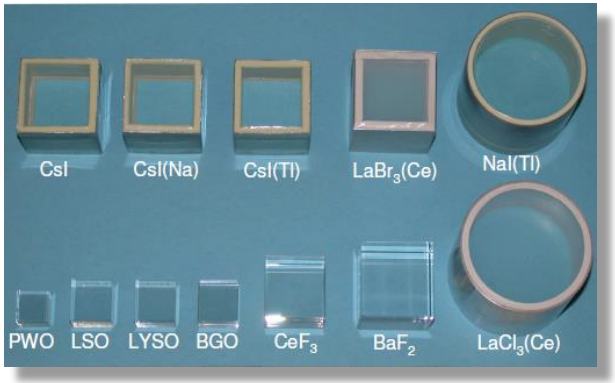
Glass Scintillators

V. Berdnikov, H. Gan, T. Horn, I.L. Pegg

Selection of Inorganic Scintillators

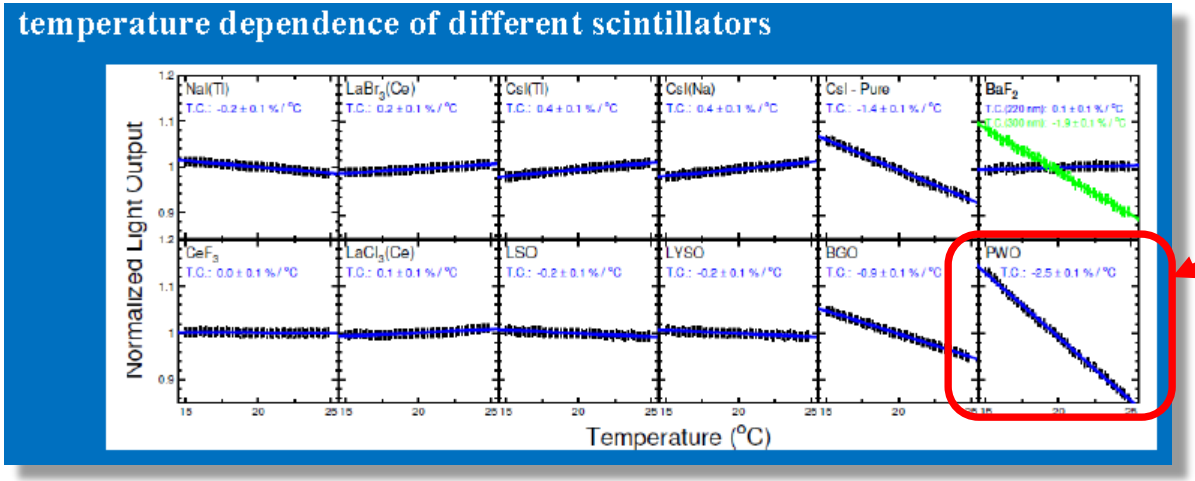
Material/ Parameter	Density (g/cm^3)	Melt. Point (°C)	Rad. Length (cm)	Moliere Radius (cm)	Refr. Index	Emission peak	Decay time (ns)	Light Yield (γ/MeV)	Rad. Hard. (krad)	Radiation type	Z_{Eff}
BaF_2	4.89	1280	2.03 2.06	3.10 3.40	1.50	300 220	650 0.9	16000 2000	>50	Scint.	52.7
CeF_3	6.16	1460	1.70 1.68	2.41 2.60	1.62 1.68	340 300	5 30	2800	>100	Scint.	50.8
(BGO)$Bi_4Ge_3O_{12}$	7.13	1050	1.12	2.23 2.30	2.15	480	300	8000 4000	>1000	.98 scint, .02 Č	83
(PWO)$PbWO_4$	8.30	1123	0.89 0.92	2.00	2.20	560 420	50 10	40 240	>1000	.90 scint. .10 Č	75.6
PbF_2	7.77	824	0.93	2.21	1.82	280 310	<30	2-6	50	Pure Č	77
(BSO):$CeBi_4Si_3O_{12}$	6.80	1030	1.85	≈5	2.06	470 505	≈100	1000 4000	>10	Scint.	75
(LSO):$CeLu_2SiO_5$	7.40	2050	1.14	2.07	1.82	420	40	30000	>1000	.98 sint ..02 Č	64.8
(LYSO):Ce$[LuY]_2SiO_5$	7.40	2050	1.14	2.07	1.82	420	40	30000	>1000	.98 scint. .02 Č	64.8



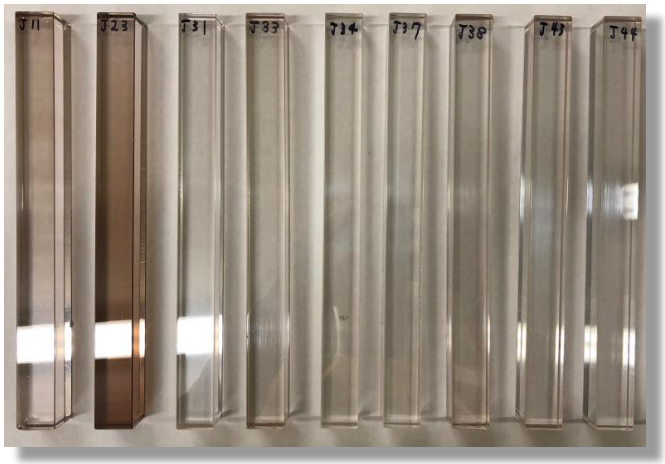
Identical Volume: X_0^3

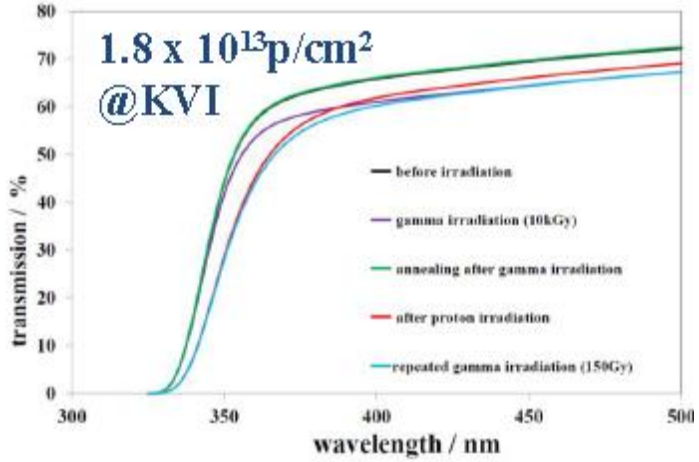
Crystals in EMCal: $PbWO_4$

- $PbWO_4$ material of choice for many EMCals – high density, fast response, large and granular solid angle, etc., but also limitations, e.g. hadron radiation damage



$PbWO_4$ light yield temperature dependence: 2%/°C

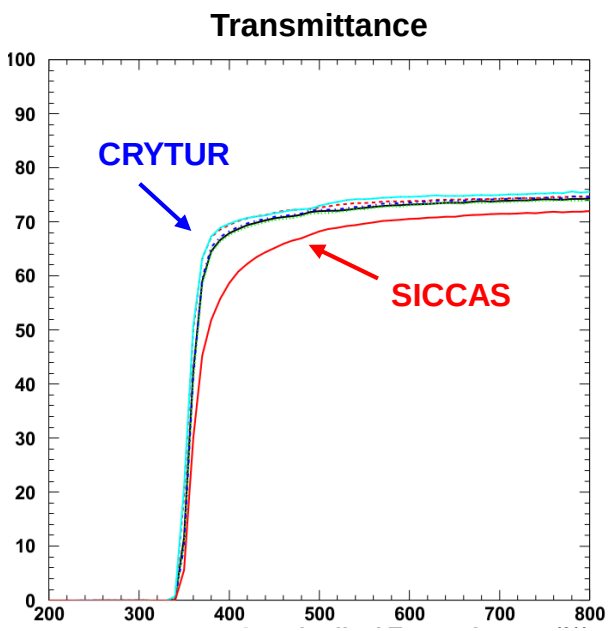


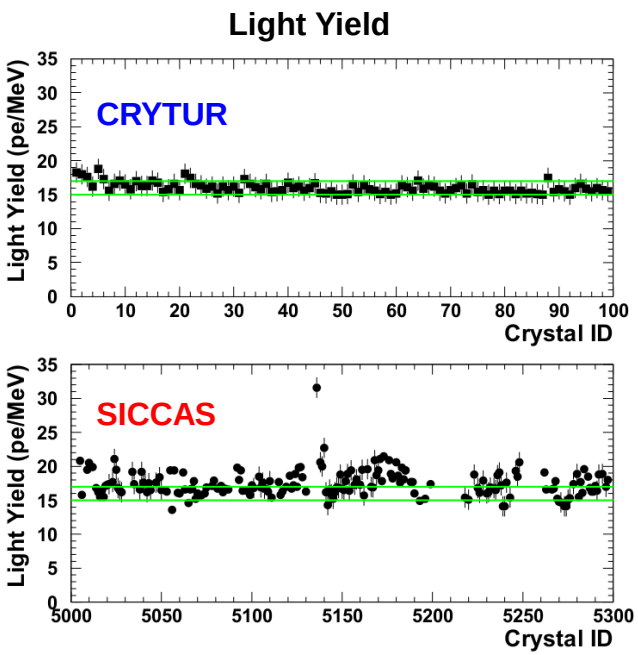


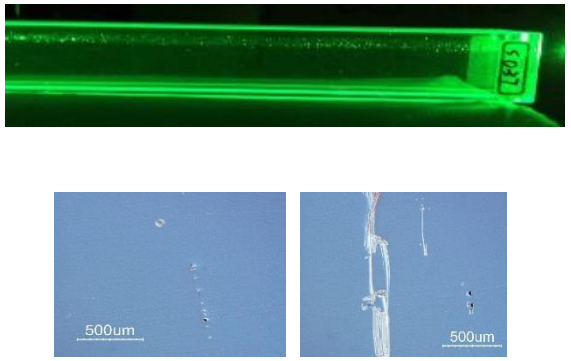
$PbWO_4$ radiation resistance

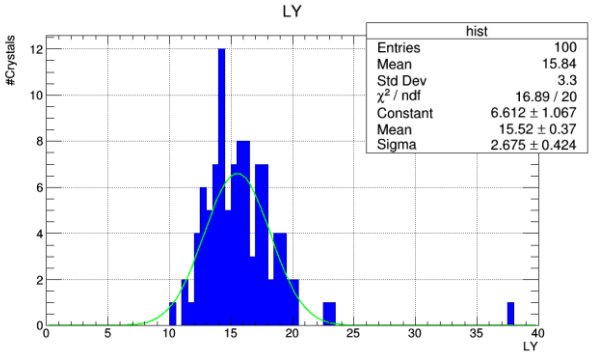
Crystals in EMCal: $PbWO_4$

- Another consideration: expensive ($15-25/cm^3) and manufacturing uncertainty
  - Despite progress (work with SICCAS and now also CRYTUR) still a struggle to work with vendors to get reliable $PbWO_4$ crystals that would be compatible with experiment requirements, e.g. NPS, EIC EMCal









Glass-based Scintillators for Detector Applications

An alternative active calorimeter material that is more cost effective and easier to manufacture than, e.g. crystals

Material/ Parameter	Density (g/cm^3)	Rad. Length (cm)	Moliere Radius (cm)	Interact Length (cm)	Refr. Index	Emission peak	Decay time (ns)	Light Yield (γ/MeV)	Rad. Hard. (krad)	Radiation type	Z_{Eff}
(PWO)$PbWO_4$	8.30	0.89 0.92	2.00	20.7 18.0	2.20	560 420	50 10	40 240	>1000	.90 scint. .10 Č	75.6
($BaO*2SiO_2$):Ce glass	3.7	3.6	2-3	~20		440, 460	22 72 450	>100	10 *(no tests >10krad yet)*	Scint.	51
($BaO*2SiO_2$):Ce glass loaded with Gd	4.7-5.4	2.2		~20		440, 460	50 86-120 330-400	>100	10 *(no tests >10krad yet)*	Scint.	58

Also: ($BaO*2SiO_2$):Ce shows no temperature dependence

The Vitreous State Laboratory – unique expertise

- Premier materials science facility with unique capabilities and expertise in glass R&D

- Current R&D program includes
  - Nuclear and hazardous waste stabilization
  - Glass and ceramic materials development
    - Formulation optimization
    - Characterization
    - Property-composition models
  - Materials corrosion and characterization
  - Off-gas treatment
  - Water treatment, ion exchange
  - Cements, flyash
  - Geopolymers
  - Biophysics
  - Nano-materials
  - Thermoelectrics
  - Spintronics
  - Scintillation detectors

The Vitreous State Laboratory – unique facility

- Designing, constructing and testing large glass production systems
  - VSL Joule Heated Ceramic Melter (JHCM) Systems:
    - The largest array of JHCM test systems in the US
    - The largest JHCM test platform in the US

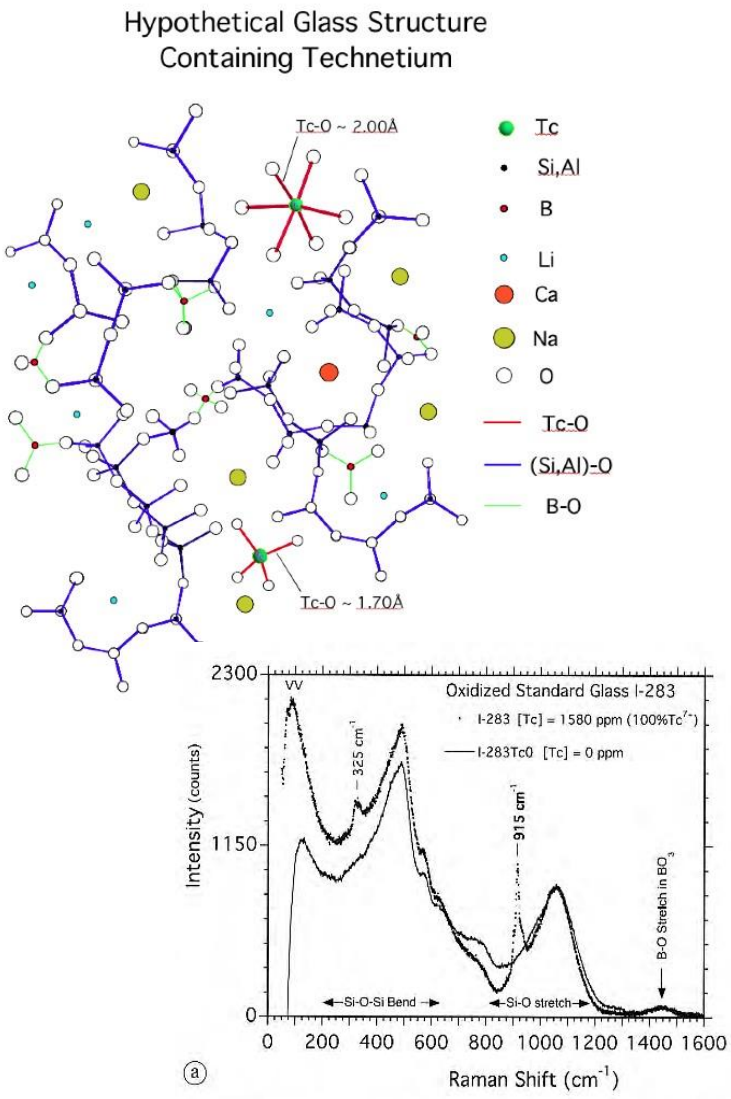
PILOT SYSTEM SCALE-UP

DM10 and DM100 JHCM Systems at VSL

VSL DM1200 HLW Pilot Melter System

About 400,000 kg glass made from about 1 million kg feed

XAS Studies on Silicate Glasses



- Na: $Na^{+}O_{3\text{-}7}$: Na-O = 2.30 -2.60 Å
- Mn: $Mn^{2+}O_{4\text{-}5}$: Mn-O = 2.07 Å, Mn-Mn = 3.48 Å
- Cu: $Cu^{2+}O_4$: Cu-O = 1.96 Å, Cu-Cu = 2.98 Å
- Sr: $Sr^{2+}O_{4\text{-}5}$: Sr-O = 2.53 Å
- Zr: $Zr^{4+}O_{6\text{-}7}$: Zr-O = 2.08 Å
- Mo: $Mo^{6+}O_4$: Mo-O = 1.75 Å
- Ag: $Ag^{+}O_2$: Ag-O = 2.10 – 2.20 Å
- I: $I^{-}(Na,I)_4$: I-Li = 2.80 Å, I-Na = 3.04 Å
- Re: $Re^{7+}O_4$: Re-O = 1.74 Å
- Bi: $Bi^{3+}O_3$: Bi-O = 2.13 Å
- S: $S^{6+}O_4$ surrounded by network modifiers; S^{2-}; S-S
- Cl: Cl-O = 2.70 Å; Cl-Cl = 2.44 Å; Cl-Na; Cl-Ca
- V: $V^{5+}O_4$; minor $V^{4+}O_5$ under reducing conditions
- Cr: redox sensitive: $Cr^{6+}O_4$ Cr-O = 1.64 Å; $Cr^{3+}O_6$ Cr-O = 2.00 Å; $Cr^{2+}O_4$ Cr-O ~ 2.02 Å
- Tc: redox sensitive, $Tc^{4+}O_6$ Tc-O = 2.00Å; $Tc^{7+}O_4$ Tc-O = 1.75 Å; evidence of Tc-Tc = 2.56 Å in hydrated, altered glass
- Sn: $Sn^{4+}O_6$ (minor $Sn^{2+}O_4$) Sn-O = 2.03 Å; Sn-Sn = 3.50 Å
- Al: $Al^{3+}O_4$: Al-O: 1.77 Å
- Si: $Si^{4+}O_4$: various polymerizations
- Zn: $Zn^{2+}O_4$: Zr-O: 1.96 Å, Zn-Si 2nd nearest-neighbor evidence

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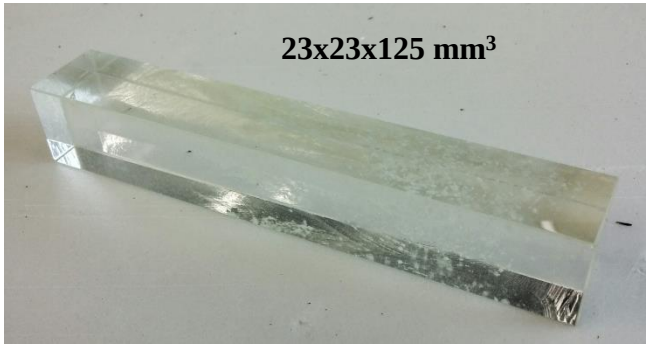
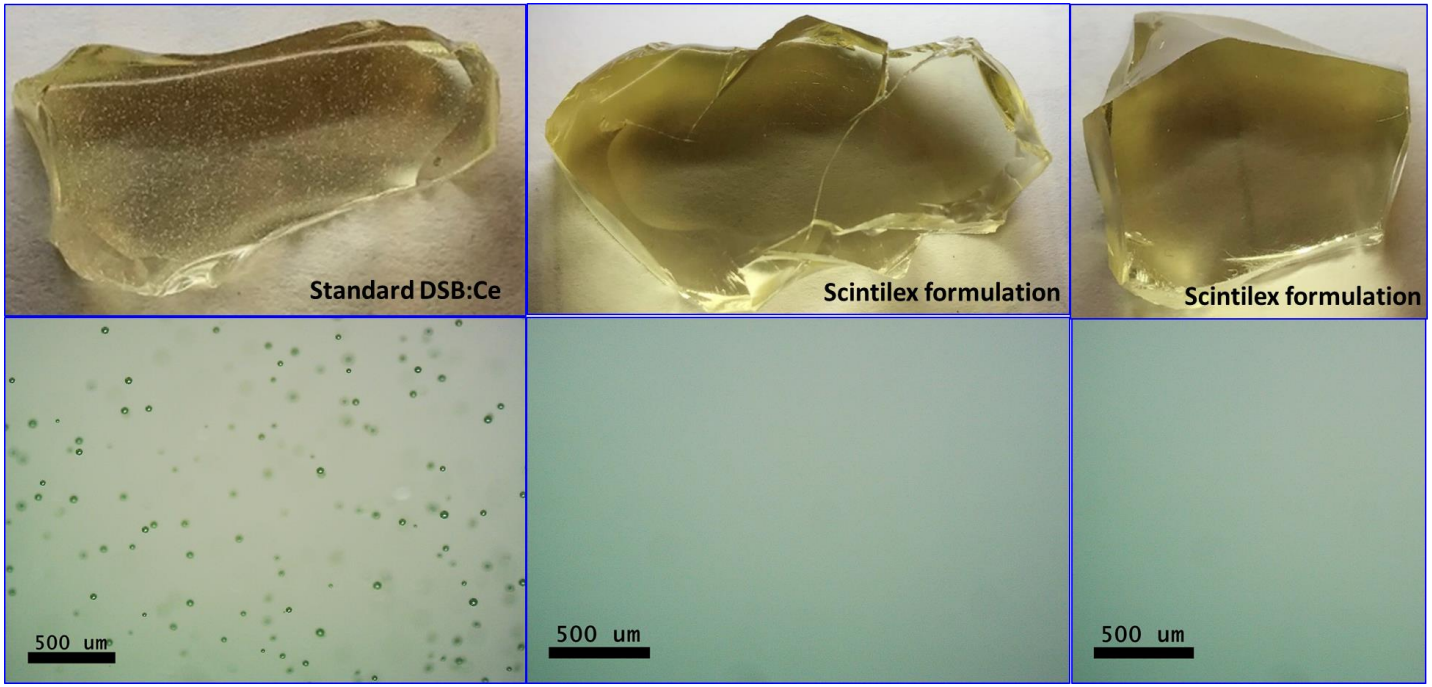
Shortcomings of earlier work:

- Macro defects, which can become increasingly acute on scale-up
- Sensitivity to electromagnetic probes

Status of New Glass/Ceramic Scintillator Material



- Uniformity remains a concern – manufacturing process requires optimization – progress with new method at CUA/VSL/Scintilex



Sample made at CUA/VSL based on previous DSB:Ce work

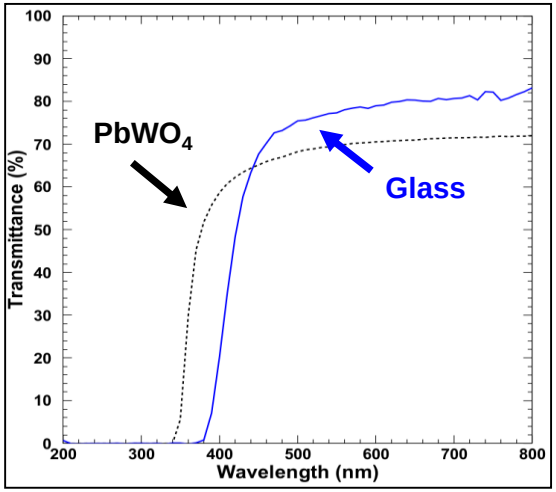
Samples made at CUA/VSL/Scintilex with our new method

Status of New Glass/Ceramic Scintillator Material

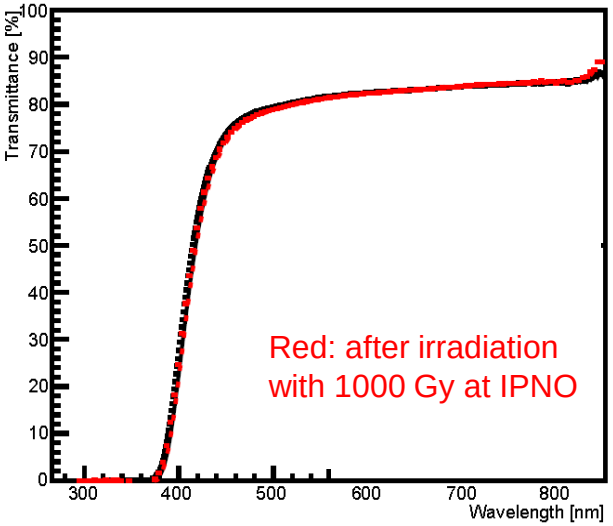
- Glass scintillators being developed at *VSL/CUA/Scintilex*
  - Optical properties comparable or better than $PbWO_4$
  - Preliminary tests on radiation damage look promising
  - Ongoing optimization work

Scintilex samples

Transmittance



Radiation Hardness

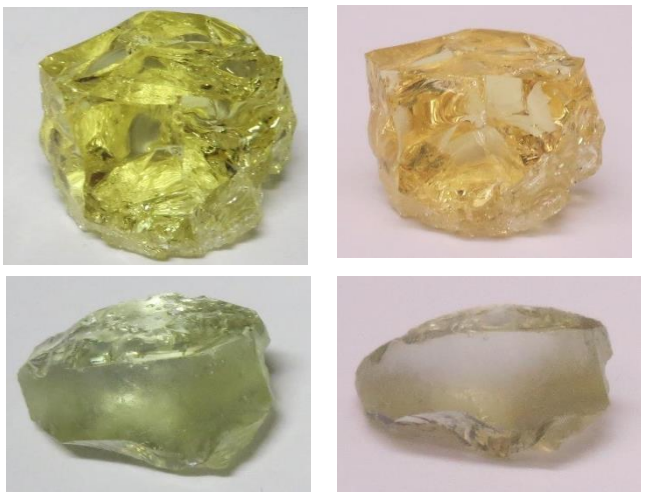


Light Yield

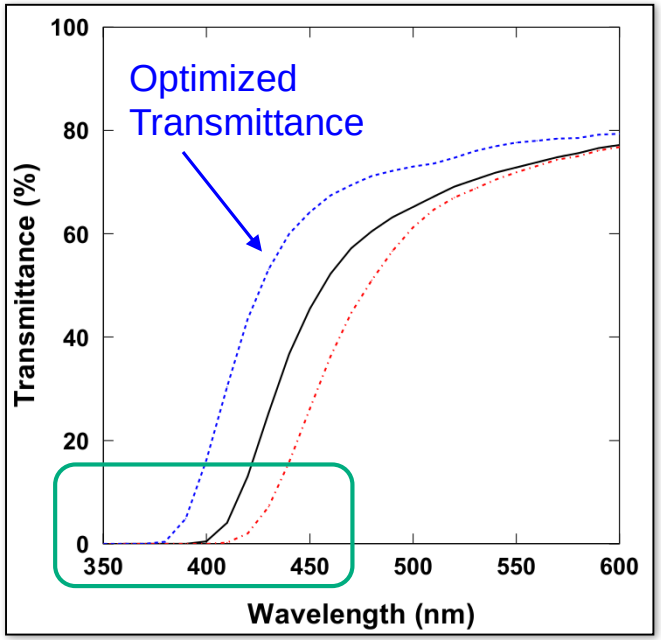
Material/ Parameter	$PbWO_4$	Sample 1	Sample 2	Sample 3	Sample 4
Luminescence (nm)	420	440	440	440	440
Relative light output (compared to $PbWO_4$)	1	35	16	23	11

Status of New Glass/Ceramic Scintillator Material

- Color optimization – progress with new method at CUA/VSL/Scintilex



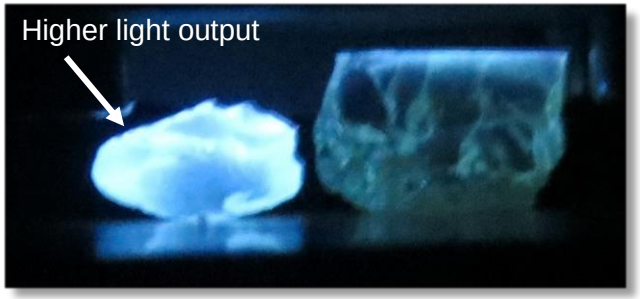
New glass has improved properties



New Scintilex formulations

More transparent

Higher light output



Excitation with 160 keV photons

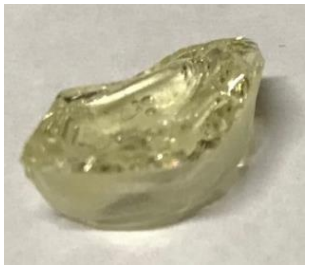
Status of New Glass/Ceramic Scintillator Material

- High dose radiation tests – progress with new method at CUA/VSL/Scintilex

VSL-Scintilex-G1 | **VSL-Scintilex-G2** | **VSL-Scintilex-G3**



Before irradiation


After 2min 160KeV Xray at >3k Gy/min



After additional 2-5 min 160 KeV Xray at >3k Gy/min

Outlook

- Finalize optimization of glass composition - patent
- Larger samples for full characterization tests
  - Transmittance
  - Light Yield
  - Radiation hardness
  - Decay kinetics
  - Other
- Monte Carlo simulations to determine compactness of detectors
- Possible prototype tests in Hall D this fall

Summary

- $PbWO_4$ crystals are ideal for precision EMCal, but also have limitations – and are expensive

- Glass-based scintillators are cost-effective alternative to crystals, in particular EMCal regions with relaxed resolution requirements
  - Initial small samples produced at CUA/VSL have 35x light yield of $PbWO_4$
  - New method also eliminates bubbles, a major problem in earlier work
  - Ongoing optimizations