Supplementary Information

Moderate-temperature fabrication of $BaZrS_3$ thin films via dithiocarbamate-based solution processing and oxygen-sink boron sulfurization

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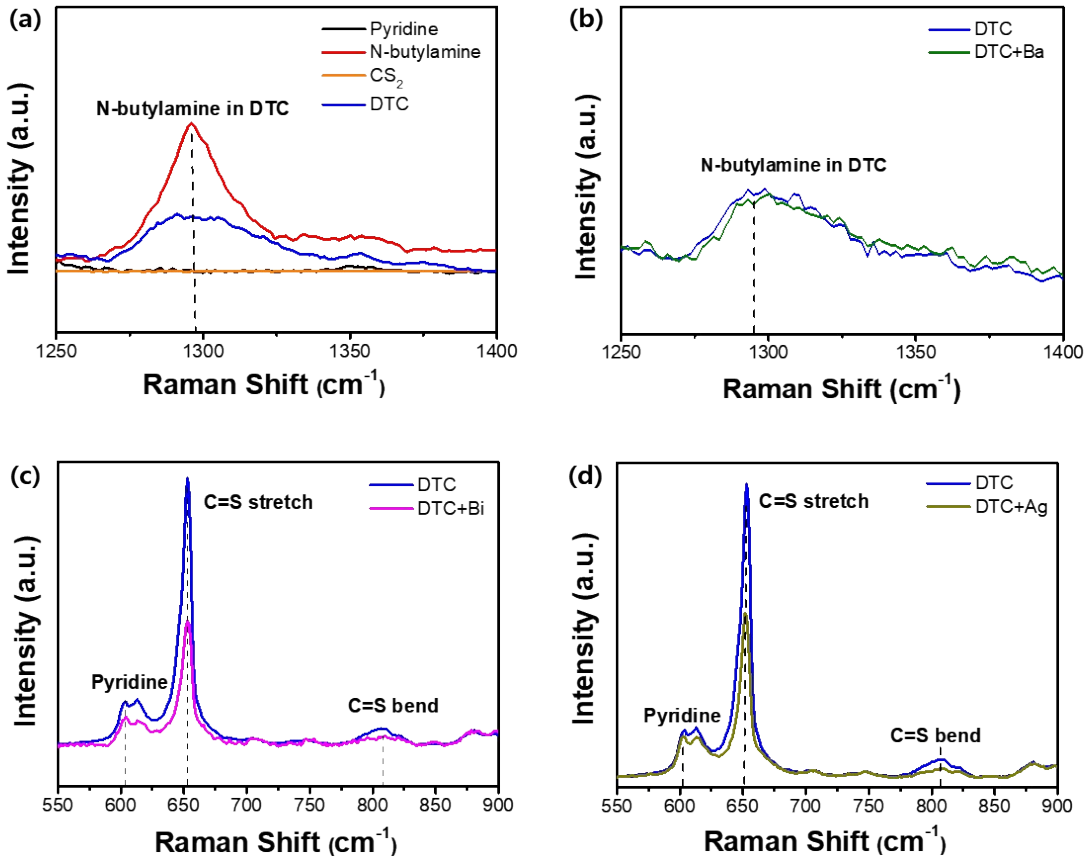

Fig. S1. Liquid Raman spectroscopy (Raman spectra) of solutions. (a) Raman spectra of pure *N*-butylamine, Pyridine, CS_2, and DTC. (b) Raman spectra of DTC+Ba solution compared with DTC in range of 1250-1400 cm^{-1}. (c) Raman spectra of DTC+Bi solution compared with DTC in range of 550-900 cm^{-1}. (d) Raman spectra of DTC+Ag solution compared with DTC in range of 550-900 cm^{-1}.

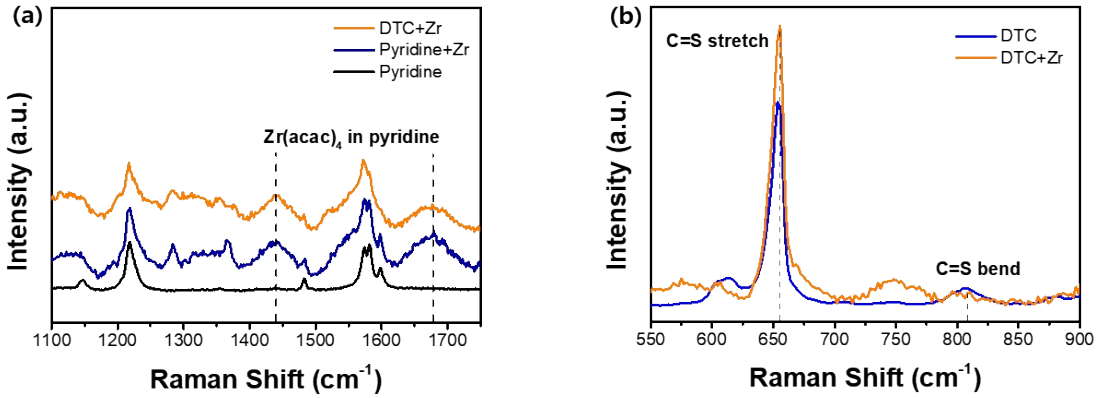

Fig. S2. Raman spectra in Zr-DTC solution. (a) Raman spectra of DTC+Zr solution compared with pyridine and pyridine+Zr solution. (b) Raman spectra of DTC+Zr solution compared with DTC solution.

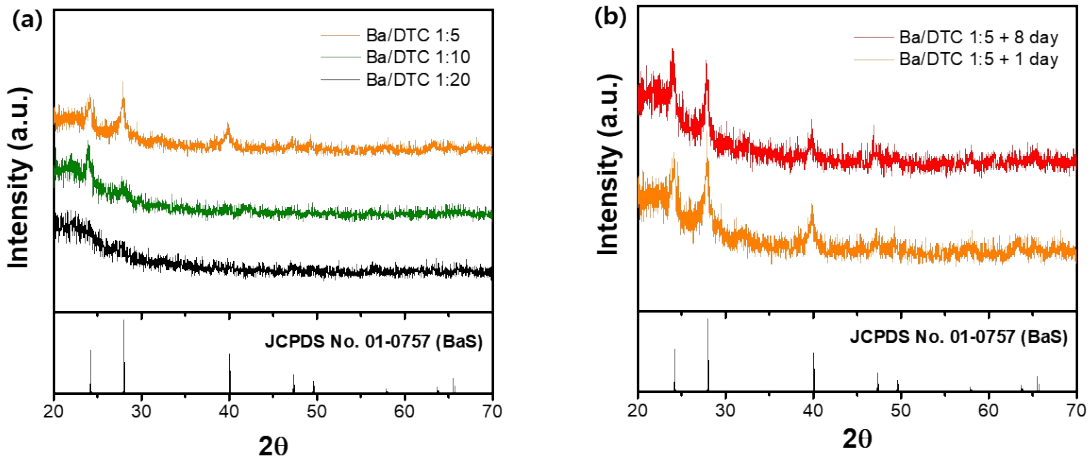

Fig. S3. X-ray diffraction (XRD) patterns of BaS films with different Ba/DTC ratios and different aging times. (a) XRD patterns of BaS films with different Ba/DTC ratios (1:5, 1:10, and 1:20). (b) XRD patterns of BaS films from two Ba-DTC solutions with different aging times (1 day and 8 days).

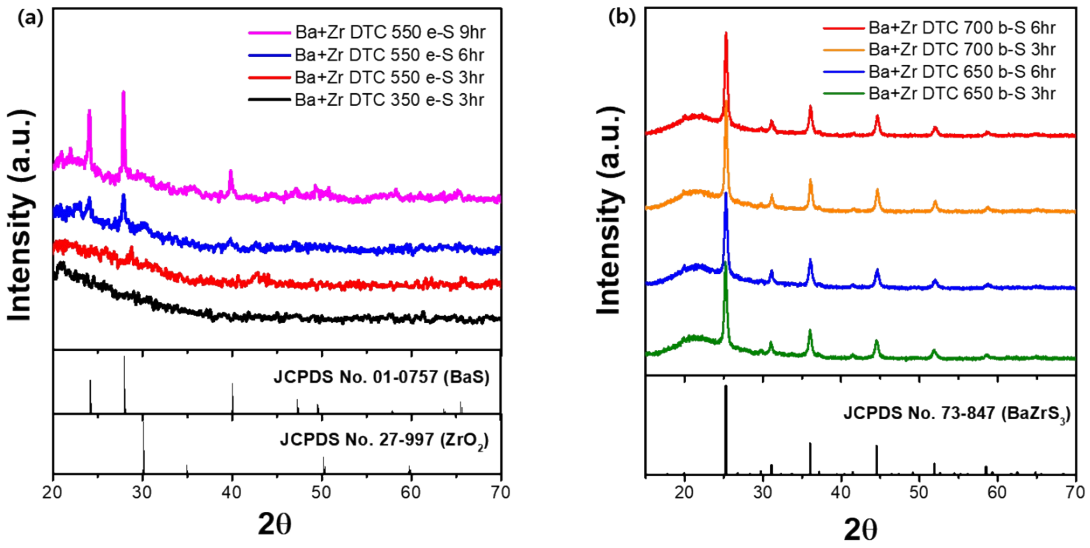

Fig. S4. X-ray diffraction (XRD) patterns of Ba+Zr DTC film sulfurized with two methods (e-S and b-S). (a) XRD patterns of Ba+Zr DTC film sulfurized with e-S method at different temperatures (350 and 550 ℃) and different sulfurization times (3hr, 6hr, and 9hr). (b) XRD patterns of Ba+Zr DTC film sulfurized with b-S method at different temperatures (650 and 700 ℃) and different sulfurization times (3hr and 6hr).

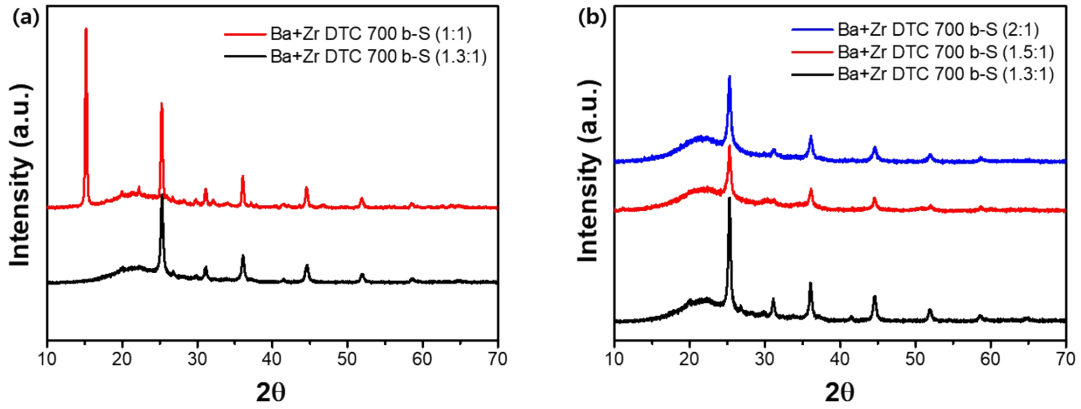

Fig. S5. X-ray diffraction (XRD) patterns of Ba+Zr DTC film sulfurized at 700 ℃ boron sulfurization method from various Ba/Zr ratio solutions. (a) XRD patterns of two samples with different Ba/Zr ratios (Ba:Zr = 1:1 and 1.3:1). (b) XRD patterns of three samples with different Ba/Zr ratios (Ba:Zr = 1.3:1, 1.5:1, and 2:1).

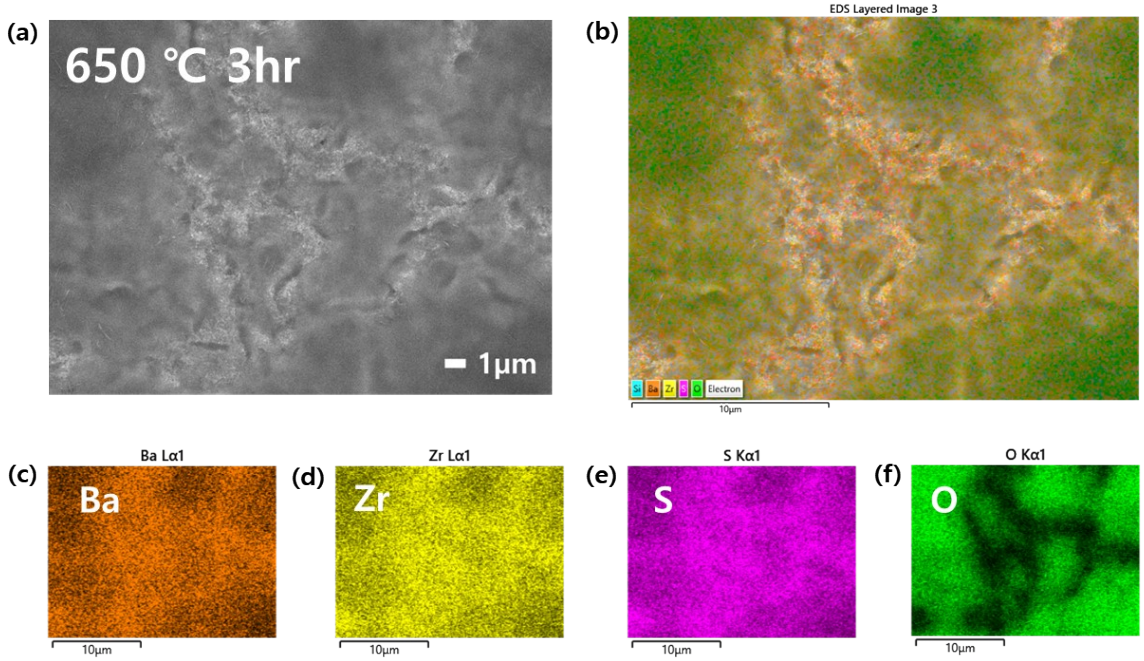

Fig. S6. Top-view scanning electron microscopy (SEM) image of $BaZrS_3$ thin film sulfurized at 650 ℃ (3hr) with boron sulfurization method. (a) Surface SEM images of $BaZrS_3$ thin film. (b) Combined energy dispersive X-ray (EDX) mapping images of Ba, Zr, S, and O. (c-f) EDX mapping of $BaZrS_3$ thin films with Ba, Zr, S, and O components in order.

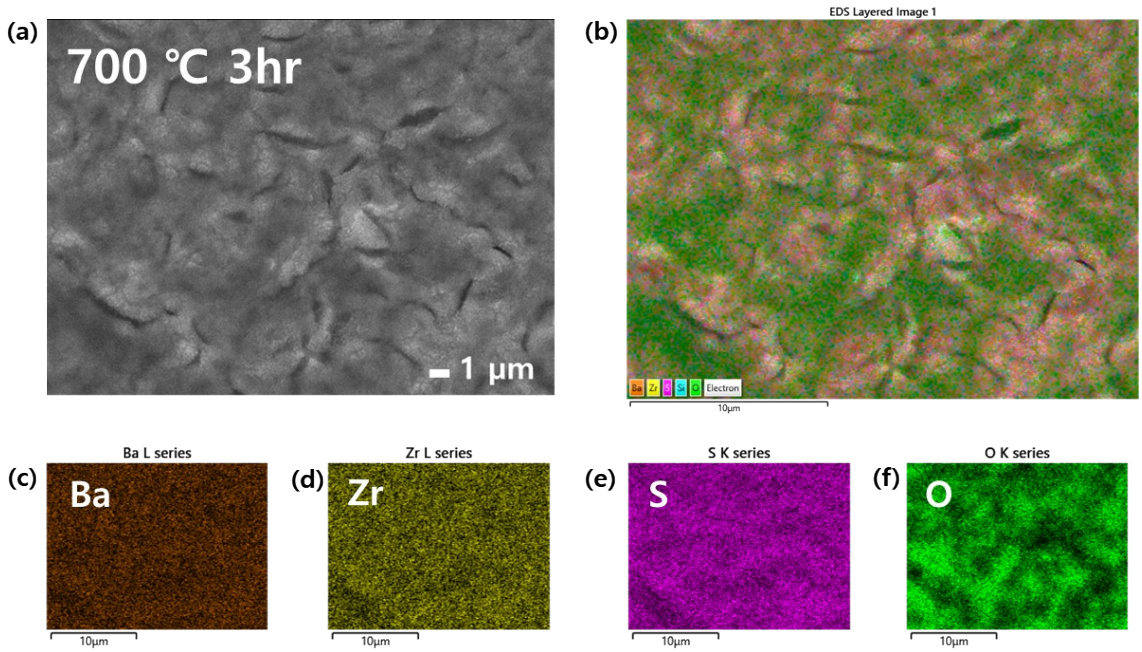

Fig. S7. Top-view scanning electron microscopy (SEM) image of $BaZrS_3$ thin film sulfurized at 700 ℃ (3hr) with boron sulfurization method. (a) Surface SEM images of $BaZrS_3$ thin film. (b) Combined energy dispersive X-ray (EDX) mapping images of Ba, Zr, S, and O. (c-f) EDX mapping of $BaZrS_3$ thin films with Ba, Zr, S, and O components in order.

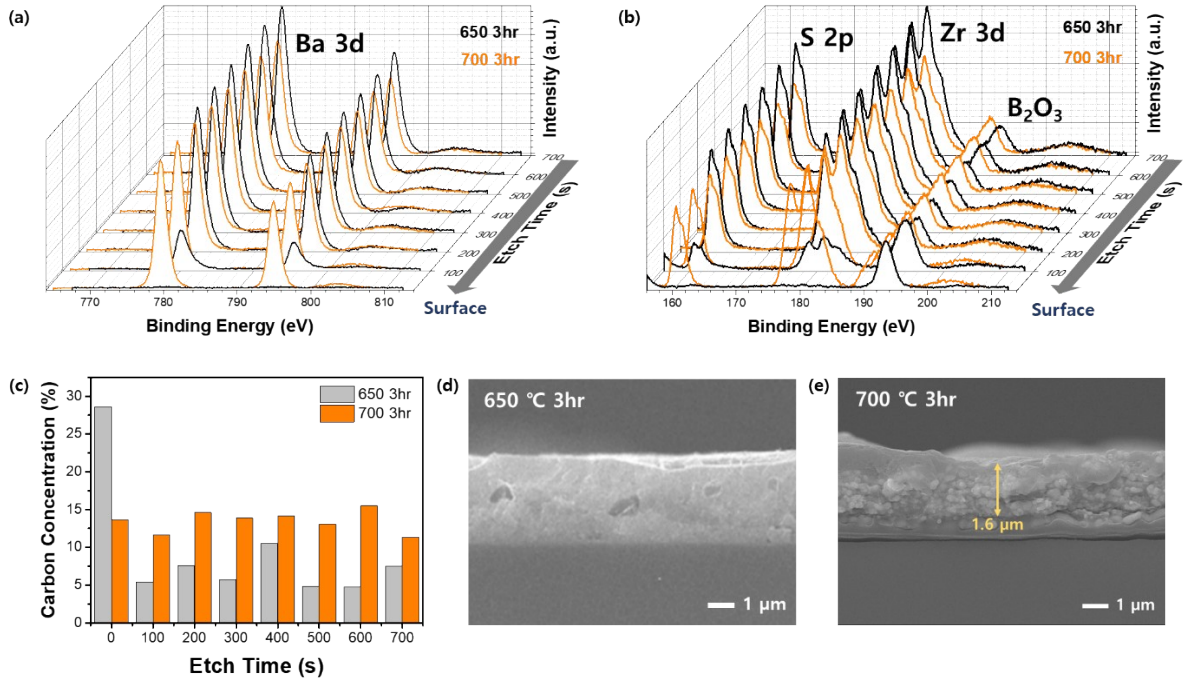

Fig. S8. Elemental distributions and concentration at various etch time with cross-sectional scanning electron microscopy (SEM) images of two samples. (a) Distributions of Ba 3d signals in depth-profile X-ray photoelectron spectroscopy (XPS) for two temperature conditions (650 and 700 ℃). (b) Distributions of S 2p, Zr 3d, and B_2O_3 signals in depth-profile XPS for two temperature conditions (650 and 700 ℃). (c) Carbon concentrations (%) at various etch times for two temperature conditions (650 and 700 ℃). (d-e) Cross-sectional SEM images of $BaZrS_3$ samples for two temperature conditions (650 and 700 ℃).

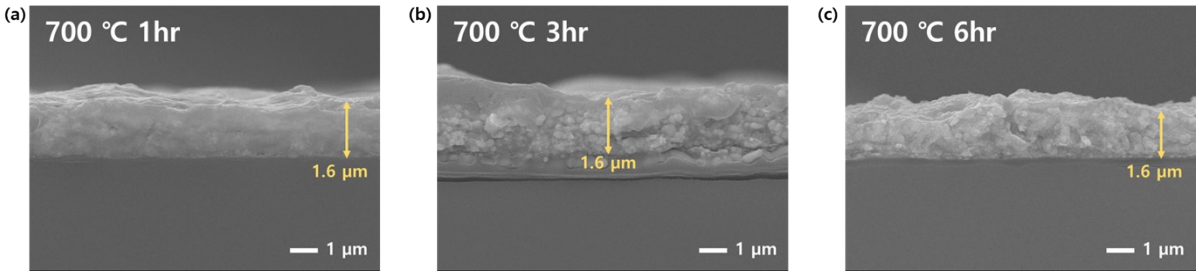

Fig. S9. Cross-sectional scanning electron microscopy (SEM) images of $BaZrS_3$ thin films at 700 ℃ (a) 1hr, (b) 3hr, and (c) 6hr with boron sulfurization method.