"ANGELL RUSHES IN WHERE FOOLS FEAR TO TREAD"

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Charles Austen Angell (1933-2021)
(Courtesy: Mary Zhu)

CHARLES Austen Angell was a renowned Australian and American physical chemist, the most versatile of his generation. With his Midas touch, he transformed our understanding of some of the most ubiquitous substances into something exotic.

He was a visionary explorer of glasses and the limits of the liquid state – the forms of matter that challenge researchers to date – and also dabbled in other, starkly disparate realms of science. His pioneering experimental discoveries helped shape the ways chemists, physicists and engineers employ to probe these materials, both for fundamental research purposes and in modern applications such as battery technology.

The internationally-acclaimed luminary in the fields of glasses and various types of liquids (including the most abundant, water), died on 12 March 2021, aged 87, at Phoenix, Arizona, after battling with cancer for the past 17 years.

Solids are classified as crystalline solids, whose components are arranged in regular repeating three-dimensional arrays (called the crystal lattice) and the amorphous solids (from the Greek word "*ámorphos*", meaning "*shapeless")* whose components are arranged randomly, with no fixed repeating pattern in any direction. If the liquid phase of almost any substance is cooled fast enough such that it prevents the atoms from arranging themselves into a regular array, it can solidify into an amorphous solid.

Some solids are intrinsically amorphous. Unlike crystalline solids, their X-ray patterns are poorly defined due to the random arrangement of the particles and they melt over a wide range of temperature, instead of having a sharp, well-defined melting point like their crystalline counterparts, whose orderly nature makes the intermolecular forces uniform such that the same amount of thermal energy is needed to break every interaction simultaneously. For instance, when molten SiO_2 is cooled rapidly at nearly 400 K/second, it forms quartz glass where the SiO_2 tetrahedrons are arranged randomly whereas amorphous aluminium forms only when the liquid is cooled at an extraordinary rate of 4×10^{13} K/second!

Being dense and disordered, the behaviours of liquids and glasses cannot be explained using the theories of crystals, which are dense but ordered, or gases, which are disordered but not dense. Therefore, long-standing questions keep lurking in the minds of scientists – about the nature of the transition from liquid to glass and how the apparently common liquids like water behave at low temperatures. Angell, thanks to his inherent inquisitiveness, forayed deeply into these lesser-known areas of material science and his imperishable concepts and contributions now define the field.

Angell was born in 1933 and raised in Canberra, Australia. His father was a pathologist and avid hobbyist and, as fondly recalled by Angell, used to cast aluminium objects from aircraft scrap, melted in their dining-room fireplace, which got him *"interested in hot liquids at an early age"*.Thereafter, he went on to devote his career to the study of those liquids, spanning across more than five decades.

He studied chemical metallurgy at the University of Melbourne, Australia and then worked on molten salts at the University of Pennsylvania in Philadelphia. He completed his PhD with John Tomlinson at the Imperial College, London in 1961 and his thesis earned him the biennial Armstrong Medal for graduate research. Returning to his alma mater in Melbourne, he was fascinated by fibres that he obtained from molten potassium nitrate and calcium nitrate, simple inorganic molecules that readily form glasses on rapid cooling. From then, he was *"hooked on glass-formers"*.

Angell then moved to Purdue University, Indiana in 1966. Thus began his work on aqueous solutions, many of which easily solidify to form glasses. But his favourite target was the most ubiquitous of all liquids – water, in which the transition to glass is notoriously difficult to achieve. As the only natural substance existing in three states of matter – solid, liquid and gas – within the range of temperatures available on Earth, water remains poorly understood but is otherwise capable of exhibiting intricate complexities when properly scrutinised under various extreme physical conditions. But for Austen Angell, it held tempting secrets that he yearned to unlock to the world. So, in his hands, water was pushed to extreme physical limits to probe into its peculiar properties.

With his postdoc student, Robin Speedy, he observed that water becomes

Quartz crystal (Crystalline solid) (left) and Quartz glass (Amorphous solid) (right) *(Courtesy: Wikipedia and Enterprise Q)*

more unusual as it is super-cooled (kept liquid below its freezing point). As temperatures fall further, properties such as specific heat and compressibility rise even faster, suggesting the presence of a thermodynamic limit beyond which super-cooled water cannot remain a liquid, an idea now known as the *Speedy-Angell conjecture.* This triggered an outburst of interest in super-cooled and glassy water, some of which continue even today, including the possibility of transitioning from one liquid phase to another with a lower density, thereby carrying forward his undiminished legacy.

With his impeccable ability to perceive patterns connecting widely-differing systems, he charted the *Angell plot*, portraying the change in viscosity of liquids with temperature for different glass-transition temperatures, allowing the behaviours of glass-forming liquids to be easily compared. It revealed that liquids form a continuous spectrum, from *"strong liquids"* whose viscosity vary gradually while approaching glass transition to *"fragile liquids"* which display precipitous change in viscosity as glass transition is neared. The glaring simplicity of his classification has re-shaped research on glass formers, from practical materials engineering to the theoretical modelling of glasses by physicists.

He is also credited with the development of the *Decoupling Index* concept, which reveals the freedom of conducting species to move independent of the supporting medium (liquid, polymer or glass). He and his group had developed the widest electrochemical window solvent on record (*ethyl-methyl sulfone*, 5.9V). He applied his interest in the diffusion and conduction of ions in liquids and glasses to systems such as molten salts, ionic liquids, ion-conducting glasses and battery electrolytes, which continue to guide modern battery technology.

Although he loved experimenting, he could not overlook the potential of computers for long. So, with some of his colleagues in the 1970s, he was the first to conduct some of the earliest simulations of molten silica and silicates, showing a pathway that computational geologists soon followed. His collaborations in computational studies of glassy behaviour continued, in the hope of unearthing mechanisms underlying experimental results.

Charles moved to Arizona State University in Tempe in 1989 and since then, he had remained a dedicated faculty member till his demise. After learning about the micrometre-sized fluid inclusions in quartz from local geochemists, he, along with his wife and colleague, Jenny Green, exploited these to study water under kilobars of negative pressure (tension instead of compression).

Angell exported his ideas developed from his extensive experiments on glasses to protein folding, cryopreservation and plastic crystals. He stayed active till the end, learning about various devices which may use phase transition of liquids for memory storage.

Such was his tenacity and intellectual rigour that he has over 520 publications to his name, which are highly cited among his peers! Four scientific societies – the American Ceramic Society, the American Chemical Society, the Materials Research Society and the Electrochemical Society gave him their internationally-contested awards. He was elected chair of three prestigious Gordon conferences. He was especially proud of an "outstanding reviewer" award given to him by the American Physical Society in 2009. He was also feted by the University College London (Bragg Lecturer, 2015), Otto Schott Research Award (2018), the Galileo Galilei Award (2018) and the Gothenburg Lise Meitner Award (2019). Overwhelmed, he playfully remarked that the attention made him *"feel like a sultan!"*

Throughout his working career, he continued to act as a beacon light to countless students and young researchers, actively connecting with them and promoting their work while leaving an indelible impression on everyone who had been fortunate enough to come across his pioneering personality in the course of their lives.

An irrepressible explorer at heart, he loved to travel, to the benefit of countless conferences. In addition to being an extremely collegial and influential scientist, a venerated teacher, an intellectual giant and a hero to the scientific community, he was also incredibly warm, gregarious and a wonderful human being – as unanimously echoed by people who knew him across the world.

Combined with his generosity, charisma, humour and humility, his love of discovery was infectious! He fearlessly speculated and linked new and disparate ideas, perennially providing fresh perspective through his talks. His blazing trail of seminal research into a plethora of versatile topics such as liquids and glasses, besides bravely venturing into the geochemical, biophysical and battery electrolyte spaces, has carved out a path through the difficult terrains of science for his contemporaries to follow; whose works will be buoyed by Angell's spearheading research. Thus, aptly quipped Speedy, his long-time colleague: *"Angell rushes in where fools fear to tread."*

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