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Charge dependent condensation of macro-ions at air-water interfaces[1] MRINAL BERA, MARK ANTONIO, Chemical Sciences and Engineering Division, Argonne National Laboratory, Argonne, IL-60439, USA — Ordering of ions at and near air-water interfaces is a century old problem for researchers and has implications on a host of physical, chemical and biological processes. The dynamic nature of water surface and the surface fluctuations created by thermally excited capillary waves have always limited measurement of near surface ionic-distributions. We demonstrate that this limitation can be overcome by using macro-ions of sizes larger than the capillary wave roughness $\sim$3Å. Our attempts to measure distributions of inorganic macro-ions in the form of Keggin heteropolyanions (HPAs) of sizes $\sim$10Å have unraveled novel charge-dependent condensation of macro-ions beneath air-water interfaces. Our results demonstrate that HPAs with -3 charges condense readily beneath air-water interfaces. This is in contrast to the absence of surface preference for HPAs with -4 charges. The similarity of HPA-HPA separations near air-water interfaces and in bulk crystal structures suggests the presence of the planar Zundel ions ($H_5O_2^+$), which interact with HPAs and the water surface to facilitate the charge dependent condensation beneath the air-water interfaces.

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