Supporting Information

Regulation mechanism of trivalent cations on friction coefficient of poly(vinylphosphonic acid) (PVPA) superlubricity system

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S1. Microscopic observation of surface morphology of PTFE balls after sliding

The surface morphologies of wear scars of PTFE balls were tested using a stereomicroscope (BX53M, OLYMPUS, Japan).

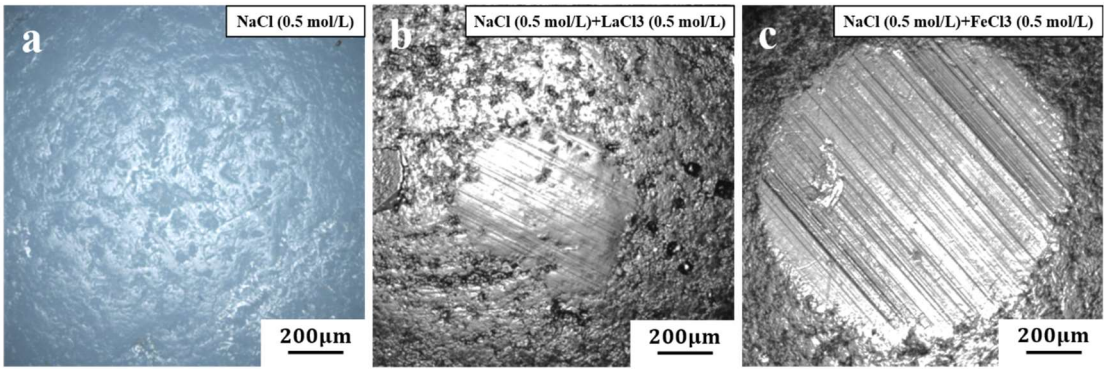

Figure S1. Wear morphologies of PTFE balls lubricated by (a) NaCl solution, (b) NaCl/LaCl3 mixed solution and (c) NaCl/FeCl3 mixed solution.

S2. Calculation of the interaction energy

According to the method mentioned in the main text, a complete cation introduction system model can be established in two steps using molecular dynamics simulation. This model includes various molecular interactions, including the interaction between the introduced cation and surrounding water molecules, the interaction between the introduced cation and the surface, the interaction between the basic cation Na^+ and the surface, the interaction between the introduced cation and Na^+, etc. For the calculation of the interaction energy between the introduced cation and the surrounding water molecules, the excess molecules or ions in the model should first be removed. The effect of the removal on the system energies is evidenced by the disappearance of the energies of the molecules or ions that were removed and the interaction energies with the surrounding molecules or ions. Notably, the removal behavior does not affect the specific value of the interaction energy between the introduced cation and surrounding water molecules. Following the removal of irrelevant molecules and ions, the energy in the system mainly comes from the introduced cation, the surrounding water molecules and the interaction

between the introduced cation and surrounding water molecules, denoted as E_{tot}. A copy of such a system is made, water molecules are removed from the system. At this time, the energy of the system only comes from the introduced cation, denoted as E_1. The introduced cation in the replicated system is removed in the same way to obtain the energy of water molecules, denoted as E_2. According to Equation (2) in the main text, the interaction energy between the introduced cation and surrounding water molecules can be calculated as follows:

$$E_{int} = E_{tot} - (E_1 + E_2) \quad \text{(S1)}$$

Various molecular interactions in the cation introduction system model can be obtained by this method.