Electronic Supplementary Information

One-Pot synthesis polyoxomolybdates anionic intercalated layered double hydroxides and their application in ultra-deep desulfurization of fuels at mild condition

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Single-cryatal X-ray diffraction data for compounds using an Oxford Diffraction Gemini R Ultra diffractometer with graphite-monochromated Mo-K*a* radiation (λ = 0.71073 Å) by using ω–scan technique. The structure were solved and refined by full-matrix least squares on F2 using the SHELXL-2017 program.[1] Hydrogen atoms on C and N atoms using the theory of hydrogenation method and refined using the O-H distance restrained to a target value of 0.85Å. The entry of CCDC–1830449 contains the supplementary crystallographic data for the compounds. H atoms are omitted for clarity.

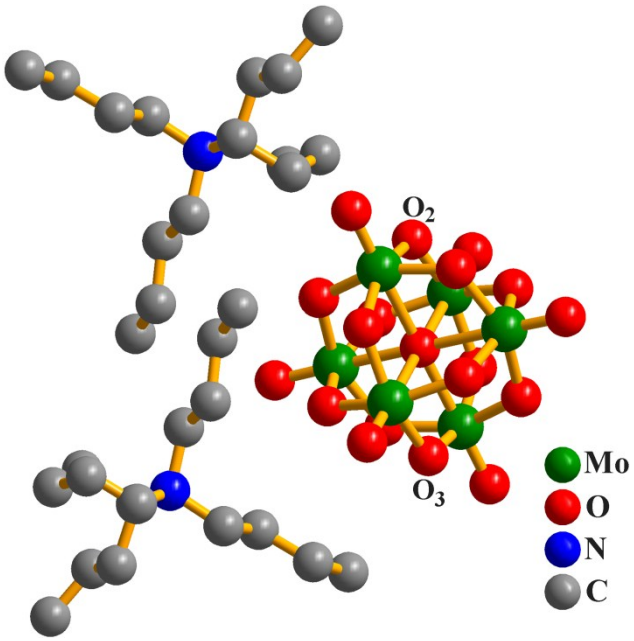

Fig.S1 The crystal structure of the $[(n\text{-}C_4H_9)_4N]_2[Mo_6O_{19}]$.

Table.S1 The crystal bond length of the $[(n\text{-}C_4H_9)_4N]_2[Mo_6O_{19}]$.

Mo(1)-O(16)	1.680(5)
Mo(2)-O(15)	1.679(4)
Mo(3)-O(19)	1.689(5)
Mo(4)-O(8)	1.917(4)
Mo(5)-O(17)	1.924(5)
Mo(6)-O(18)	1.688(5)
N(1)-C(16)	1.513(9)
N(2)-C(43)	1.516(10)

Table.S2 The crystal bond angle of $[(n\text{-}C_4H_9)_4N]_2[Mo_6O_{19}]$.

O(16)-Mo(1)-O(6)	103.8(3)
O(15)-Mo(2)-O(12)	103.6(2)
O(11)-Mo(3)-O(14)	153.5(2)
O(8)-Mo(4)-O(7)	87.5(2)
O(17)-Mo(5)-O(6)	153.7(2)
O(14)-Mo(6)-O(2)	76.38(16)
Mo(3)-O(2)-Mo(6)	90.44(13)
Mo(5)-O(2)-Mo(4)	89.57(12)
Mo(4)-O(1)-Mo(5)	116.0(2)
C(5)-N(2)-C(10)	105.6(5)
C(8)-C(5)-N(2)	116.2(6)

Table.S3 The BET surface areas 、 pore volume and pore size of the$Mg_3Al\text{-}Mo_6$ catalyst. [a] The atomic ratio of Mg and Al was determind by ICP measurement.

Catalyst	Mg:Al[a]	S_{BET}/m^2g^{-1}	Pore volume (cm^3g^{-1})	Pore size/nm
$Mg_3Al\text{-}Mo_6$	0.77:0.23	77.45	0.092	4.79

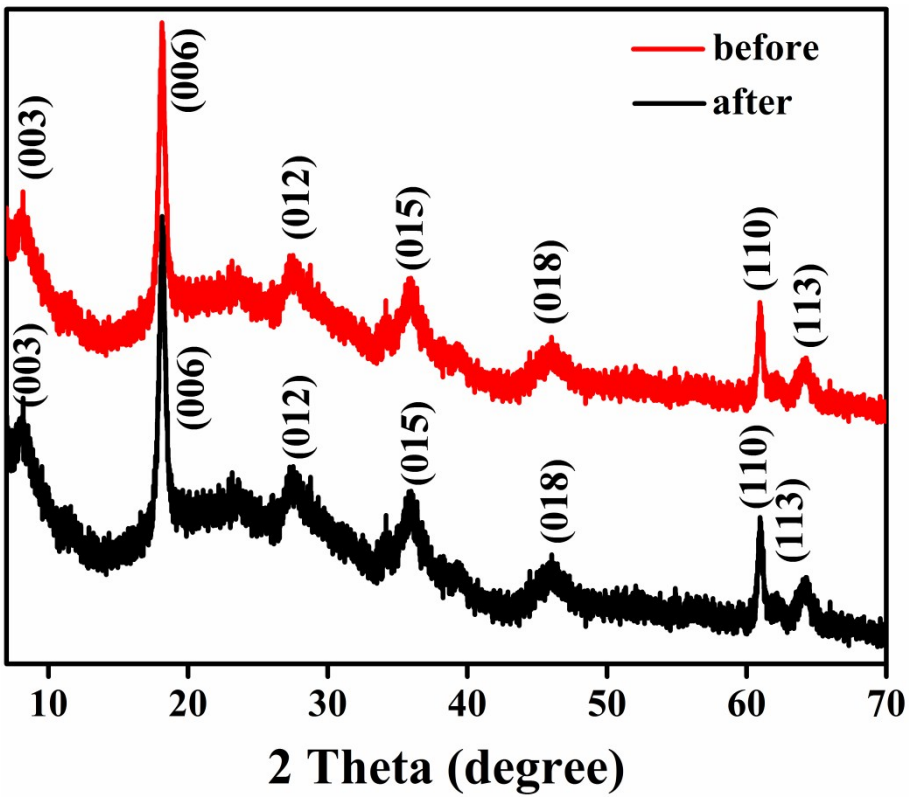

Fig.S2 XRD patterns of the recyclable Mg_3Al-Mo_6 heterogeneous catalyst.

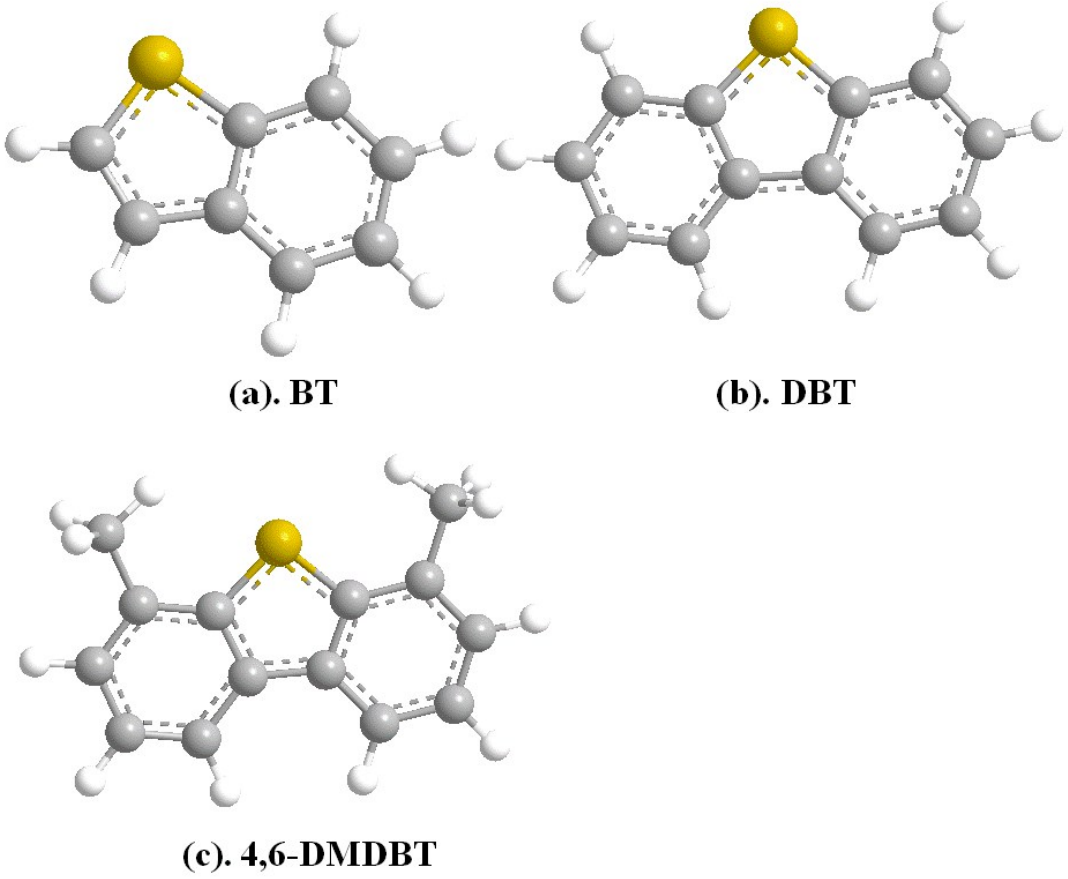

Fig.S3 Spatial structures of different sulfur-containing compounds. (a).BT, (b)DBT and (c)4,6-DMDBT.

Table.S4 The removal of DBT in various ODS systems with different n(O/S)

Catalyst	n(O/S)	Sulfur removal %
HPW-CeO_2	6	96.1[2]
HPW-TUD-1	8	98.1[3]
Ti-MSM	10	99.8[4]
Mg_3Al-IL-EuW_{10}	8	100[5]
$FePcCl_{16}$	40	100[6]
$Na_7H_2LnW_{10}O_{36} \cdot 32H_2O$	5	100[7]

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