CHAPTER 12.35

Sulfur

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Sulfur is an abundant, multivalent, nonmetal chemical element (symbol S and atomic number 16 on the periodic table). Under normal conditions, sulfur atoms form cyclic octatomic molecules with the chemical formula S_8. At room temperature, elemental sulfur presents as a bright-yellow crystalline solid. Chemically, sulfur reacts as either an oxidizing or reducing agent. Sulfur oxidizes most metals and several nonmetals, including carbon. This leads to its negative charge in most organosulfur compounds. Used as a reducing agent, sulfur reduces several oxidants including oxygen and fluorine.

Sulfur occurs naturally as the pure element (native sulfur); as sulfide and sulfate minerals; and in volcanic gases as sulfur dioxide (SO_2), hydrogen sulfide (H_2S), and sometimes sulfur vapor. Plentiful in native form, sulfur and its uses were known in ancient times in China, Egypt, India, and Greece. Fumes from burning sulfur, which emits a blue flame, were used as bleaches and fumigants, and sulfur-containing medicinal mixtures were used as balms and antiparasitics. Sulfur is referred to in the Bible as *brimstone* (*burn stone* in English) and the name is still in use today, although archaic. In 1777, the French chemist Antoine Lavoisier argued and convinced his scientific community that sulfur was a basic rather than compound element.

Historically, elemental sulfur was extracted from volcanic deposits as native sulfur where it occurred in lumps or veins and occasionally in beautifully crystalline forms. More recently, sulfur has been extracted from salt domes, where it sometimes occurs in nearly pure form, using the Frasch (superheated water) process (Tuller 1954) or it is physically mined as lump sulfur from Poland. Physically mining sulfur, however, has become obsolete because of the high cost of extraction and lower-cost alternative sources. Today, almost all elemental sulfur is produced as a by-product of removing sulfur from natural gas and crude oil.

Sulfur's largest commercial use (mostly after conversion to sulfuric acid) is to produce sulfate and phosphate fertilizers, because of the high requirement of all plant life for sulfur and phosphorus (USGS 1999). Sulfuric acid is also a primary industrial chemical. About 10% of sulfuric acid is produced as the by-product of nonferrous metal smelting, mainly copper, nickel, and, to a lesser extent, lead and zinc. An additional 10% of sulfuric acid production is in the form of recycled/recovered acid from petroleum alkylation units or chemicals production, such as methyl methacrylate (MMA).

Other well-known uses for sulfur are insecticides and fungicides. And although matches are associated with sulfur, they have been almost entirely replaced with butane lighters. Many sulfur compounds are odoriferous, and the smell of odorized natural gas, skunk scent, grapefruit, and garlic is caused by the presence of organosulfur compounds. Hydrogen sulfide produced by living organisms imparts the distinctive odor to rotting eggs and other biological processes.

As an essential element for life, sulfur is extensively used in biochemical processes. Sulfur compounds serve as both fuels (electron donors for CO_2 fixation; Brune 1995) and respiratory (anaerobic) materials in metabolic reactions (electron acceptors; Zhang et al. 2018). Sulfur in organic form is present in the vitamins biotin and *thiamine*, the latter named for the Greek word for *sulfur*. Organically bonded sulfur is a component of all proteins, such as the amino acids cysteine and methionine.

The information presented in this chapter provides an overview of sulfur and its uses, trade, and resources. Almost all sulfur is now the by-product of natural gas and oil production and refining. Large-scale mining of sulfur is almost nonexistent and practiced only by small artisanal miners and in a few legacy locations.

Excellent and up-to-date information is available from the U.S. Geological Survey (USGS), Department of the Interior, which publishes a comprehensive annual summary of sulfur. The *Mineral Commodity Summaries* are updated annually and currently include data through 2015. The *Minerals Yearbook* is currently updated to 2013. Many of the consumption and use figures presented in this chapter are derived from these open USGS sources (1932–2018). In addition, there are other online resources that offer useful references and color images.

PHYSICAL PROPERTIES

Sulfur forms polyatomic molecules with different chemical formulas. The best-known allotrope is octasulfur, cyclo-S_8.

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Octasulfur is a soft, bright-yellow solid with only a faint odor, similar to that of matches. It melts at 115.2°C, boils at 444.6°C, and sublimes easily (Lumen Learning, n.d.). Amorphous or *plastic* sulfur is produced by rapid cooling of molten sulfur—for example, by pouring it into cold water. At 95.2°C, cyclo-octasulfur changes from α-octasulfur to the β-polymorph. The structure of the S_8 ring is virtually unchanged by this phase change. Between its melting and boiling temperatures, octasulfur changes its allotrope again, turning from β-octasulfur to γ-sulfur, again accompanied by a lower density but increased viscosity because of the formation of polymers. At even higher temperatures, however, the viscosity decreases as depolymerization occurs. Molten sulfur becomes dark red at temperatures higher than 200°C. The density of sulfur is approximately 2 $g \cdot cm^{-3}$, depending on the allotrope; all of its stable allotropes are excellent electrical insulators (Lumen Learning, n.d.).

Liquid sulfur is always handled between 130° and 140°C with a viscosity of 7–10 cP. Below this temperature, the viscosity increases gradually; above 155°C, viscosity rapidly increases and the sulfur becomes unpumpable.

CHEMICAL PROPERTIES

Notable for its peculiar stifling odor, sulfur dioxide is formed from vaporized sulfur. Sulfur vaporizes at 445°C and autoignites at approximately 235°C. Insoluble in water, sulfur is soluble in carbon disulfide and, to a lesser extent, in other nonpolar organic solvents, such as benzene and toluene. The first and the second ionization energies of sulfur are 999.6 and 2,252 $kJ \cdot mol^{-1}$, respectively. The +2 oxidation state is rare, however, with +4 and +6 being more common. Because of electron transfer between orbitals, the fourth and sixth ionization energies are 4,556 and 8,495.8 $kJ \cdot mol^{-1}$, and these states are only stable with strong oxidants such as fluorine, oxygen, and chlorine.

Both sulfur dioxide and sulfuric acid are corrosive to many metals and materials. Sulfuric acid, however, has the unusual property of being significantly more corrosive when diluted with water than when nearly pure. Although dilute sulfuric acid requires exotic alloy steels for its containment, concentrated sulfuric acid can be safely stored in carbon steel tanks. This is because sulfuric acid forms a stable iron sulfate film on the carbon steel, and this restricts corrosion.

Polymer-lined tank trucks or railcars, often in unit trains, are commonly used for handling and transporting sulfuric acid. Barges, ships, and occasionally pipelines are used to transport large shipments. To prevent serious chemical burns from contact with the acid, special precautions and protective suits are required when handling sulfuric acid.

NATURAL OCCURRENCES

Sulfur is created inside massive stars, at a depth where the temperature exceeds 2.5×10^9 K, by the fusion of one nucleus of silicon plus one nucleus of helium (Cameron 2013). This synthesis, the alpha process, produces elements in abundance. Sulfur is the 10th most common element in the universe.

According to Harraz (2015), "On Earth, elemental sulfur can be found near hot springs and volcanic regions in many parts of the world, especially along the Pacific Ring of Fire; such volcanic deposits are currently mined in Indonesia, Chile, and Japan," but are of little commercial importance. "Historically, Sicily was a large source of sulfur during the Industrial Revolution."

Native sulfur is geologically produced from sulfate minerals, such as gypsum (calcium sulfate), by anaerobic bacteria in salt domes in the presence of methane and other hydrocarbons. "Significant deposits in salt domes occur along the coast of the Gulf of Mexico, and in evaporative deposits in Eastern Europe and Western Asia" (Harraz 2015). Until recently, sulfur deposits from salt domes were the basis for most commercial production in the United States, Russia, Turkmenistan, and Ukraine. Such sources are now of little commercial importance, and most are no longer worked.

Common naturally occurring sulfur compounds include the sulfide minerals, such as pyrite (iron sulfide), chalcopyrite (copper-iron sulfide), nickel sulfides, cinnabar (mercury sulfide), galena (lead sulfide), sphalerite (zinc sulfide), and stibnite (antimony sulfide); and the sulfates, such as gypsum (calcium sulfate), alunite (potassium aluminum sulfate), and barite (barium sulfate) (Harraz 2015). In some organic-rich environments, sulfates and iron compounds are reduced to iron sulfide by bacterial action yielding small pyrite or marcasite nodules. These are common in the southern United States but are only a curiosity.

SULFUR EMISSIONS AND THE SULFUR CYCLE

The discharge of sulfur compounds and their fate in the atmosphere are important to the world's climate. Many views on the sulfur balance in the atmosphere exist, but the consensus is that anthropogenic emissions of sulfur range between 60 and 100 Mt/yr (million metric tons per year), and these account for approximately 70% of all sulfur emissions. Approximately 65% of the discharged sulfur is from power generation. Organic sulfides from oceanic plankton are estimated to emit between 15 and 40 Mt/yr of sulfur equivalent. Smaller sulfur emissions come from natural fires and biological processes in soils and wetlands. Volcanos discharge between 5 and more than 20 Mt/yr of sulfur, mainly as sulfur dioxide. Volcanic emissions of sulfur dioxide and hydrogen sulfide from an individual eruption can be significant. It is well known that large volcanic eruptions, for example, the 1991 Pinatubo eruption in the Philippines, cause short-term lowering of global temperatures.

Sulfur dioxide has a short life in the atmosphere, with some estimates calculating that it oxidizes to sulfur trioxide (SO_3) in a day. The sulfur trioxide reacts with water vapor to form sulfuric acid mist, which in turn is removed from the atmosphere through precipitation (acid rain) or by absorption on dust particles and sedimentation. Volcanic sulfur emissions may be more persistent than most anthropogenic emissions because of their release at higher altitudes. Hydrogen sulfide emissions are similarly oxidized in the atmosphere and removed.

SULFUR PRODUCTION

Sulfur may be found in its elemental form, not associated with other elements, and historically was obtained in this way. Natural sulfide minerals—metal sulfides—have been sources of sulfur via their oxidation to produce sulfur dioxide followed by conversion to sulfuric acid. Today, virtually all sulfur production is as a by-product of other industrial processes such as oil refining and natural gas production. In these processes, sulfur often occurs as undesired or detrimental sulfide compounds that must be removed, mainly as hydrogen sulfide. The hydrogen sulfide is then converted to elemental sulfur using the Claus process.

The Frasch process has been used for more than a century to extract elemental sulfur from sulfur-bearing porous limestones in the salt dome caprocks of Texas and Louisiana (United States). The caprocks are calcite (limestone) and the basement is halite (salt) overlain by anhydrite ($CaSO_4$). Typically, a 150-mm-diameter hole is drilled from the surface to a depth of approximately 300–900 m, and a string of three concentric pipes is lowered into the hole. The 150-mm-diameter outer pipe is the casing, and the inner pipes are ~75 mm and 25 mm in diameter, respectively. Water heated to approximately 165°C is pumped down the annulus at high pressure between the two larger pipes. The casing rests on anhydrite and is perforated near the bottom and again at some distance up into the sulfur-bearing calcite bed. An annular collar seals the space between the two larger pipes and is positioned between the sets of perforations. Hot water emerging from the upper perforations melts the sulfur. The molten sulfur forms a pool that submerges the lower set of perforations, allowing the liquid sulfur to enter the perforations and rise through the inner annulus. Hydrostatic pressure exerted by the column of hot water causes the liquid sulfur to rise in the annulus. Because sulfur is denser than water, it will only rise about halfway. Compressed air is discharged from the bottom of the 25-mm pipe and mixes with the molten sulfur, sufficiently lowering the density of the two-phase fluid so that it reaches the surface, where it is deaerated and collected.

In another method, superheated water is pumped into a native sulfur deposit to melt the sulfur, and then compressed air is injected to return the 99.5% pure melted product to the surface. Throughout the 20th century, this procedure produced elemental sulfur that required no further purification. However, because of a limited number of such sulfur deposits and the high operating cost, this process has not been employed in a major way anywhere in the world since 2002 (Harraz 2015).

Today, most sulfur is produced by its removal from petroleum, natural gas, and related fossil resources, mainly as hydrogen sulfide but also as more complex organosulfur compounds. Organosulfur compounds, which are undesirable impurities in petroleum, may be upgraded by subjecting them to hydrodesulfurization. This action cleaves the C–S bonds as shown in the following reaction (Harraz 2015):

$$R\text{–}S\text{–}R + 2H_2 \rightarrow 2RH + H_2S \quad \textbf{(EQ 1)}$$

This process forms hydrogen sulfide, which also occurs in natural gas, and it is converted into elemental sulfur by the Claus process. Half of the hydrogen sulfide is oxidized to sulfur dioxide in the Claus process. The two are then reacted at high temperature so that comproportionation occurs:

$$3O_2 + 2H_2S \rightarrow 2SO_2 + 2H_2O \quad \textbf{(EQ 2)}$$

$$SO_2 + 2H_2S \rightarrow 3S + 2H_2O \quad \textbf{(EQ 3)}$$

Large stockpiles of prospective elemental sulfur exist within the Athabasca oil sands in western Canada, owing to the high sulfur content of the sands. Marketing this sulfur, however, is not currently economic because of high shipping costs.

BY-PRODUCT SULFUR AND MINING

In the 1930s, the first sulfuric acid plant treating smelter gases was installed at the Kennecott Utah Copper smelter near Salt Lake City, Utah (United States). This small plant produced only 50 t/d of sulfuric acid, which was sold to the local explosives industry. By the 1950s, the nonferrous industry was installing sulfuric acid plants to reduce emissions—although the impact was small—and produced a useful by-product. For years, sulfide smelting industry emitted most of the sulfur dioxide into the atmosphere through highly visible smoke stacks. Figure 1 shows a typical nonferrous smelter up until the 1980s.

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Figure 1 Large U.S. copper smelter, circa 1978

Although the United States had promulgated regulations to reduce emissions from industrial sources for many years, it was not until the 1970 amendment to the Clean Air Act of 1963 that companies were forced to seriously consider how to reduce sulfur and other air pollutant emissions. By the late 1980s, most U.S.-based smelters had either modernized or shut down. The nonferrous industry in the United States went into a major decline starting around 1970. Many mines and smelters closed, some because of the end of economic mine production and others because of the high cost of meeting ever more stringent air quality regulations. In the United States in 1970, there were more than 30 copper smelters; today, there are only 3.

Worldwide, the recovery of sulfuric acid from nonferrous smelters has dramatically increased over the last 40 years. Starting in the 1970s, several new smelters were constructed in Japan and Europe as part of the post–World War II industrialization. These smelters all achieved high sulfur capture using sulfuric acid plants and more modern smelting technologies than was practiced in the United States and elsewhere. Sulfur capture was greater than 90% and approached 98% at some smelters. The by-product acid found a ready local market.

In the United States, sulfuric acid production at copper, nickel, lead, molybdenum, and zinc roasters and smelters accounted for approximately 7% of total domestic production of sulfur in all forms and totaled the equivalent of 620,000 t of elemental sulfur in 2010 (Apodaca 2015–2017). Three acid plants operated in conjunction with copper smelters, and several more were by-product operations at lead, molybdenum, and zinc smelting and roasting operations. The three largest by-product sulfuric acid plants, in size and capacity, were associated with copper smelters and accounted for 86% of the by-product sulfuric acid output (Apodaca 2015–2017). The copper producers Asarco, Rio Tinto Kennecott Utah, and Freeport-McMoRan Copper and Gold each operated a sulfuric acid plant at their primary copper smelters.

Except in some countries such as Chile, Russia, and parts of Africa, most nonferrous smelters around the world are now achieving greater than 90% sulfur capture. The latest copper smelting technology, for example, can capture more than 99.9% of the input sulfur, making copper smelting a minor source of sulfur dioxide emissions in almost every developed location. In contrast, some smelters in Russia have almost no sulfur capture because of the enormous problems of sulfuric acid transport in the Arctic north (e.g., at Norilsk in Siberia). Some of these smelting complexes emit more than 2 Mt/yr of sulfur dioxide compared to a few hundred metric tons per year at the most advanced smelters. Chile, the world's largest copper producer, has lagged in sulfur emissions control, and many smelters are still achieving less than 80% sulfur capture, making these among the dirtiest smelters in the developed world. China, in contrast, has embraced environmental control in their newest smelters and achieves world-class performance. But many small and dirty smelters remain in China, seemingly immune to normal market and regulatory forces. Today, China treats more than 40% of the copper concentrate produced at the world's mines.

Although production of sulfuric acid from smelter and roaster off-gas is relatively easy, the capital cost is significant, and the product is difficult to ship for long distances unless there is ready access to barges or ships. Generally, the production of sulfuric acid at smelters is not a money-making proposition, although there are many exceptions to this generality. In the United States, the acid produced by the three copper smelters in the West is consumed locally, often for ore leaching, but some is shipped as far as Florida for use in phosphate production.

In Japan, where all the copper smelters achieve very high sulfur capture, the acid produced now exceeds domestic requirements. This has necessitated shipping sulfuric acid to Chile or Peru, where much of the copper concentrate that is smelted in Japan originates, where it is used in oxide copper leaching. It is anticipated that China will soon have a surplus of sulfuric acid, which could further depress sulfuric acid demand and prices.

U.S. CONSUMPTION

Apparent domestic consumption of sulfur in all forms has remained nearly stable with growth in the 2%–4% range, depending on fertilizer demand, which is often cyclical. Of the sulfur consumed, 65% is obtained from domestic sources as elemental sulfur (60%) and by-product acid (5%). The remaining 35% is supplied by imports of recovered elemental sulfur (26%) and sulfuric acid (9%) (Apodaca 2015–2017).

Sulfur differs from most other major mineral commodities in that its primary use is as a chemical reagent rather than as a component of a finished product. This use generally requires that it be converted to an intermediate chemical product prior to its initial use by industry. The leading sulfur end use, sulfuric acid, represents 66% of reported consumption with an identified end use. Although reported as elemental sulfur in many sources, it is reasonable to assume that nearly all the sulfur consumption reportedly used in petroleum refining was first converted to sulfuric acid, bringing sulfur used to produce sulfuric acid to 85% of the total sulfur consumption (Apodaca 2015–2017).

Practically all of the sulfuric acid production in the world is through the contact process, where sulfur dioxide (SO_2) is oxidized to sulfur trioxide (SO_3) over a vanadium-based catalyst and absorbed into circulating strong sulfuric acid. The source of SO_2 gas is either off-gas from a nonferrous smelter or generated by burning sulfur (or a sulfur-containing compound) in a furnace. Since the mid-1970s, most new acid plants have been built in the double-contact, double-absorption configuration to achieve reduced tail gas emissions of SO_2.

Because of its desirable properties, sulfuric acid retains its position as the most universally used mineral acid and the most produced and consumed inorganic chemical, by volume. Data based on USGS surveys of sulfur and sulfuric acid producers show that reported U.S. consumption of sulfur in sulfuric acid (100% basis) between 2012 and 2013 increased by 8%, and total reported sulfur consumption increased by 7% (Apodaca 2015–2017). The reported increase in sulfuric acid consumption can be attributed to a threefold increase in sulfuric acid use in petroleum refining and other petroleum and coal products. Some inconsistencies exist in reported sulfur consumption and use figures, so this information is not considered particularly accurate.

Agriculture is the leading sulfur-consuming industry, mainly for the production of fertilizers, primarily phosphate fertilizer. Approximately 45% of domestic phosphate fertilizer production is exported. The second-ranked end use for sulfur is in petroleum refining and other petroleum and coal processing. Demand for sulfuric acid in copper ore leaching, which is the third-ranked end use, has decreased in recent years.

Adding to the problem of quantifying the production of sulfur and sulfur chemicals is the practice of recycling some sulfuric acid, mainly that used in petroleum refining and explosives and chemical production. Some acid is recycled internally by companies and some is sent to recyclers who regenerate sulfuric acid. Data on this part of the industry is very hard to obtain and verify and could comprise 10% of the total sulfuric acid production.

PRICES

The price of sulfur is highly influenced by location and cyclical demand for fertilizer. Prices vary greatly on a regional and international basis, and it is recommended that previously mentioned USGS publications be referred to in any effort to understand current trends.

During periods of poor agricultural commodity prices, the agribusiness sector and small farmers elect to not use fertilizers. This can be sustained for a few years, but the drop in yield and recovering commodity prices typically spurs a demand in fertilizer. Usually, the highest reported prices in the United States are from Tampa, Florida, because of the large demand for sulfur for fertilizer production using local phosphate deposits in the central Florida area.

At year-end 2013, U.S. West Coast prices ranged from US$93 to $98/t (Apodaca 2015–2017). Nearly all the elemental sulfur produced in some regions, such as the West Coast, is processed at sulfur-forming plants to form sulfur prills, which are necessary to make solid sulfur acceptable for shipping overseas. The cost for prilling is significant. Recently, world sulfur prices generally were higher than domestic prices. This is evidenced by the Abu Dhabi National Oil Company (ADNOC) price. Although prices can vary according to locations, providers, and types of sulfur produced, the ADNOC contract price is recognized as an indicator of world sulfur price trends. The ADNOC price averaged approximately US$120/t in 2013, with price ranging from a low of US$70/t in August and as high of US$160/t in April (Apodaca 2015–2017). As a rule, the sulfuric acid price is typically one-third the sulfur price.

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CHAPTER 12.36

Talc

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INTRODUCTION

The objective of this chapter is twofold: (1) to review the traditional mineral processing technology used by the talc industry and (2) to introduce and discuss the new technologies that are being implemented by talc producers in response to a changed marketplace.

Fifty years ago or so, talc processing was an easy manufacturing process because of an abundance of high-quality talc ores and relatively simple end-user applications. For instance, talc products were used as fillers in unsophisticated ceramic articles or in cosmetics and body powder. Nevertheless, the talc industry was reasonably profitable.

Today, the markets for talc have completely changed. Additionally, the good-quality ores are being depleted around the world, especially in China. Many simple end uses for talc are gone and have been replaced by applications that require the talc products they buy to be produced using state-of-the-art quality control programs and to comply with difficult-to-meet specifications. The talc industry worldwide continues to consolidate. The profitability of talc companies will be poor unless a substantial portion of their total revenues comes from value-added talc products that are targeted to enhance important properties of sophisticated end products manufactured by the plastics, specialty technical ceramics, and coatings industries.

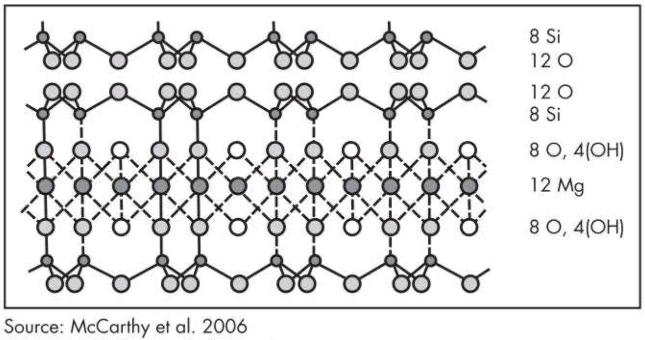
Even with major new world-class talc deposits being developed in Afghanistan and Pakistan in the last decade, it is challenging for talc producers to find talc ores that—when beneficiated the traditional way—would yield end products that meet the new market requirements. Consequently, talc producers have made significant investments to upgrade beneficiation, micronizing, and compaction capability to fulfill the market demands and also to alleviate the pressure of having to high-grade mines. These separation technologies, together with advances in wet grinding, fine particle sizing, surface coatings of talc particles, and talc synthesis, are discussed in this chapter.

The mining, processing, and applications of value-added talc products is a sophisticated process involving geologists, mining engineers, mineral processing engineers, chemical engineers, surface chemists, applications specialists, and a solid commercial group. In some instances, the applications are unique to talc and even to specific talc deposits. Indeed, for the most part, the market for talc is heavily dependent on the scientific research knowledge and technical skills of the producer. Without such application development efforts, new markets are very difficult to penetrate. This chapter borrows some content from the “Talc” chapter written by McCarthy et al. (2006) in *Industrial Minerals and Rocks* published by the Society for Mining, Metallurgy & Exploration (SME).

The Mineral Talc

Talc is a crystalline hydrated magnesium silicate of the general formula $3MgO\text{-}4SiO_2\text{-}H_2O$ (see Figure 1), which theoretically is 31.7% MgO, 64.5% SiO_2, and 4.8% H_2O. It is one of the layer silicates such as kaolin and mica. Talc ores can occur as a relatively pure mineral or as a mixture with other minerals. In its pure form, talc is colorless or appears white, but when the ore contains other minerals, it can also appear green, light green, yellowish, pink, or even black. Accompanying minerals include magnesite, dolomite, chlorite, calcite, and quartz.

The adjacent layers of silica are very weakly bonded with only van der Waals forces, and this allows talc to be easily sheared along this plane. This gives talc its natural slippery



Source: McCarthy et al. 2006

Figure 1 Talc crystal structure

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