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Disorder-Order Transition in Melt-Quenched MOF Glass: Insight from ZIF-4

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Introduction

- Metal-organic frameworks (MOFs) are a new family of glass-formers that adopt networked structures of metal ions connected by organic ligands via coordination bond1-3.
- Zeolitic imidazolate frameworks (ZIFs), a subclass of MOFs that possess high chemical and thermal stability, have attracted increasing attention..

- The majority of MOFs decompose directly upon heating, instead of melting. But a few of MOFs can be vitrified, e.g., ZIF-4, if it can be melted before decomposed1.

- Similarity in structural bonding units between ZIF-4 and SiO2.

Powder XRD

No Bragg peaks occur, hence there is no long-range order, but nano-ordered domains might appear.

Raman

- There are no peaks for ZIF-4 glass.
- Raman peaks occur for the heat-treated ZIF-4 glass.
- These confirm that the crystals appear at nano-scale, which are larger than medium-range order structure.

- The FWHM decrease with annealing temperature.
- This implies an increase of the degree of order and the ordered domain size.

Liquid-state NMR

- Local environment around the 13C is almost the same.
- 1H resonances of crystal, glass and “recrystallized” sample appear at different chemical shifts.
- These indicate that heat-treatment causes the structural change.

Conclusion

- Disorder-order transition in ZIF-4 glass is discovered.
- After heat treatment, some ordered domains are formed. However, ZIF-4 glass is still in amorphous state.
- The mechanism of this disorder-order transition remains elusive.
- New techniques should be applied to characterize structural features during the transition, such as neutron scattering.

Why neutron scattering?

Disorder-order transition in ZIF-4 glass has been studied by performing heat-treatments and structural analyses. Neutron techniques are expected to help to answer these questions:
- At which length scale the order-disorder transition occur in ZIF-4 glass?
- Does the heat treatment induce change of coordination bonds (Zn-N) in terms of bonding length and angle, and recovery of the distorted network?