

Friction-triggered self-reactive deep eutectic solvents for tunable lubrication under ultra-heavy load through in situ formation of ultra-thick multi-gradient tribofilm

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Fig. S1 Photographs of three friction-triggered self-reactive DESs samples before and after vacuum drying. The top row shows the samples before drying, and the bottom

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row shows the samples after drying.

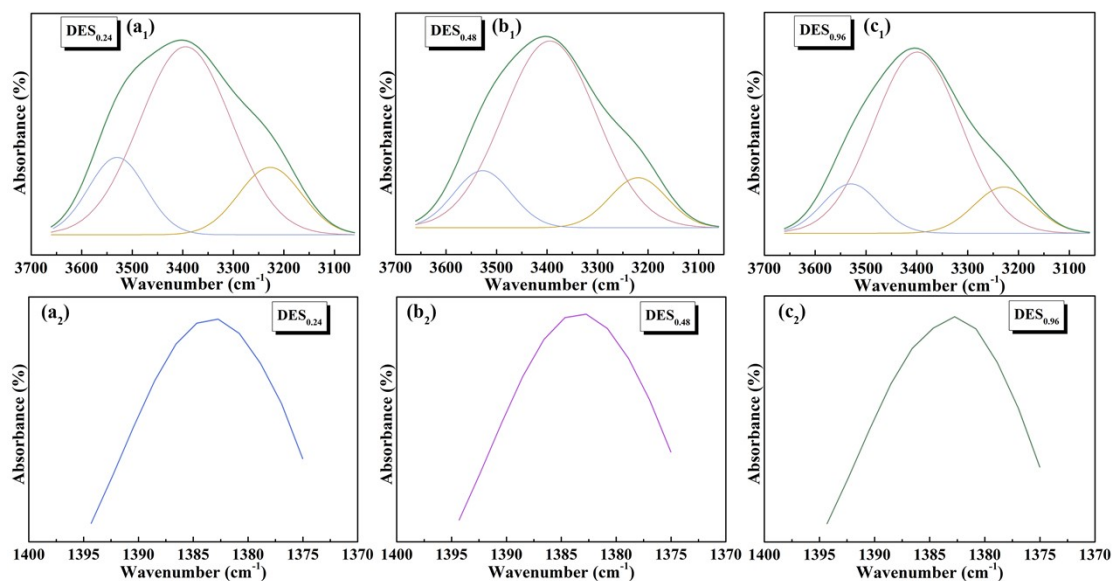


Fig. S2 FTIR spectra focusing on the part of hydrogen bonding interactions in the region of 3000-3600 cm^{-1} and 1395-1375 cm^{-1} .

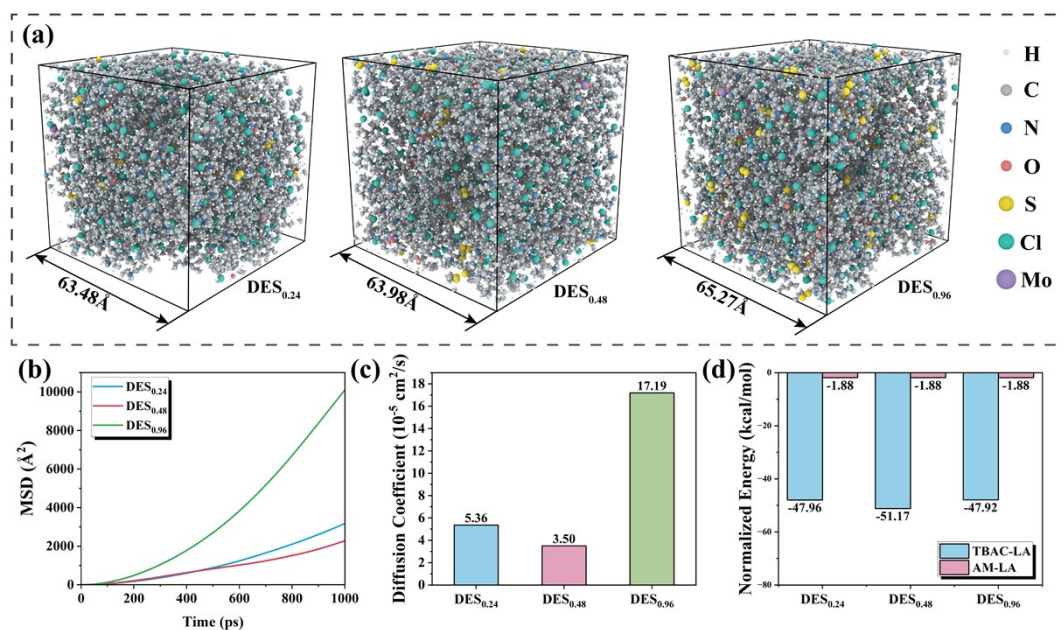


Fig. S3 (a) Simulation configuration of the friction-triggered self-reactive DESs, (b) Mean square displacement (MSD) as a function of time for $\text{DES}_{0.24}$, $\text{DES}_{0.48}$, and $\text{DES}_{0.96}$, (c) Diffusion coefficients calculated from the MSD curves, (d) Normalized interaction energies between TBAC-LA, AM-LA, and LA-LA with the three DESs.

Table S1 The average number and proportion of hydrogen bonds in three friction-triggered self-reactive DESs.

DES	DES _{0.24}		DES _{0.48}		DES _{0.96}	
	Average Count	Percentage (%)	Average Count	Percentage (%)	Average Count	Percentage (%)
Cl-LA	21.7526	36.29%	45.4418	52.86%	88.5707	67.54%
Cl-NH ₄ ⁺	35.1354	58.62%	36.6737	42.66%	32.4213	24.72%
LA-NH ₄ ⁺	1.0465	1.75%	2.8541	3.32%	4.0025	3.05%
LA-MoO ₄	1	1.67%	1.0001	1.16%	2.9815	2.27%
MoO ₄ -NH ₄ ⁺	1	1.67%	0	0.00%	2.8636	2.18%
LA-LA	0.0075	0.01%	0	0.00%	0.3003	0.23%
ALL	59.9420		85.9697		131.1399	

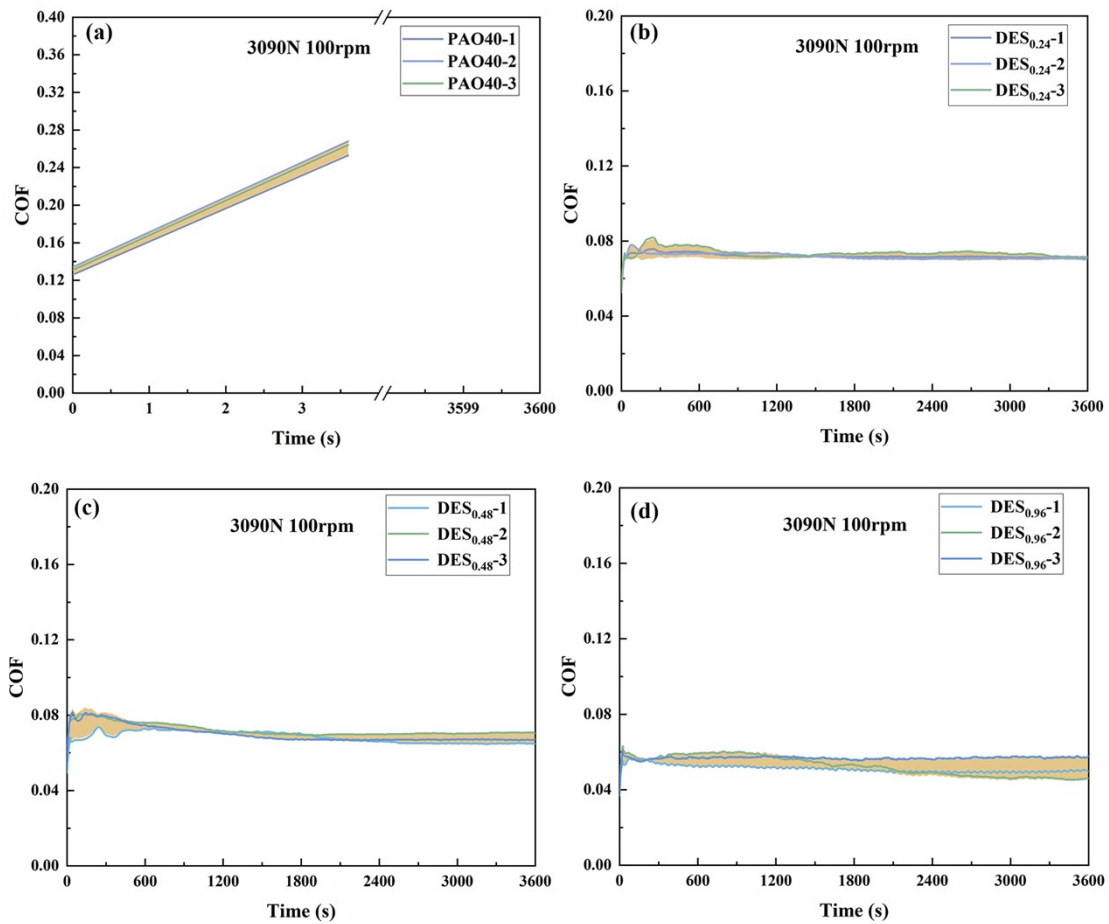


Fig. S4 COF curves as a function of time and the corresponding error ranges obtained from three repeated friction tests under 3090 N for (a) PAO 40, (b) DES_{0.24}, (c) DES_{0.48}, and (d) DES_{0.96}.

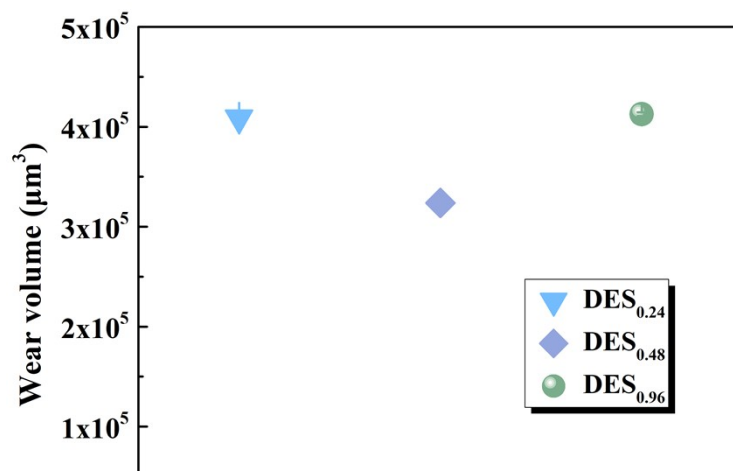


Fig. S5 The wear volume of the friction-triggered self-reactive DESs under 3090 N and 100 rpm.

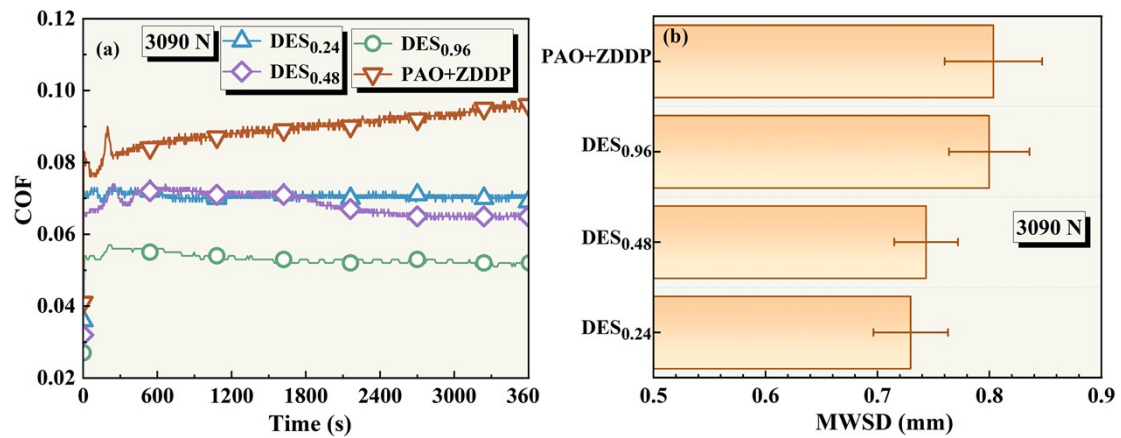


Fig. S6 (a) Friction curves and (b) MWSD of friction-triggered self-reactive DESs at 100 rpm and 3090 N.

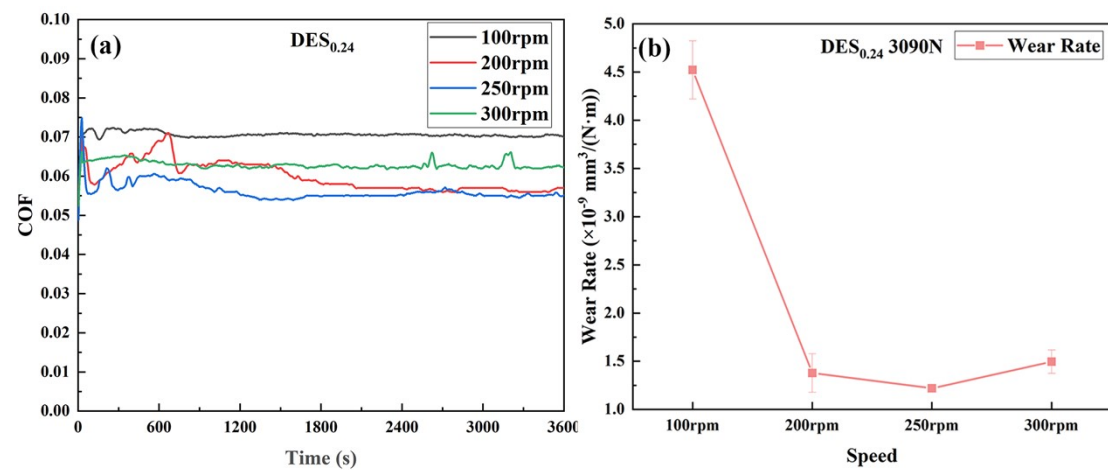


Fig. S7 (a) Evolution of COF values over time for DES_{0.24} at rotational speeds ranging from 100 r/min to 300 r/min and (b) corresponding WSD values.

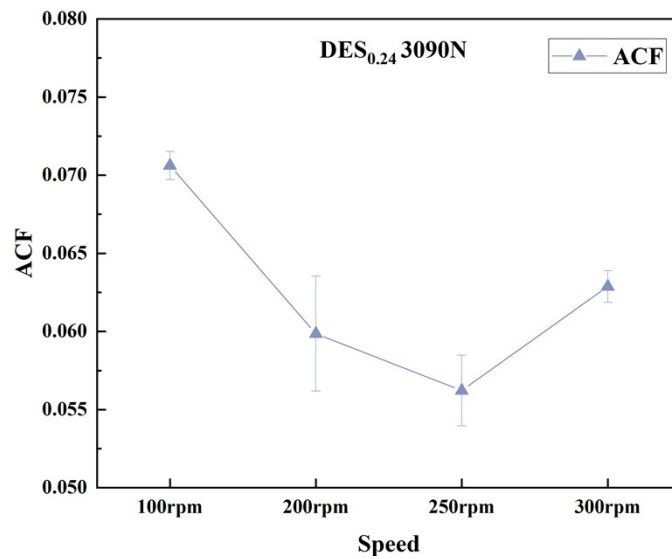


Fig. S8 Average COF values of DES_{0.24} at different rotational speeds.

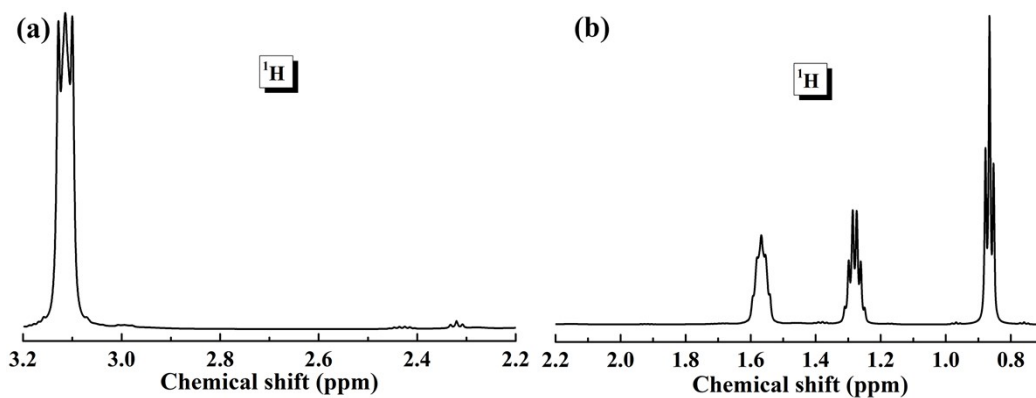


Fig. S9 ¹H NMR of the DES_{0.96} after the friction experiment.

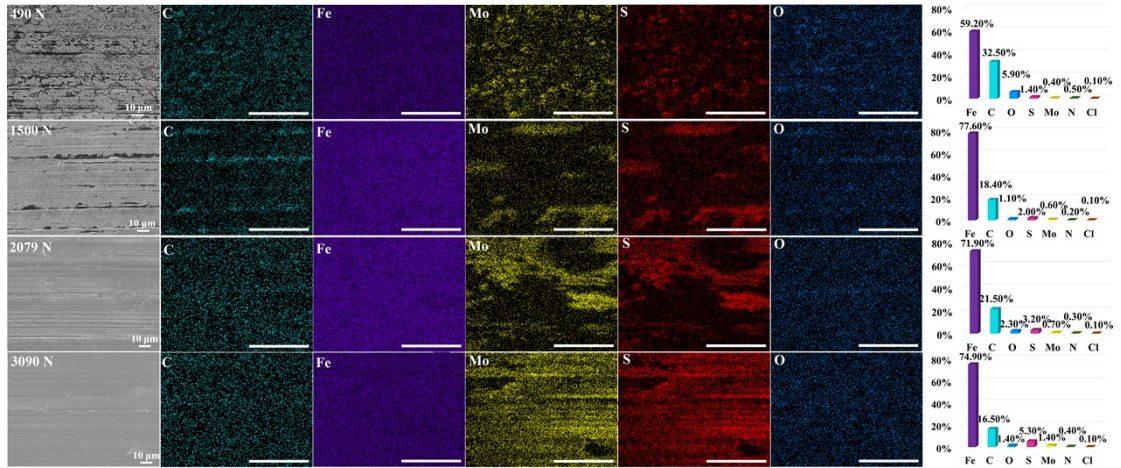


Fig. S10 The surface topographies, EDS mappings and atomic percent of the wear scars achieved by friction-triggered self-reactive $DES_{0.96}$ at 100 rpm and 490 N, 1500 N, 2079 N and 3090 N.

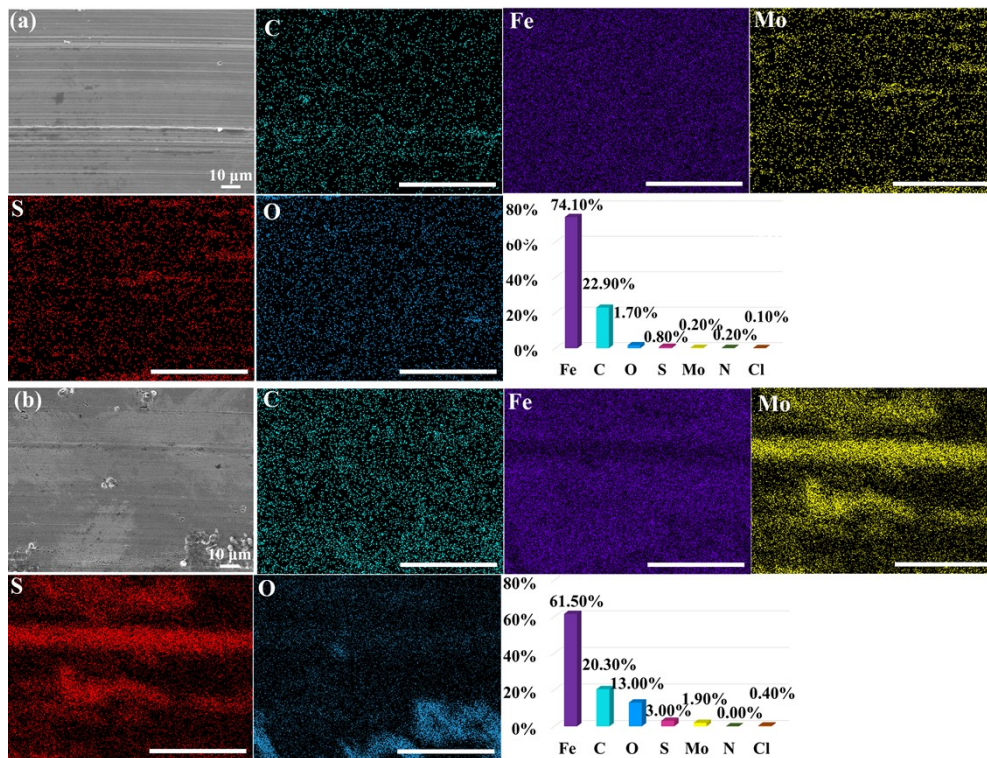


Fig. S11 The surface topographies, EDS mappings and atomic percent of the wear scars achieved by friction-triggered self-reactive (a) $DES_{0.96}$ at 1200 rpm and 490 N, and (b) $DES_{0.24}$ at 100 rpm and 3090 N.

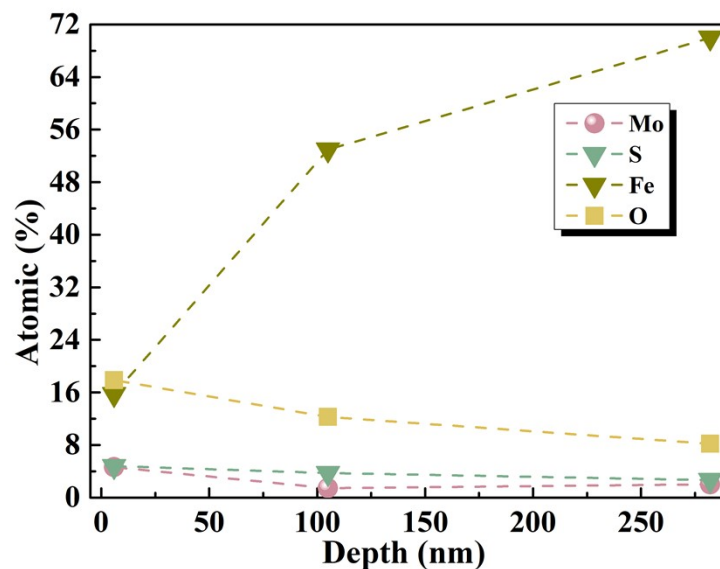


Fig. S12 Fe, O, Mo and S atomic concentrations at different depths, obtained by XPS sputter test on the wear track of DES_{0.24}.

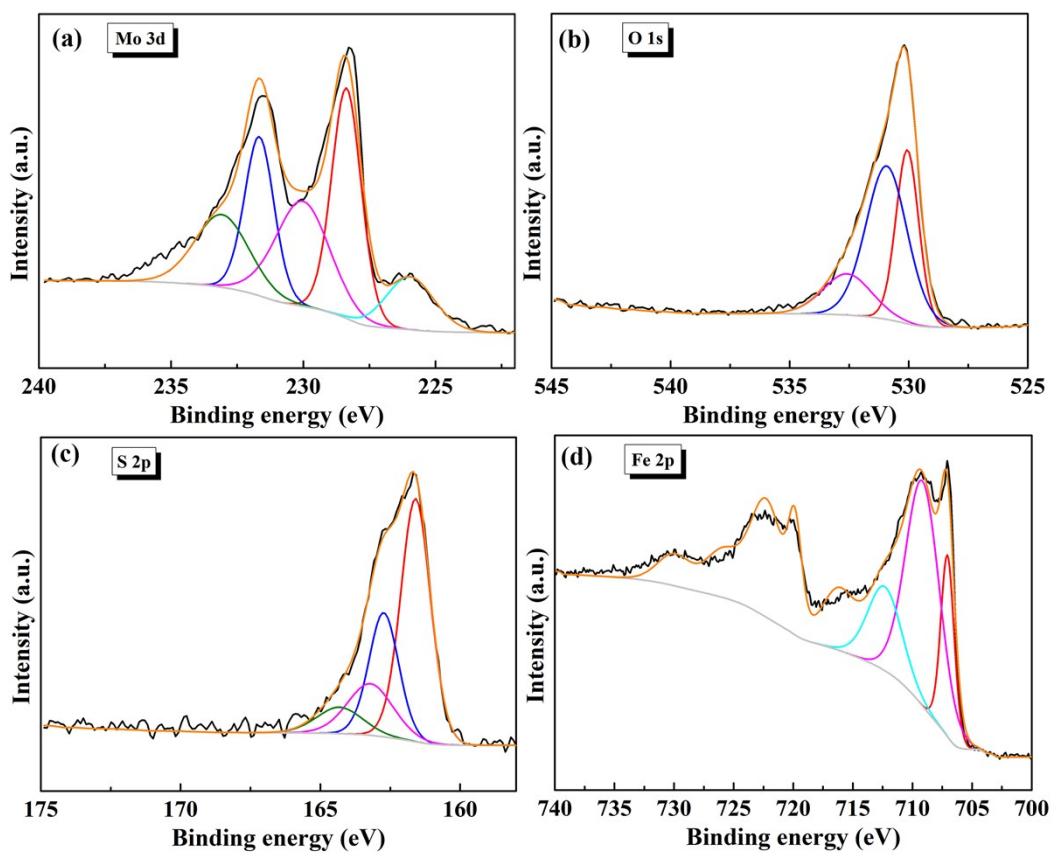


Fig. S13 Mo 3d, S 2p, O 1 s and Fe 2p high-resolution XPS spectra on the worn

surface at a sputtering depth of 5 nm.

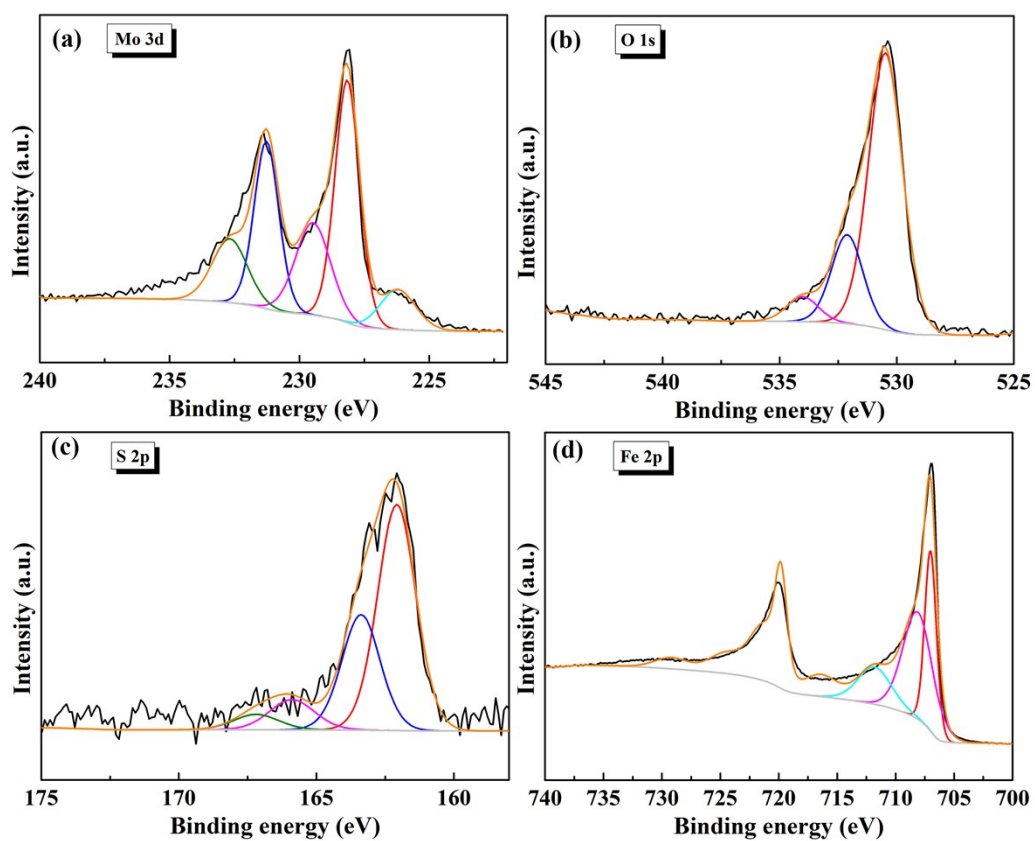


Fig. S14 Mo 3d, S 2p, O 1 s and Fe 2p high-resolution XPS spectra on the worn surface at a sputtering depth of 105 nm.

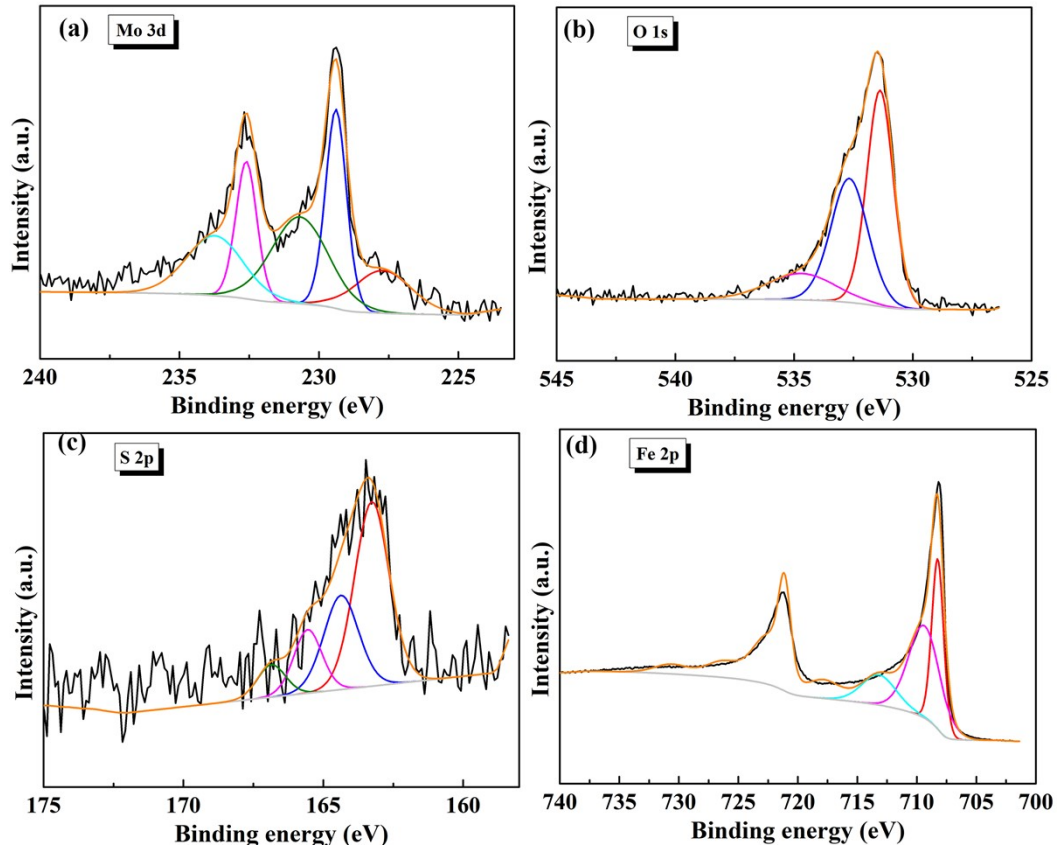


Fig. 15 Mo 3d, S 2p, O 1 s and Fe 2p high-resolution XPS spectra on the worn surface at a sputtering depth of 282 nm.

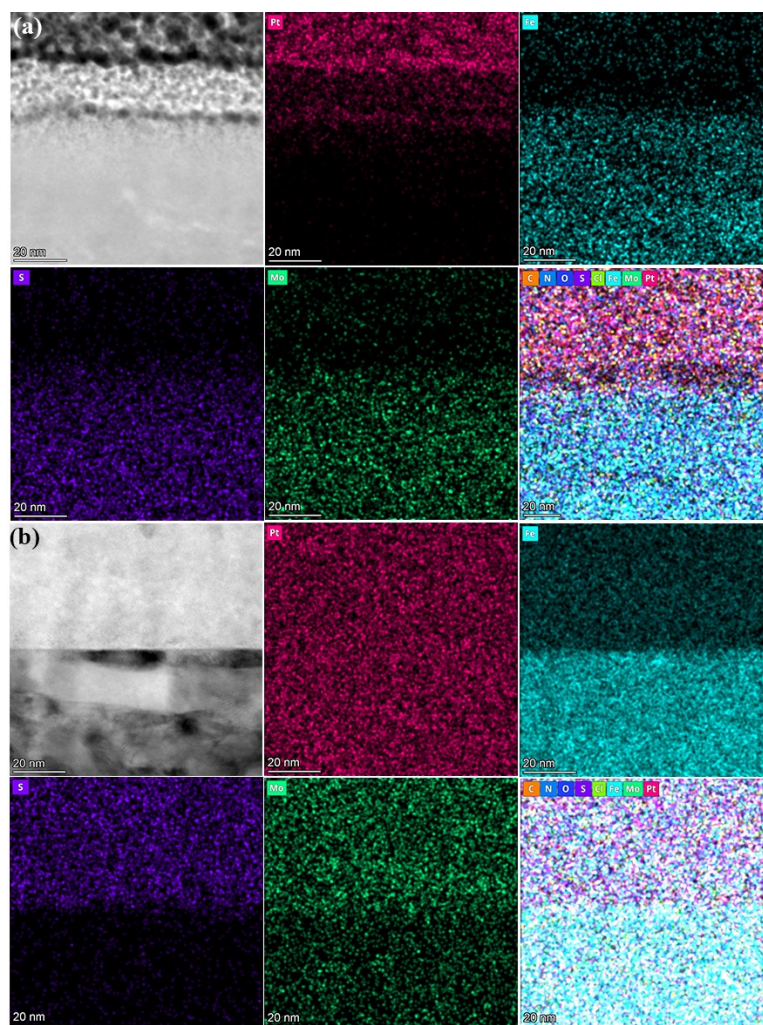


Fig. S16 FIB-TEM analysis results of the friction-induced tribofilm through chemical reaction between HBDs and HBA in the self-reactive DES at 490 N and 100 rpm: (a) the interface between the upper film and the Pt protective layer, (b) the interface between the lower tribofilm and the steel substrate.

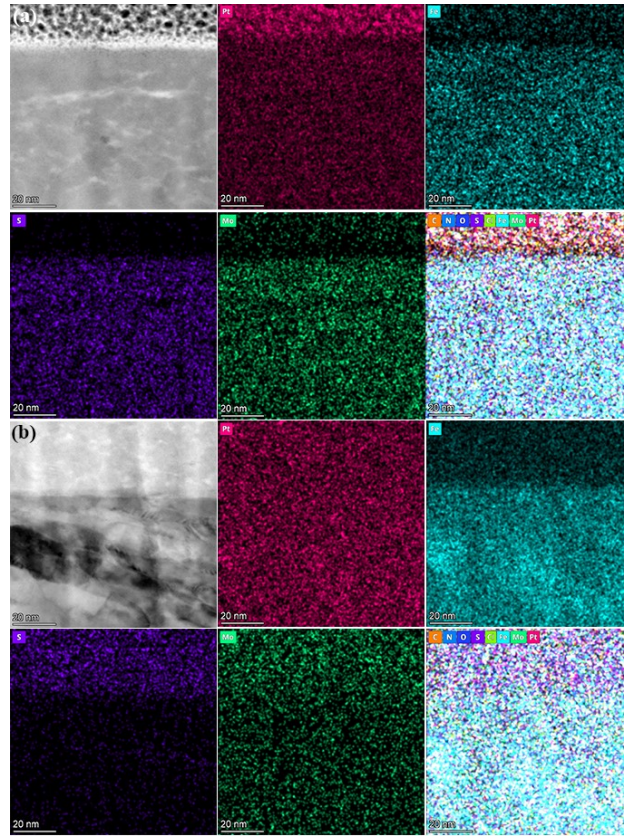


Fig. S17 FIB-TEM analysis results of the friction-induced tribofilm through chemical reaction between HBDs and HBA in the self-reactive DES at 3090 N and 100 rpm: (a) the interface between the upper film and the Pt protective layer, (b) the interface between the lower tribofilm and the steel substrate.