

Lecture 17

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
- 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.
- 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
 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 $\sim 50\text{-}500\text{ ns}$, which is the characteristic time of scintillation light.

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”
- 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.
- Example of an efficiency calculation for NaI:

On average it takes $\sim 3X$ the band gap energy to create electron hole pair, for NaI this is $\sim 20\text{ eV}$, for 1 MeV absorption, 5×10^4 electron hole pairs are formed.

With an absolute scintillation efficiency of 12%, 1 MeV would yield about 1.2×10^5 total light energy, with average photon energy of 4 eV , there is the production of $\sim 4 \times 10^4$ photons. So there is ~ 1 photon for every electron-hole pair formed.

- 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 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 of 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 ns
- Phosphorescence with a characteristic decay time of 0.15 s.
- 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.

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
- Can be grown in a columnar microstructure where individual columns behave as optically isolated scintillators (diameters $\sim 5 \mu\text{m}$)
- 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 components ($0.64 \mu\text{s} - 64\%$ and $3.34 \mu\text{s} - 36\%$)

CsI(Na):

- Emission spectrum is similar to NaI(Tl)
- Has a slow decay (with two components 0.46 and $4.18 \mu\text{s}$)
- Hygroscopic

LiI(Eu)

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

Other slow (>200ns) inorganic crystals

BGO: ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$)

- 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
- 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)

CdWO_4 : (cadmium Tungstenate)

- Light yield is $\sim 40\%$ 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)

$\text{ZnS}(\text{Ag})$

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

$\text{CaF}_2(\text{Eu})$:

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

Unactivated fast inorganics with low light yield

BaF_2 :

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

Pure CsI:

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

CeF_3 : (Cerium Fluoride)

- light output $\sim 5\%$ of NaI(Tl)
- decay time of ~ 27 ns for visible spectrum; but $\sim 5\text{ns}$ for UV

Cerium-activated “fast” inorganics

GSO: (Gd_2SiO_5) – Gadolinium 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
- has a long rise time of 10-20ns depending on cerium concentration caused by slow population processes for fluorescent states
- light yield $\sim 20\%$ of NaI(Tl)
- shows no appreciable radiation damage for γ -ray exposure up to 10^7 Gy

YAlO₃ (YAP): Yttrium Aluminum Perovskite

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

Y₃Al₅O₁₂ (YAG): Yttrium Aluminum Garnet

- light yield $\sim 50\%$ 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

Lu₂(SiO₄)O (LSO) : Lutetium Oxyorthosilicate

- light yield $\sim 75\%$ 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

Lu₂AlO₃ (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)
- 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 \sim few ns
- Low scintillation efficiency
- List of gas scintillator properties are shown in Table 8.4 (at atmospheric pressure)

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
- 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 show effects at 10^6 Gy)
- Rough list of increasing radiation resistance: CsF, BGO, YAP, CeF₃ and BaF₂.