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## **In situ chemical imaging of environmental, energy, and biological liquid surfaces and interfaces: toward multimodal and mesoscale imaging**

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The surfaces of liquid phases and films have unique kinetics and thermodynamics, distinct from the bulk. However, major surface analytical techniques are mostly vacuum-based and direct applications for volatile liquid studies are difficult. We developed a vacuum compatible microfluidic interface, System for Analysis at the Liquid Vacuum Interface (SALVI), to enable direct observations of liquid surfaces and liquid-solid interactions using time-of-flight secondary ion mass spectrometry (ToF-SIMS). The unique aspects of this 2014 R&D 100 award winner include the following: 1) the detection window is an aperture of 2-3  $\mu\text{m}$  in diameter allowing direct imaging of the liquid surface, 2) surface tension is used to hold the liquid within the aperture, and 3) SALVI is portable among multiple analytical platforms. SALVI is composed of a silicon nitride (SiN) membrane as the detection area and a microchannel made of polydimethylsiloxane (PDMS). Its applications ToF-SIMS and scanning electron microscopy (SEM) as an analytical tool were evaluated using a variety of aqueous solutions and complex liquid mixtures, some of which contain nanoparticles. SALVI was also used to investigate the solvent structure of switchable ionic liquids. Recently, we demonstrated in situ probing of the electrode-electrolyte solution interface (or solid-electrolyte interface, SEI) in SIMS using an electrochemical microreactor consisting of three electrodes. It provides the first direct observation of the surface and diffused layer of SEI in a liquid with chemical speciation using dynamic ToF-SIMS. Moreover, SALVI was extended for studying live biofilms and single mammalian cells using correlative imaging by more than one spectroscopy and microscopy technique, each offering different spatial and temporal scales. That is, collecting data on different information level from an identical area in the same sample ideally could lead to a more holistic view of the hierarchical structural organization of complex systems in the real world. Selected results from our latest development will be presented, showcasing new directions and applications of multimodal imaging of environmental surfaces and interfaces and studying chemistry from the bottom up, all based on microfluidics. SALVI, a portable microfluidic reactor, sets the analytical foundation toward chemical imaging of complex phenomena occurring in multiple time and length scales, or the mesoscale, underpinning changes at the molecular level in the liquid surface and solid-liquid interface.