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Low Characteristic Temperature Glass Ceramic for LED Lighting

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Outline

- Introduction
- Experimental Procedure
  - Phosphor embedding into softened glass
- Results and Discussion
  - Low characteristic temperatures
  - Glass structure and bonding
  - Optical performance
- Conclusion

Introduction

- Long life time (~10^5 hours, even at 50°C still has $4*10^4$ hour)
- Low on/off time (~10^{-9} sec)
- Low power loss
- High shock resistant
- Small size (less than 2 mm)
- Easy focus
- Low flicker
- No pollution
- High monochromatic

Source: http://www.digitalversus.com / Low energy consumption: how to choose your LED lights

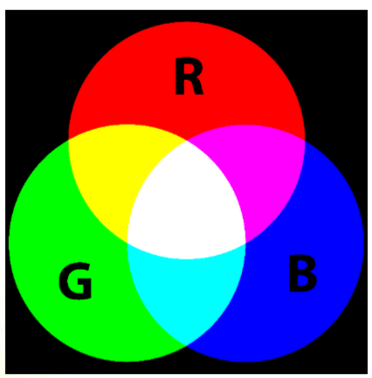
Introduction

- White light" usually means a multi-color mixed light.

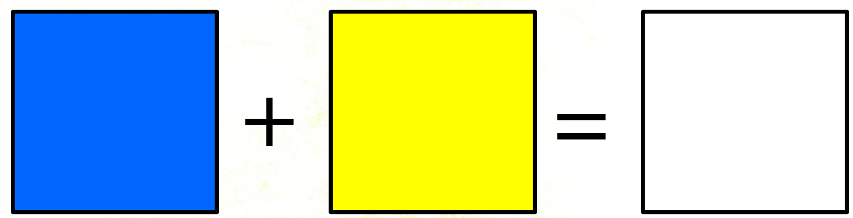
1. To mix red, green, blue

   and light to get a tri-chromatic white light.

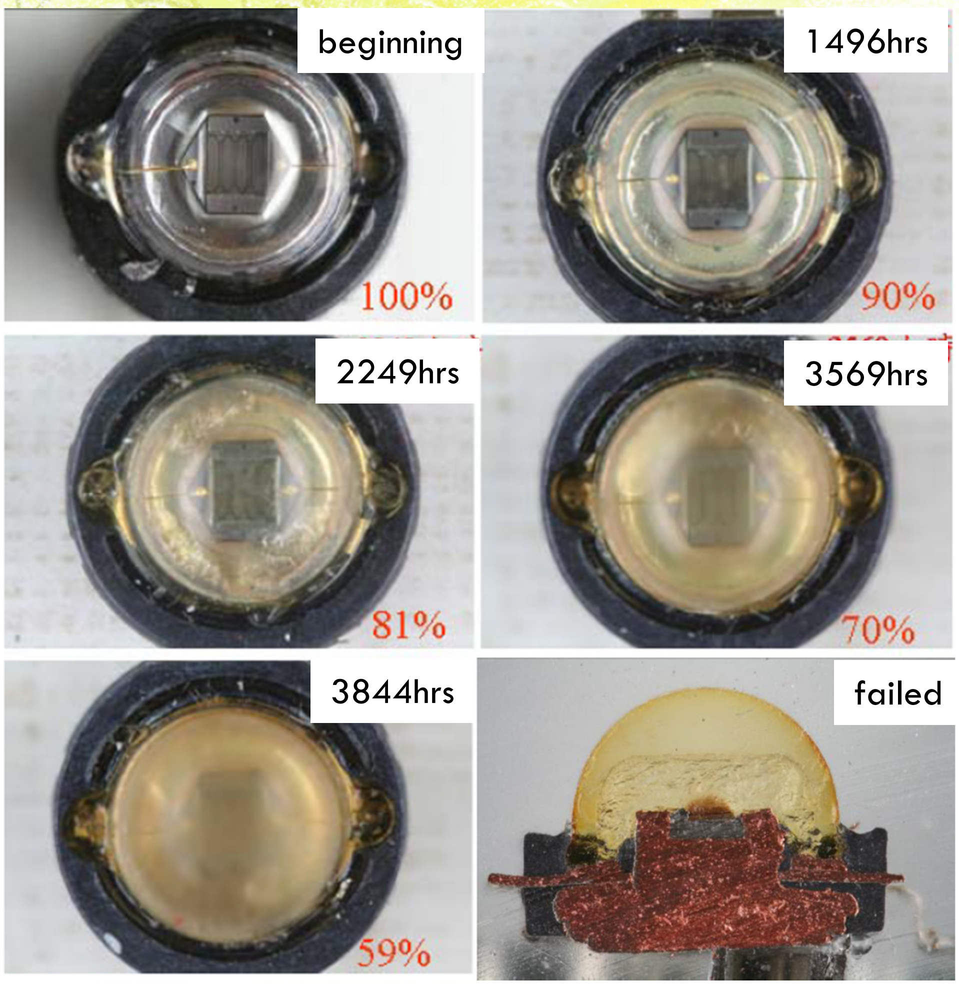
1. Using blue light and yellow light to obtain a di-chromatic white light, two complementary colors combine to form white light.

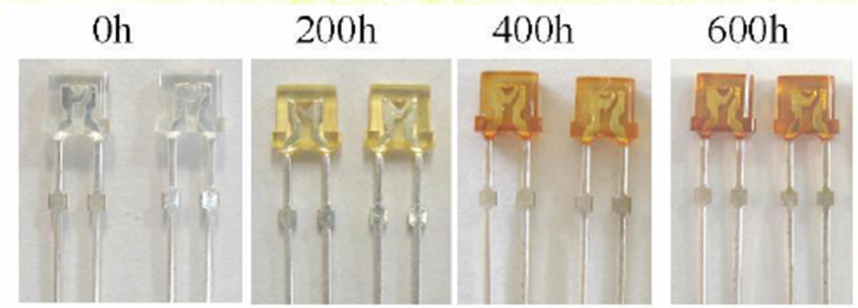


Source: BPI COLOR



Critical issues





Source : F. Shunsuke, YAG glass-ceramic phosphor for white LED (I)

- The blue LED chip excite light making the temperature of chip increase.
- The epoxy degenerated significantly due to thermal annealing, leading to decrease the life time and luminous efficiency.

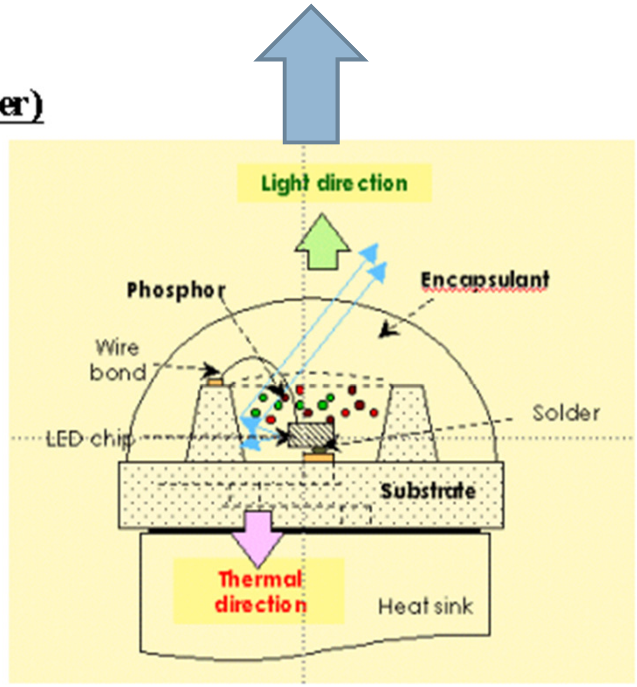
Source :C.C Tsai, The Reliability Study of Optical Power and Radiation Pattern for High-Power Light-Emitting Diodes Modules in Aging Test

Critical issues Materials

Higher Power, Enhanced Efficiency, elongated Lifetime …
Applications, like Car, Building, Spotlight, Lighthouse…

Phosphors (Wavelength Converter)

- High excitation in NUV or Blue
- High quantum efficiency
- High color rendering index
- High thermal stability
- Low scattering loss

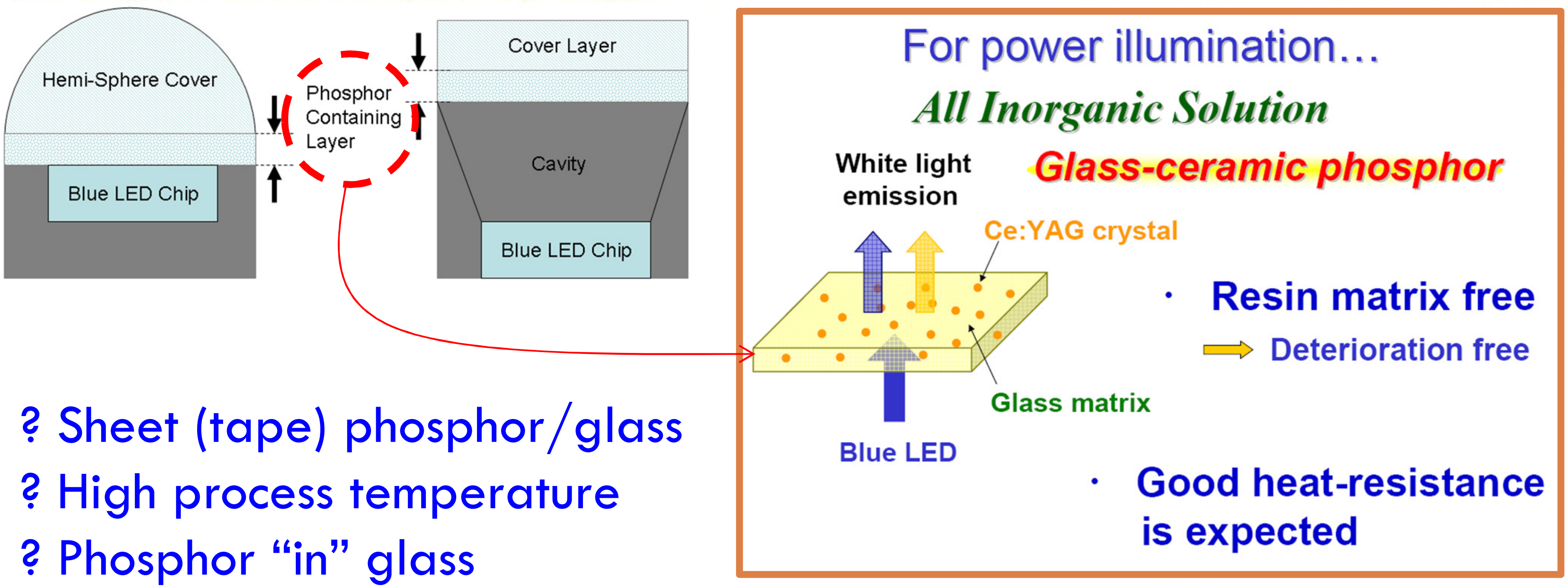


Encapsulants (Packaging Materials)

- High refractive index /High VLT
- High thermal stability
- High UV stability
- Moisture resistant
- Environmental-Friendly/Reworkable

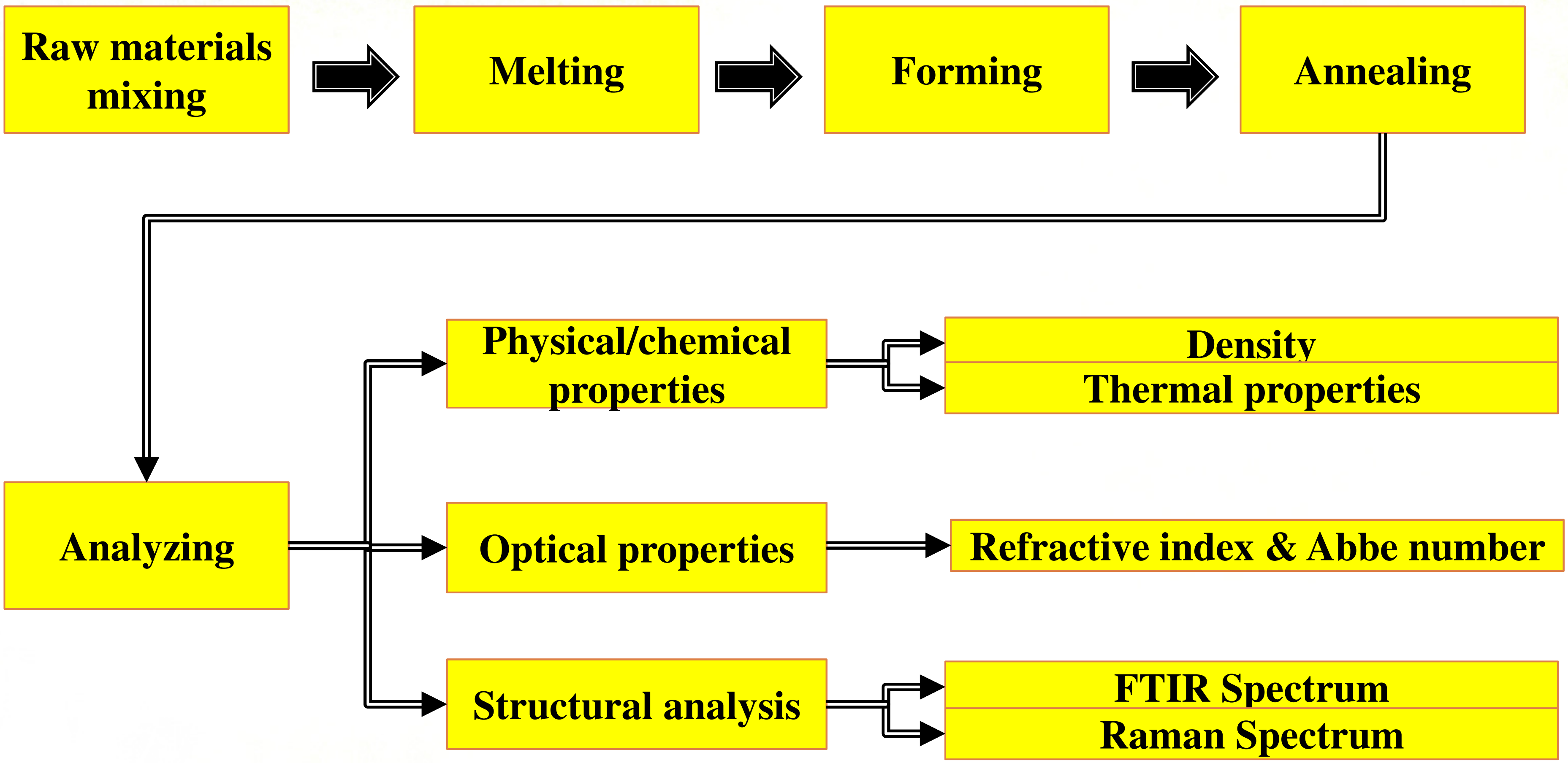
The replacement of polymer-based encapsulate by quality **"glass materials"**

Challenges



- Sheet (tape) phosphor/glass
- High process temperature
- Phosphor “in” glass
- Match in physical/thermal/optical properties like Density, CTE, Refractive Index…

Experimental Design for Glass

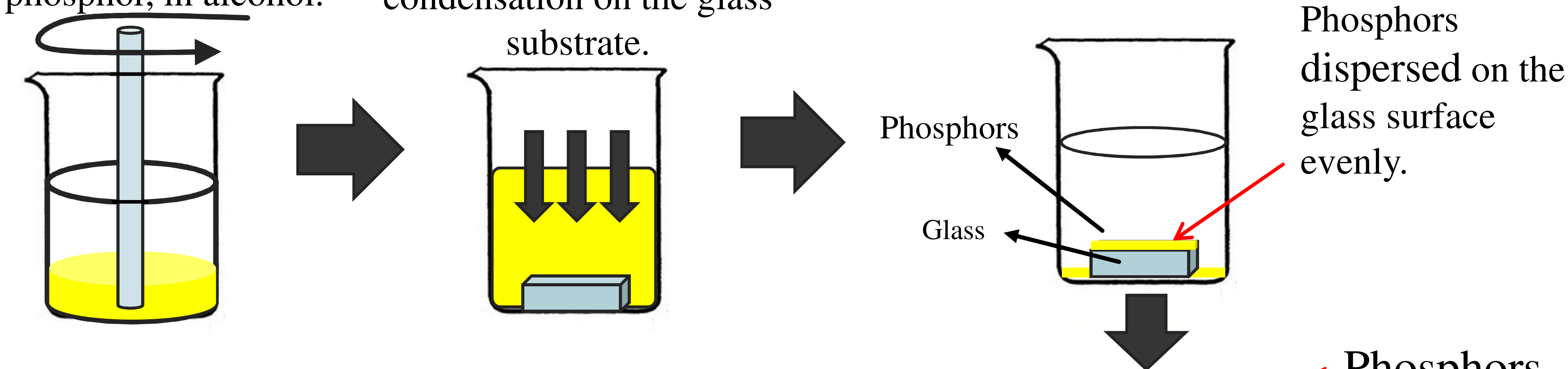


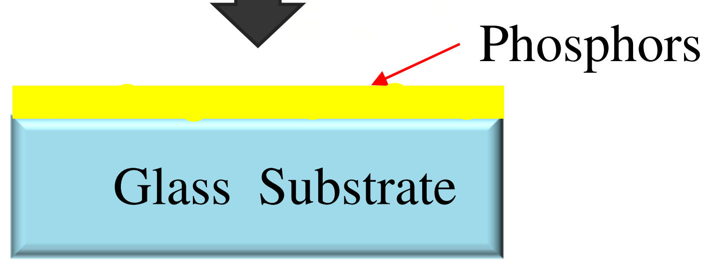
Experimental procedures

Coating → ***Heat treatment*** → ***Annealing*** → ***LED lighting measuring***

Suspension of YAG phosphor, in alcohol.

YAG phosphor condensation on the glass substrate.



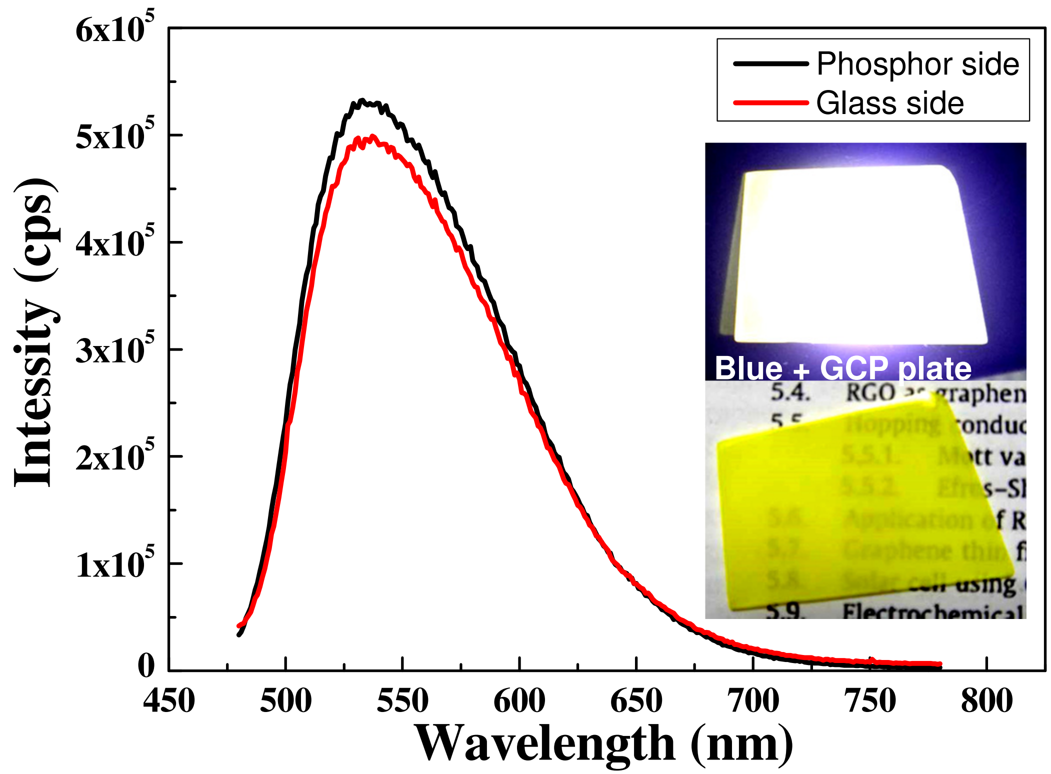


The incorporation of phosphors in the low-characteristic-temperature glass is conducted by the embedding of phosphor powders at softened glass surface.

YAG GCP under Blue light

➢ *Glass Ceramics-Phosphor Optical Performance*

- *Glass Ceramics-Phosphor & Phosphor Emission Spectrum*

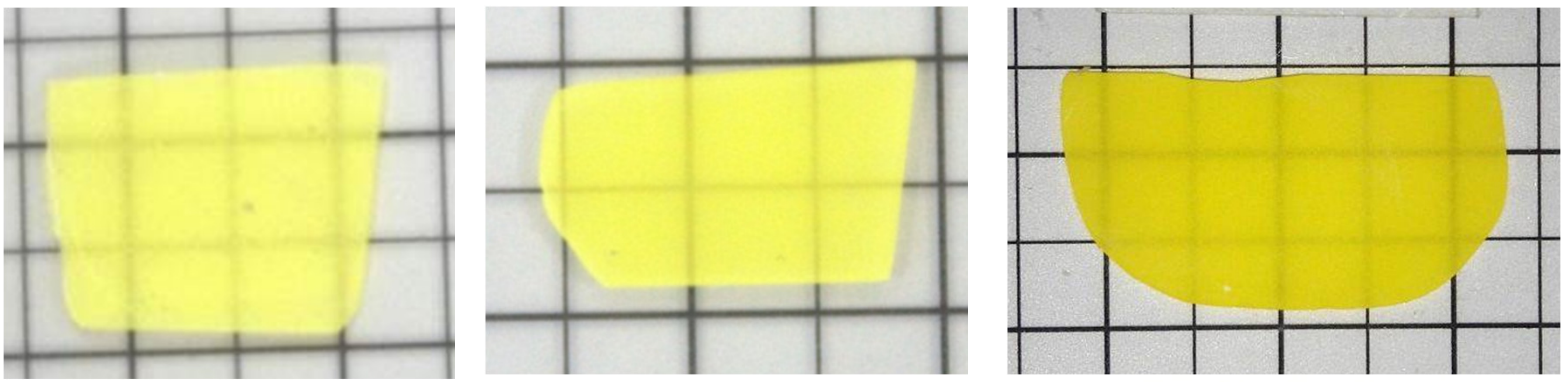


- The result shows an intense emission peak at 550 nm for the GCP sample, which is a unanimous result as the YAG phosphors of emission spectra.

YAG GCP by Various Glass Systems

- *Glass Ceramics-Phosphor*
  - *Appearances*

(a) Phosphate-SrK **(b) Phosphate- NbLi** **(c) Borosilicate-BiZn**



- Phosphate systems are not well-wetted with phosphors. Shallow incorporation and limited YAG into glasses are detrimental for lighting.
- Borosilicate glasses are chemically stable, yet higher thermal characteristic temperatures are expected.

YAG GCP by Various Glass Systems

➢ *Glass Substrate Characteristics Measure*

- *Thermal & Optical properties*

Glass systems	T_g(°C)	T_d(°C)	T_c(°C)	CTE(10^{-6}/K)	T_c - T_d (°C)	Refractive Index	Abbe number
P-40Sr10K	618	661	853	4.5	192	1.616	38.93
P-30Sr20K	579	614	889	6.3	275	1.601	40.29
P-20Sr30K	543	582	917	8.6	335	1.584	45.97
P-25Nb0Li	529	570	905	5.8	335	1.791	27.75
P-20Nb5Li	533	569	856	7.0	287	1.759	32.42
P-15Nb10Li	527	567	794	7.4	227	1.720	35.15
B-30Zn6Bi	549	586	784	5.9	198	1.774	37.90
B-27Zn9Bi	552	574	775	6.2	201	1.796	34.96
B-24Zn12Bi	547	569	770	6.1	201	1.816	32.03

- The greater T_s and the smaller Td of the glass substrate, leading to a layer operating temperature range ($T_c - T_d$) for the glass ceramic preparation was expected.
- In this study, the glass systems all exhibit a ($T_c - T_d$) over 200 °C, which is quite sufficient in glass ceramic fabrication.

Designed Glass systems

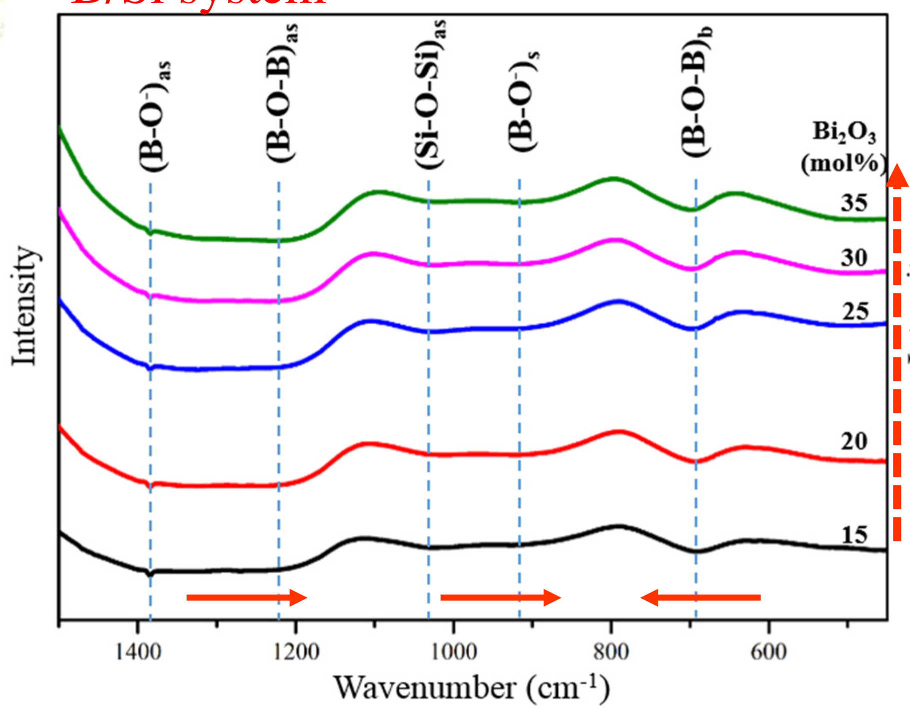
Glass systems	T_g(ºC)	T_d(ºC)	T_c(ºC)	CTE(10^{-6}/K)	T_c - T_d (ºC)	Refractive index	Abbe number
50B0Si	464	495	627	6.97	132	1.89	28.32
45B5Si	461	494	684	7.49	172	1.89	26.63
40B10Si	461	489	690	7.86	201	1.89	23.7
35B15Si	466	487	737	6.21	250	1.90	24.02
30B20Si	474	497	738	6.49	241	1.90	26.1
25B25Si	474	499	764	6.55	265	1.90	24.3
15Bi35Zn	483	524	737	6.02	213	1.85	28.32
20Bi30Zn	472	508	684	7.05	176	1.85	27.6
25Bi25Zn	461	494	666	7.49	172	1.88	24.11
30Bi20Zn	455	488	639	7.67	151	1.91	24.36
35Bi15Zn	447	476	626	6.79	150	1.94	22.53

➢ Working range(WR): $T_c - T_d$ (ºC) : Glass can be shaped (worked) without devitrification

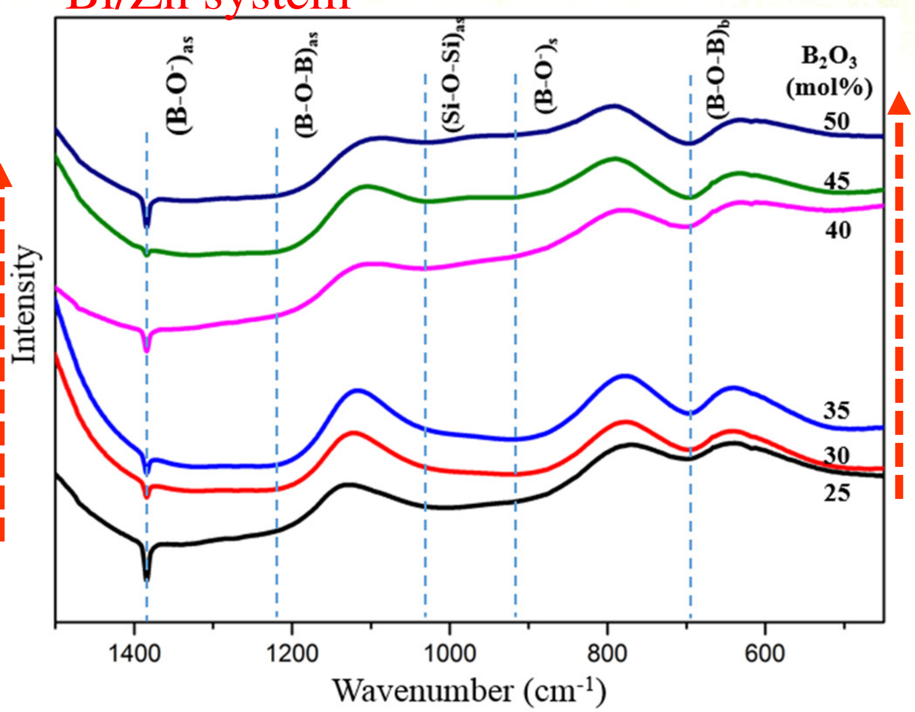
- More NBOs lead to a more relaxed structure when the Bi_2O_3 content increases.
- All working ranges are grater than 150ºC.
- When Bi_2O_3 increases, refractive index increases.
- The refractive index of all developed glass systems are greater than 1.85.

Glass structure FTIR spectra

B/Si system



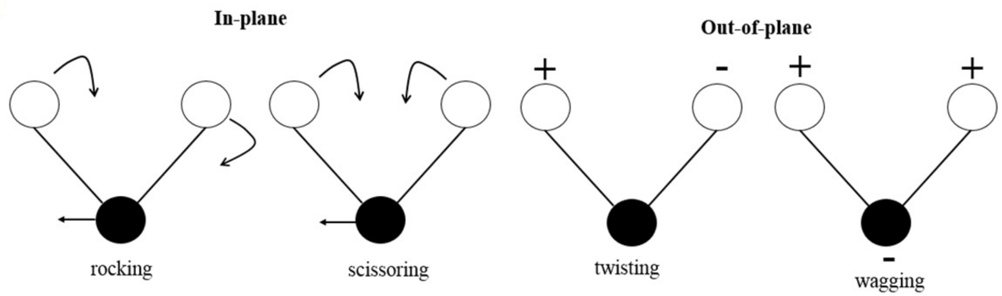
Bi/Zn system



- B-O$^-$ reflections shift toward low wavenumbers as Bi_2O_3 increases .

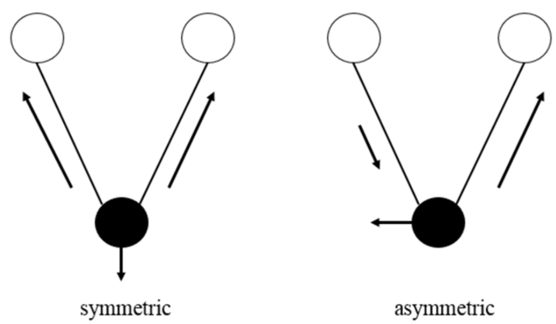
Molecular absorption spectrometry

- *Bending vibration*



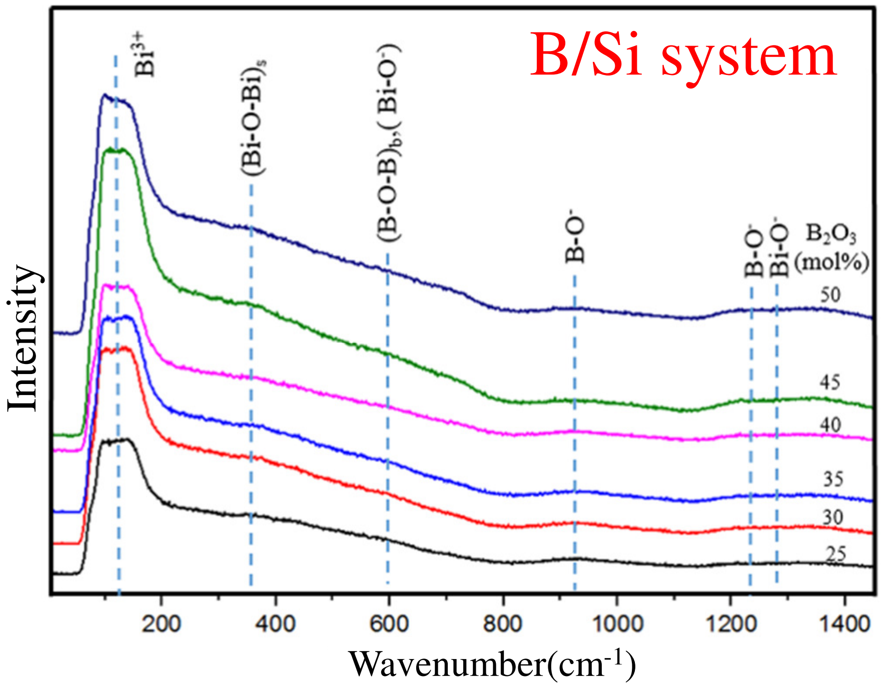
- Bending vibration means a change in the angle but no change in the length between two bonds at molecular vibration.

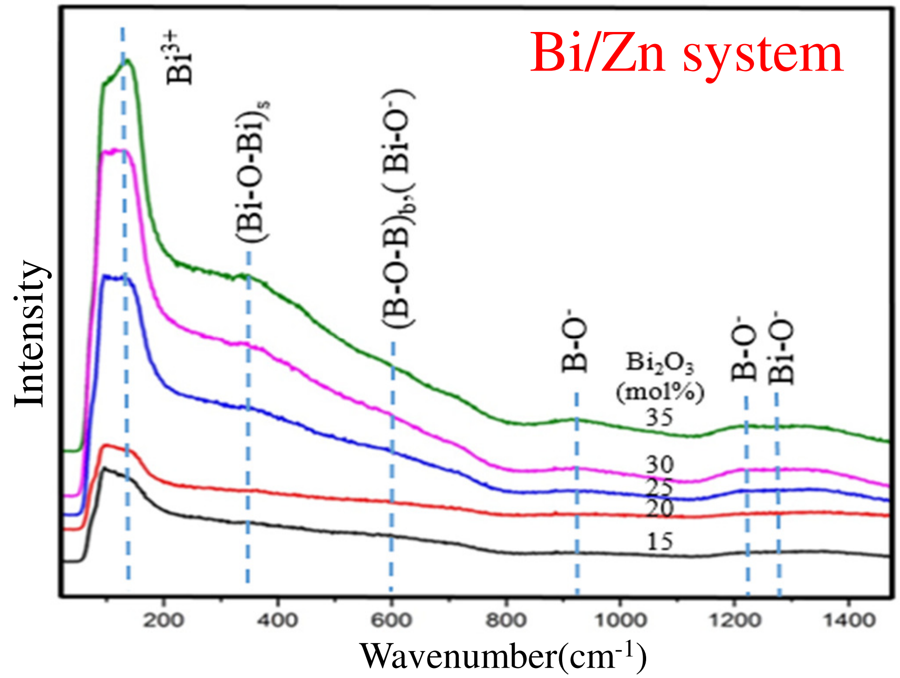
- *Stretching vibration*



- Stretching vibration means the variation in bonding length between two atoms. Atom stretches along bond axis and no change between bond angle.
- Stretching vibrations are divided into two parts: symmetric stretching and asymmetric stretching.

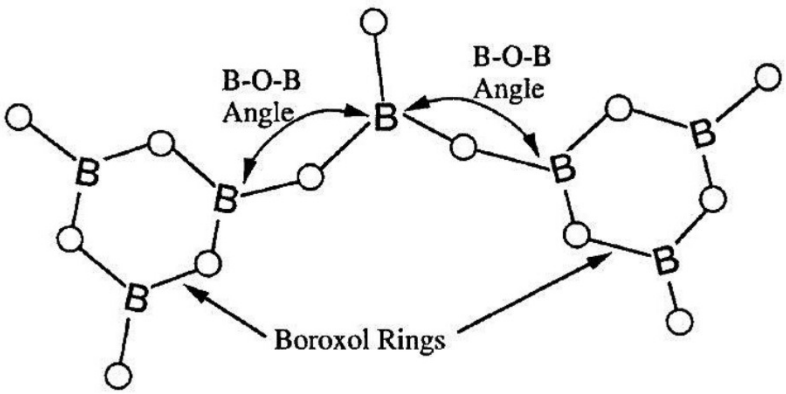
Glass structure Raman spectra



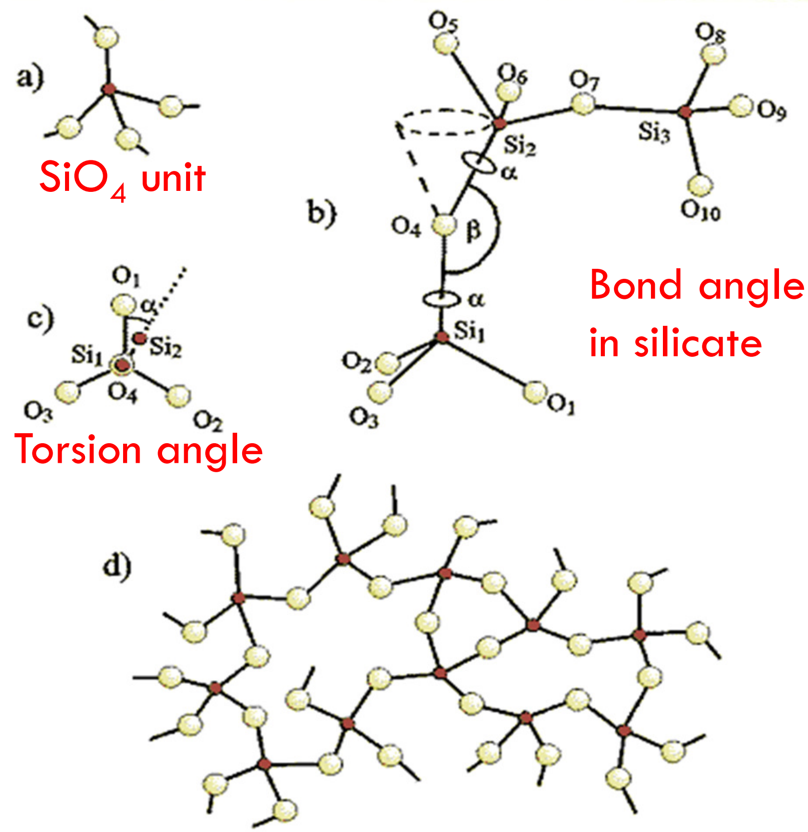


- All bonds become stronger with increasing Bi_2O_3 content.
- $(B\text{-}O^-)_{as}$ and $(Bi\text{-}O^-)_s$ vibrations get stronger with Bi addition, indicating the increase in NBOs.
- The glass structure becomes relaxed when Bi_2O_3 increases.

Structure of Glass systems



*Typical borate structure with boroxol rings.
*Stable $[BO_3]$ plane unit dominated.
*Further stabilization when other oxides are added to form $[BO_4]$ tetrahedron.



SiO_4 unit

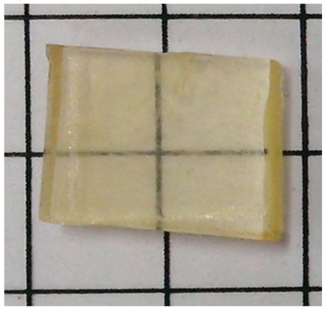
Torsion angle

Bond angle in silicate

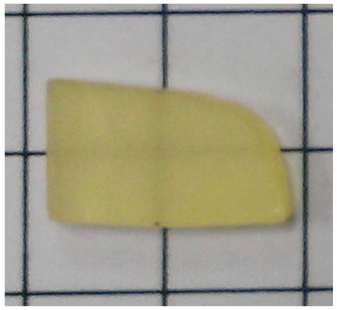
Network of silicate glass

GCP Appearances

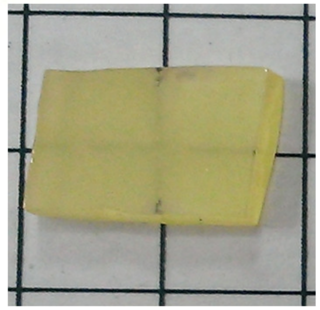
BiZn-borosilicate at 520°C



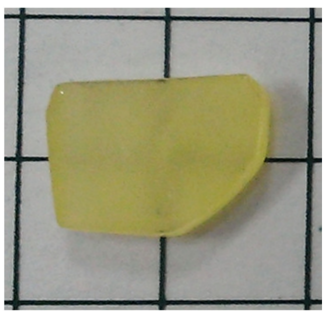
5 min



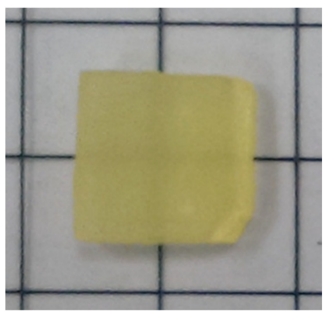
10 min



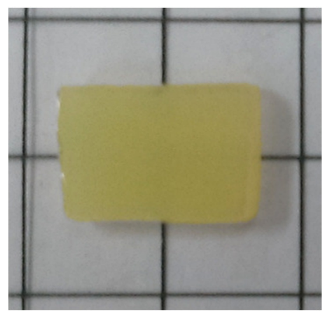
20 min



30 min

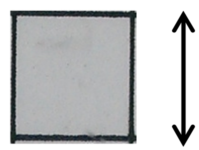


60 min

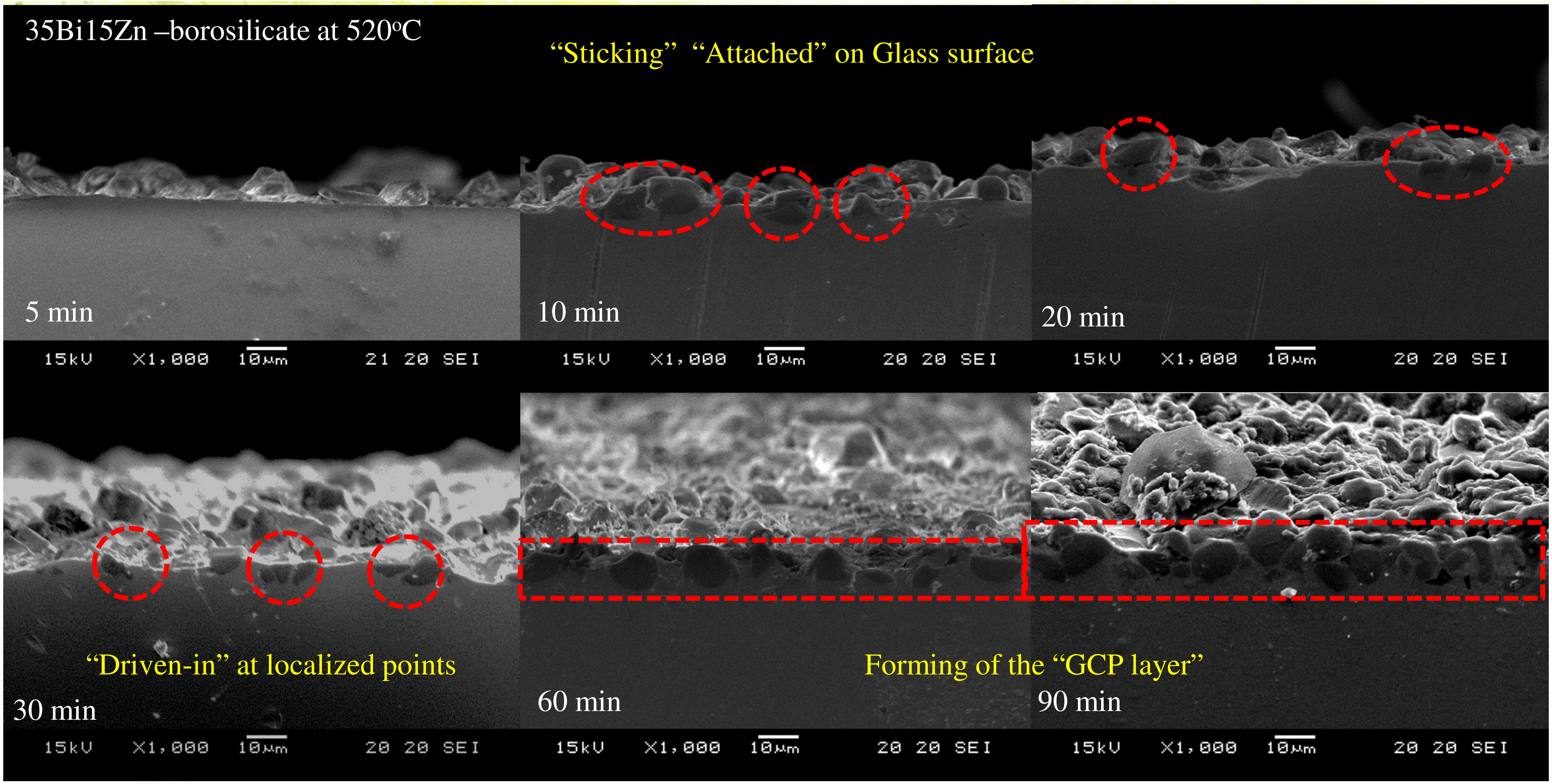


90 min

- The YAG phosphor embedded into the 35Bi15Zn glass substrate at 520°C for various time.
- As heat treatment time increased, the surface of the GCPs become darker yellow.

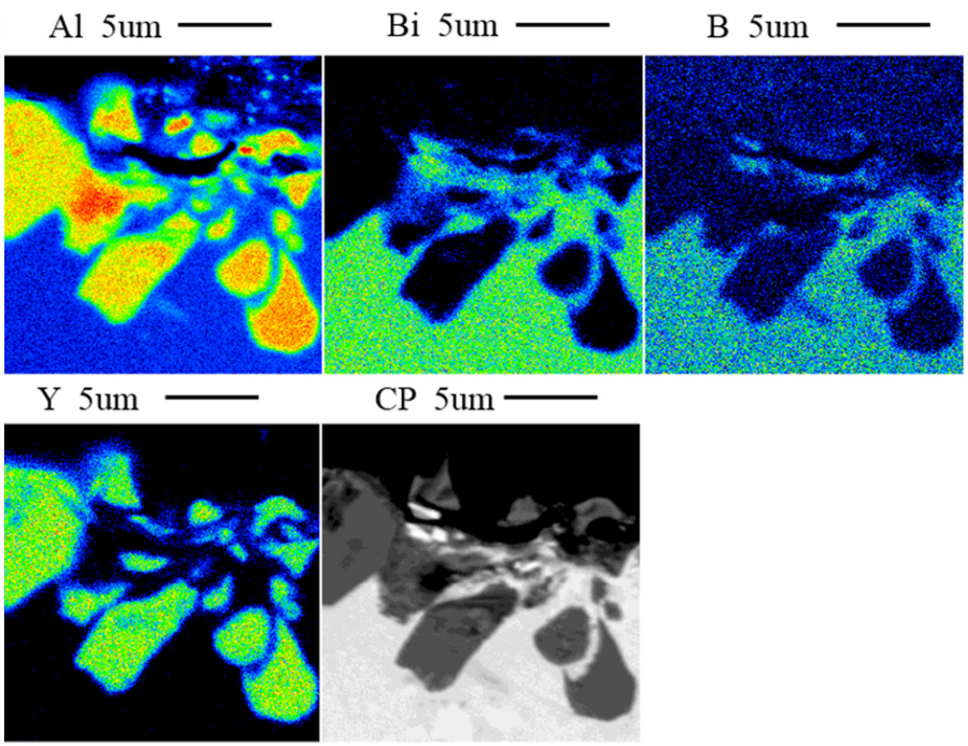


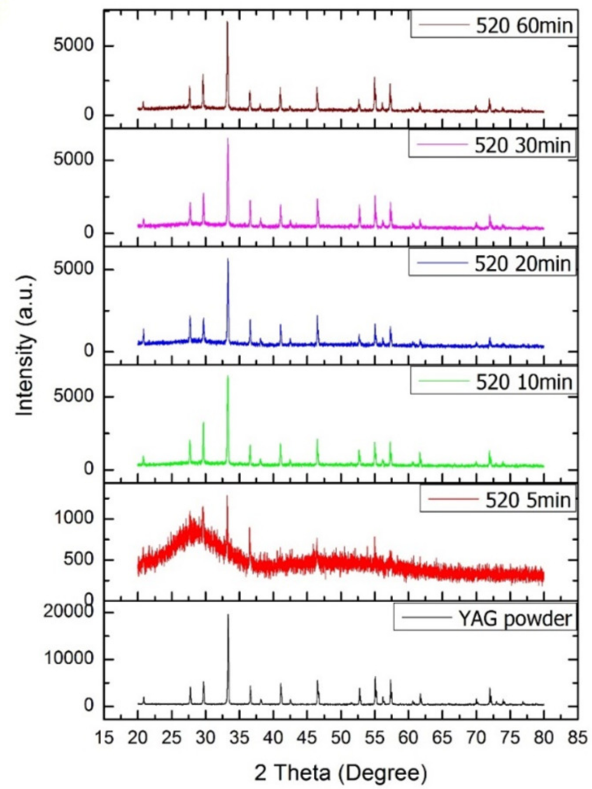
GCP Cross-sectional SEM view



GCP Chemical and Phase Stability

YAG GCP:35Bi15Zn-borosilicate at 520°C for 90 min

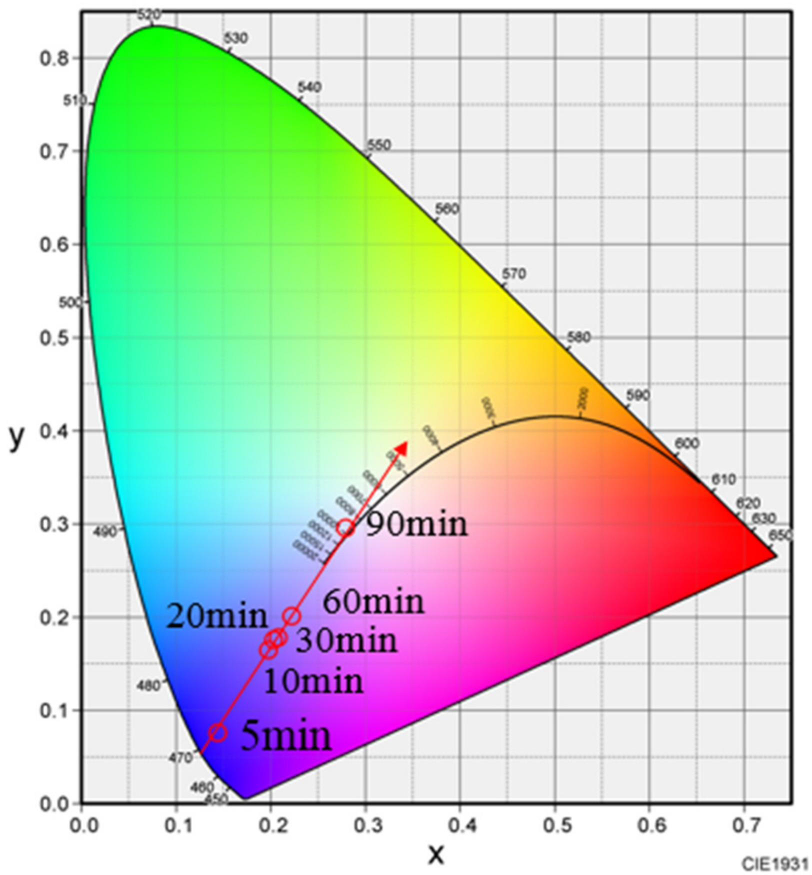




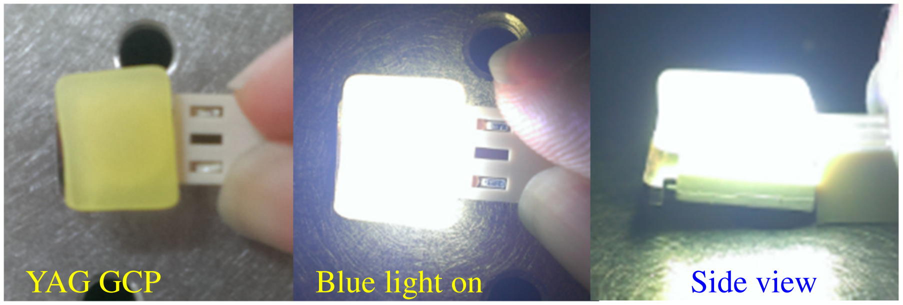
- EPMA color mapping: showing that YAG phosphor and glass substrate are stable without chemical reaction.
- X-ray patterns: showing a stable YAG phase after embedding.

GCP CIE coordinate

YAG GCP:35Bi15Zn-borosilicate at 520°C



35Bi15Zn-borocilicate at 520°C for 90min



- The emission lights were turned from blue, light blue and white according to phosphor driven-in depth and distribution.
- The white light could be reached with the YAG embedded 35Bi15Zn borosilicate GCP and the emission light was adjustable by controlling heat treatment time.

Conclusion Remarks

- The developed borosilicate glasses with functional element incorporation, including Bi, Li, Sr, K, Nb, and Zn, exhibit low characteristic temperature and high refractive index and are applicable in fabricating phosphor embedded glass ceramic layer for Phosphor-Conversion LED lighting system.
- The addition of Bi and Zn induces the relaxation of the borosilicate structure and lower down the characteristic temperatures. Furthermore a large working temperature range of 150°C and above can be obtained.
- The phosphor-embedded glass layer with controllable phosphor distribution can be manipulated through heat treatment temperature and time design associated with the softening temperatures of the glass systems.
- The feasibility of the glass ceramic phosphor, GCP, layer on LED lighting package is verified. White light can be approached using GCP/blue-LED with designed phosphor distributed glass system.

Acknowledgement

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and Thank You!

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