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Figure 1. Frank Wigglesworth Clarke. From Wikipedia (https:// en.wikipedia.org/wiki/Frank_Wigglesworth_Clarke#/media/ File:U.S._Government_FW_Clarke_Public_Domain.jpg; NOAA Photo Collection, U.S. Department of Commerce, public domain).





Mastering the Art of Silicate Mineralogy

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he Chemistry of Earth's Crust

Even by the extravagant standards of the nineteenth century, Frank Wigglesworth Clarke (1847–1931) (fig. 1) exhibited a startling passion for the tabulation of scientific measurements. He served for more than four decades as chief chemist with the U.S. Geological Survey, simultaneously acting as honorary curator of minerals for the Smithsonian (Schaller 1931; Dennis 1932). His legacy of more than three hundred papers includes five editions of his signature work, *The Data of Geochemistry*, an exhaustive anthology of the compositions of lake, river, spring, and ocean waters, volcanic gases, molten magmas, minerals, metallic ores, hydrocarbons, and igneous, metamorphic, and sedimentary rocks—exceeding 800 pages by the final version (Clarke 1924). His output also comprises *The Constants of Nature* (Clarke 1873), *Weights, Measures, and Money of All Nations* (Clarke 1877), *A Recalculation of the Atomic Weights* (Clarke 1910), and—perhaps not surprising for a person who seemed most at ease with numbers—*How to Play Solitaire* (Clarke 1870).

Clarke was much honored in his lifetime, and today he is esteemed for a particularly daring calculation—the earli-

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See Figure 27



Figure 2. Composition of Earth's crust. Only eight elements account for 98.5 percent of the crust by mass. Data are from Klein and Dutrow (2002).

est scientific estimate of the chemical composition of Earth's crust (Clarke 1889). A little historical context illuminates the audacity of this undertaking. A century before Clarke's seminal paper, Antoine Lavoisier (1743-1794) provided the first rigorous definition of chemical elements (Lavoisier 1789), and he recognized only thirty-three species (including heat and light). Thanks to the dogged pursuit of new and exotic minerals by geologically inclined surveyors across the globe, by 1889 the number of known elements had more than doubled to 70, of the 98 elements observed in nature today. Thus, at the end of the nineteenth century, the elemental complexity of Earth was just coming into focus, but much of the crust, with all its petrological diversity, had never been surveyed by humans. In addition to continental areas that had resisted exploration by the turn of the century, the seafloor was a veritable terra incognita. Even now, an estimated 80 percent of the oceanic crust remains unmapped (Garrison and Ellis 2018).

Clarke (1889, p. 133) acknowledged that "[a]t first sight, it would seem to be impossible to determine the average composition" due to the magnitude of Earth's crust and its extreme heterogeneity. "[Y]et, upon consideration," he concluded, "the question is not seriously complicated." In his characteristically methodical fashion, he started with a series of simplifying assumptions. He posited a thickness of 10 miles for Earth's "known crust." That estimate is less than the range of 20 to 45 miles (about 30 to 70 kilometers) cited today for continental crust but greater than the 3 to 4 miles (6 to 7 kilometers) for the more abundant oceanic crust. As Clarke surmised, a 10-mile-thick stratum was of sufficient volume that much of the befuddling lithological variety—"all veins, pockets, patches, organic substances, etc."-become "trivial constituents" (p. 133). Second, the assortment of sedimentary rocks so vital for the construction of Earth's geologic time scale in the first half of the nineteenth century (Knoll 2021) could also be ignored. They are, after all, merely the weathered detritus of the crystalline rocks.

Although Clarke would tinker with his calculations throughout the remainder of his career, in 1889 his inventory suggested that oxygen (O) composes about 47 weight percent and silicon (Si) about 28 weight percent of Earth's crust, essentially indistinguishable from values that we cite today (e.g., Klein and Dutrow 2002). It was a tour-de-force, and today he is memorialized through the F. W. Clarke Medal, the top award given to early career scientists by the Geochemical Society. Because his dissection of Earth's surface into its component chemicals revealed that two elements (O and Si) make up 75 weight percent of

outer Earth, we might correctly expect that silicon oxidesor *silicates*—will dominate the mineralogy of Earth's surface. Clarke determined that when 7 more elements are factored in (aluminum, iron, calcium, magnesium, potassium, sodium, and hydrogen), 98.5 weight percent of Earth's crust is accounted for (fig. 2). These elements are themselves often bonded with O and Si, and Clarke (1895, p. 7) opens The Constitution of the Silicates with another sweeping inference: "In the solid crust of the earth the silicates are by far the most important constituents. They form at least nine-tenths of the entire known mass [bold added]" Not surprisingly in light of their abundance, silicates also exhibit the greatest diversity of species relative to the other major mineral classes, such as sulfates, phosphates, and carbonates. F. W. Clarke's ceaseless harvesting of geochemical data established the silicate anatomy of Earth's exoskeleton.

Significance of a Silicate Crust

Why does it matter? The fanatical enthusiasm with which Clarke assembled facts and figures into comprehensive monographs is unfashionable today because modern technology makes the compilation of vast databases all too easy. Clarke himself seemed aware of potential criticism, and he justified collecting troves of chemical data by emphasizing their essential role in understanding geochemical and geophysical processes. "Each rock," he wrote in his Data of Geochemistry (1924, p. 10), "may be regarded . . . as a chemical system, in which, by various agencies, chemical changes can be brought about. Every such change implies a disturbance of equilibrium, with the ultimate formation of a new system." Significantly, when Clarke (1889) first proposed the composition of Earth's surface, the existence of atoms was still a matter of vigorous dispute (Chalmers 2019). Clarke (1903) proclaimed his firm belief in atomism based on the detailed mineral analyses on which he and his colleagues



Figure 3. Pseudomorphism of goethite [FeOOH] after pyrite [FeS₂], 7 cm across, from Pelican Point area, Utah County, Utah. John Rakovan specimen and photo.

labored. These studies confirmed the existence of mineral *isomorphism* (crystals having the same structures but different compositions, such as galena [PbS] and halite [NaCl]), *solid solution* (isomorphism within the same mineral family, as with olivines ranging from forsterite $[Mg_2SiO_4]$ to fayalite [Fe₂SiO₄]), and *pseudomorphism* (the replacement of one mineral by another with the retention of the original habit) (fig. 3). Clarke (1903) regarded all these phenomena as explicable only if atoms exist as discrete particles that can substitute for each other when the chemical environment changes.

Indeed, Clarke's support of atomic theory would be validated within a decade by the X-ray scattering experiments of Max von Laue and William Lawrence Bragg in 1912 (Authier 2013). But in the early 1900s the silicate composition of Earth's exterior played a critical and rarely acknowledged role in the development of the most important revolution in the Earth sciences—the birth of plate tectonics. At the same time that European scientists were mapping crystals with X-rays and developing quantum theory to create a language for the smallest dimensions of the physical world, a less-heralded group was building an intellectual framework for the holistic study of the entire Earth, from its core to the outermost atmosphere (Greene 2000). Chief among these was Alfred Wegener (1880-1930), a German scientist trained in geology, geophysics, atmospheric sciences, and astronomy (fig. 4). Though often inaptly characterized as a meteorologist, Wegener never devoted his energies to weather prediction, with the worrisome exception of his service to the German army during World War I to forecast the optimal deployment of chemical weapons. For most of his working life, Wegener attempted to integrate planetary processes over vast scales in a search for connections among the atmosphere, oceans, and lithosphere, pioneering a field that today would be called "Earth systems science."

In early 1911, Wegener was admiring a neighbor's new edition of a large-format atlas that illustrated the submarine shelves rimming the major landmasses. In a revelatory flash, he deduced from the puzzlelike fit of those shelves that a much larger supercontinent must have split apart in the distant past to generate our modern geography. Wegener spent the remainder of his life gathering evidence for the reality of Pangaea and continental drift, charting fossil and rock correlations across oceans and, with the aid of his father-in-law Wladimir Köppen (1846–1940), documenting extreme variations in continental paleoclimate as the landmasses wandered (Greene 2015).

Wegener analogized the plowing of landmasses through lithosphere to icebergs gliding in seawater, but to fortify the comparison, he needed to model the chemistry of Earth's crust. Wegener was heavily influenced by Eduard Suess (1831-1914), an Austrian geologist who speculated that a concentrically layered Earth contained an iron-nickel core enveloped by a magnesium silicate shell (Sima, for siliconmagnesium), which in turn was blanketed by an aluminum silicate crust (Sial, for silicon-aluminum). The chemical compositions and, by extension, the densities of the continental and seafloor silicates deduced by Clarke were essential to Wegener's efforts to develop a mechanism for continental drift. Wegener incorrectly presumed that lighter continental Sial crust (the icebergs) floated in denser oceanic Sima crust (the water), and modern geodynamicists ungenerously criticize him for getting the details wrong (fig. 5). Today we know that both continental and seafloor crusts are aluminosilicate (Sial), and they each float on a dense magnesium silicate mantle (Sima). But Wegener was visionary in inferring that a dichotomy between light continental and dense oceanic crust was mechanically important, and his way of looking at crustal silicate buoyancy within Earth's mantle (based on a principle called *isostasy*) is foundational in plate tectonic theory. By building on Wegener's groundbreaking insights, geologists now know that Earth's silicate exoskeleton controls surface processes through lateral displacements



Figure 4. Alfred Wegener overwintering in the Greenland base "Borg" during J. P. Koch's 1912–1913 expedition. From Wikipedia (https://en.wikipedia.org/wiki/Alfred_Wegener#/media/ File:Wegener_Expedition-1930_008.jpg; archive of the Alfred Wegener Institute, public domain).



Figure 5. Wegener's conception of the internal structure of Earth based on the theories of Eduard Suess. The iron (Fe)-nickel (Ni) core is abbreviated Nife. Today the boundary between the silicon magnesium (Sima) mantle and the core is placed at 5,885 kilometers, not 1,500 kilometers. Silicon aluminum (Sial or Sal) crust was hypothesized to float on the denser Sima mantle. From Wegener (1912). Used with permission of Springer.

of brittle plates, accounting for phenomena ranging from earthquakes to volcanism to perhaps even the emergence of life about 3.5 billion years ago (Kasting 2021).

Answering the questions of when, how, and why silicate rocks were distilled from primordial Earth is at the vanguard of petrological research today. A consensus has emerged that Earth's iron core formed early, perhaps even as the planetesimals were aggregating 4.55 billion years ago to construct our planet (Wood, Walter, and Wade 2006). This so-called Iron Catastrophe resulted from interior heating that smelted ironrich minerals into dense droplets of liquid iron that settled to Earth's center, creating a residual mantle of magnesium silicate that constitutes 84 percent of Earth's volume. The most abundant of these minerals in the lower mantle, and therefore over the entire Earth, is presumed to be bridgmanite [(Mg,Fe)SiO₃], a silicate member of the perovskite fam-



Figure 6. Atomic structure of bridgmanite $[(Mg,Fe)SiO_3]$, the most abundant mineral in the Earth. Red balls: Oxygen. Blue balls: Silicon. Yellow ball: Magnesium/Iron.

ily that might constitute 39 percent of Earth's volume (fig. 6). Though it was synthesized in the 1970s, the discovery and naming of Earth's most common mineral awaited a recent high-resolution analysis of a meteorite that landed in 1879 (Tschauner et al. 2014).

The way in which an aluminosilicate veneer was extracted from this magnesiosilicate mantle is a matter of hot dispute. Scientists are even unsure whether the formation of Earth's crust, which comprises less than 1 percent of our planet, was a cause of, or a consequence of, the inception of plate tectonics (Hawkesworth, Cawood, and Dhuime 2020). Today we recognize approximately 1,400 distinct silicate minerals in Earth's crust, and geochemists have developed a vast array of isotopic clocks to investigate the timeline for their appearance. Silicate minerals such as zircon [$ZrSiO_4$], which can store ele-

mental clues to crustal genesis over billions of years, serve as one of our most important resources for unraveling Earth's history (fig. 7). Can scientists use the chronology of silicate arrivals to discover the origin of large-scale processes in the way that paleontologists have used fossils to infer global events? Inspired by the "Mineral Evolution" paradigm of Robert Hazen (Carnegie Institution for Science), scientists are now asking whether the appearance of specific silicate



Figure 7. Zircon, $2.5 \times 1.7 \times 1.6$ cm, from Astor Valley, Astore District, Gilgit-Baltistan, Pakistan. Rob Lavinsky specimen and photo.

minerals can be tied to the episodic emergence of transformative geophysical, geochemical, and geobiological forces (Hazen 2012).

Mapping Silicate Minerals at the Atomic Scale

Physical distinctions among the families of silicate minerals have been recognized for millennia—presumably, as long as hominins have used them for technology and adornment. Stone tools from West Turkana, Kenya, date as far back as 3.3 million years, predating *Homo habilis* (Harmand et al. 2015). For the next several million years, one type of stone, microcrystalline quartz, dominated the materials industry, and humanity's increasing sophistication in fashioning flint was mirrored by physiological changes that enhanced our ability to fabricate tools (Marzke 2013). In this fashion, silicate minerals guided human evolution.

Rigorous measurements of the physical properties of minerals began with the work of Abraham Gottlob Werner (1749-1817), a German geologist who developed a mineral classification based upon finely delineated external traits. Werner (1774) characterized minerals by their appearance to each sense-sight, touch, smell, sound, and taste-and he divided each of these characters into subcharacters and sub-subcharacters. Mineral qualities discernable by touch, for example, included hardness, solidity, frangibility (brittleness), flexibility, and adhesion to the tongue. The color gray was divided into nine shades (lead-gray, pearl-gray, smokegray, ash gray, and so on). This system proved effective in distinguishing quartz from muscovite (fig. 8), but it was less capable of clarifying subtle distinctions among mineral species within the same family, such as the amphiboles tremolite and actinolite (fig. 9).

A chemically based taxonomy emerged antagonistically to correct the shortcomings of Werner's "natural system," and the Swedish mineralogist Jöns Jacob Berzelius (1779-1848) was the most important champion of this new school. Berzelius's name is unfamiliar to many, but he belongs in the pantheon of favorite scientists for every reader of this magazine (fig. 10). In the early 1800s, Berzelius invented chemical symbols for the elements and introduced the modern chemical formula. He also discovered four elements, and silicon is one of them (or more accurately, Berzelius was the first to isolate silicon in 1823, confirming a hypothesis by the British chemist Sir Humphry Davy that quartz is a compound and not an element). Berzelius argued that compounds represent a marriage between positively and negatively charged chemical species, establishing electrochemical dualism as a cornerstone of modern inorganic chemistry (Levere 2001). Berzelius also recognized that the carriers of negative charge are not always single elements, such as oxygen (O²⁻), but they may involve charged clusters of atoms. He coined the term radical to describe the union of, for example, sulfur and oxygen as the negatively charged sulfate group: $(SO_4)^{2-}$.

Those who organize their minerals by the "Dana System" should know that Berzelius actually was first to propose a mineral classification based on these *negative* chemical components (Berzelius 1826). Only in the fourth edition of his *A System of Mineralogy* (Dana 1854) did Yale professor James Dwight Dana (1813–1895) finally reject Werner's "natural" system in favor of a Berzelian taxonomy (Heaney 2016). Despite Dana's reluctance to convert, his midcentury treatise explicitly articulates the menagerie of negatively charged Si-O radicals that define the silicate minerals. Orthosilicates such as forsterite and zircon were known to exhibit Si:O ra-



Figure 8A (above). Quartz crystal, 10.2 cm wide, from La Gardette mine, Bourg d'Oisans, Isere, France. Bill and Will Larson specimen, Jeff Scovil photo.

Figure 8B (right). Muscovite, 9.5 cm high, from the Pederneira mine, São José da Safira, Minas Gerais, Brazil. Scott Rudolph specimen, Jeff Scovil photo.





Figure 9A. Tremolite, 3.6 cm wide, from Minden, Ontario, Canada.True North Minerals specimen, Jeff Scovil photo.



Figure 9B. Actinolite, 14.2 cm wide, from Arkaroola Bore, Australia. South Australia Museum specimen, Jeff Scovil photo.

tios of 1 to 4 (the $[SiO_4]^{4-}$ radical), whereas in pyroxenes the ratio is 1 to 3 (the $[SiO_4]^{2-}$ radical). Wet chemical analysis, the measurement of mineral formulas through assays of a mineral's dissolved residue, increased in sophistication, and the silicate mineral groups as we know them today—micas, amphiboles, pyroxenes, and so forth—were rigorously differentiated in mineral classifications at the turn of the century.

Nevertheless, the geometrical configurations of Si and O that yielded different ratios for silicate radicals remained a mystery. It was obvious that the three-dimensional arrangement of the atoms in Si-O radicals in some way dictated the physical properties of silicate minerals. Members of the mica family, for example, share unmistakable similarities, such as their pseudohexagonal tabular crystal habits, basal cleavage, and tenacity. Despite the undoubted connection between Si:O ratios and external character, early efforts to relate the proportions of Si and O to a structural model that captured those physical traits were unsuccessful. Mineralogists were hamstrung not only by the unknown nature of the atom, but also by an intellectual framework that imposed principles of solution chemistry on their conception of crystals. Antoine Lavoisier named the element oxygen for its supposed ability to transform metals into acids (oxy from the Greek for "acid" and gen for "birth" equals acid creator). Lavoisier inferred that the combination of oxygen with carbon, nitrogen, and sulfur generated carbonic (H₂CO₃), nitric (HNO₃), and sulfuric (H₂SO₄) acids. By extension, it seemed logical that



Figure 10. Statue of Jöns Jacob Berzelius in the center of Berzelii Park, Stockholm. © Jim.henderson photo (https://creative commons.org/licenses/by-sa/4.0/deed.en).

the coupling of oxygen with silicon would produce acids, and mineral treatises even in the early 1900s were permeated with acid-based theory. (Today, igneous petrologists commonly refer to highly silicic magmas as "acidic" and lowsilica magmas as "basic" in a throwback to this notion.)

The bridge between the external characters of silicate minerals and chemical composition was provided by the physicist William Lawrence Bragg (1890-1971; fig. 11), who collaborated with his father, William Henry Bragg (1862-1942), to unravel the atomic structures of silicates using X-ray diffraction. As one of the principal architects of X-ray diffraction analysis, W. L. Bragg initially focused on chemically simple minerals, such as halite [NaCl], diamond [C], calcite [CaCO₃], and pyrite [FeS], whose structures were solved in the 1910s. As Bragg and West (1927, p. 450) observed, "The difficulty of analysis [of silicates] is due to the complexity of the molecules which enter into their composition, and the low symmetry of most of the crystals; a direct attack on the problem of their structure by the usual type of X-ray examination is not easy." Nevertheless, as both the instruments and the theory for X-ray diffraction were refined by crystallographic successes, Bragg and his collaborators made significant headway through the 1920s, and patterns of behavior among the silicate structures began to emerge.

First, Bragg realized that acid-base theory had little usefulness in silicate crystal chemistry. Unlike other scientists,



Figure 11. William Lawrence Bragg. From Wikipedia (https:// commons.wikimedia.org/wiki/File:Wl-bragg.jpg; published in 1916 in Sweden in Les Prix Nobel 1915; public domain).

Bragg and West (1927, p. 453) noted that "we refer to silicon as a positive ion . . . and not as being the centre of an acid group." In contrast to the tenets of solution chemistry, Bragg realized that even complex silicates should be conceptualized as rigid frameworks of negatively charged oxygen ions (called oxygen anions) that create spaces infilled by smaller silicon ions with a positive charge (or silicon cations). In all of the crustal silicates that Bragg examined, the number of oxygen anions that coordinated a single silicon cation was a small integer that was thrillingly constant: 4 (fig. 12). Because these 4 oxygen anions mutually repel each other, they adopt a geometric configuration that maintains equal distances among them, forming a tetrahedron, the Platonic solid constructed from 4 equilateral triangles (read John Rakovan's Word to the Wise column for more on coordination polyhedra; Rakovan 2023). Bragg marveled that, despite the vast variety of silicate minerals, the modularity of these localized SiO₄ clusters was a recurrent theme, and the tetrahedral shapes outlined by oxygen were nearly ideal. X-ray diffraction of dozens of silicate minerals revealed that the distance between O anions along the tetrahedral edges was always close to 2.6 Ångstroms (or Å, where 1 Å is one



Figure 12. Tetrahedral coordination of Si by 4 O atoms observed in the atomic structures of virtually all crustal silicate minerals.

Compositions of garnets with formula $A_3^{2+}B_2^{3+}Si_3O_{12}$.

| Name | Α | В | Formula |
|-------------|----|----|---|
| Pyrope | Mg | Al | Mg ₃ Al ₂ Si ₃ O ₁₂ |
| Almandine | Fe | Al | Fe ₃ Al ₂ Si ₃ O ₁₂ |
| Spessartine | Mn | Al | Mn ₃ Al ₂ Si ₃ O ₁₂ |
| Grossular | Ca | Al | Ca,Al,Si,O ₁₂ |
| Andradite | Ca | Fe | Ca ₃ Fe ₂ Si ₃ O ₁₂ |
| Uvarovite | Ca | Cr | $Ca_3Cr_2Si_3O_{12}$ |

ten-billionth of a meter). Similarly, the distance between a Si cation at the tetrahedral center and the 4 O anions at the corners (or tetrahedral apices) was always close to 1.6 Å. Even today, when mineralogists assess the validity of a proposed silicate structure, they will reject the model if those O-O and Si-O interatomic distances deviate from these values.

Bragg and West (1927) credit Ferruccio Zambonini (1880–1932) and Edgar T. Wherry (1885–1982)—the former an Italian mineralogist and the latter an American cofounder of the Mineralogical Society of America—with a spectacularly significant insight about the crystal chemistry of silicate minerals: Other metal ions will readily substitute into the sites created by coordinating oxygen anions as long as the size of the hole matches the diameter of the cation. Therefore, a mineral family, such as garnet or pyroxene, exhibits a rigid network of O atoms into which different metal atoms substitute to generate distinct mineral species (see table; fig. 13). In a flash, the existence of silicate "solid solution" (or isomorphism in Bragg's terminology) was solved.

The most important of these substitutions in silicate minerals is the replacement of silicon by aluminum. These two metals are situated adjacent to each other in the Periodic Table, and so their ionic sizes are nearly identical. Moreover, as F. W. Clarke determined, Al is the next most abundant element in Earth's crust after Si. Consequently, aluminosilicates are exceedingly common-in fact, the most common minerals in Earth's crust. Feldspars are minerals with compositions that fall within a chemical triad: orthoclase [KAl-Si₂O₂]—albite [NaAlSi₂O₂]—anorthite [CaAl₂Si₂O₂]. As was first articulated in 1928 by the Austrian mineralogist Felix Karl Ludwig Machatschki (1895-1970), Al cations in feldspars and most other silicates will adopt a tetrahedral coordination in tandem with silicon cations. In this way, AlO₄ units become an integral part of the silicate framework (fig. 14). As an additional complication, the great German mineralogist Paul von Groth (1843-1927) had speculated in the early 1900s that isomorphism can involve not just single atoms but groups of atoms substituting for each other, as long as charge is conserved (Zambonini and Washington 1923). Along the join between albite and anorthite, for example, Na¹⁺ and Si⁴⁺ (total charge of 5+) will swap as a couple for Ca^{2+} and Al^{3+} (total charge of 5+), creating the solid solution known as plagioclase. Plagioclase is the most abundant mineral in basalt, which is the rock that paves the ocean bottom. Because oceans cover 71 percent of our planet's surface, the aluminosilicate plagioclase series [NaAlSi₂O₈-CaAl₂Si₂O₈] is the most abundant mineral group (39 weight percent) in Earth's crust (fig. 15).



Figure 13A. The structure of garnet can be conceived as a network of polyhedra framed by oxygen anions (red spheres) with formula $A_3^{2+}B_2^{3+}S_{13}O_{12}$. Different metal cations can substitute into the centers of the polyhedra. The atomic structure of spessartine garnet is shown with A = Mn and B = Al, yielding Mn_3Al_2Si_3O_{12}.



Figure 13B. Spessartine, 3 cm high, from Shengus, Gilgit-Baltistan, Pakistan. Alex Venzke specimen, Jeff Scovil photo.



Figure 14A (left). Albite, 12.1 cm wide, from Virgem da Lapa, Minas Gerais, Brazil. Steve Smale specimen, Jeff Scovil photo. *Figure 14B (right).* Atomic structure of low albite [NaAlSi₃O₈]. Yellow spheres: Na cations. Dark blue tetrahedra contain Si cations. Light blue tetrahedra contain Al cations. Aluminum-filled tetrahedra reticulate with silicon-filled tetrahedra to create a fully polymerized network.



Figure 15A (left). Pillow basalts on the Pacific seafloor near Hawaii. From Wikipedia (https://en.wikipedia.org/wiki/Basalt#/media/File:Pillow_basalt_crop_l.jpg; courtesy of NOAA, public domain).

Figure 15B (right). Cross-polarized light microscope image of a thin section of basalt from the Black Rock Desert in Central Utah. The gray and white striped crystals that constitute the groundmass are plagioclase. The large crystals are olivine phenocrysts. © Tiffany Rivera, NSF EAR 1940305 (https://creativecommons.org/licenses/by-sa/4.0/deed.en).

Polymerization as the Bridge between Micro and Macro

By 1930, Bragg and his coworkers had solved the structures of enough silicate minerals that the connection between atomic architecture and the physico-chemical properties of the major rock-forming minerals at long last could be grasped. He published a synopsis of his insights in Nature (Bragg 1930a) and a 70-page anthology in the journal Zeitschrift für Kristallographie (Bragg 1930b). The final brick in the conceptual edifice was the realization that silicate mineral groups are differentiated by the way in which SiO₄ tetrahedra connect to each other. Whereas radicals such as the $(CO_3)^{2-}$ or the $(SO_4)^{2-}$ groups always are isolated from one another within carbonate and sulfate atomic structures, [(Si,Al)O₄] radicals are considerably more social. Just as groups of carbon atoms create repeated links to generate the large panoply of organic macromolecules that sustain all life (e.g., proteins, carbohydrates, fats, and nucleic acids), Bragg (1930a, p. 510) recognized that "[i]n the silicates, there often is a similar indefinitely extended linking, but it is always one in which an oxygen atom is interposed between two silicon atoms." (See fig. 16.) In other words, silicates are polymers (another term introduced by Berzelius to describe macrostructures generated by the repetition of a chemical module). The nature of the polymerization network is specific to each silicate family, and the geometry of the polymerization scheme more than any other feature endows a silicate mineral with its physical and chemical properties.

Why do silicates polymerize? An answer to this question had already been suggested by Linus Pauling (1901-1994; fig. 17), a (mostly) friendly rival of W. L. Bragg. Readers of a certain age may remember Pauling from the 1970s as a misguided advocate for the health benefits of massive doses of vitamin C. Scientists nevertheless revere him as the principal engineer of quantum chemistry and molecular biology



Figure 16. Four-ringed network of silica tetrahedra linked via bridging oxygen anions to create part of a polymerized network in the zeolite ZM5.



Figure 17. Linus Pauling in 1955 with an inset of his Nobel Prize in Chemistry. From Wikipedia (https://en.wikipedia.org/wiki/ Linus_Pauling#/media/File:Linus_Pauling_1955a.jpg; The Big T, yearbook of Caltech, public domain).

(Hager 1995). Whereas W. L. Bragg is still the youngest Nobel laureate in the sciences (he was twenty-five years old in 1915), Pauling won two Nobel prizes—one for chemistry (1954) and the other for peace (1962). Despite the complexity of the disciplines that Pauling conceived, he had a gift for explanatory metaphors that enabled high school students to understand sophisticated science.

To account for polymerization, Pauling invented a simple way of quantifying the strength of a bond between a cation and an anion (Pauling 1929). To calculate a Pauling bond strength, one merely divides the charge of the cation by the number of anions that coordinate the cation. It works because the greater the charge of the cation (e.g., Ca²⁺ versus K¹⁺), the greater the electrostatic attraction between the cation and the anion (that is known as Coulomb's law). As more O²⁻ anions are clustered around a single cation, however, basic geometry dictates that the distance between the centers of the cation and each coordinating anion must increase (fig. 18). The attractive forces between charged atoms decrease by the square of the distance between them (Coulomb's law again), so larger coordination polyhedra lead to weaker interatomic bonds. There are no real physical units associated with the Pauling bond strength, though some mineralogy textbooks pretend otherwise through confections such as "evu's," for "electrovalency units." The Pauling bond strength is a simple ratio that captures the essence of the interatomic attractions in a crystal, and it offers surprisingly deep insights into chemical behavior.

For example, Pauling noted that metal-oxygen bond strengths for isolated radicals such as the carbonate $[(CO_2)^{2-}]$ and sulfate $[(SO_4)^{2-})]$ groups were always greater than unity. Carbon is located in column IV of the Periodic Table and



Figure 18. As the number of oxygen anions that coordinate a cation increases (known as the coordination number, or CN), the size of the interstitial space increases, leading to an increase in the distance between the center of the cation and the center of any oxygen anion.

will donate 4 electrons to achieve a noble gas state, achieving a charge of 4+. In the carbonate group, three oxygen anions coordinate each carbon in equilateral triangular geometry. Thus, the Pauling bond strength for the C-O bond is 4/3. Sulfur is a Group VI element, and so each S donates 6 electrons to gain a charge of 6+. In light of the tetrahedral coordination of oxygen atoms about sulfur in the sulfate group, the Pauling bond strength of the S-O bond is 6/4 (fig. 19). Pauling intuited that when these bond strengths exceed unity, electrons are strongly localized between the cation and the oxygen anions, typical of the sharing of electrons that characterizes *covalent bonding* between atoms.

Halite (NaCl) offers a counterpoint. Sodium is an alkali metal in the first column of the Periodic Table, and it donates only a single electron to achieve the noble gas state of neon. In halite, each Na⁺ cation is coordinated to 6 Cl⁻ anions. Thus, the Pauling bond strength for the Na-Cl bond is 1/6, and that jibes with the low Mohs hardness for halite of about 2. Pauling suggested that bond strengths below



Figure 19. The Pauling bond strength (B.S.) for the sulfuroxygen bond is greater than unity, whereas the bond strength for the silicon-oxygen bond equals one exactly.

unity are suggestive of the swapping rather than sharing of electrons between elements. Consequently, these bonds are highly *ionic*, and the crystals can be modeled through the packing of positively and negatively charged spheres.

So what is the Pauling bond strength for the Si-O bond that yokes the atoms in Earth's crust? As a Group IV element just below C in the Periodic Table, Si donates 4 electrons and adopts a charge of 4+. As previously noted, in (virtually) all crustal silicates, the coordination by oxygen is tetrahedral, so the bond strength is 4/4, or exactly 1 (fig. 19). Pauling argued that the precise balancing of electric charge on either side of a metal-oxygen-metal linkage promotes the polymerization of the radical. For example, borates with $(B^{3+}O_2)^{3-}$ radicals also exhibit unitary bond strengths (3/3). Both borates and silicates are excellent glass-formers because the exact electron balance (or homopolarity) on either side of the bridging oxygen atoms in the B-O-B and Si-O-Si bonds promotes polymerization. The original recipe for Pyrex involved a polymerized mixture of $(BO_3)^{3-}$ and $(SiO_4)^{4-}$ groups with alkali metals, creating the important class of borosilicate glasses (Smedskjaer, Youngman, and Mauro 2014).

Bragg (1930b) offers a catalog of different silicate polymerization schemes, and they gratifyingly mirror the silicate classes identified decades earlier through chemical and physical interrogation. A former student of Bragg's named Karl Hugo Strunz (1910-2006) codified those divisions further. Some readers may be familiar with the Nickel-Strunz mineral classification, which was a refinement of the Berzelian/Dana system and is supported by the world's superintendent of mineral taxonomy, the International Mineralogical Association's Committee for New Minerals, Nomenclature, and Classification. For better or worse (and judging by the difficulty it poses to my mineralogy students, perhaps the latter), it was Strunz (1938) who introduced the Greek prefixes by which we often distinguish the major silicate groups. Let's consider these in order of increasing degree of polymerization.

Nesosilicates

Named from the Greek *nēsos*, or "island," and perhaps giving rise to the word *insular*, the silica tetrahedral units in this group are completely isolated from one another. None of the oxygen anions bridge between two silicon atoms, and thus silicate polymerization is absent. The ionic nature of these crystal structures allows for a dense, close packing of anions, and many nesosilicates are compatible with highpressure regimes such as the upper mantle of Earth. The basic silicate unit is represented as $[SiO_4]^{4-}$, and nesosilicate minerals include olivine $[(Mg,Fe)_2SiO_4]$ (fig. 20), garnets [such as pyrope, $Mg_3Al_2(SiO_4)_3$], zircon $[ZrSiO_4]$, and topaz $[Al_2SiO_4(F,OH)_2]$.

Sorosilicates

One must presume that the ancient Greeks had no word for "bow tie," the shape created when two tetrahedra are attached at one apex, because *sōros* translates to "heap" or "pile." Mineral structures within this group contain two silicate tetrahedra that share only a single oxygen. The silicate



Figure 20A (left). In nesosilicates such as forsterite $[Mg_2SiO_4]$, the silica tetrahedra are isolated from each other. *Figure 20B (right).* Forsterite crystal, 3.8 cm high, from Myanmar. Gene and Roselind Meieran specimen, Jeff Scovil photo.



Figure 21A (left). In sorosilicates such as zoisite $[Ca_2Al_3O(SiO_4)(Si_2O_7)(OH)]$, disilicate groups share a single bridging oxygen anion, and in some instances isolated silica tetrahedra also are present.

Figure 21B (right). A gem-quality zoisite, tanzanite, 7.6 cm high, from Arusha, Merelani Hills, Tanzania. Barry Kitt specimen, Jeff Scovil photo.

unit is $[Si_2O_7]^{6-}$, and it is useful to note that the Si:O ratio is 2:7 rather than 2:8 because the bridging oxygen is shared between two silicon cations. Consequently, each Si "owns" only half of that bridging oxygen, so it cannot be doubly counted. The formulas for sorosilicates are always too complicated to memorize: lawsonite $[CaAl_2(Si_2O_7)(OH)_2 \cdot H_2O]$, zoisite $[Ca_2Al_3O(SiO_4)(Si_2O_7)(OH)]$ (fig. 21), and vesuvianite $[Ca_1$ $_0(Mg,Fe)_2Al_4(SiO_4)_5(Si_2O_7)_2(OH)_4]$ are examples.

Cyclosilicates

With this group, the relationship between polymerization scheme and crystal habit becomes apparent. *Cyclo* (or Greek *kýklos*) stands for circle, or ring, and that precisely describes the geometry of the silicate polymers in this class. In cyclosilicates, three (benitoite), four (papagoite), or, most commonly, six silicate tetrahedra join as rings such that the Si:O ratio is 1:3, with $[Si_nO_{3n}]^{2n-}$ as the silicate unit. Beryl $[Be_3Al_2(Si_6O_{18})]$ and



Figure 22A (above). In cyclosilicates such as beryl [Be,Al,(Si,O₁₀)], silicate groups join as rings that are isolated from other tetrahedra. Figure 22B (right). A gem-quality beryl, variety aquamarine, 10.3 cm high, from Shigar Valley, Gilgit-Baltistan, Pakistan. Steve Smale specimen, Jeff Scovil photo.

tourmaline [(Na,Ca)(Al,Li,Mg)₃(Al,Fe,Mn)₆(Si₆O₁₈) (BO₃)₃ (OH),] are well-known cyclosilicate gem materials (fig. 22). They both are built from six-membered rings of silicate tetrahedra, and the prismatic hexagonal and trigonal habits of beryl and tourmaline, respectively, embody the underlying symmetry of the atomic structures.

Inosilicates

В

Even an admiring mineralogist might concede that Strunz faltered here, because the inosilicates (from inos, Greek for "fiber") encompass two silicate classes that each merits primary categorization. Pyroxenes are single-chain silicates characterized by the infinite polymerization of sil-



Figure 23A (left). In single-chain inosilicates, such as the pyroxene diopside $[CaMgSi_2O_6]$, silicate tetrahedra polymerize in a linear fashion.

Figure 23B (right). A 6.5 × 4.5 × 2-cm cluster of diopside crystals from Khugiani, Nangarhar Province, Afghanistan. From Wikipedia (https://en.wikipedia.org/wiki/File:Diopside-jmix07-156a.jpg; Rob Lavinsky photo).



Figure 24A (left). In double-chain inosilicates, such as the amphibole tremolite $[Ca_2Mg_5Si_8O_{22}(OH)_2]$, two-strands of silicate tetrahedra polymerize in a linear fashion.

Figure 24B (right). Tremolite single crystal, 5.2 × 2.0 × 1.3 cm, from Khash and Kuran Wa Junjan districts, Badakhshan, Afghanistan. Rob Lavinsky specimen and photo.

icate tetrahedra via a linear linking, with [Si_nO_{3n}]²ⁿ⁻ as the silicate radical. In amphiboles, two of these tetrahedral links condense into a double-chained silicate, represented by the radical $[Si_{4n}O_{11n}]^{6n-}$. The existence of the single-chain silicate polymers that form the backbone of pyroxenes actually was predicted prior to their confirmation using X-ray diffraction by Machatschki (1928), who inferred that a linear linking of $[(SiO_4)^{4-}]$ tetrahedra would mathematically generate a [SiO₃]²⁻ residue. Moreover, the chainlike skeleton of pyroxenes and amphiboles explains the elongated habit of these minerals. In inosilicates, the silicate chain runs parallel to the long axis of the crystal, a relationship that is most completely manifested in amphiboles, whose habits may range from acicular to asbestiform. Common pyroxenes include enstatite [MgSiO₃] and diopside [CaMgSi₂O₆], and corresponding amphiboles are anthophyllite [Mg₂Si₈O₂₂(OH)₂] and tremolite $[Ca_2Mg_5Si_8O_{22}(OH)_2]$ (figs. 23 and 24).

Phyllosilicates

One measure of the power of a scientific model is its predictive capacity, and again it happened that the sheetlike polymerization scheme for phyllosilicates (from the Greek *phýllon* for "leaf") was predicted by W. L. Bragg prior to his experimental confirmation. Bragg (1930a, p. 510) observed that "No compounds of this type have yet been analysed, but it is interesting . . . [that] . . . linking tetrahedral groups into sheets leads to an arrangement which has hexagonal symmetry, and the dimensions of the network are precisely those of the basal plane of mica . . ." Unbeknownst to Bragg—we presume!—two months beforehand Linus Pauling (1930) had published the structures of talc, pyrophyllite, and the micas as ascertained by X-ray diffraction, and they concurred with Bragg's intuition. Phyllosilicates share a $[(Si_{2n}O_{5n})^{2n}]$ radical, and they are all hydrated, typically via the hydroxyl (OH⁻) group. In addition to the micas, such as muscovite $[KAl_2(Si_3Al)O_{10}(OH)_2]$ (fig. 25) and biotite $[K(Mg,Fe)_3(Si_3Al)O_{10}(OH)_2]$, phyllosilicates include the serpentines $[Mg_3Si_2O_5(OH)_4]$, the chlorites $[(Mg,Fe)_3(Si_Al)_4O_{10}(OH)_2 \cdot (Mg,Fe)_3(OH)_6]$, and the technologically and environmentally important group of clay minerals, such as kaolinite $[Al_2Si_2O_5(OH)_4]$ and montmorillonite $[(Na,Ca)_{0.33}(Al,Mg)_2 Si_4O_{10}(OH)_2 \cdot nH_2O]$.

Tectosilicates

Tekton is the Greek term for "artisan" or "builder," and it finds its way into our language in "architect" and "plate tectonics." Mineralogically, it defines the most fully polymerized silicate class, also known as the framework silicates. The five primary silicate classes (neso-, soro-, ino-, phyllo-, and tectosilicates) contain roughly equal numbers of distinct mineral species, but tectosilicates are easily the most abundant by mass in Earth's crust, representing 75 weight percent of surficial minerals. With the exception of the SiO₂ minerals (also known as the *silica* family), all tectosilicates are aluminosilicates, and $[Al_xSi_yO_{(2x+2y)}]^{x-}$ is the chemical module. In addition to quartz (SiO_2) and the feldspars (such as orthoclase $[KAISi_3O_8]$; fig. 26), tectosilicates are represented by the feldspathoids (such as sodalite $[Na_8(AISiO_4)_6Cl_2]$) and the vast zeolite family (such as analcime $[NaAISi_2O_6 \cdot H_2O]$).

Polymerization and Silicate Crystallization and Dissolution

Polymerization does not merely account for a silicate mineral's physical properties. It also lies at the heart of the pathways by which silicate minerals materialize and break down. At the same time that Bragg and coworkers were





Figure 25B (right). Muscovite and microcline, 5 cm high, from Mantena, Minas Gerais, Brazil. John Rakovan specimen and photo.

using X-rays to unravel the atomic structures of minerals, experimental petrologist Norman Levi Bowen (1887–1956; fig. 27) was investigating the sequence by which silicic magmas evolve into igneous rocks. After receiving a PhD from the Massachusetts Institute of Technology in 1912, Bowen was hired by the Carnegie Institution in Washington, D.C., where he worked until 1937. Systematically, he melted igneous rocks of different compositions in a high-temperature kiln, and then he slowly cooled the melt to a temperature of interest. While the melt was pinned at that temperature, crystals would form and equilibrate with the liquid, at which point Bowen quenched the mass by dropping it in a bucket of





Figure 26B. Carlsbad-twinned orthoclase, 5.7 cm high, from Kienbergsee, Unterbränd, Baden-Wuertemburg, Germany. Gregor Markl specimen, Jeff Scovil photo.



Figure 27. Norman Levi Bowen of the Geophysical Laboratory, Carnegie Institution for Science. From Wikipedia (https://en.wikipedia.org/wiki/Norman_L._Bowen#/media/ File:Norman_Levi_Bowen_(1887-1956).jpg; public domain).

water. The result was a glass, representing the cooled liquid, hosting silicate crystals that were stable at the temperature of equilibration.

Examination of these quenched products revealed a surprising sequence: The first crystals to form, a bit below 1300°C, were olivine $[(Mg,Fe)_2SiO_4]$. If the melt was equilibrated at a lower temperature, however, the olivine reacted with the molten silica to generate a new mineral, pyroxene, via the reaction $[(Mg,Fe)_2SiO_4] + SiO_{2(liquid)} \rightarrow 2(Mg,Fe)SiO_3$. As long as SiO₂ was available in the magma to react, a sequence of minerals appeared and disappeared with decreasing temperature. Bowen's reaction series charts a canonical

crystallization path for silicic magmas, and here is the full sequence for the so-called discontinuous leg: olivine \rightarrow pyroxene \rightarrow amphibole \rightarrow biotite \rightarrow muscovite, feldspar, and quartz (fig. 28).

The astute reader will notice that this mineralization series tracks a progression in silicate polymerization: nesosilicate \rightarrow inosilicate \rightarrow phyllosilicate \rightarrow tectosilicate. Bowen's reaction sequences are not merely observed in laboratory experiments. They are written in stone when magma chambers undergo "fractional crystallization," such as when gravitational settling removes the dense crystals from the liquid as they form. The sinking of one generation of minerals after the next yields a stratified body with olivine-rich rocks (dunites) at the bottom to mixtures of pyroxene and feldspar (pyroxenites) in the middle and mostly plagioclaserich rocks (gabbros) at the top. If the starting magma is sufficiently silica-rich, these "layered igneous complexes" even contain a final crystallization zone of quartz, muscovite, and alkali feldspars, yielding a window of granite. The Bushveld complex in South Africa is a massive, 2-billion-year-old example of a stratified polymerization sequence preserved as an igneous complex. These bodies are targets for economic geologists because they often concentrate platinum-group metals and other ores, such as tin, chromium, titanium, and vanadium (fig. 29). In a confluence of mineralogical and petrological revolutions, Bowen published his findings in a seminal textbook, The Evolution of Igneous Rocks (Bowen 1928), just as Bragg, Pauling, and coworkers were charting the patterns of silicate polymerization in minerals.

Conversely, polymerization dictates the order in which silicate minerals weather. Samuel S. Goldich (1909–2000) completed a PhD at the University of Minnesota in 1936, and his study of the weathering of a granite gneiss, two diabase rocks, and an amphibolite revealed a striking pattern in their vulnerability to the elements (Goldich 1938). Olivine is most susceptible, followed in order by pyroxene, amphibole,



Bowen's Reaction Series

Figure 28. Schematic representation of Bowen's reaction series. © Steven Earle, https://openpress.usask.ca/physicalgeology/chapter/7-2-crystallization-of-magma-2/ (https://creativecommons.org/licenses/by/4.0/).



Figure 29. The Bushveld Igneous Complex at the Mononon River near Seelpoort, South Africa. Black bands are chromitite interbedded with Ca-plagioclase-rich anorthosite rocks (light gray). © Kevin Walsh (https://creativecommons.org/licenses/by/2.0/deed.en).

micas, feldspar, and quartz. This inversion of Bowen's reaction series is known as the Goldich dissolution series, and it makes sense. In order to break down a mineral, water molecules look for weakly defended points of attack. All those homopolar bonds in a highly polymerized network of Si cations inhabit a chemical nirvana and are immune to the wiles of the polarized water molecule. Goldich's series is embedded in every handful of sand from a mature beach replete with quartz, alkali feldspar, and mica grains-heavily polymerized minerals that are resistant to dissolution. Unlike Bowen, Bragg, and Pauling, Goldich received few professional accolades during his lifetime. His eulogists are unusually candid about his dyspeptic personality; one colleague described him as "a sugar-coated pill turned inside out" and "a thorn in everyone's side" (Dodd 2009, pp. 3-4). Nevertheless, Goldich is the yin to Bowen's yang, and his investigations are foundational to the science of soil formation.

Parting Thoughts

It is easy to frame the story of silicates as a heroic narrative that climaxes in the 1920s with the discovery of polymerization as the integration of crystallography, macroscopic mineral properties, and igneous magma dynamics. A modern mineralogist must feel gratitude for the work of Bragg and Pauling, as one can only imagine the elation that nineteenthcentury scientists would have experienced by the simplicity of the solution. It is hard to read the papers of Berzelius and Dana, who struggled so nobly to understand the relationship between silicate chemistry and mineral properties but never reached that promised land. Polymerization turns silicate mineralogy into a game of Legos.

But there are two problems with this narrative. The first is that the game is hardly over. The second is that the game is still not simple. We now know that the "discontinuous" leg of Bowen's reaction series is less segmented than originally thought. Bowen presumed that the recrystallization steps from neso- to ino- to phyllosilicates involved discrete stages of structural reincarnation. As a graduate student at Harvard in the 1970s, however, David Veblen discovered "mineralogical mules" that straddle the single-chain, double-chain, and sheet silicate structures (fig. 30) (Veblen 1991). How many silicate chains must be united to qualify as a sheet? Three? Ten? One hundred? As with organic macromolecules, polymerization networks in silicate minerals can achieve a baroque intricacy, to the degree that classifying some structures within the Strunz system is fraught with ambiguity. Sergei Krivovichev, a professor at the University of St. Petersburg, has quantified the complexity of about four thousand known mineral structures by measuring the number of bits required to store the total structural information content for a given species (Krivovichev 2013). Of the twenty most complex minerals, seventeen are silicates. Moreover, when complex polymer networks are seen in minerals that grow only as large as a few nanometers in diameter, as is the case with many clays and zeolites, crystallographers almost need a new language to describe geology at the edge of crystallinity.

In addition, silicates play a central role in one of the most existential crises that we confront today. To what extent does silicate crust control climate? Global geochemists Maureen Raymo and William Ruddiman (1992) proposed that the uplift of the Tibetan Plateau was the driver of worldwide



Figure 30. Jimthompsonite $[(Mg,Fe)_{10}Si_{12}O_{22}(OH)_4]$ contains triple chains of silicate tetrahedra and is a hybrid of a sheet and a chain silicate.

cooling during the past 40 million years as the result of the drawdown of atmospheric CO₂ by reaction with newly exposed silicate rocks: $CaSiO_3 + CO_2 \rightarrow CaCO_3 + SiO_2$. This reaction is written schematically and not intended literally; wollastonite (CaSiO₂) is not a major crustal mineral, and the composition is intended as a stand-in for any reactive Casilicate, particularly basaltic plagioclase, the (Na,Ca)-rich feldspar. This hypothesis has forced geologists to assess the degree to which active tectonics controls climate through silicate weathering today and in the past. Going deep into Earth's history, did the emergence of silicate crust, which likely modeled the Bowen's reaction sequence over hundreds of millions of years (Hazen et al. in press), also promote changes in global climate? Our knowledge of silicate crystallography allows scientists to map feedbacks between crustal mineralogy, atmospheric dynamics, and oceanic processes as part of the total Earth system. A century after silicate structures were finally resolved, silicate mineralogy remains at the forefront of Earth science research.

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Tourmaline from Tourmaline King Mine, Pala Mining District, California, U.S.A, 21 cm tall, Photo by Jeff Scovil

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