Supporting Information

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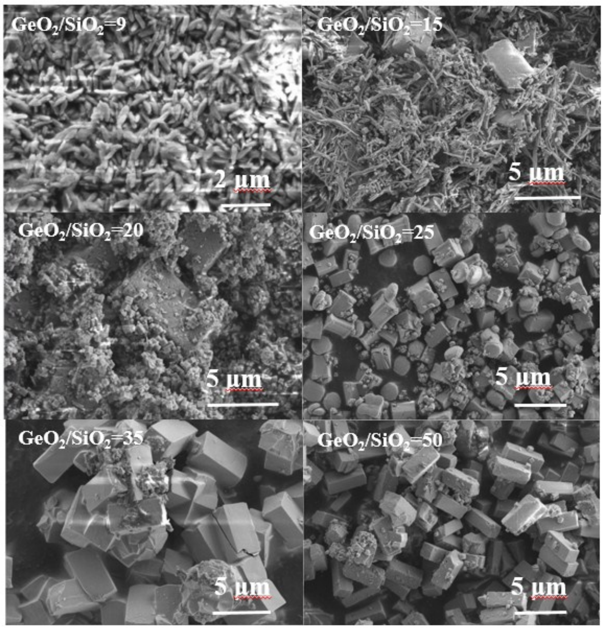

Fig. S1a SEM images of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 130℃. The sample with GeO_2/SiO_2=9 has regular ITQ-17 morphology (rod-like morphology) and the crystal size is less than 2μm. The sample with GeO_2/SiO_2=15 has two morphologies (except for little impurity), meaning a mixture of ITQ-17 and BUCT-1. However, the samples with GeO_2/SiO_2=20-50 have large cuboid morphology (except for little impurity) and the crystal sizes of all these samples are almost 5 μm.

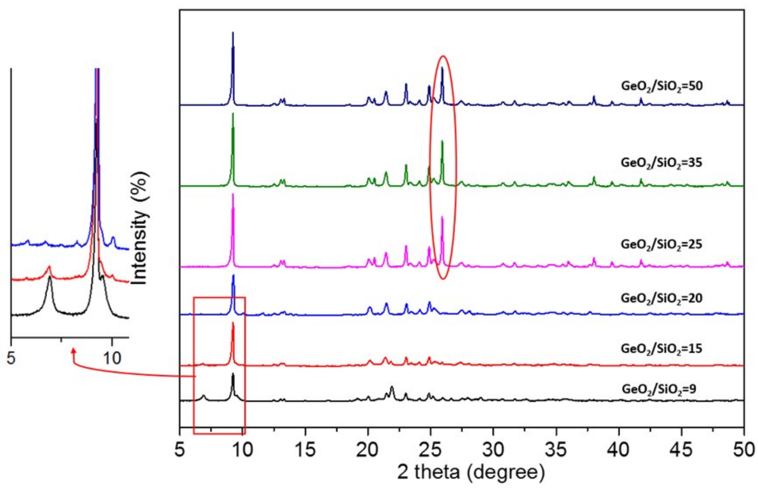

Fig. S1b XRD patterns of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 130℃. The sample with GeO_2/SiO_2=9 has typical ITQ-17 peaks. The pattern of sample with GeO_2/SiO_2=15 is magnified to see clearly and there is a small peak at 2θ=6.78°, belonging to ITQ-17. The other four samples (with GeO_2/SiO_2=20-50) have typical patterns of BUCT-1 except for the obvious impurity peaks (marked) of α-quartz germanium oxide.

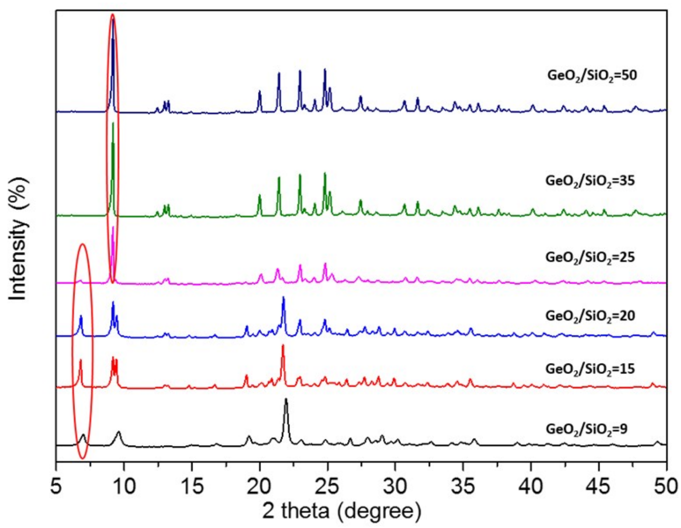

Fig. S2 XRD patterns of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 140℃. The samples with GeO_2/SiO_2=9-25 all have ITQ-17 peaks before 2θ=10° (marked), though the first peak of sample with GeO_2/SiO_2=25 is very weak. The samples with GeO_2/SiO_2=25-50 all have BUCT-1 peaks at 2θ=9.17°, which means they all contain BUCT-1 phase.

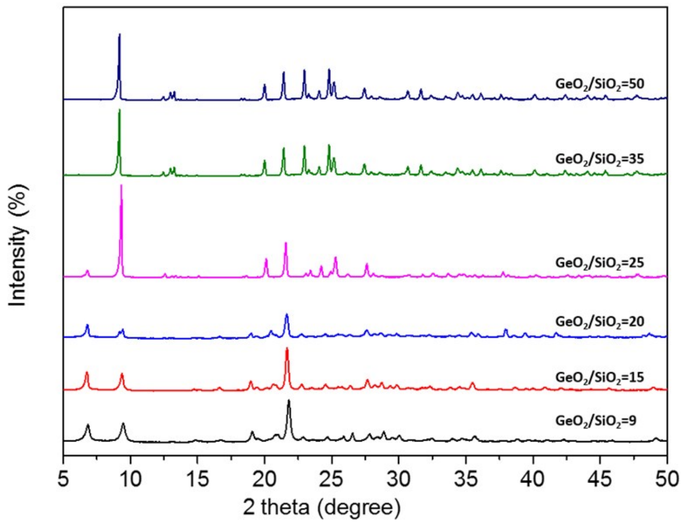

Fig. S3 XRD patterns of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 150℃. The samples with GeO_2/SiO_2=9-20 all have pure ITQ-17 peaks and the samples with GeO_2/SiO_2=35-50 have pure BUCT-1 peaks. However, the sample with GeO_2/SiO_2=25 has characteristic peaks of both ITQ-17 and BUCT-1.

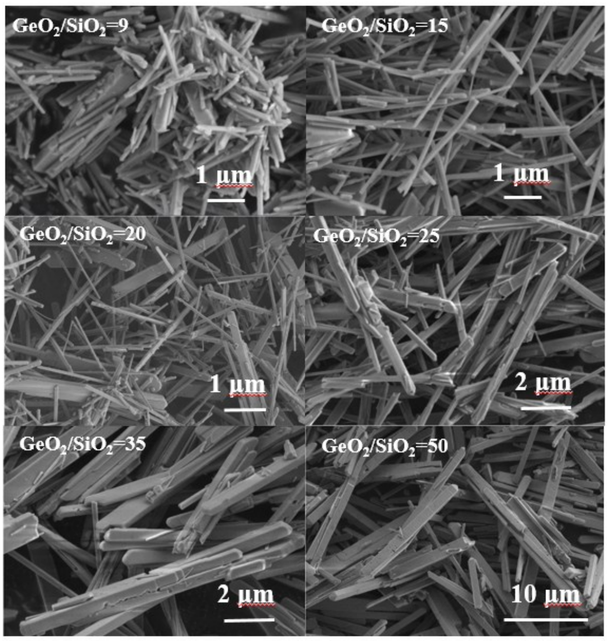

Fig. S4a SEM images of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 170℃. All the samples have rod-like morphology, so it's difficult to distinguish ITQ-17 from α-quartz germanium oxide.

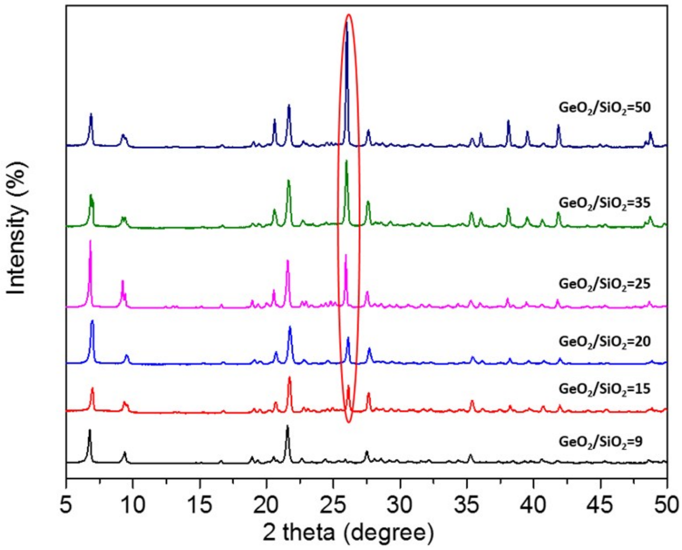

Fig. S4b XRD patterns of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 170℃. Even though all the samples have typical ITQ-17 peaks, only the sample with GeO_2/SiO_2=9 is pure. Moreover, the intensity of the α-quartz germanium oxide impurity (marked) increases with the increase of GeO_2/SiO_2 ratio.

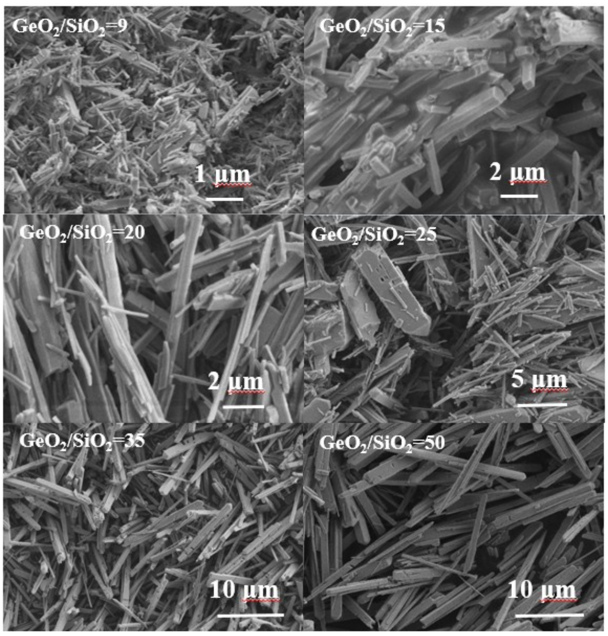

Fig. S5a SEM images of samples with the same H_2O/TO_2 ratio (=1.5) but different

GeO_2/SiO_2 ratios at 180℃. All the samples have rod-like morphology, so it's difficult to distinguish ITQ-17 from α-quartz germanium oxide.

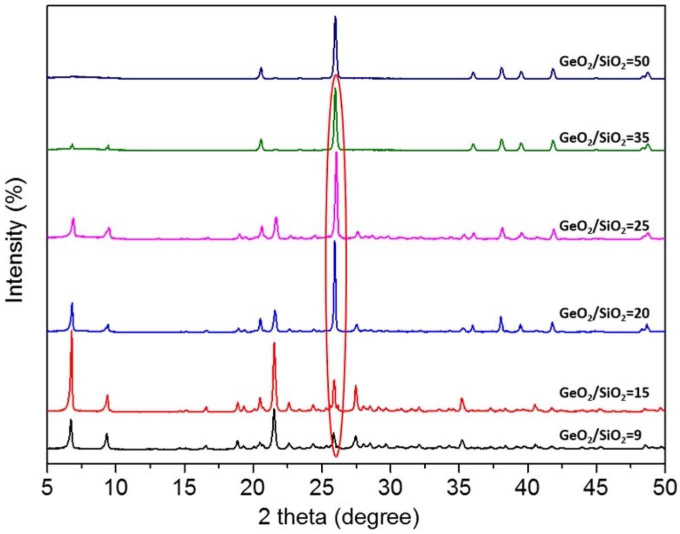

Fig. S5b XRD patterns of samples with the same H_2O/TO_2 ratio (=1.5) but different GeO_2/SiO_2 ratios at 180℃. The samples with GeO_2/SiO_2=9-35 have typical ITQ-17 peaks but they also have the peaks of α-quartz germanium oxide impurity (marked) and the intensity of the impurity increases with the increase of GeO_2/SiO_2 ratio. The sample with GeO_2/SiO_2=50 has pure α-quartz type germanium oxide peaks.

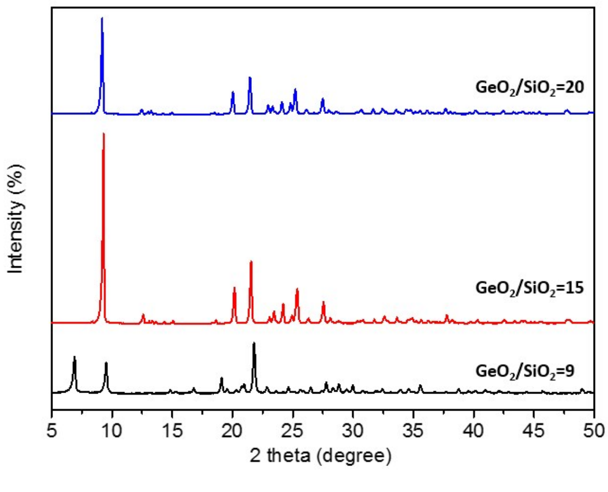

Fig. S6 XRD patterns of samples with the same H_2O/TO_2 ratio (=3) but different

GeO_2/SiO_2 ratios at 120℃. The sample with GeO_2/SiO_2=9 has typical ITQ-17 peaks. However, the other two samples (with GeO_2/SiO_2=15 and 20) have BUCT-1 pattern. The yield of the sample with GeO_2/SiO_2=25 was too low to test.

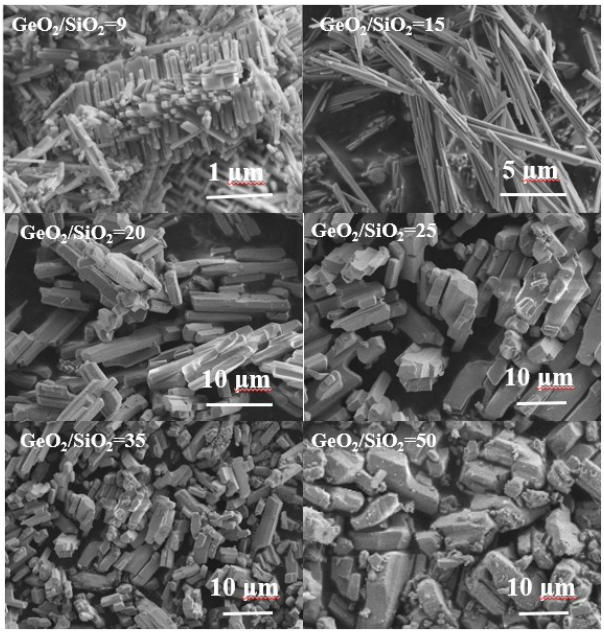

Fig. S7a SEM images of samples with the same H_2O/TO_2 ratio (=3) but different GeO_2/SiO_2 ratios at 130℃. Sample with GeO_2/SiO_2=9 and15 have regular pure ITQ-17 morphology (rod-like morphology) but the crystal size of the former (1 μm) is smaller than the latter (5 μm). For samples with GeO_2/SiO_2 =20-50, they all have cuboid morphology for BUCT-1.

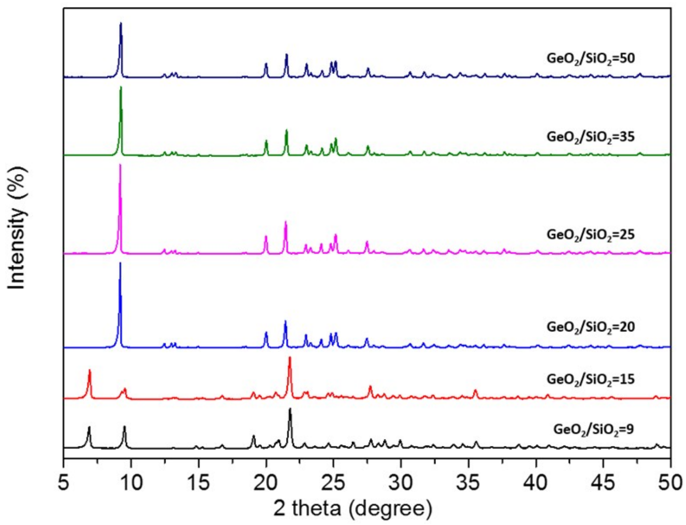

Fig. S7b XRD patterns of samples with the same H_2O/TO_2 ratio (=3) but different GeO_2/SiO_2 ratios at 130℃. The sample with GeO_2/SiO_2=9 and 15 have typical ITQ-17 peaks. However, the other four samples (with GeO_2/SiO_2=20-50) have typical BUCT-1 peaks. In addition, the impurity peaks of germanium oxide is too weak to be distinguished for the samples with GeO_2/SiO_2=15 and 20.

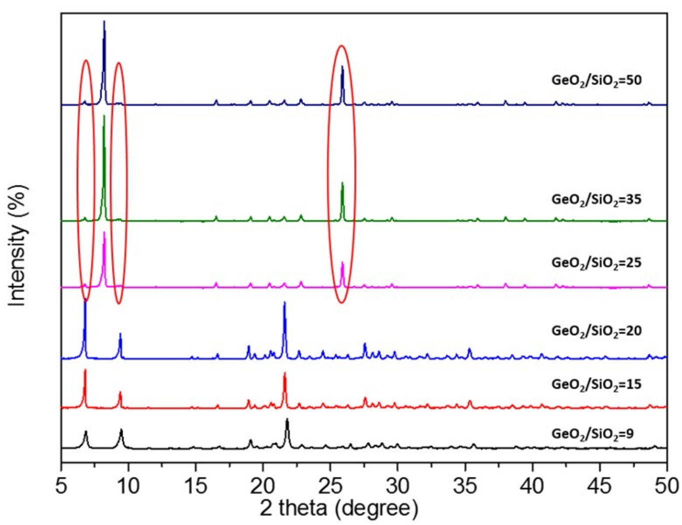

Fig. S8 XRD patterns of samples with the same H_2O/TO_2 ratio (=3) but different GeO_2/SiO_2 ratios at 160℃. The samples with GeO_2/SiO_2=9-20 have typical ITQ-17 peaks. However, the samples with GeO_2/SiO_2=25-50 have peaks of both ITQ-17 and

BUCT-2, though their peaks at 2θ=6.8° and 9.4° (ITQ-17) are very weak. Besides, there are obvious impurity peaks of α-quartz germanium oxide for samples with GeO_2/SiO_2=25-50.

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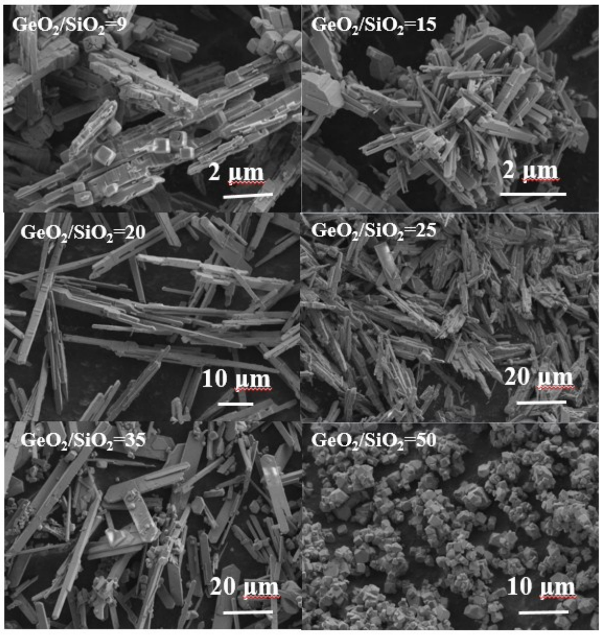

Fig. S9a SEM images of samples with the same H_2O/TO_2 ratio (=3) but different GeO_2/SiO_2 ratios at 170℃. The samples with GeO_2/SiO_2=9 and 15 have typical ITQ-17 morphology (rod-like morphology) with α-quartz germanium oxide impurities. The samples with GeO_2/SiO_2=20-35 have both rod-like and flake-like morphologies, belonging to ITQ-17 and BUCT-2 respectively, except for germanium oxide impurities. The sample with GeO_2/SiO_2=50 has the morphology of pure α-quartz germanium oxide.

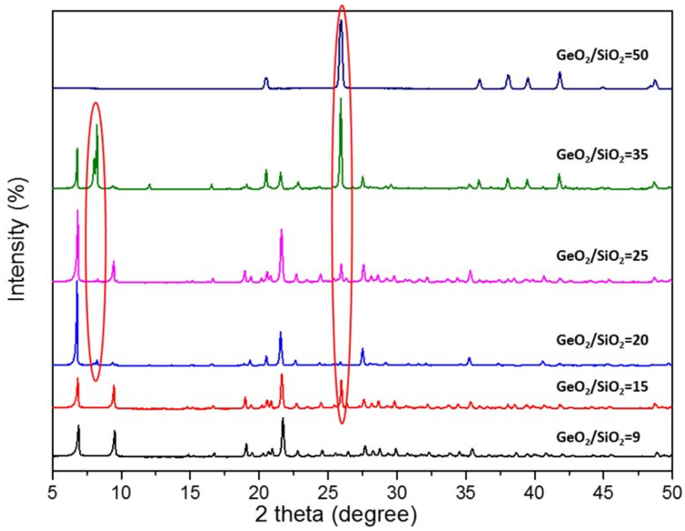

Fig. S9b XRD patterns of samples with the same H_2O/TO_2 ratio (=3) but different GeO_2/SiO_2 ratios at 170℃. The samples with GeO_2/SiO_2=9-15 have typical ITQ-17 peaks and the samples with GeO_2/SiO_2=25-50 have peaks of both ITQ-17 and BUCT-2 (marked). However, except for the samples with GeO_2/SiO_2=9, the rest all have α-quartz germanium oxide impurities (marked). Besides, the samples with GeO_2/SiO_2=50 is pure α-quartz germanium oxide.

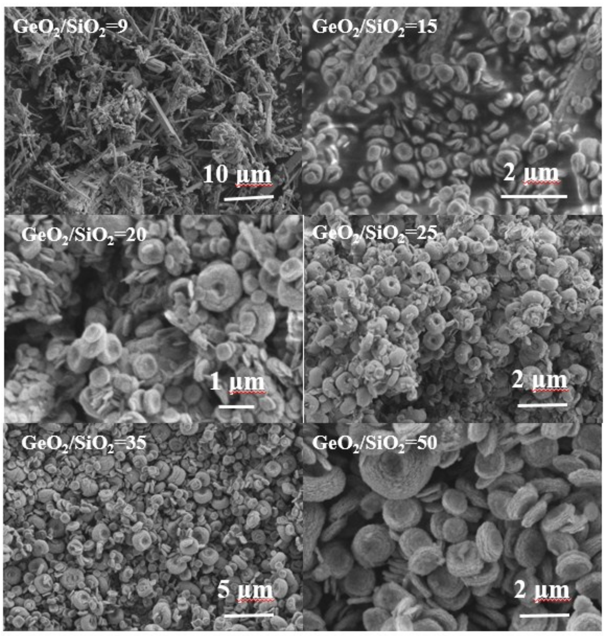

Fig. S10a SEM images of samples with the same H_2O/TO_2 ratio (=3) but different

GeO_2/SiO_2 ratios at 180℃. The sample with GeO_2/SiO_2=9 has two morphologies of both rod-like and flake-like, belonging to ITQ-17 and BUCT-2 respectively. It also has impurities with cubic morphology for α-quartz germanium oxide and bird nest morphology for germanium oxide (argutite). The sample with GeO_2/SiO_2=15 has little ITQ-17 and lots of impurities with bird nest morphology. The rest samples all have pure bird nest morphologies.

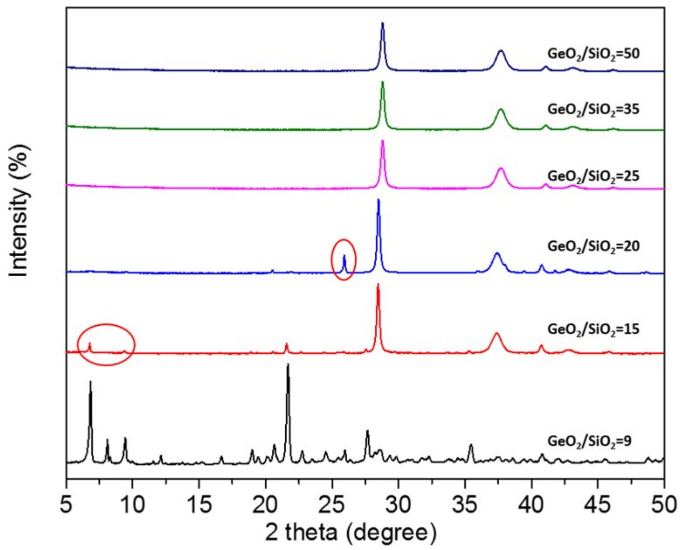

Fig. S10b XRD patterns of samples with the same H_2O/TO_2 ratio (=3) but different GeO_2/SiO_2 ratios at 180℃. The sample with GeO_2/SiO_2=9 has both ITQ-17 and BUCT-2 peaks, but the impurity peaks are hard to be distinguished. The sample with GeO_2/SiO_2=15 has very weak peaks of ITQ-17 (marked) but strong peaks of germanium oxide (argutite). What is interesting is that the sample with GeO_2/SiO_2=15 has two kinds of germanium oxide peaks, though the α-quartz type is very weak. The rest of samples are all pure germanium oxide (argutite).