Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Segregation of ions at the interface: Molecular dynamics studies of bulk and liquid – vapor interface structure of equimolar binary mixtures of ionic liquids

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Supplementary Information

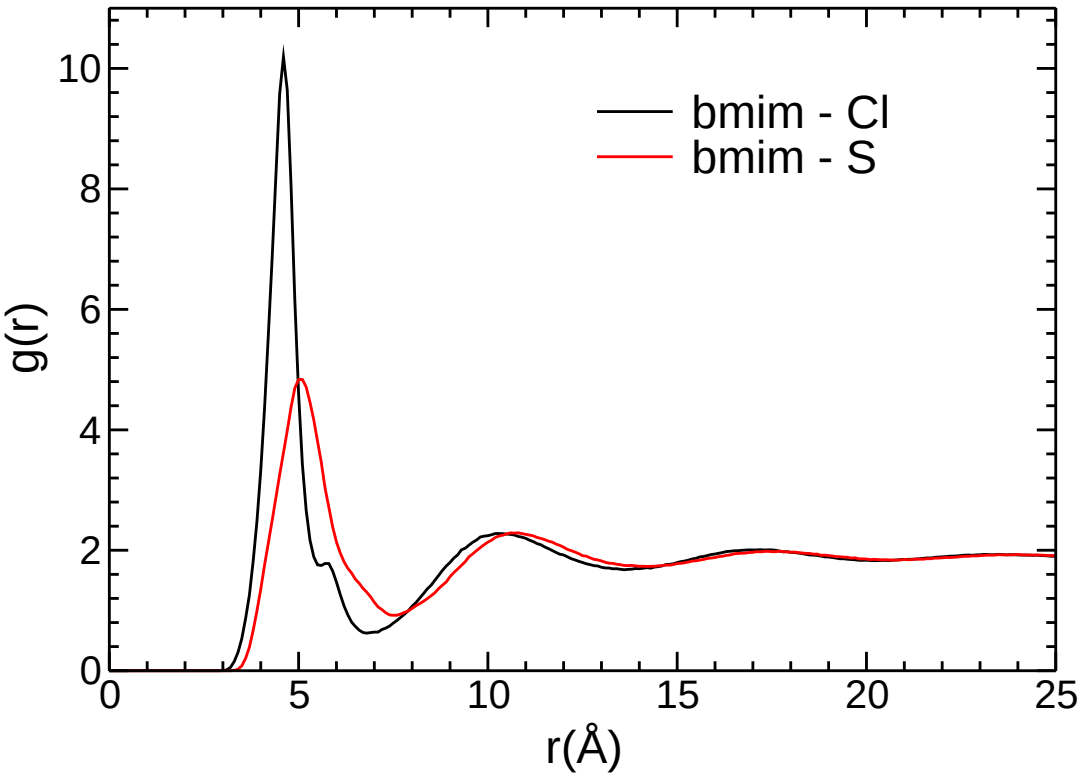

Figure S1: RDFs of the chloride anions and the sulfur atoms of the triflate anions around the geometric center of the imidazolium ring in [Cl] – [TfO] mixture at 500 K.

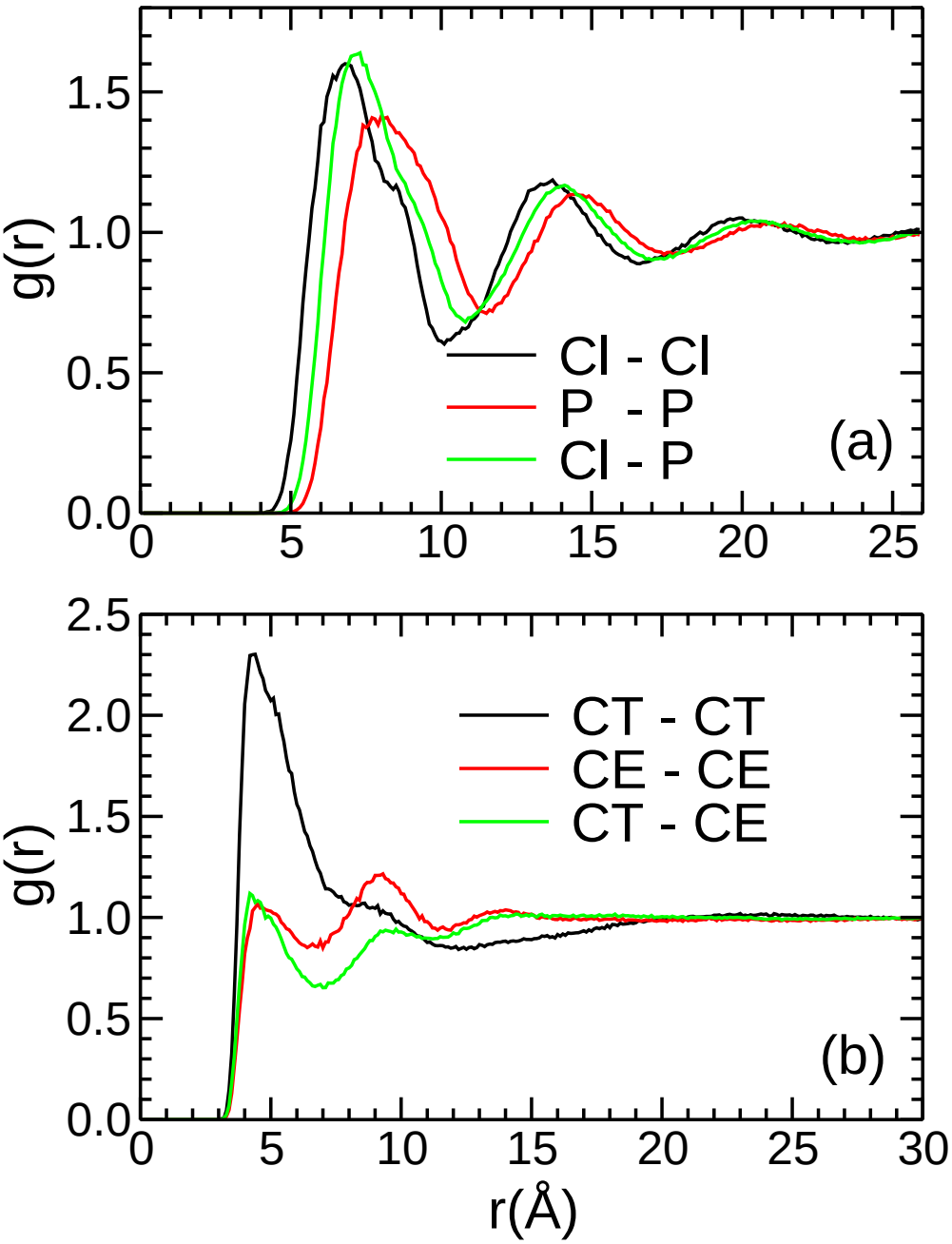

Figure S2: RDFs of (a) different anion sites in [Cl] – [PF_6] and (b) terminal carbon atoms of the alkyl chains belonging to distinct types of cations in [emim] – [omim] mixture at 500 K.

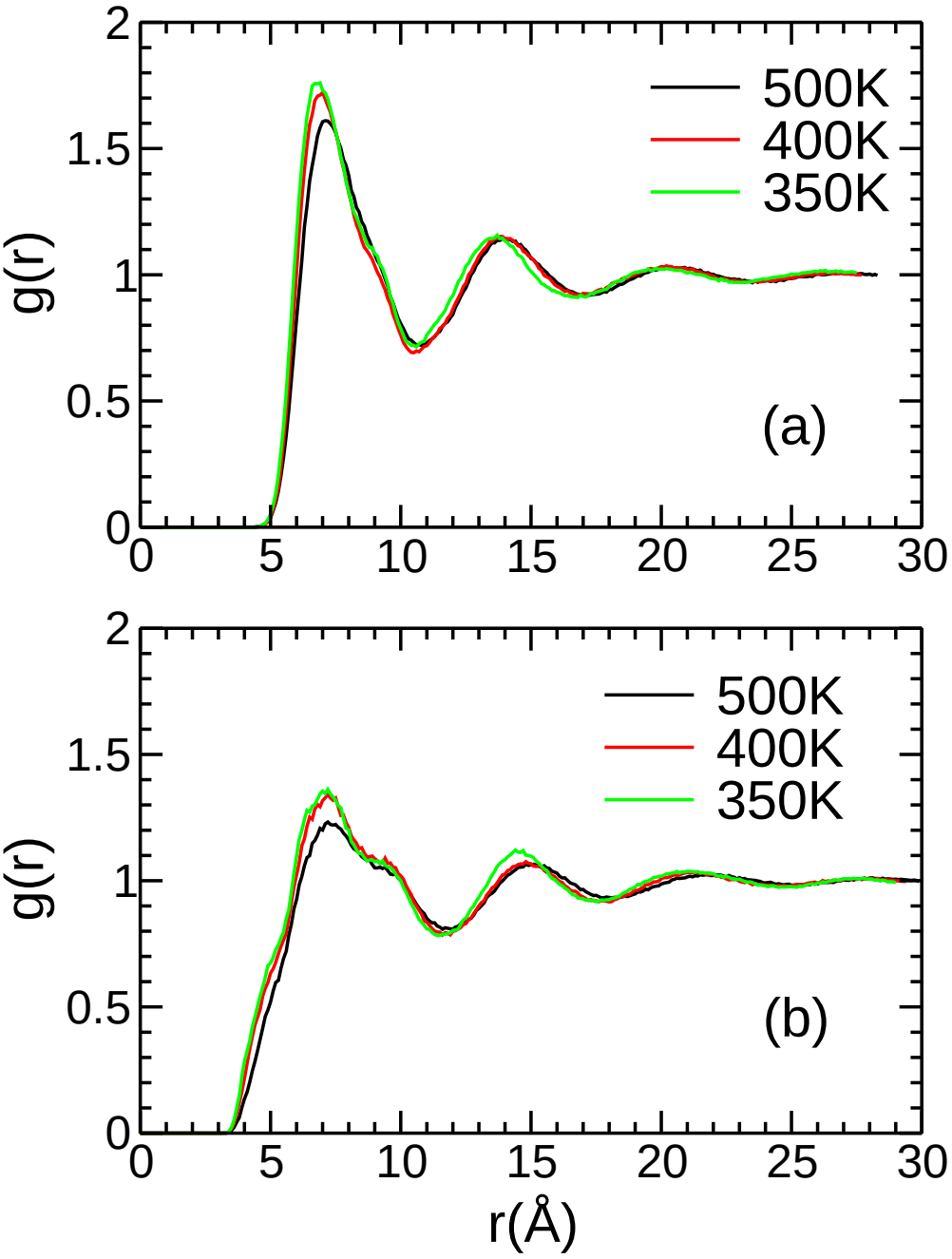

Figure S3: RDFs of (a) sulfur atoms of triflate anions around chloride anions in [Cl] – [TfO] system and (b) head group of [emim] cations around head groups of [omim] cations in [emim] – [omim] system at different temperatures.

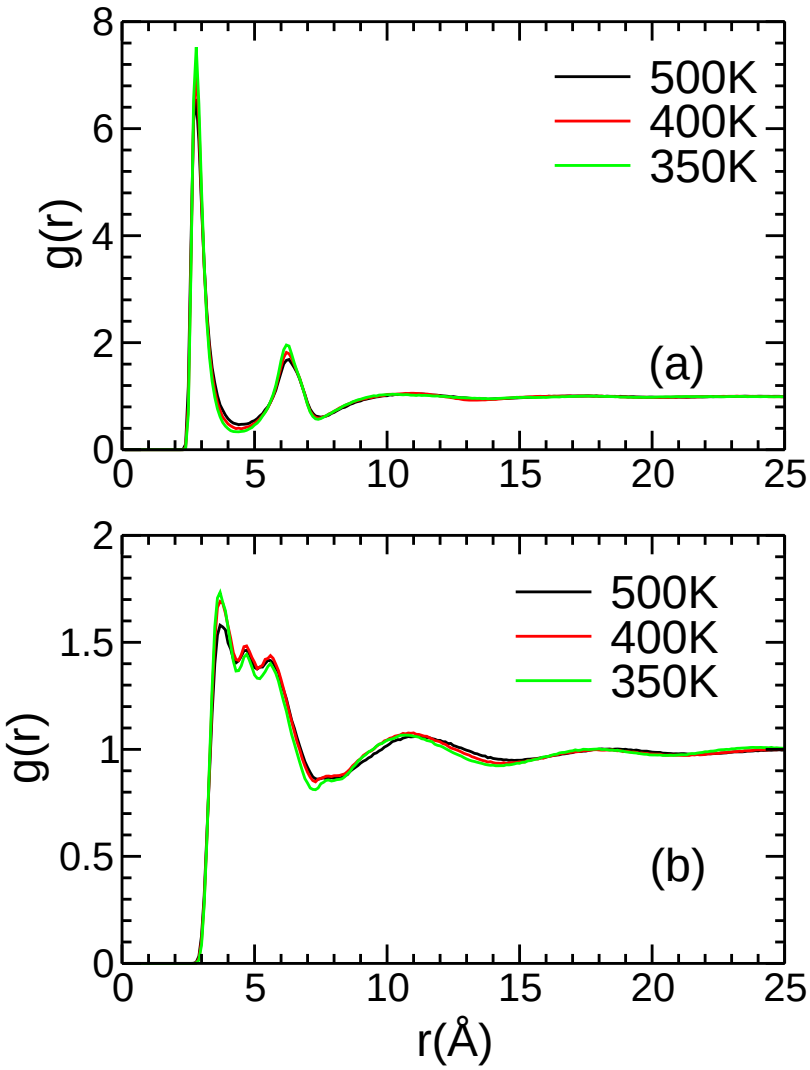

Figure S4: RDFs of (a) the chloride anions around the most acidic hydrogen atom of the imidazolium ring in [Cl] – [TfO] system and (b) oxygen atoms of the triflate anions around the terminal carbon atoms of the ethyl chain [emim] – [omim] system at different temperatures.

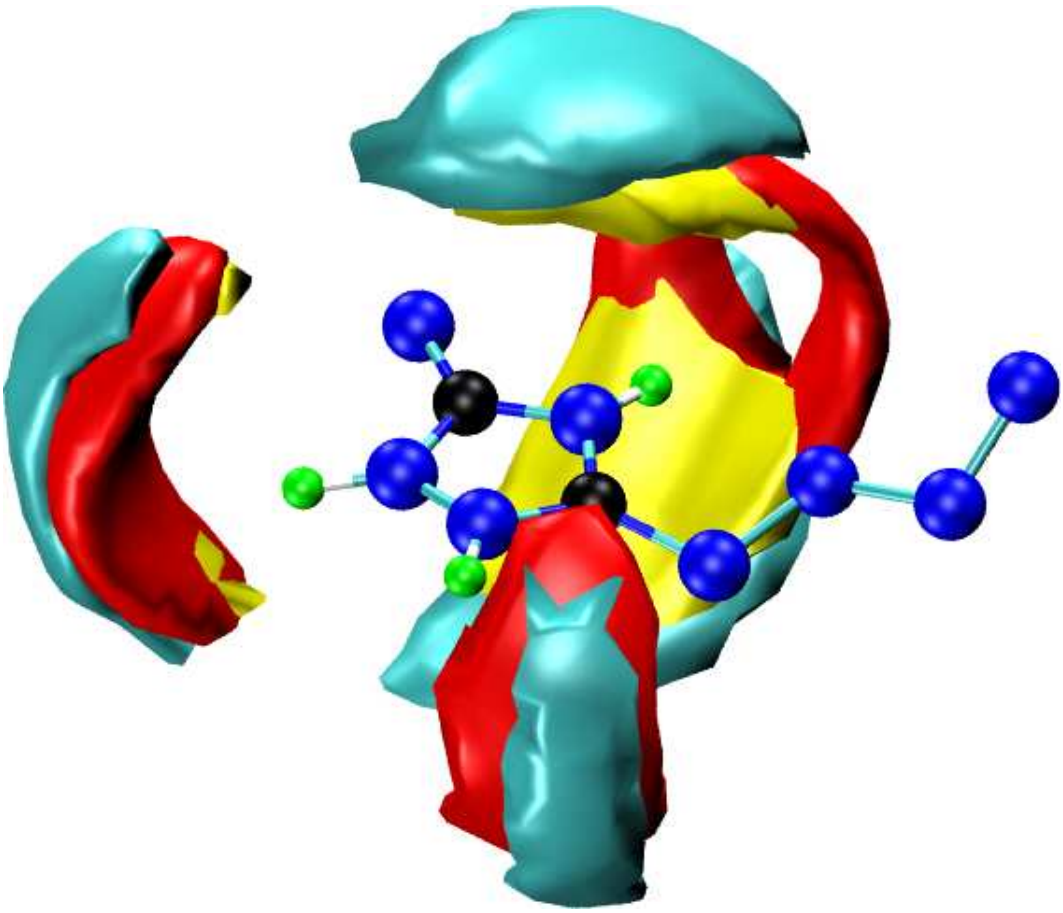

Figure S5: SDFs of chloride anions (in red), oxygen atoms (in yellow) and sulfur atom (in cyan) of the [TfO] anions, that are within 6Å from the cation, around the geometric center of the imidazolium ring in [Cl] – [TfO] mixture at 500 K. The isosurface density shown corresponds to 8.5, 4 and 2 times the average density of chloride ions, oxygen atoms and sulfur atom of the triflate anions respectively. Hydrogen atoms on the alkyl groups are not shown.

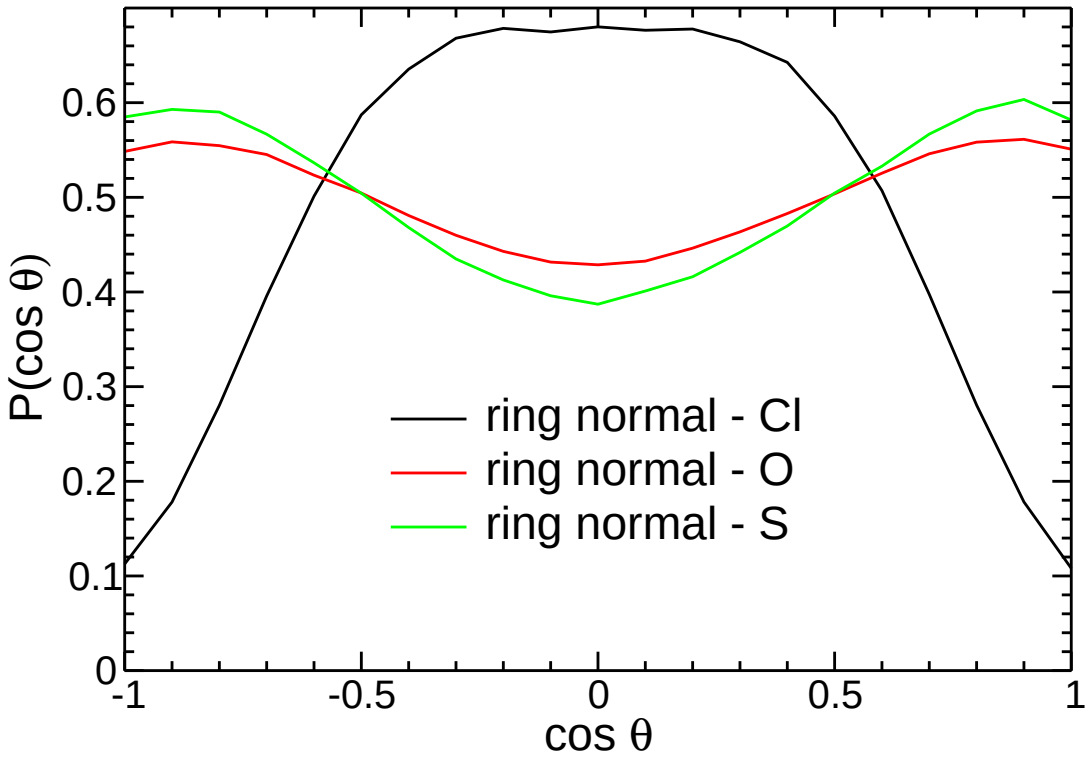

Figure S6: Distribution of the angles between the ring normal vector and the vector connecting the geometric center of the imidazolium ring and the chloride ion or the sulfur atom of the [TfO] anion in [Cl] – [TfO] mixture at 500 K. Only those anions that are within 6Å from the cation are considered.

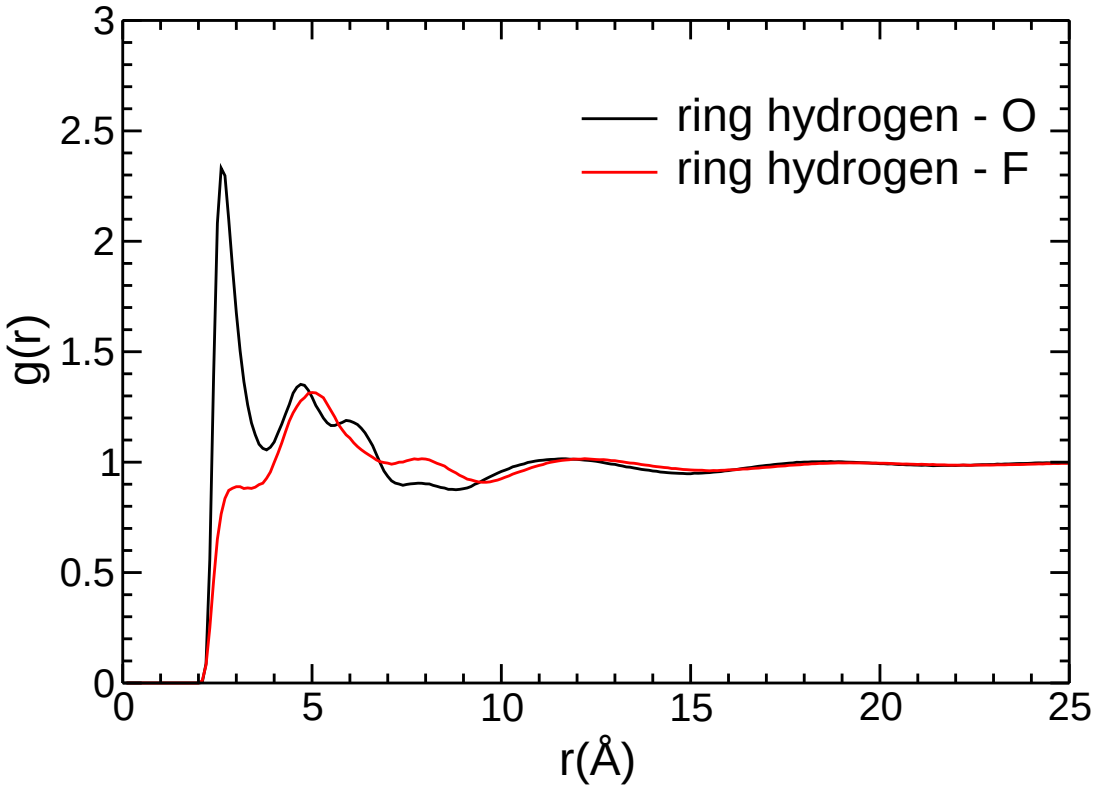

Figure S7: RDFs of oxygen and fluorine atoms of the triflate anions around the imidazolium ring hydrogen atoms in [emim] – [omim] system at 500 K.

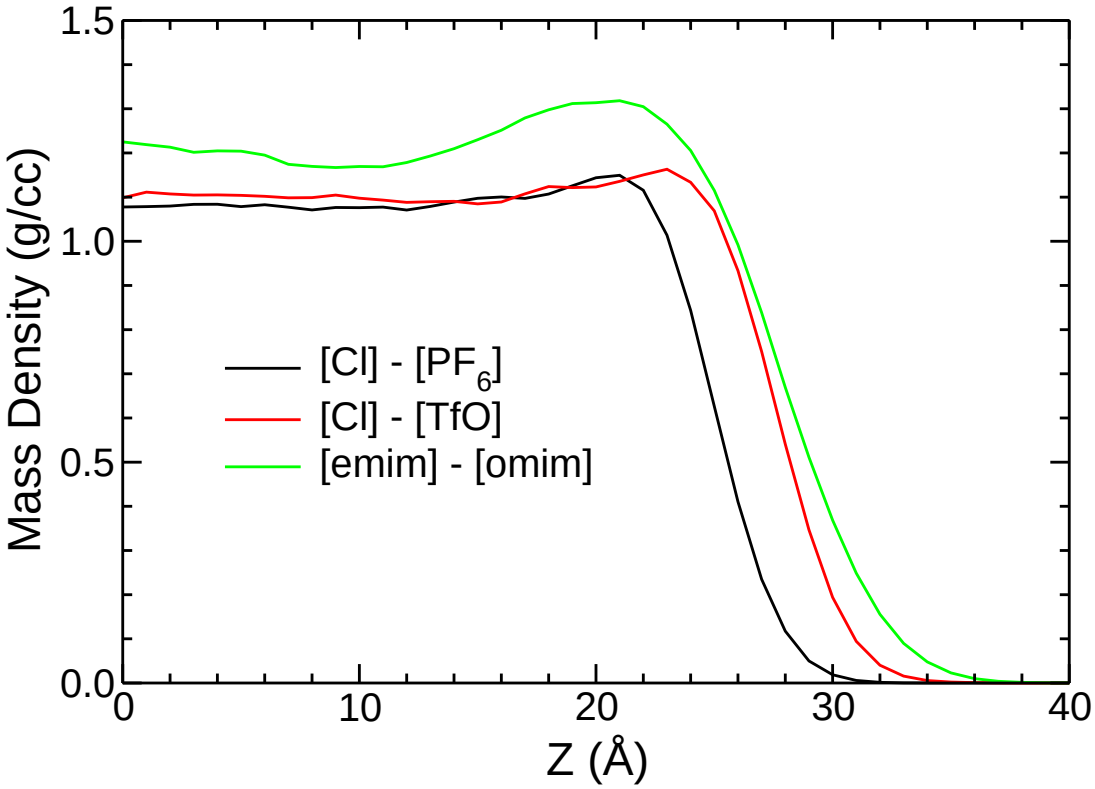

Figure S8: Mass density profiles along the interface normal (z-axis) in binary ionic liquid mixtures at 500 K.

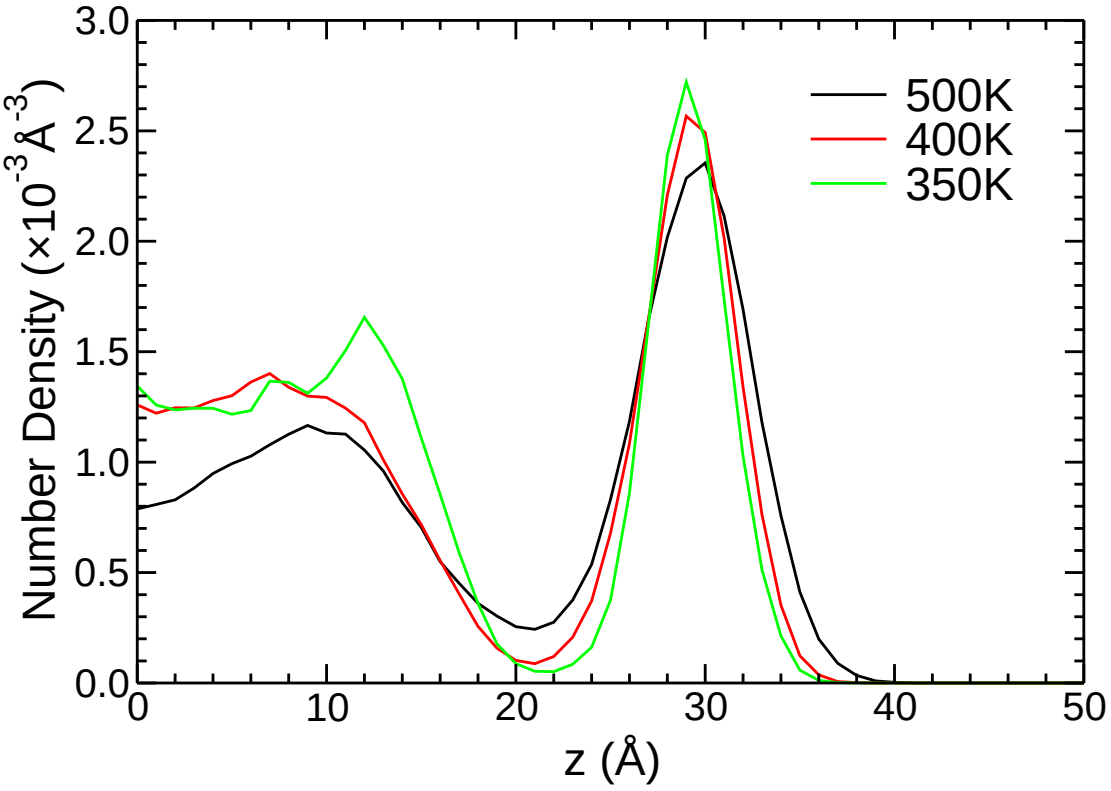

Figure S9: Number density profiles of terminal carbon atoms of the octyl chains in [emim] – [omim] mixture along the interface normal (z-axis) at different temperatures.

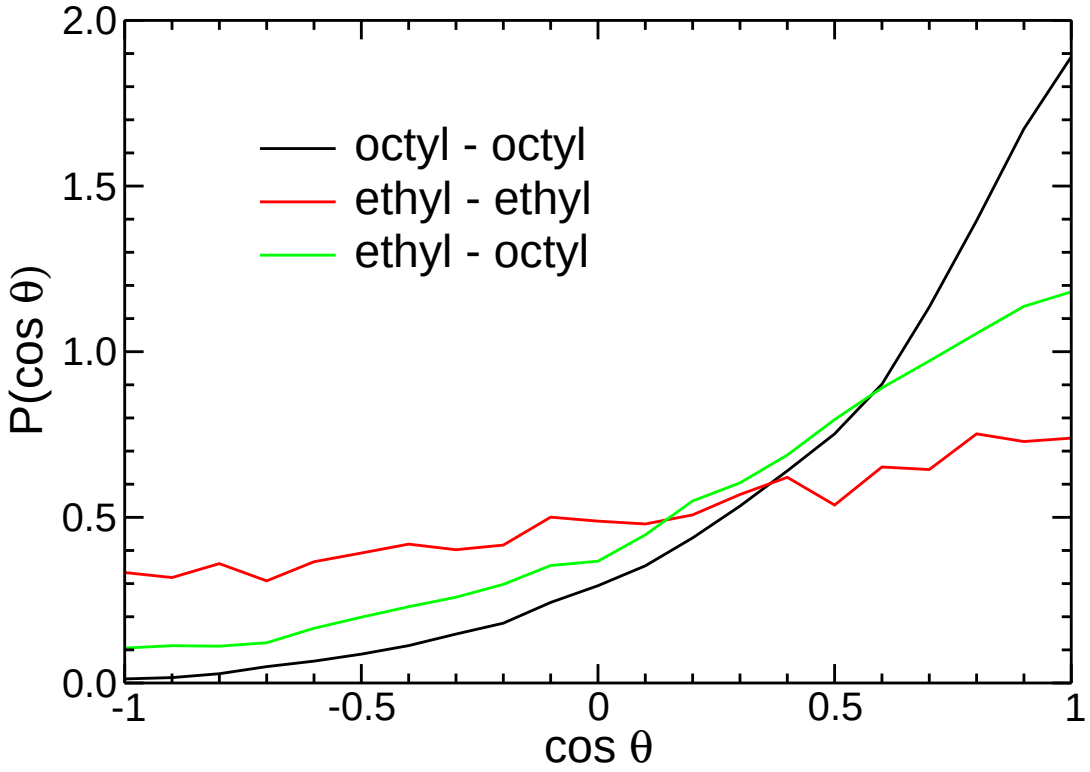

Figure S10: Orientation of ethyl and octyl chains of cations that are present in the interfacial region of [emim] – [omim] mixture at 500 K.

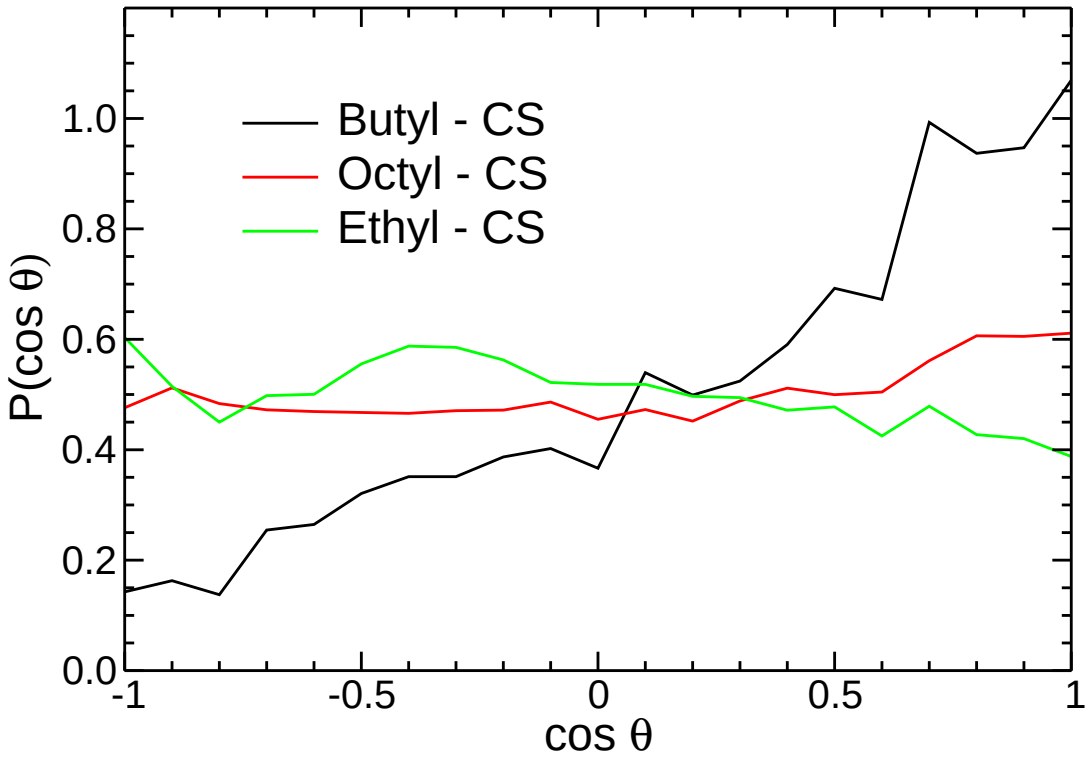

Figure S11: Distribution of the angles between the alkyl chains of the cations and the carbon – sulfur bond vectors of the triflate anions present in the interfacial layer of IL mixtures at 500 K.

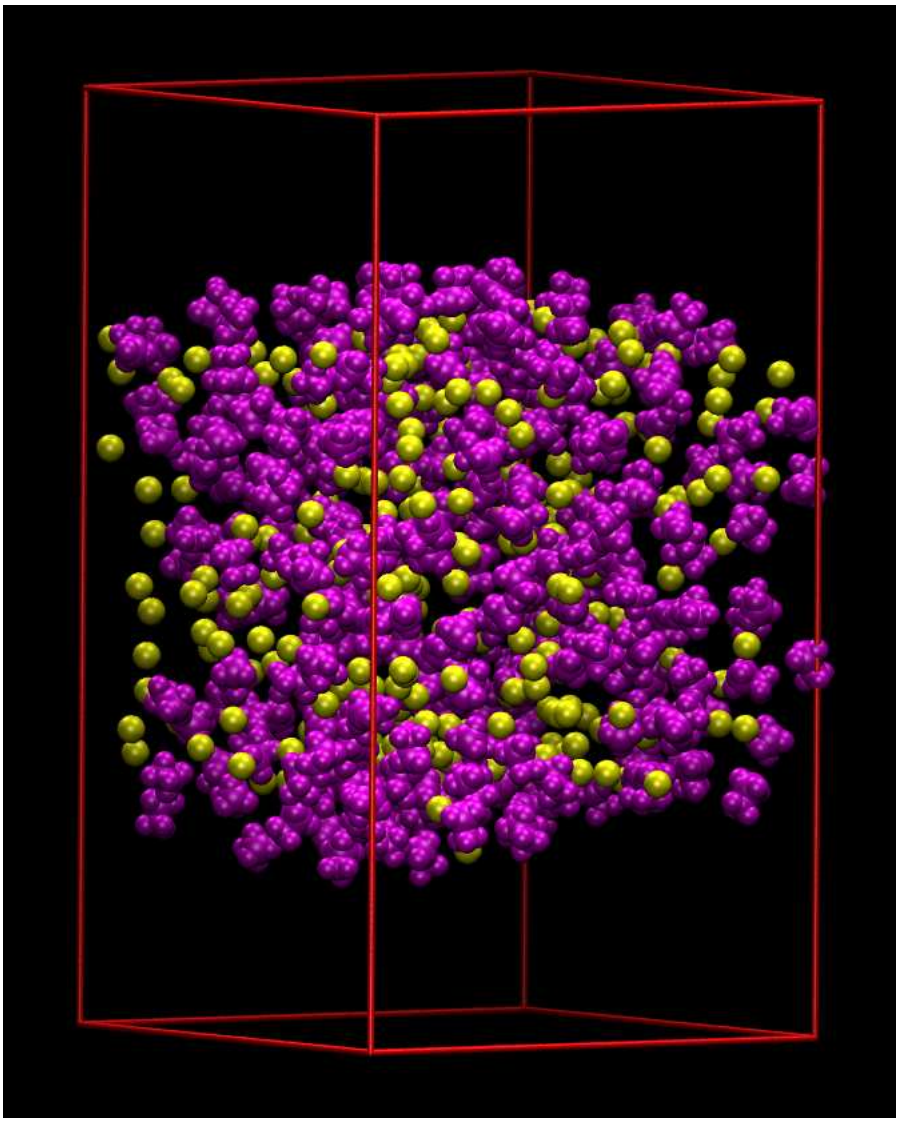

Figure S12: Snapshot of the liquid – vapor interface of [Cl] – [TfO] mixture after 20 ns simulation. Only the anions are shown for the ease of visualization. [Cl] anions are shown in yellow and [TfO] anions are shown in magenta.

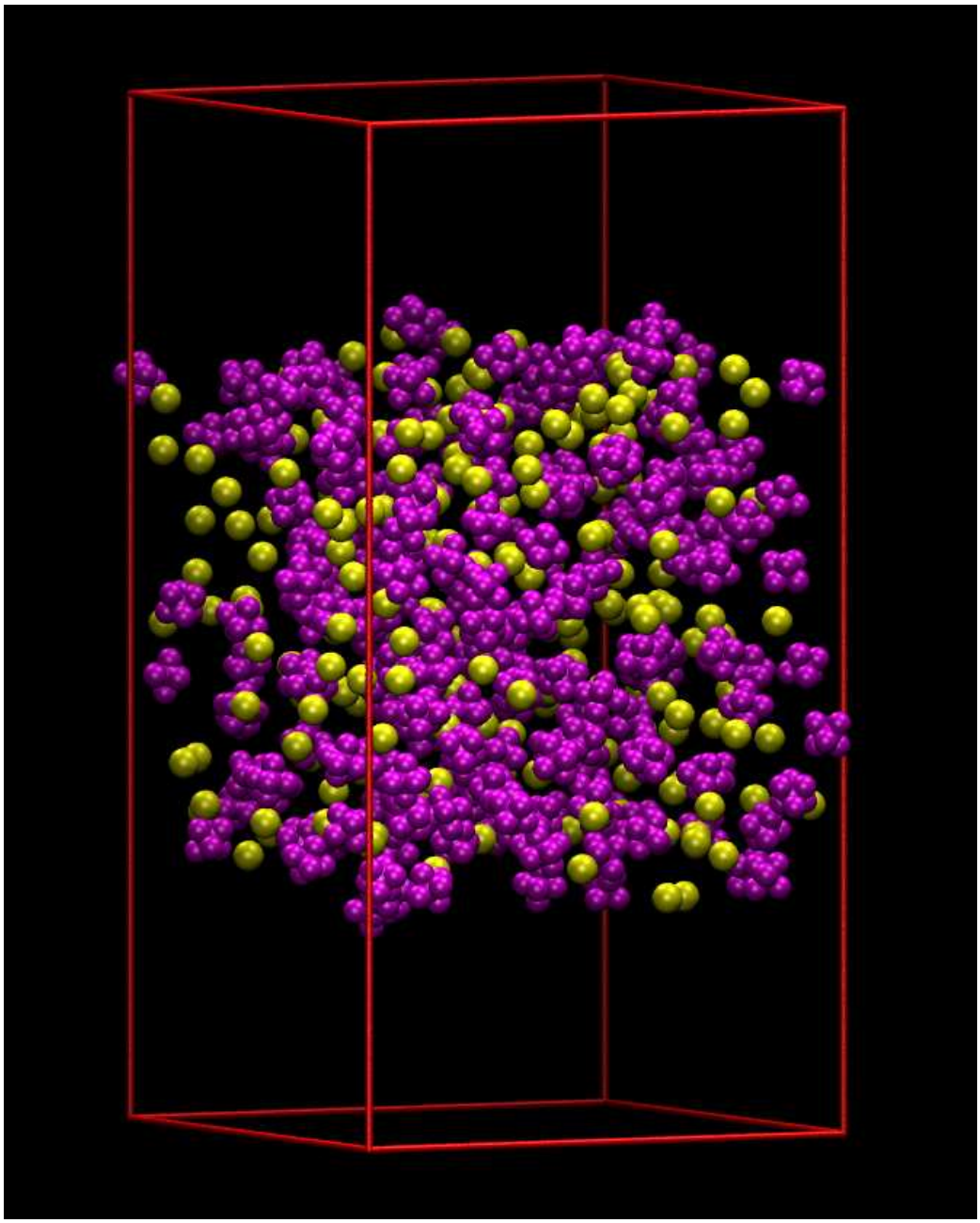

Figure S13: Snapshot of the liquid – vapor interface of [Cl] – [PF_6] mixture after 20 ns simulation. Only the anions are shown for the ease of visualization. [Cl] anions are shown in yellow and [PF_6] anions are shown in magenta.

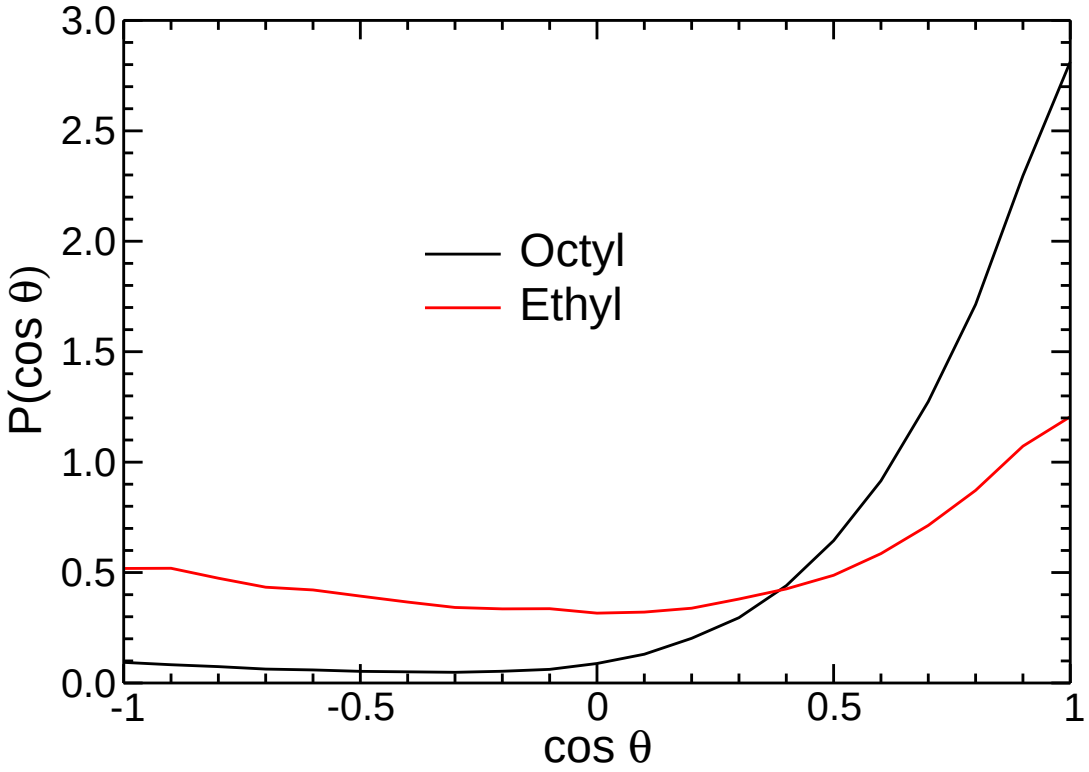

Figure S14: Distribution of the angles between the interface normal (z-axis) and the alkyl chains of the [emim] – [omim] mixture at 500 K.

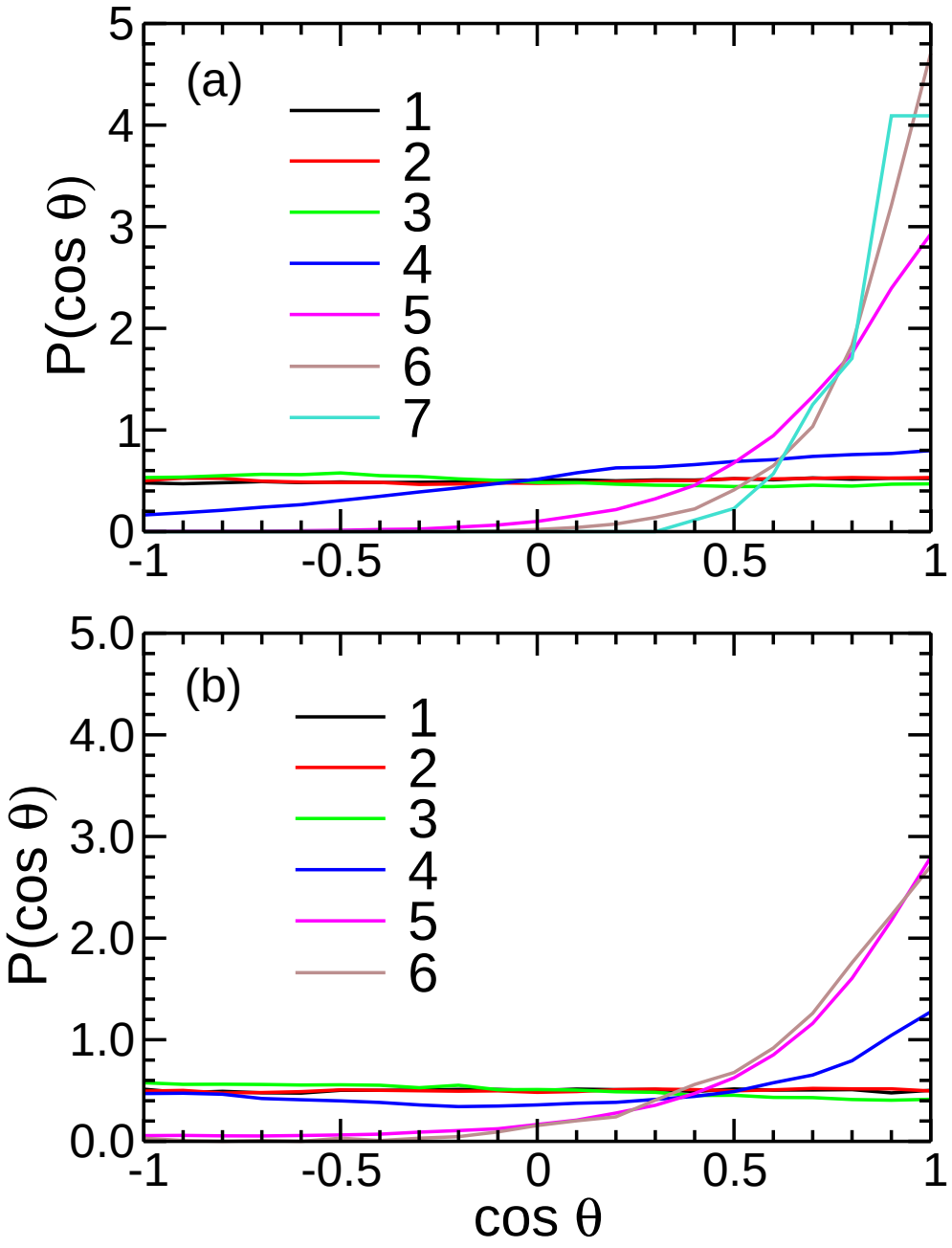

Figure S15: Distribution of the angles between the interface normal (z-axis) and (a) the butyl chains of the cations and (b) carbon – sulfur bond vectors of the triflate anions in various slabs along the interface normal in [Cl] – [TfO] mixture at 500 K.