteristics of alkali halide scintillators

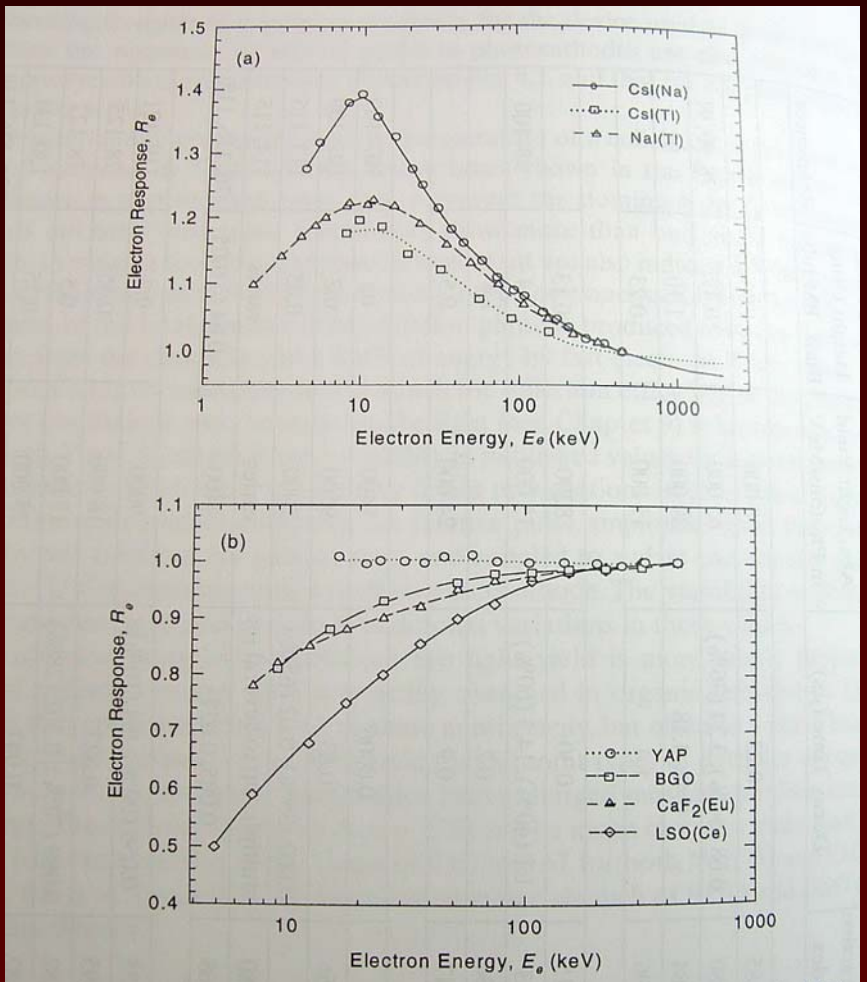

Figure 8.8 The relative scintillation response per unit energy deposited for fast electrons plotted as a function of energy for the scintillation materials shown. The curves are normalized to unity at 445 keV. Perfectly linear response would correspond to a horizontal line on this plot. (From Mengesha et al.[80])

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Characteristics of alkali halide scintillators

- At high counting rates , the phosphorescence will build up over multiple pulses (through overlap)
- As with other scintillators the light output drops off with increasing temperature (Fig 8.9)
- Scintillation decay time is also a function of temperature (Fig 8.10) where the response time increases with temperature.

Characteristics of alkali halide scintillators

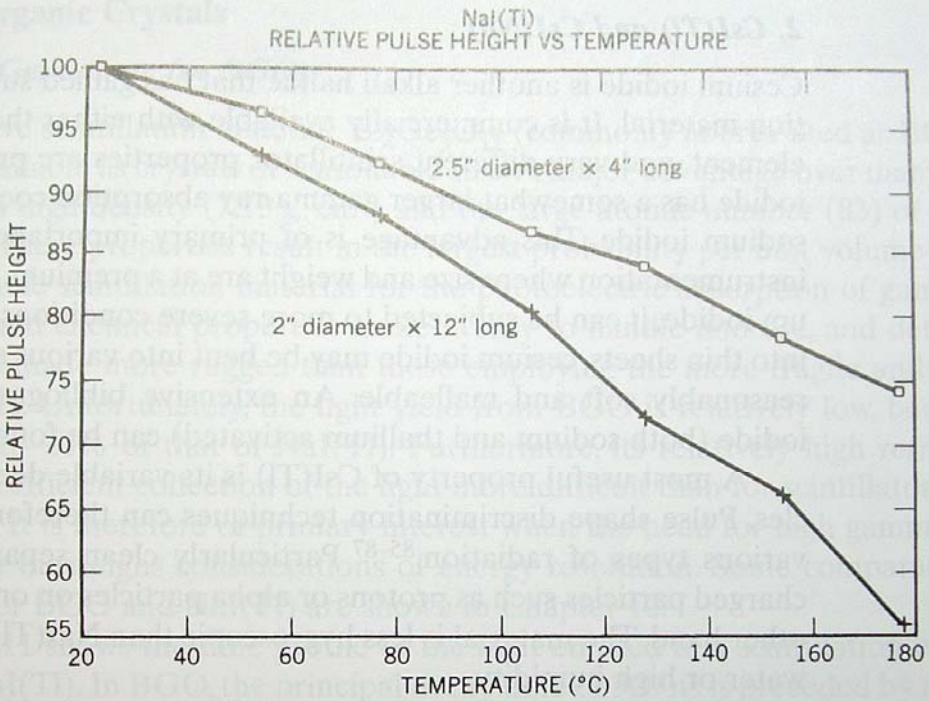

Figure 8.9 The temperature dependence of the light yield measured from two NaI(Tl) crystals. The measurements were made using an oven equipped with a light pipe, and the temperature of the photomultiplier tube was held constant. The difference in behavior between the two crystals is probably due to changes in surface reflectivity. (Data courtesy R. Dayton, Bicron Corporation, Newbury, Ohio.)

Characteristics of alkali halide scintillators

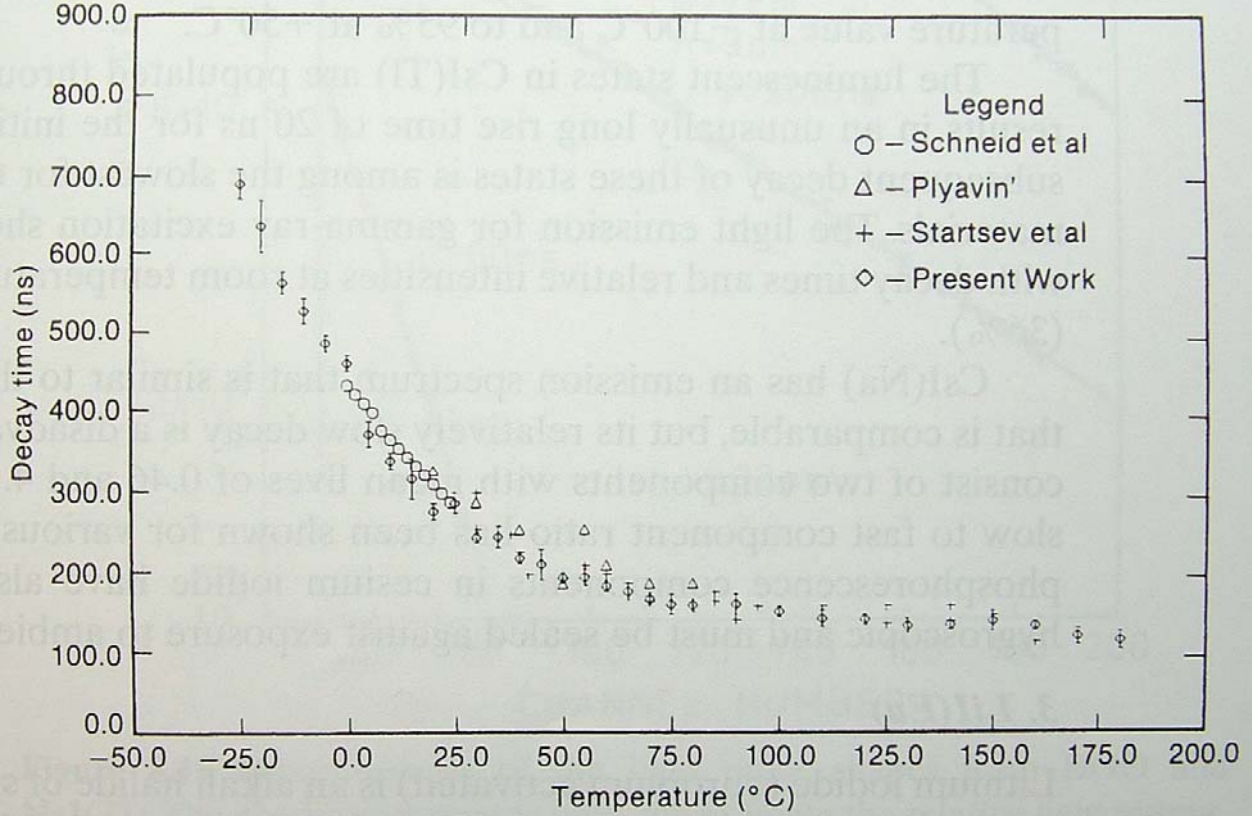

Figure 8.10 Temperature dependence of the scintillation decay time in NaI(Tl). (From Schweitzer and Ziehl.[83])

Characteristics of alkali halide scintillators

CsI(Tl):

- When cut into thin sheets, it may be bent into various shapes without fracture, so fairly malleable and reasonably soft
- Has various decay times for differing excitation particles, and can use pulse shape discrimination
- It is also hygroscopic

Characteristics of alkali halide scintillators

- Can be grown in a columnar microstructure where individual columns behave as optically isolated scintillators (diameters ~ 5 μm)
- Have a broad emission spectrum at a much longer wavelength than that of NaI(Tl)
- Has a relatively long rise time (20 ns) and slow decay time with two primary compounds (0.64 μs – 64% and 3.34 μs 36%)

Characteristics of alkali halide scintillators

CsI(Na):

- Emission spectrum is similar to NaI(Tl)
- Has a slow decay (with two components 0.46 and 4.18 μs)
- Hygroscopic

Characteristics of alkali halide scintillators

LiI(Eu)

- Used in neutron detection (low energy neutrons) through $^{6}Li(n,\alpha)$ reaction
- So formed using Li enriched with ^{6}Li

Other slow (>200ns) inorganic crystals

BGO: ($Bi_4Ge_3O_{12}$)

- High density (7.13 g/cc), high atomic number (83) and can produce crystals of reasonable size
- This results in the largest probability per unit volume of interaction for any commonly available scintillation material for photoelectric absorption of γ-rays

Other slow (>200ns) inorganic crystals

- Low light yield (10-20% of NaI(Tl)) (compared in Fig 8.11)
- Has a relatively high index of refraction (2.15) making light collection difficult
- Most of the long decay components do not lead to after glow
- Luminescence is associated with the Bi+3 ion (so no activator)
- Again light output decreases with increasing temperature (Fig 8.12)

Other slow (>200ns) inorganic crystals

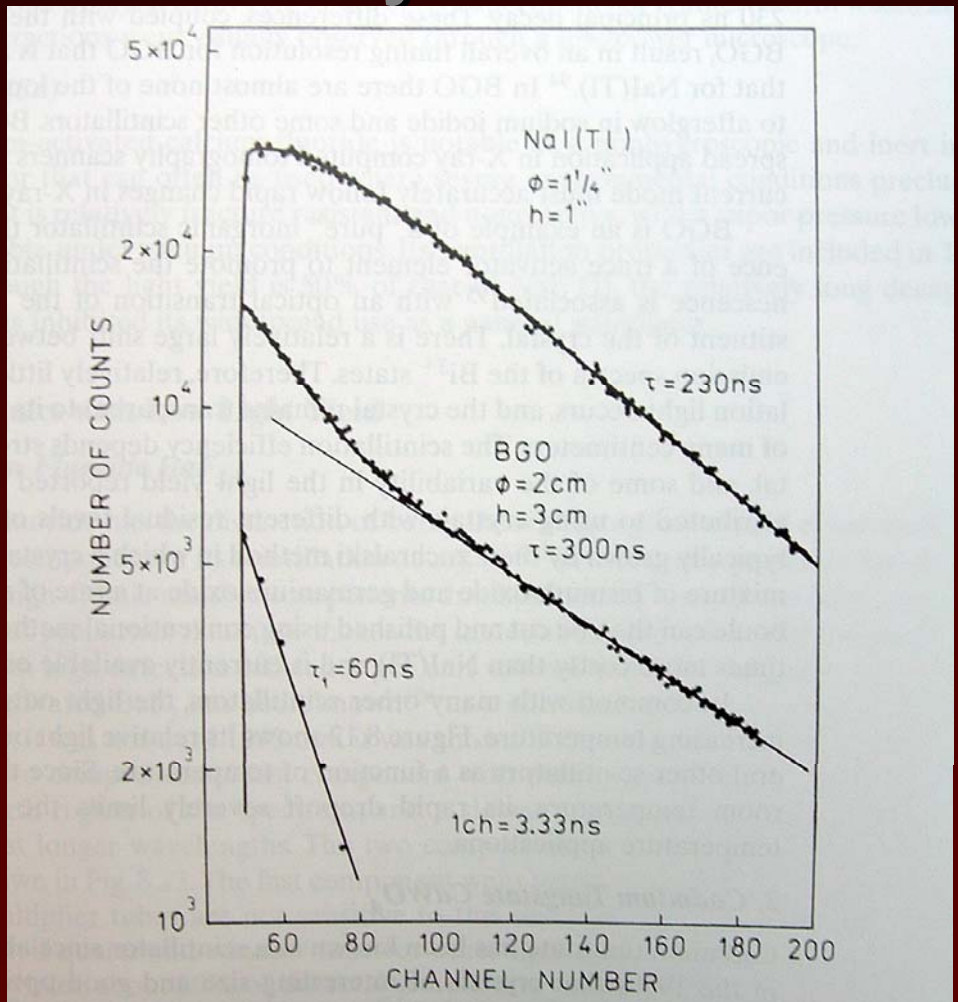

Figure 8.11 Measurements of the light pulse shapes from BGO and NaI(Tl). The abscissa represents time, the ordinate the relative light output. The BGO yield is represented as the sum of separate decay components with 60 and 300 ns decay times. (From Moszynski et al.[94])

Other slow (>200ns) inorganic crystals

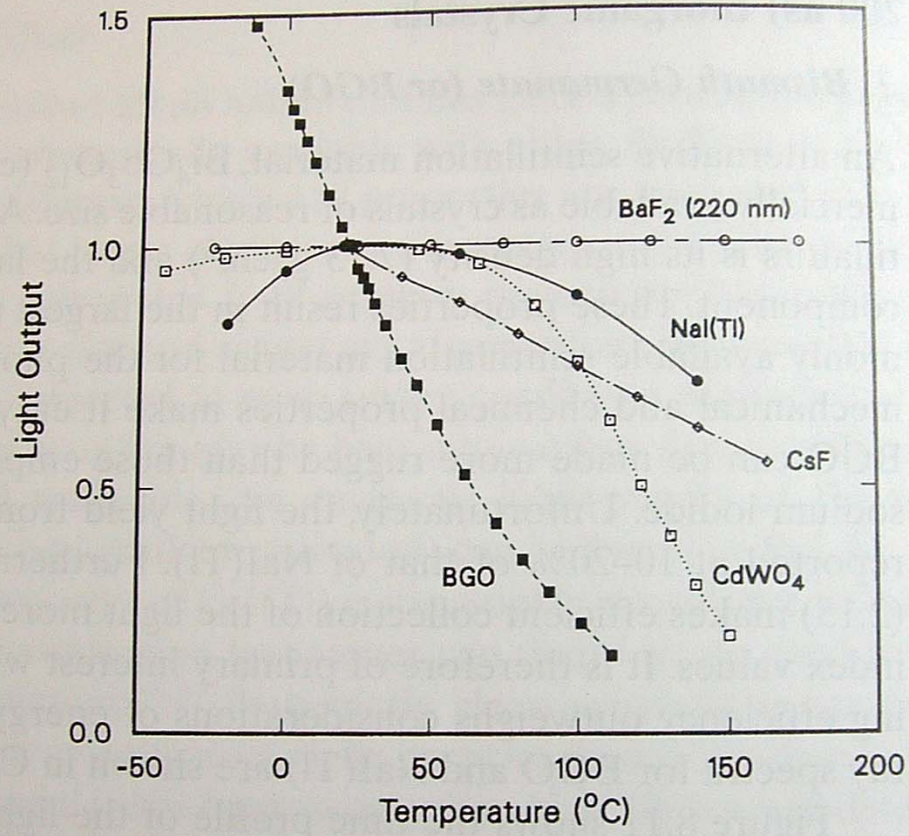

Figure 8.12 The dependence of the light output of some common scintillators as a function of temperature. Only the fast (220 nm) component from BaF_2 is included (From Melcher.[97])

Other slow (>200ns) inorganic crystals

- $CdWO_4$: (cadmium Tungstenate)
- Light yield is ~ 40% of NaI(Tl)
- has a high density and atomic number(effective)
- has a long decay time (1.1 μs 40%; 14.5 μs 60%)
- high index of refraction (2.3)
- free of long lived phosphorescence
- used in application where decay time is not an issue (CT,CR)

Other slow (>200ns) inorganic crystals

ZnS(Ag)

- only available in polycrystalline powder, so limited use in thin screens for α detection

Other slow (>200ns) inorganic crystals

CaF_2(Eu):

- non hygroscopic and inert
- light yield ~ 50% of NaI(Tl)
- Long decay time (~900ns), limiting the usefulness of this material as a general scintillator.

Unactivated fast inorganics with low light yield

BaF_2:

- light yield ~ 20% of NaI(Tl)
- two components emission spectrum (0.6 ns and 630 ns at longer wavelengths)- Fig 8.13 shows spectra at differing temperatures

Unactivated fast inorganics with low light yield

Pure CsI:

- fast components with an effective decay time ~ 10 ns in UV
- slower components emit in visible spectrum (~ several μs) and may be related to impurities in the crystal
- light output ~ 5-8 % of NaI(Tl

Unactivated fast inorganics with low light yield

CeF_3 : (Cerium Flouride)

- light output ~ 5% of NaI(Tl)
- decay time of ~ 27 ns for visible spectrum; but ~ 5ns for UV

Cerium-activated “fast” inorganics

GSO: (Gd_2SiO_5) – Godolinium Silicate

- large atomic number of Gd is advantageous for γ-ray spectroscopy
- scintillation decay time depends on cerium doping level: ~ 56 ns at concentration of 0.5 mole%
- longer decay time component ~ 400 ns with 10% intensity

Cerium-activated “fast” inorganics

- has a long rise time of 10-20ns depending on cerium concentration caused by slow population processes for fluorescent states
- light yield ~ 20% of NaI(Tl)
- shows no appreciable radiation damage for γ-ray exposure up to 10^7 Gy

Cerium-activated “fast” inorganics

$YAlO_3$ (YAP): Yttrium Aluminum Perovskite

- light yield ~ 40-50% of NaI(Tl)
- principle decay time of 27 ns, with a slower component of decay time (10μs at 10% intensity)
- performance deteriorates with larger physical size

Cerium-activated “fast” inorganics

$Y_3Al_5O_{12}$ (YAG): Yttrium Aluminum Garnet

- light yield ~ 50% of NaI(Tl), long wavelengths (relative)
- principle decay time ~ 88 ns (72%) and 302 ns (28%) which reverse to 34% and 66% for α-particles

Cerium-activated “fast” inorganics

$Lu_2(SiO_4)O$(LSO) : Lutetium Oxyorthosilicate

- light yield ~ 75% of NaI(Tl)
- decay time ~ 47 ns
- high atomic mass of 71(Lu) makes it attractive, but cost and manufacturing issues make it less desirable
- also contains the natural radioactive element of Lu-176
- Energy resolution of 7.5 – 10 % differs based on detector size, smaller produces better energy resolution

Cerium-activated “fast” inorganics

Lu_2AlO_3 (LuAP): Lutetium Orthoaluminate

- Principle decay time 17 ns
- Light yield > 50% of NaI(Tl)
- Limited to thickness < 1 cm due to strong self absorption

Glass scintillators

- Silicate glasses containing lithium and activated with cerium used as neutron detectors
- Modern scintillation glasses : SiO_2, LiO_2,Al_2O_3,MgO and Ce_2O_3 with BaO added sometimes to increase the glass density
- Low light yield
- Can be used in extreme environs (corrosive or high temperature)

Glass scintillators

- Decay time ~ 50-75 ns
- May contain naturally radioactive thorium or potassium
- When used for neutron detection, lithium content is enriched to 95%
- When activated with terbium instead of Cerium, light decay time ~ 3.5 ms, but light output is much higher

Scintillator gases

- Generally noble gases (Ar, Kr, Xe, He)
- The incident radiation or charged particles create excited molecules. Photons are emitted in de excitation, from the two lowest molecular excited states
- Transition times ~ few ns
- Low scintillation efficiency
- List of gas scintillator properties are shown in Table 8.4 (at atmospheric pressure)

Scintillator gases

Table 8.4 Properties of Gas Scintillators at Atmospheric Pressure

Gas	Mean Wavelength of Emission	Number of Photons with $\lambda > 200$ nm per 4.7 MeV Alpha Particle
Xenon	325 nm	3,700
Krypton	318 nm	2,100
Argon	250 nm	1,100
Helium	390 nm	1,100
Nitrogen (for comparison)	390 nm	800
NaI(Tl)	415 nm	41,000

Source: J. B. Birks, *The Theory and Practice of Scintillation Counting.* Copyright 1964 by Pergamon Press, Ltd. Used with permission.

Radiation damage effects in inorganic scintillators

- May damage by inducing one or more of the following effects

1. reduction in the transparency of the scintillator caused by the creation of color centers that absorb the scintillation light
2. Interference with process that give rise to the emission of scintillation light
3. can induce long lived emission in the form of phosphorescence

Radiation damage effects in inorganic scintillators

- effects are known to be rate dependent and radiation particle dependent
- effects can also be reversible through hour or day long annealing
- correlations are complicated but most sensitive appears to be thallium activated halides (~10 Gy; where GSO starts to slow effects at 10^6Gy)
- Rough list of increasing radiation resistance: CsF, BGO, YAP, CeF_3 and BaF_2.