SAND2007-4789C

Investigation of Strengthening Agents for Lanthanide Halide Scintillators

M. J. Harrison
F.P. Doty
Engineered Materials Dept.
Sandia National Laboratories, Livermore, CA 94550

Background

- Lanthanide halide scintillators exhibit improved scintillation characteristics over conventional scintillators, such as NaI(Tl).

- Mechanical properties of these scintillators (brittle, low stress strength) pose serious manufacturing and application problems.

- Improving the mechanical strength of these scintillators will allow the performance benefits of these scintillators to be utilized in many fields, including homeland security and medical imaging.

Objective

Utilize known methods of material strengthening to improve the mechanical strength of the lanthanide halide family of scintillators without significantly altering their scintillation properties.

Methods:

1. ~~Particle Inclusion~~
2. ~~Work Hardening~~
3. Alloying
   a. Isovalent (e.g. $Ce^{3+} \rightarrow La^{3+}$)
   b. Aliovalent (e.g. $Ce^{3+} \rightarrow Cd^{2+}$)

Sandia National Laboratories

Isovalent Alloying

- Strengthening effect related to:

1. Differences in ionic radii, i.e. greater difference yields a stronger alloy for a given concentration.
2. Concentration of isovalent species, typically a parabolic relationship.

- Typically requires a large fraction of substitution, ~10% or greater. Maximum effect near 50% normally.

Aliovalent Alloying

- Strengthening effect related to:

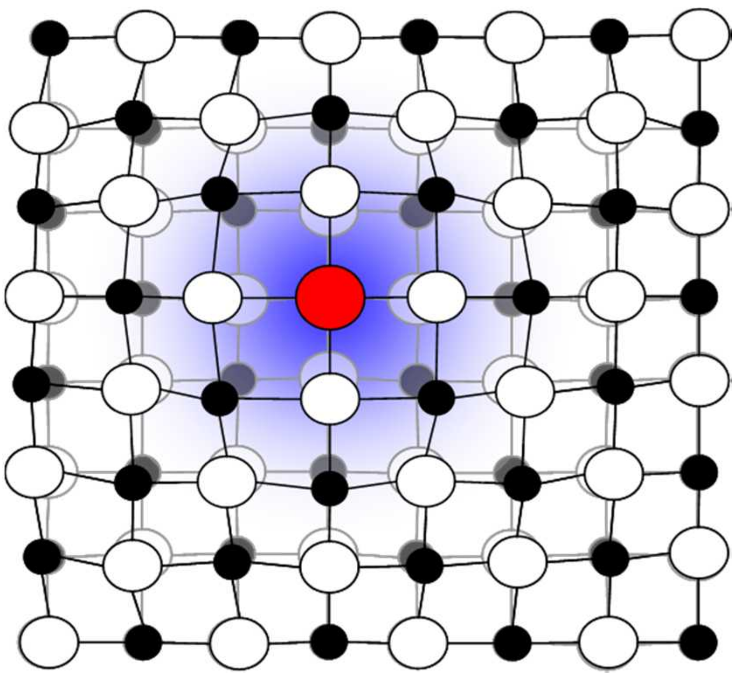
1. Differences in ionic radii.
2. Differences in valence state.
3. Square root dependence on concentration.

- Typically requires only a small fraction of substitution, on the order of 100 – 1000 ppm.

Comparison

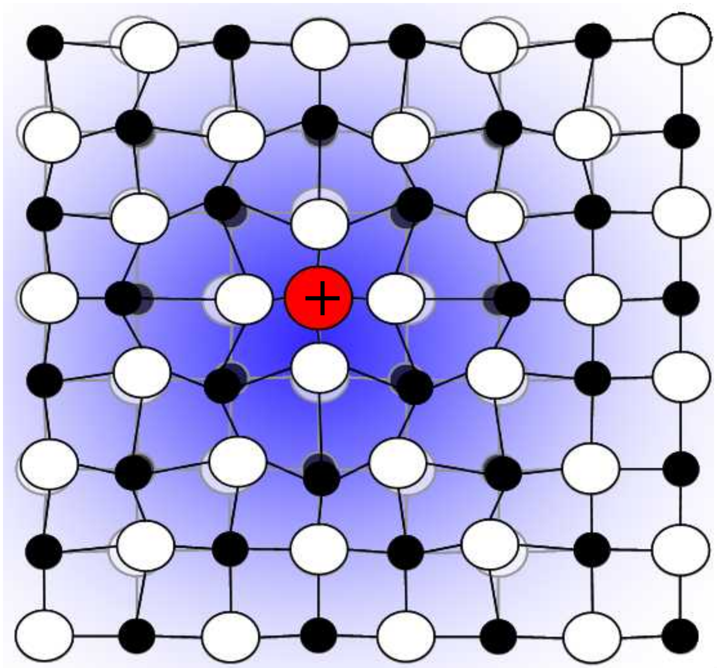
Isovalent alloying

-Short range elastic strain



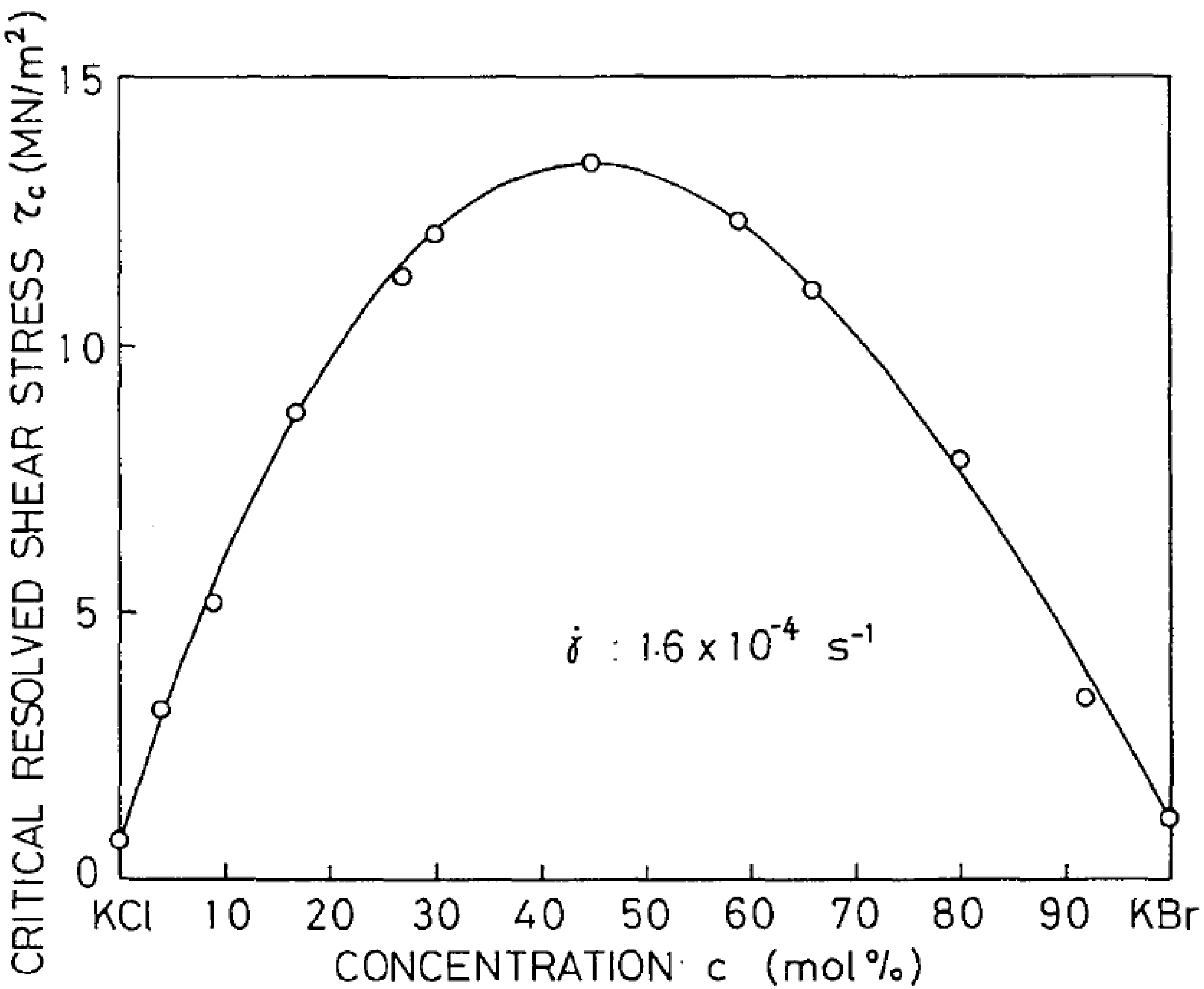
Aliovalent alloying

-Longer range effect



See for instance: T. Mitchell and A. Heuer, *Mat. Sci. Engg.* **28** (1977) 81-97.

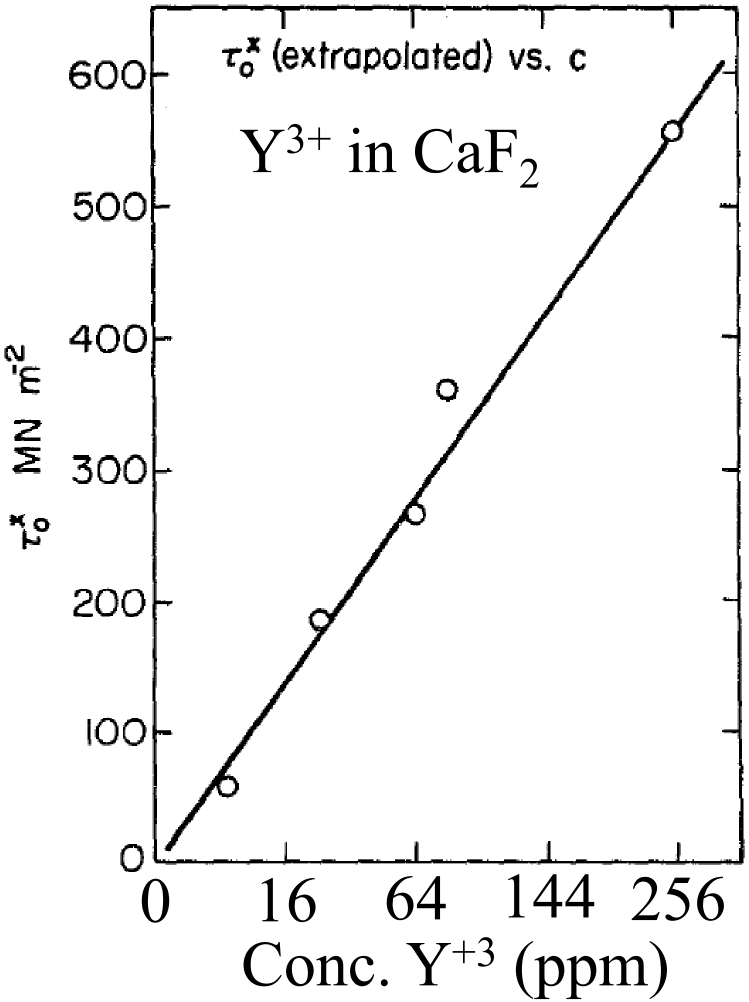
Isovalent Example



- Order of magnitude increase in CRSS requires ~30-40% molar concentration.

Figure from: Kataoka, T. and T. Yamada, *Jap. J. Appl. Phys.* **16** (1977) 1119-1126.

Aliovalent Example



- Order of magnitude increase in CRSS requires less than 300ppm!

Figure from: Sinha, M.N. and P.S. Nicholson, *J. Mat. Sci.* **12** (1977) 1451-1462.

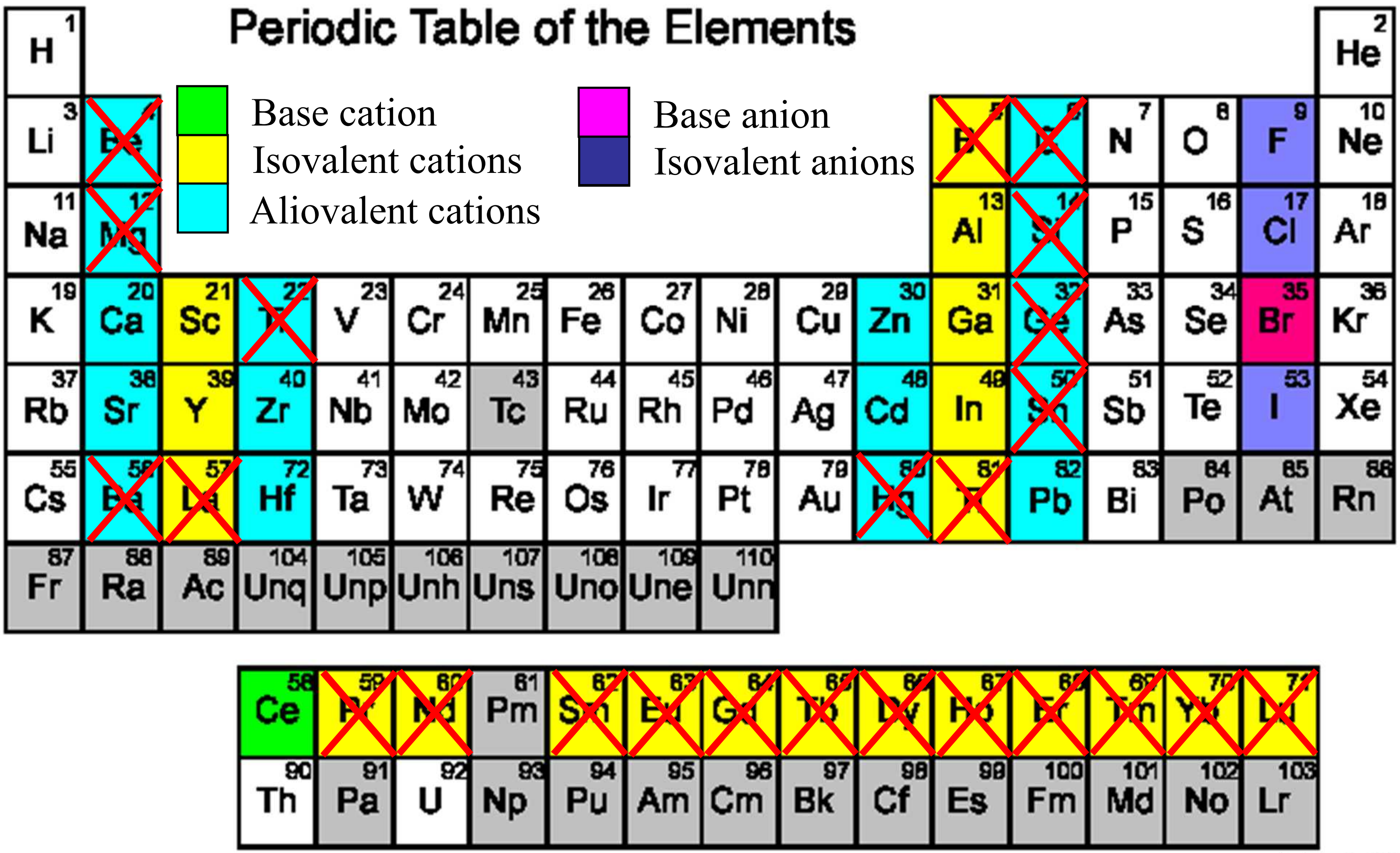
Sandia National Laboratories

Experimental Procedure

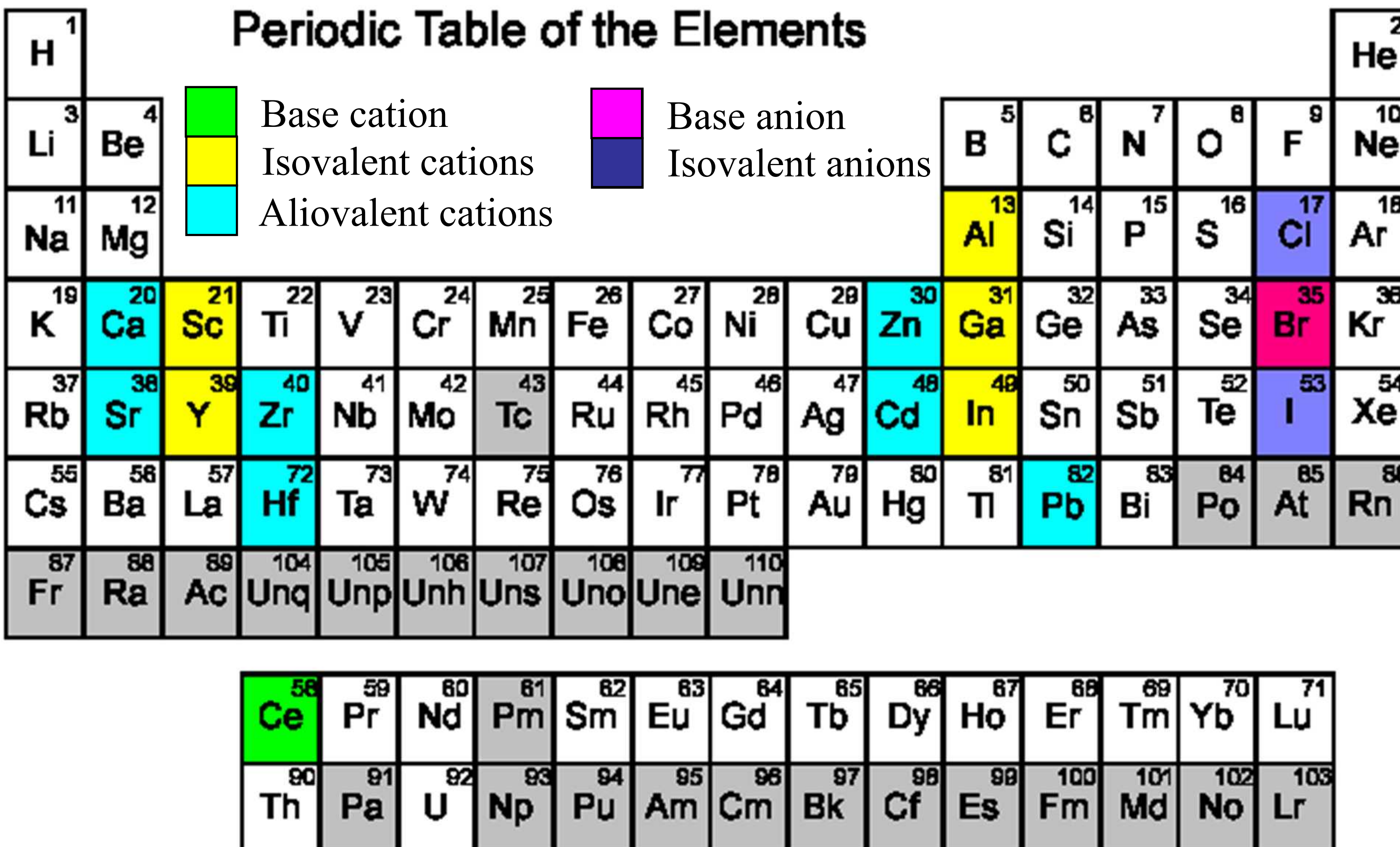
1. Select isovalent and aliovalent elements to alloy $CeBr_3$ with.
2. Prepare small (~0.5g) samples of each alloy.
    a. Isovalent – 10% molar (nominal)
    b. Aliovalent – 1% molar (large excess)
3. Observe fluorescence under hard UV excitation.
4. Measure fluorescence spectrum of each.
5. Determine whether each alloy scintillates.
6. Eliminate alloys that greatly alter the wavelength of the emitted light or do not scintillate.
7. Further investigate remaining alloys.

Sandia National Laboratories

Scope of Investigation



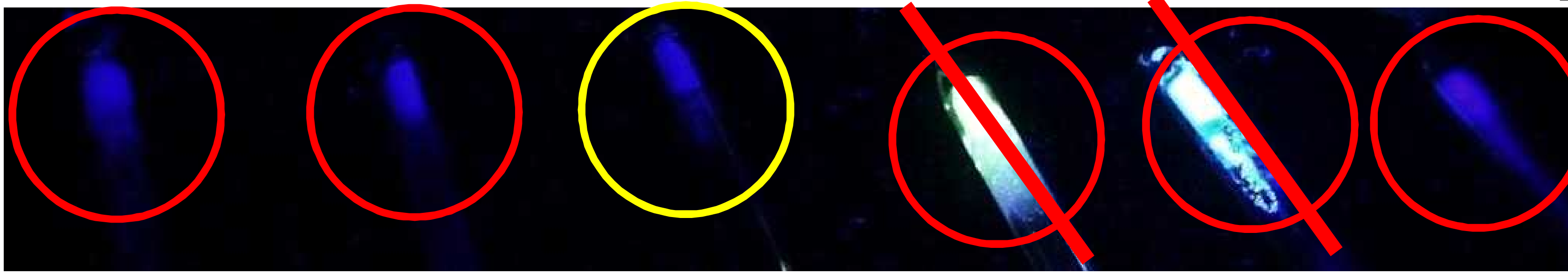
Scope of Investigation



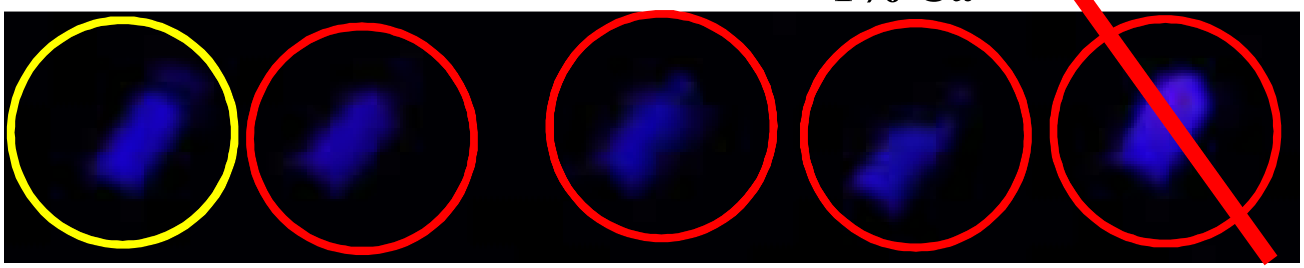
Sandia National Laboratories

Hard UV Fluorescence

10% Al^{3+} | 10% Ga^{3+} | Pure | 10% Sc^{3+} | 20% Y^{3+} | 1% $CdCl_2$

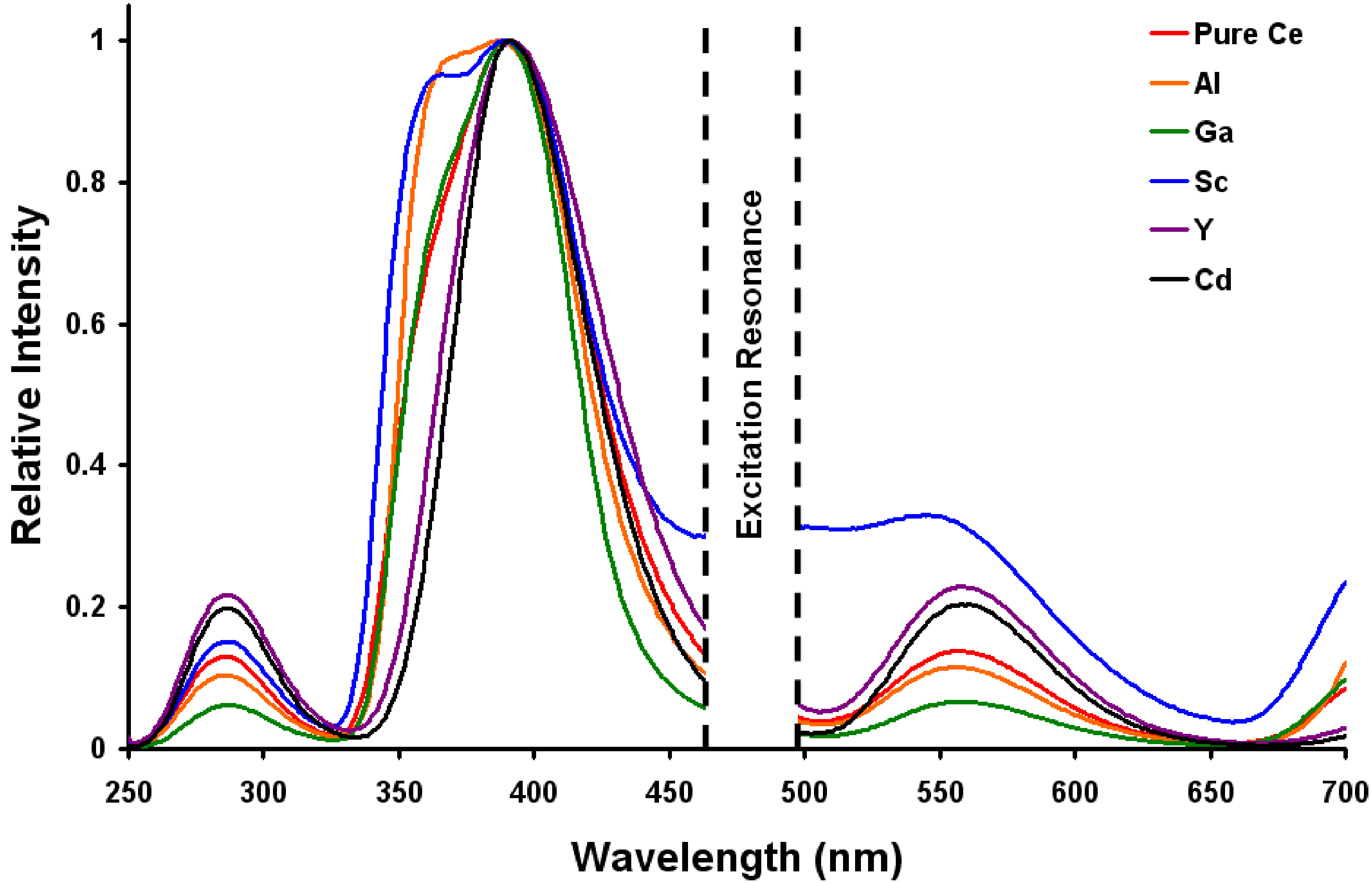


Pure | 1% Cd^{2+} | 1% Zn^{2+} | 1% Ca^{2+} | 1% Sr^{2+}

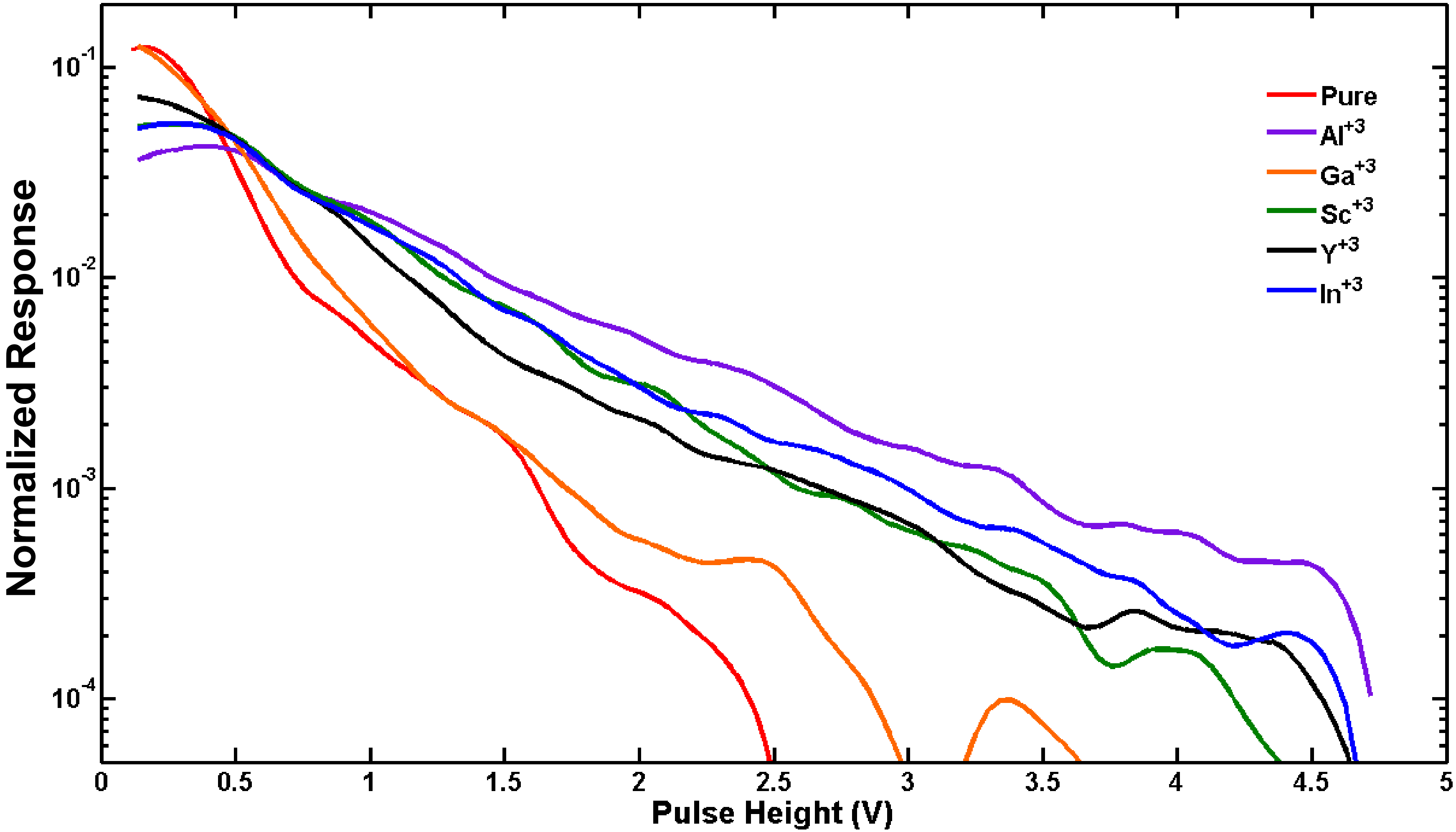


1% Hf^{4+} | 1% Zr^{4+} | 1% Pb^{2+} | 10% In^{3+} | 10% I^-

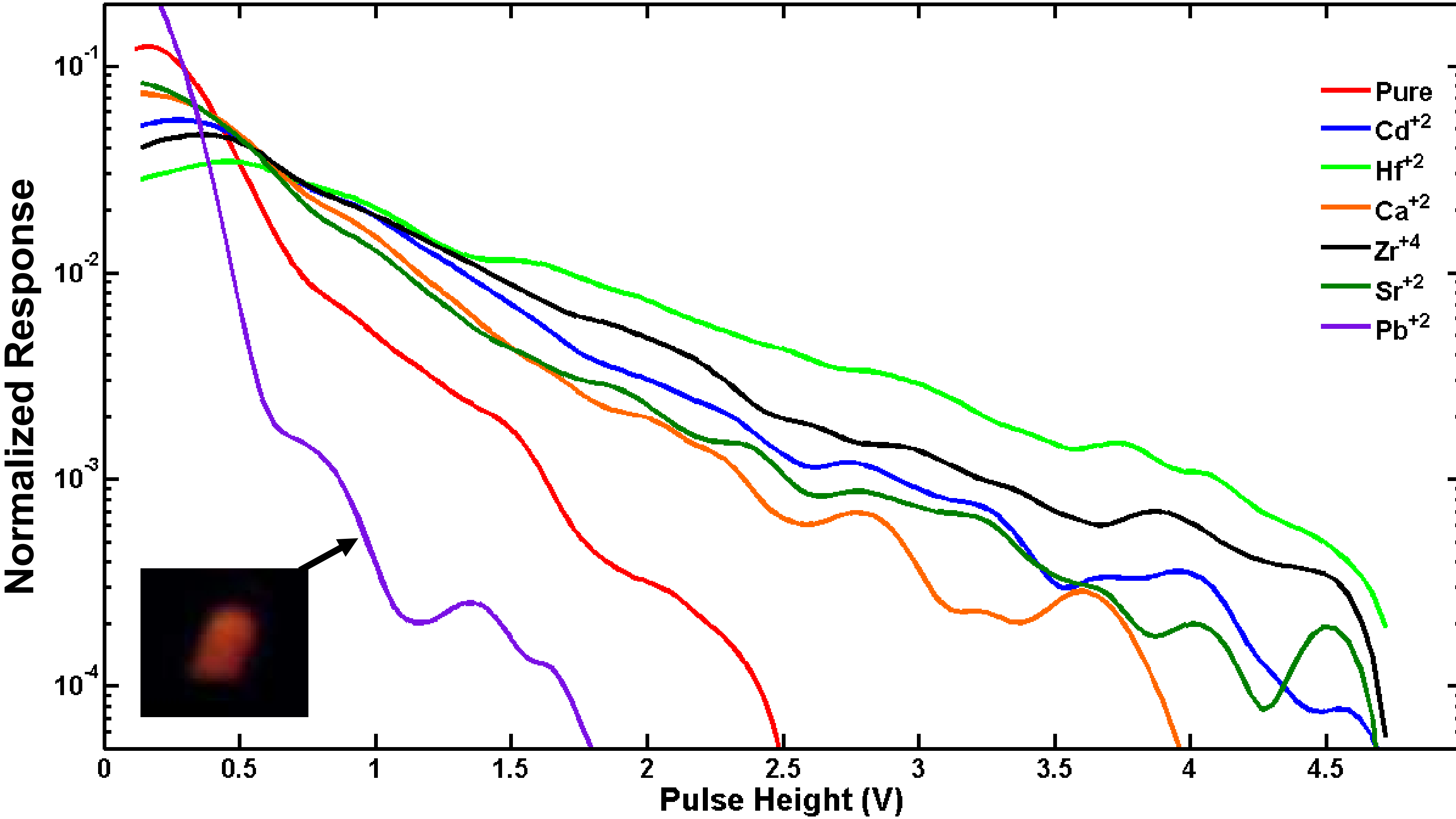
Emission Spectra



Isovalent Spectra

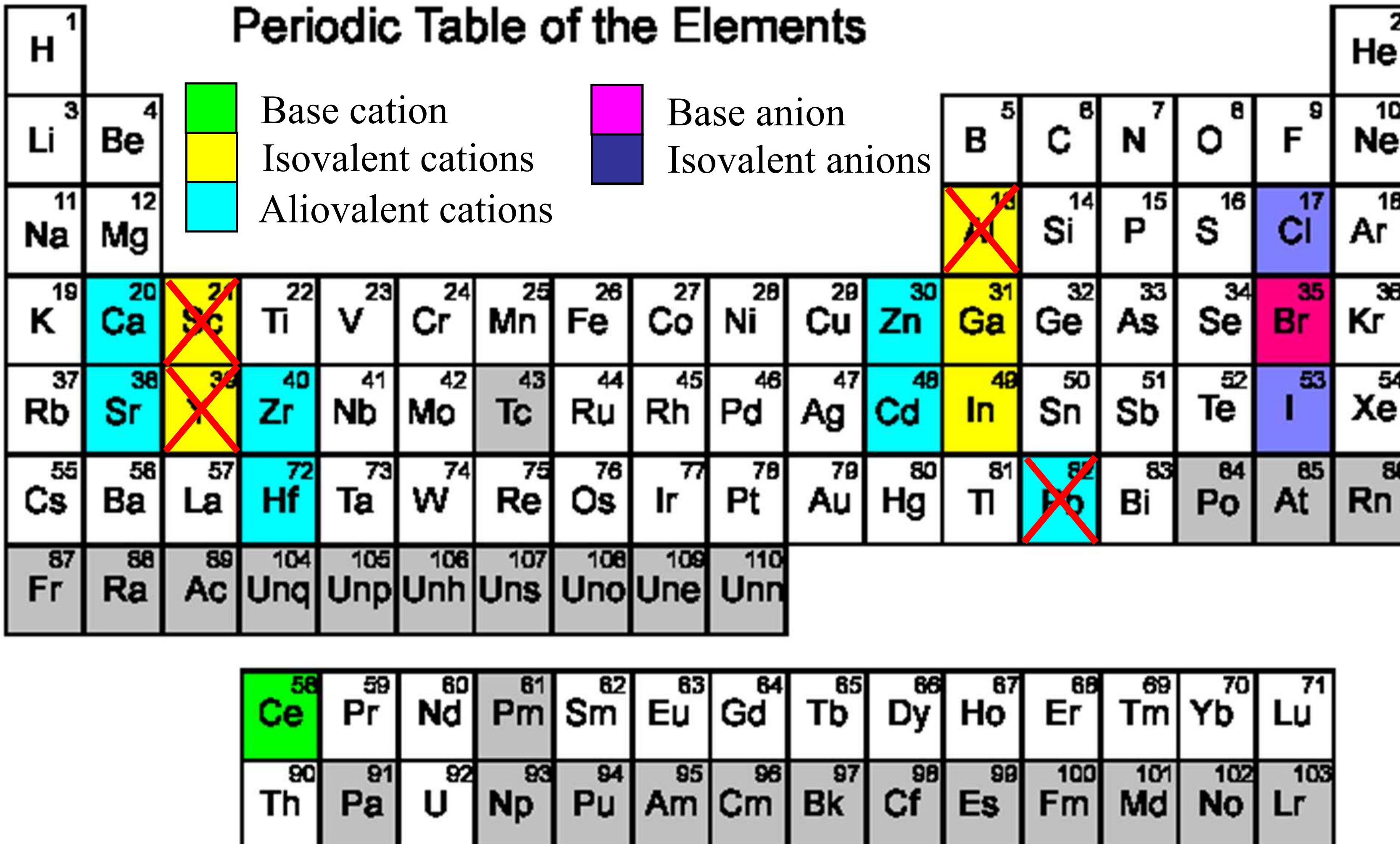


Aliovalent Spectra



Sandia National Laboratories

Results


Periodic Table of the Elements
Base cation
Isovalent cations
Aliovalent cations
Base anion
Isovalent anions
1 H
2 He
3 Li 4 Be 5 B 6 C 7 N 8 O 9 F 10 Ne
11 Na 12 Mg 13 Al 14 Si 15 P 16 S 17 Cl 18 Ar
19 K 20 Ca 21 Sc 22 Ti 23 V 24 Cr 25 Mn 26 Fe 27 Co 28 Ni 29 Cu 30 Zn 31 Ga 32 Ge 33 As 34 Se 35 Br 36 Kr
37 Rb 38 Sr 39 Y 40 Zr 41 Nb 42 Mo 43 Tc 44 Ru 45 Rh 46 Pd 47 Ag 48 Cd 49 In 50 Sn 51 Sb 52 Te 53 I 54 Xe
55 Cs 56 Ba 57 La 72 Hf 73 Ta 74 W 75 Re 76 Os 77 Ir 78 Pt 79 Au 80 Hg 81 Tl 82 Pb 83 Bi 84 Po 85 At 86 Rn
87 Fr 88 Ra 89 Ac 104 Unq 105 Unp 106 Unh 107 Uns 108 Uno 109 Une 110 Unn
58 Ce 59 Pr 60 Nd 61 Pm 62 Sm 63 Eu 64 Gd 65 Tb 66 Dy 67 Ho 68 Er 69 Tm 70 Yb 71 Lu
90 Th 91 Pa 92 U 93 Np 94 Pu 95 Am 96 Cm 97 Bk 98 Cf 99 Es 100 Fm 101 Md 102 No 103 Lr

Future Work

- Grow single crystal samples of each successful alloy with various dopant concentrations.
  - Measure absolute light yield and proportionality of light to energy deposition curve.
  - Measure mechanical strength vs. concentration.

Acknowledgements

- Tiffany Wilson for her help with the fluoroscopy measurements and Dr. Darryl Sasaki for graciously allowing us to use his fluorimeter.

- Project funded by the U.S. Department of Energy, National Nuclear Security Administration

Sandia National Laboratories