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Physical Chemistry of Gas-Liquid Interfaces: Foreword

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Foreword

Gas–liquid interfaces are all around us. In biological chemistry, oxygen and carbon dioxide molecules partition in and out of the bloodstream with every breath we take. In environmental chemistry, carbon dioxide dissolves in seawater and lowers the pH of the ocean. In industrial chemistry, the ancient Sumerians synthesized surface-active molecules as far back as 2200 BCE. All of these systems present unique avenues for chemical reactions; because of the inherent asymmetry at surfaces, the physical and chemical properties of gas–liquid interfaces differ from those of bulk gases and bulk liquids.

However, the exact definition of the gas–liquid interface is nebulous. Does the interface consist of only the top layer of molecules in the liquid or is it a broader, “fuzzier” region? J. Willard Gibbs was the first to ponder the chemical meaning of surfaces in his 1878 treatise “On the Equilibrium of Heterogeneous Substances.”¹ He first reflected on the need for a new word: *phases*, which he defined as bodies that differ in composition or thermodynamic state regardless of quantity or form. Gibbs then proceeded to craft a thermodynamic definition of the interface based on an infinitesimally thin plane of discontinuity, which he called the dividing surface. The position of the Gibbs dividing surface is arbitrary, but it is generally chosen to balance the concentrations of a component on either side.

Gibbs presented elegant mathematical constructions in isolation from any related experiments. In fact, scientists still debated the existence of atoms and molecules at that time, though Gibbs himself referred to “the exceedingly small sphere of sensible molecular action” when he considered the extent of the surface region.¹ The Gibbs dividing surface and Gibbs adsorption isotherm are still widely used today; over the course of the 20th century, molecular dynamics simulations, quantum-classical approaches, density functional theory calculations, and a plethora of surface-sensitive and surface-selective experimental techniques have all yielded results that fall within Gibbs’ framework. Moving into the 21st century, surface scientists are striving toward faster, more accurate, and more efficient theoretical calculations, as well as experimental methods capable of characterizing gas–liquid interfaces under ambient conditions.

The atmospheric chemistry community has driven many of these advances in techniques and instrumentation. Mario Molina and coworkers sparked an interest in atmospheric heterogeneous reactions in 1987 when they revealed that reactions on the surfaces of stratospheric cloud particles helped explain ozone depletion over Antarctica.^{2,3} Mario Molina shared the 1995 Nobel Prize in chemistry for his work on the ozone hole, and atmospheric chemists began exploring reactions at other interfaces, including liquid aerosol particles, cloud and fog droplets, dew, sea spray, and the surface of the ocean.

In this volume, we seek to introduce readers to modern simulations and measurements of gas–liquid interfaces, with a special focus on atmospheric systems. Our goal is to create a tutorial-style resource to complement the many excellent

reviews published over the last 15 years (e.g., the “Structure and Chemistry at Aqueous Interfaces” themed issue of *Chemical Reviews*).⁴ In keeping with the aims of the *Developments in Physical & Theoretical Chemistry* series, the volume combines perspectives from established leaders and new voices in the field. Chapters 1–3 introduce broad themes in theory, experiment, and emerging applications to atmospheric chemistry. The remaining chapters present the latest advances in narrower research areas, but they emphasize background, context, and implications more strongly than a typical journal review article does. Overall, we strove to create the type of book you would hand to a new graduate student or to a colleague just entering this particular field. We are excited about the recent developments described here, and we look forward to seeing how future studies of gas–liquid interfaces evolve.

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