

## Supporting Information

# Ionic liquid functionalized binary montmorillonite nanomaterials as water-based lubricant additives for steel/steel contact

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**S1. The synthetic route of alkanolamine-phosphate PIL and DBU-unsaturated fatty acid PIL.**

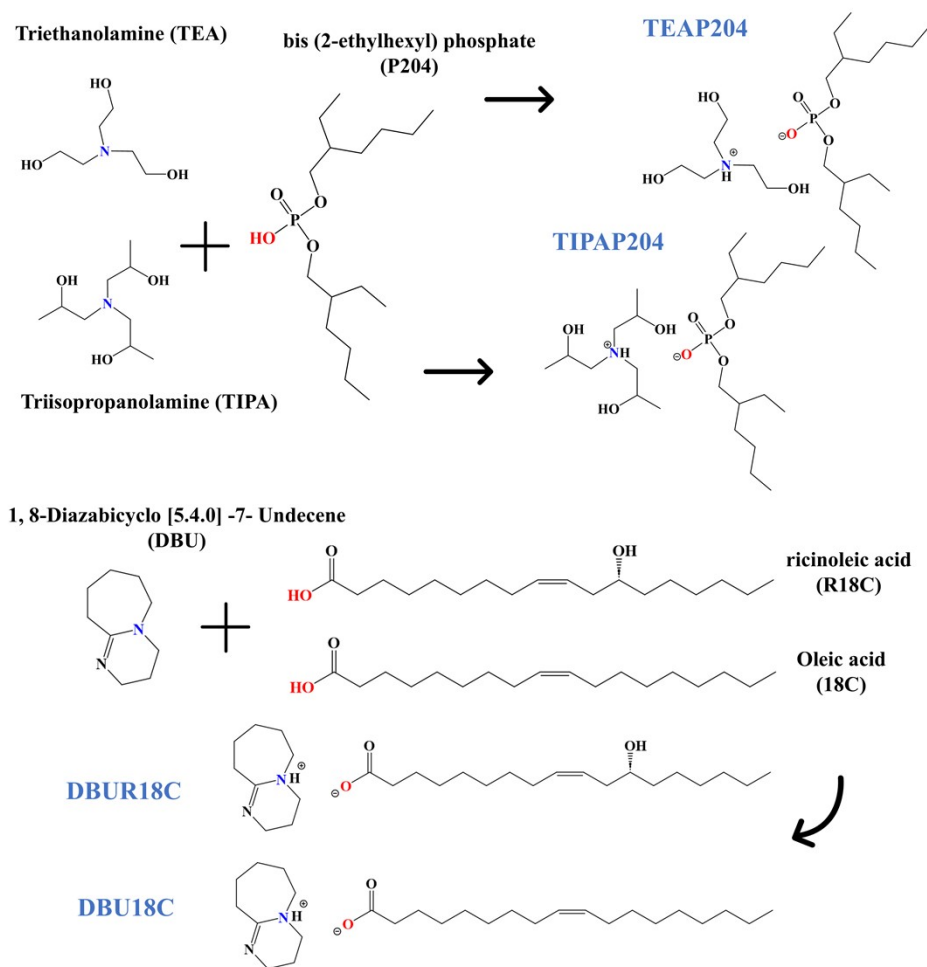


Figure S1. The synthetic route of alkanolamine-phosphate PIL and DBU-unsaturated fatty acid PIL.

The alkanolamine-phosphate PIL was made by proton transfer reactions. To a 250 mL three-necked flask, 19.1 g of triisopropanolamine or 14.9 g of triethanolamine, and 32.2 g bis (2-ethylhexyl) phosphate were added, then consequent mixture was allowed to stir in 50 ml acetonitrile solvent at 80°C for 12 h. After reaction, the product was collected using rotary evaporator and was vacuum-dried for 24 hours to remove the solvent. The synthesis of 1, 8-Diazabicyclo [5.4.0] -7- Undecene (DBU)-unsaturated fatty acid PIL follows a

process similar to that of alkanolamine-phosphate PIL, involving proton transfer between DBU and oleic acid or DBU and ricinoleic acid. And the molar ratio of their reaction is 1:1. As shown in Figure S1, according to the different raw materials, the PIL are named TEAP204, TIPAP204, DBU18C and DBUR18C, respectively.

## S2. The Fourier transform infrared spectroscopy (FTIR) of PIL.

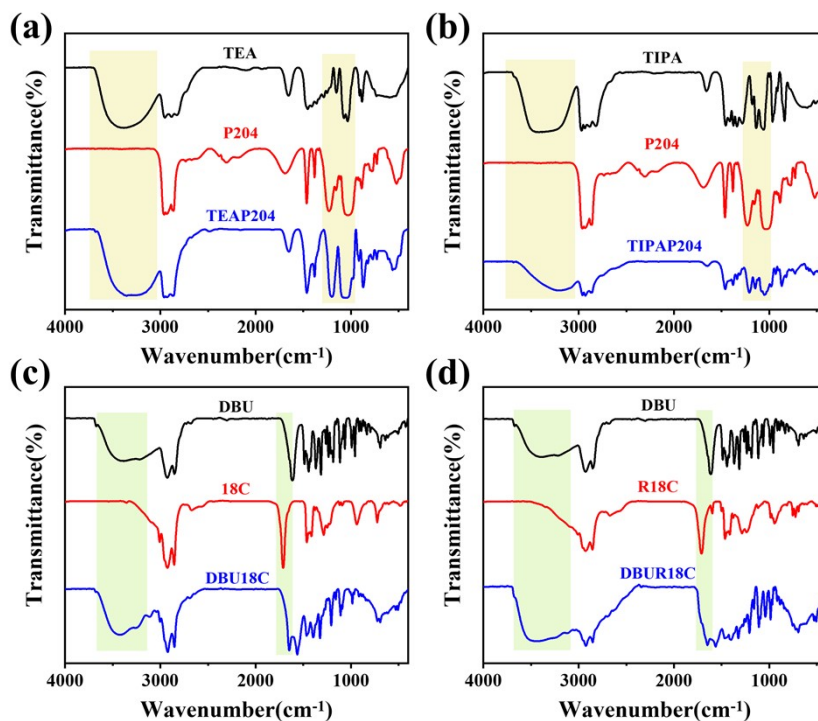


Figure S2. FT-IR spectra of (a) TEAP204, (b) TIPAP204, (c) DBU18C and (d) DBUR18C.

The FT-IR spectrum of alcoholamine-phosphate PIL is illustrated in Figure S2a, b. It is noteworthy that the broad peak at  $3400\text{ cm}^{-1}$  corresponds to the stretching vibrations of N-H and O-H. Additionally, the peaks at  $1209\text{ cm}^{-1}$  and  $1043\text{ cm}^{-1}$  indicate the presence of P=O and P-O stretching bands, which serve as confirmation for the successful synthesis of the ionic liquid.

Meanwhile, the IR spectrum of DBU-unsaturated fatty acid PIL is depicted in Figure S2c, d, showing a broad peak at  $3350\text{ cm}^{-1}$  corresponding to the stretching vibration of

N-H bonds. Additionally, a distinctive peak at  $1651\text{ cm}^{-1}$  indicative of the carbon-carbon double bond specific to oleic acid and ricinoleic acid was observed. The alterations in the IR spectrum validate the successful preparation of DBU-unsaturated fatty acid PIL.

### S3. Comparison of tribological properties with Polyethylene Glycol 200 (PEG-200).

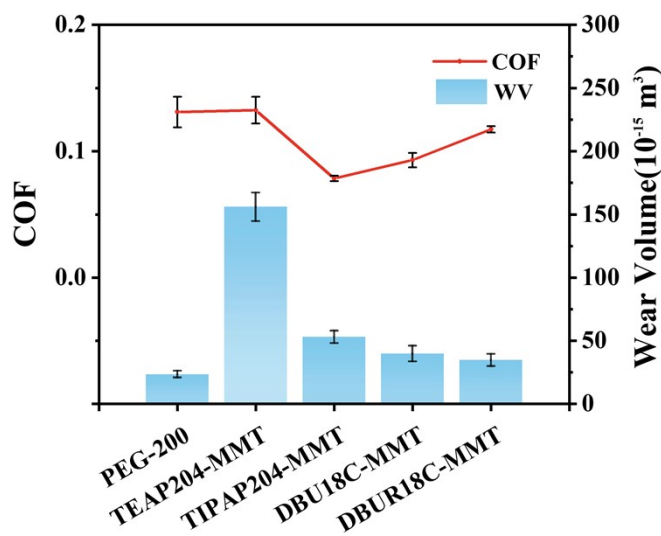


Figure S3. Average COF and WV following lubrication with PEG-200, TEAP204-MMT, TIPAP204-MMT, DBU18C-MMT and DBUR18C-MMT.

The friction coefficients (COF) and wear volumes (WV) of four lubricant samples with an additive concentration of 1.5% after undergoing friction tests under the same test conditions with PEG-200 are depicted in Figure S3. As a widely employed lubricant, the COF of PEG-200 amounts to 0.13, and the WV reaches  $23.5 \times 10^{-15}\text{ m}^3$ . By contrast, the COF of the lubricant prepared in this research is lower; however, the WV after lubrication is marginally higher than that of PEG-200.