

Supplement

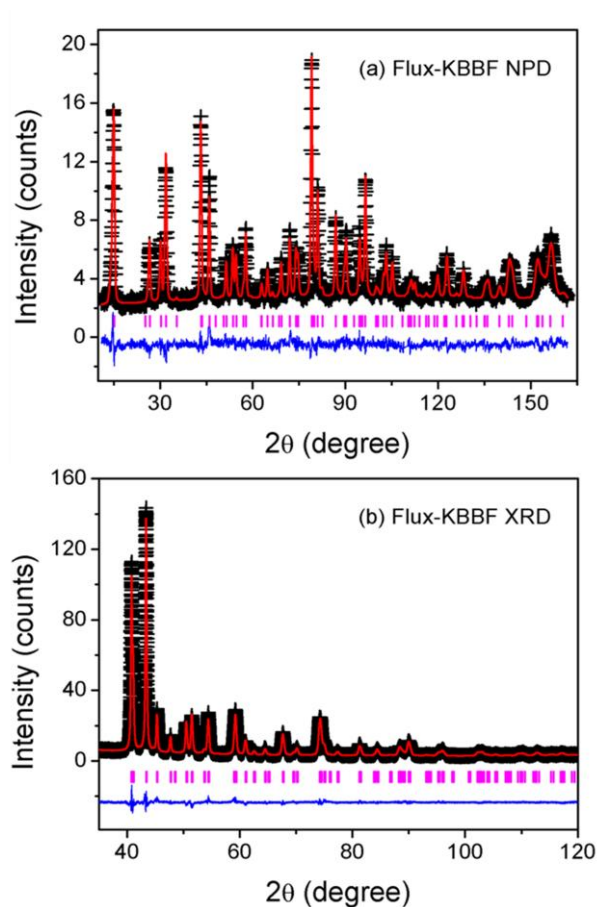


Figure S1. Combined Rietveld analysis of NPD (a) and XRD (b) for flux-KBBF powder. Plus (+) marks represent the experimental results and the solid line is from refinement. The ticks indicate the expected positions for structure *R32*. The difference between experimental and calculated results is shown beneath.

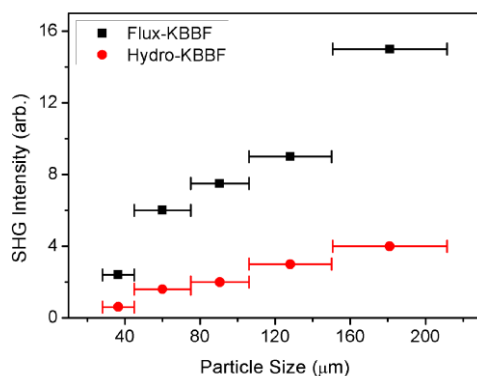


Figure S2. Powder SHG intensities of flux-KBBF and hydro-KBBF

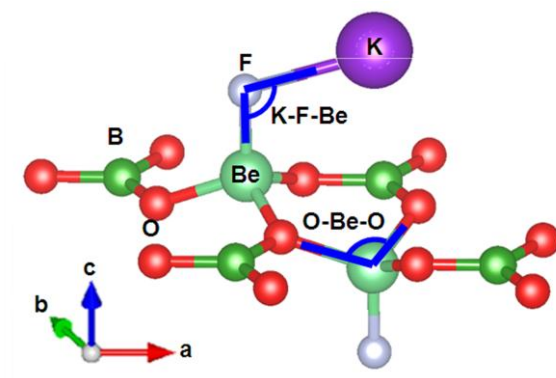


Figure S3. The sketch of the geometric relation between bonds and angles in layer $(\text{Be}_2\text{BO}_3\text{F}_2)_\infty$