Molten Salt–Assisted Screen Printing of Highly Textured Zn_2SiO_4 Films with Enhanced Deep UV Emission

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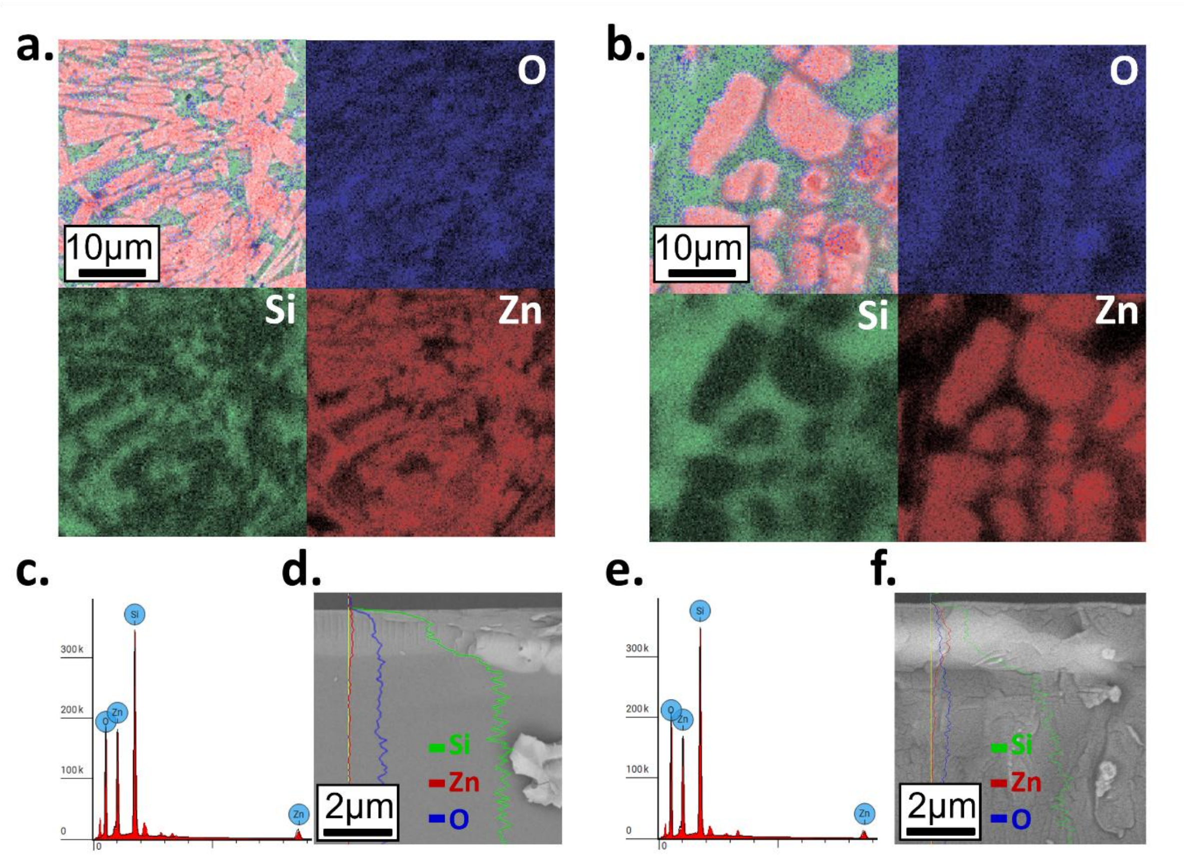

Fig. S1 EDS elemental mapping and line-scan analysis of Zn_2SiO_4 films with 1.5Zn composition: (a, b) surface EDS maps of Zn, Si, and O for films synthesized at 1100 °C and 1200 °C, respectively; (c, e) corresponding EDS spectra; (d, f) cross-sectional SEM images with EDS line scans across the film thickness for samples processed at 1100 °C and 1200 °C, respectively.

S2. Lattice matching analysis

To support the origin of the preferential (300) orientation, a lattice matching analysis was performed between Zn_2SiO_4 and the quartz (0001) substrate.

The d-spacing of the Zn_2SiO_4 (300) reflection was calculated from the XRD peak position (2θ = 22.03°) using Bragg's law:

$$d_{(300)} = \frac{\lambda}{2 \sin \theta}$$

where λ=1.5406 Å is the wavelength of Cu Kα radiation and θ is the Bragg angle, yielding $d_{(300)}$≈4.032 Å, in good agreement with the reference value (4.025 Å, JCPDS card No. 37-1485).

The quartz lattice parameters used were a = 4.913 Å and c = 5.405 Å (JCPDS 46-1045).

A lattice matching condition based on integer multiples of lattice periodicity can be expressed as:

$$4 \times d_{(300)} \approx 3 \times c_{quartz}$$

which gives a mismatch of approximately:

$$f = \frac{4 \times d_{(300)} - 3 \times c_{quartz}}{3 \times c_{quartz}} \times 100 = -0.55\%$$

This low mismatch indicates good lattice correspondence along the direction normal to the quartz (0001) surface, suggesting that the substrate provides a geometrically compatible template for oriented growth. This is consistent with the experimental observation that preferential (300) orientation is present across all compositions and processing conditions.

Raman depth profiling further reveals the formation of a cristobalite interlayer under specific processing conditions. The presence of a continuous interlayer correlates with increased Lotgering factors, indicating improved crystallographic alignment.

Using α-cristobalite lattice parameters (a ≈ 4.969 Å, c ≈ 6.926 Å, JCPDS card No. 39-1425), a representative lattice matching condition:

$$3 \times c_{Zn_2SiO_4} \approx 4 \times c_{cristobalite}$$

yields a mismatch of approximately $f \approx +0.82\%$, indicating improved lattice correspondence along this direction.

Although the lattice mismatch with cristobalite (+0.82%) is slightly higher than that with quartz (-0.55%), the improved crystallographic alignment is consistent with the formation of a continuous cristobalite interlayer, which reflects differences in the arrangement of SiO_4 units in cristobalite compared to quartz and provides a more accommodating interfacial environment for oriented growth.

These results indicate that the quartz substrate supports baseline preferential orientation, while the formation of a cristobalite interlayer enhances the degree of crystallographic alignment under favourable conditions.

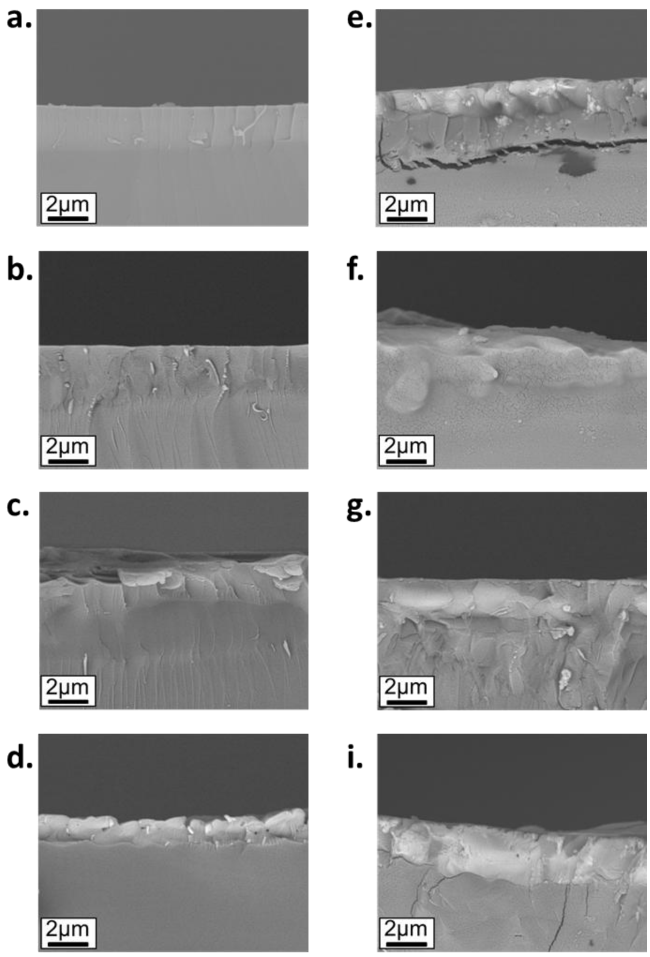

Fig. S3 Cross-sectional SEM micrographs of Zn_2SiO_4 films with varying ZnO concentrations: **(a-d)** films synthesized at 1100 °C for 0.5Zn, 1.0Zn, 1.5Zn, and 2.0Zn compositions, respectively; **(e-h)** corresponding compositions processed at 1200 °C.

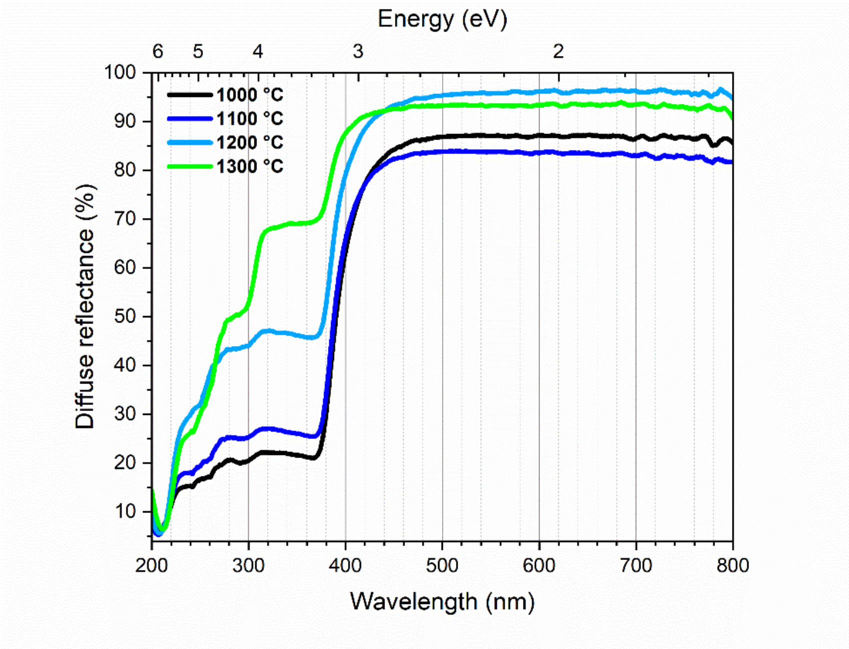

Fig. S4 Diffuse reflection spectra of Zn_2SiO_4 powder synthesized at 1000, 1100, 1200, and 1300 °C via the sol-gel route.

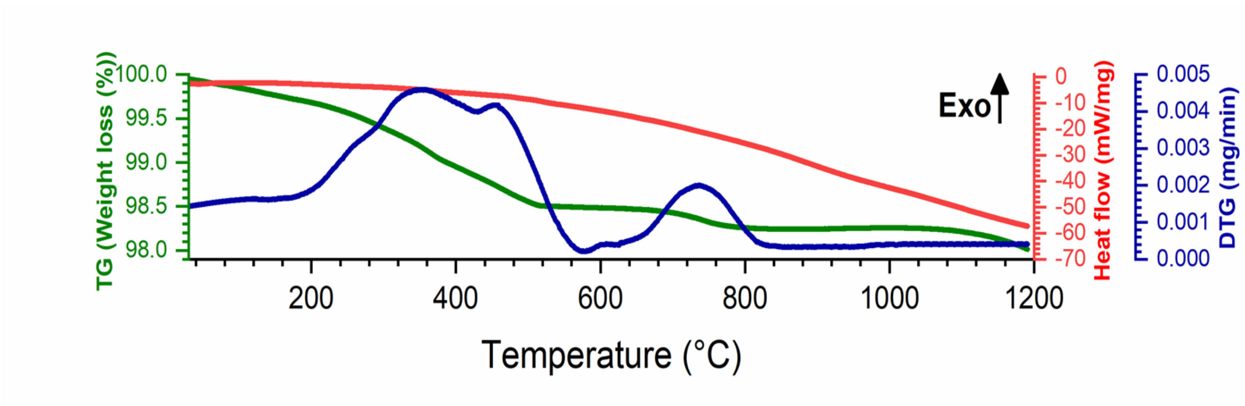

Fig. S5 Thermal analysis of fully crystallized Zn_2SiO_4 powder synthesized at 1300 °C via the molten salt route and subsequently reheated.