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# Preface

Two… four… six… eight…

In the most humble opinion of we two typical biogeochemists, everyone should have a favorite element. Ours is silicon. Did you just leap to a conclusion? Computer chips? Solar cells? They're neat, but not why we appreciate… silicon!

We heart silicon because it's at the heart of silica.

Silica is a compound, nominally $SiO_2$. Silicon dioxide you could call it, if you felt like sounding technical, or silicic acid, when it is dissolved in water. In any event, as molecules go, silica has versatility and a habit of participating in feats of derring-do.

Yes, feats of derring-do, and we don't mean the sci-fi dreams of silica-based life forms. What silica does in real life is cooler than a Horta. If you knew, you'd heart silica, too. That's what the next ten chapters, each telling a scientific tale of silica, are designed to do- make you fall in love with silica.

Silica was there, for example, when life began on Earth. In fact, it wasn't just there, it was key. It was the basis of the silicate rocks that reacted with hydrothermally warmed seawater to produce the solutes (dissolved substances) that reacted with each other and with metals to become the metabolic reactions at the core of all Terran life.

Much later on in geologic time, in the guise of stone tools made and used, silica helped to steer the evolution of human hands, cleverness, and ability to create and comprehend technology, music, mathematics, and complex compound sentences. This enabled us to become what we are today—intelligent and dexterous enough to have, for example, discovered, understood, and put to ubiquitous use in modern technology the piezoelectric properties of silica in the form of quartz crystals (if you press on them the right way, they give off electricity). Sonar, ultrasound, radios, telecommunications, you name it, it probably depends on quartz's piezoelectric effect.

During the billions of years in between the origin of metabolism and the invention of the quartz oscillator, some rudimentary animals, unicellular critters, and land plants developed the ability to biomineralize silica, producing microscopic scales, shells, skeletons, and liths featuring shapes, pores, lattices, grooves, spikes, and processes too fantastic to be matched by any mere human glassmaker (not even the venerable Blaschkas famous for their glass flowers and other equally incredible botanical and zoological glass models). More impressive yet, silica biomineralizers need no flame or furnace. They make their glass at room temperature or cooler. Incidentally, silica biomineralization is so common and widespread that, despite our best (unintentional) agricultural efforts to distill silica out of soils and into sewage systems, the world in your immediate vicinity teems with tons of microscopic, biomineralized glass.

Part of the reason that silica biomineralization is so common is that silica is everywhere. You can't throw a stone without hitting a silicate rock except maybe in the middle of a city (and even there, they're lurking immediately below the pavement, as the granite of fancy countertops, and on the outer walls of grander buildings). The water is full of dissolved silica, too, making it unsurprising that even vertebrate animals like ourselves, who don't biomineralize silica, have a true nutritional need for it. Our skeletons would be weak and malformed, our hair and skin would be a wretched mess, our organs would be falling apart, and our wounds would not heal if we hadn't kept up an adequate daily intake of dietary silica so far throughout our lives. Thank not only our water, but our beer and the plants we eat (especially grains). They're all full of easily digestibly absorbable silica.

All this silica cycling through lakes, rivers, the ocean, and the biosphere came from the dissolution of the silicate minerals that constitute silicate rocks, new ones of which are continually being produced through volcanism. The craziest thing about this slow dissolution of silicate minerals, which is known as chemical weathering, is not that it wipes out mountain belts (although it does), but that it is key to maintaining the pleasant, temperate habitability of Earth. When silicate minerals dissolve, they convert carbon dioxide from the atmosphere into carbonate salts that eventually end up in the ocean. In the longer term, this compensates for the steady release of carbon dioxide out of the magmatic interior of the Earth via such things as volcanoes and hot springs. Without silicate weathering, the surface of the Earth would have long ago baked itself sterile through a runaway greenhouse effect.

Silicate weathering will also mop up all the carbon dioxide we've spewed out via the burning of fossil fuels and forests too. Unfortunately for us, this will take more than 200,000 years. On the other hand, human beings are inveterate tinkerers. We have already found a way to speed up the chemical weathering of silicate minerals that, if it were to be deployed across broad swathes of the warm, wet tropics, could put some brakes on the global warming and climate change we've unintentionally unleashed.

Read on, please, for all the siliceous details.

Lund, Sweden

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# Acknowledgements

Between the two of us, we (the authors of this book) have acquired 60 years of scientific experience of silica by working in limnology, oceanography, and biogeochemistry and by rubbing elbows with ecologists, microbial ecologists, paleoceanographers, and other Earth scientists. That's 60 years of learning *a lot* about silica from our own work and thinking and *even more* from that of hundreds of colleagues. It would take pages to name names. But, those of you who are reading this, you know who you are, especially those of you who have been especially special to us and the world of silica. We thank you for your passion for silica, for the diligence of your work, for the strength of your insights, for the excellence of your publications and presentations, for your guidance and generosity, and for the depth of the discussions we've had with you. Because of you, it's a great field to work in. Extra hats off go to those of you who read, commented on, and caught typos in early versions of the chapters and gave permission for figures to be printed in this book.

On a more practical level, this book could not have been written without the generous support from the Knut and Alice Wallenberg Foundation and from the Swedish Research Council (VR).

The friendly, stimulating, and coffee-, tea-, and cake-fueled environs of the Lund University Geology Department were no obstacle either. We thank all of you who are and were there for making it a great place.

# Contents

# Chapter 1
# A Brief Introduction to the Players

Any undertaking on silica requires first setting a few terms straight, namely silicon, silica, silicic acid, silicate, and silicone. Even scientists get these words confused. So here it is stripped down to bare bone: *Silicon* is the element. *Silica* refers to a tetrahedron formed by one silicon atom bound to four oxygen atoms or to material consisting pretty much entirely of such tetrahedra. *Silicic acid* (also known as dissolved silica and dissolved silicate) consists of silica tetrahedra dissolved in water. *Silicate* is anything that contains silicon in a compound that acts as a negatively charged species (this means silica tetrahedra and silica tetrahedra where atoms of elements like aluminum have substituted in for some of the atoms of oxygen). *Silicone* covers a whole slew of silicon-containing organic compounds, generally man-made (and put to a variety of uses, such as lubricants, sealants, and novelty ice cube trays).

That quick rundown is enough to get you through the book, meaning you could skip straight from here to the first silica story (Chap. 2). But if you want to know a little bit more about these different silicon-containing materials and why they behave as they do, you should brave the rest of this chapter (which, by dealing with definitions, is the least exciting chapter of the book).

## 1.1 Silicon

Silicon is the name of the chemical element that sits on the periodic table between aluminum and phosphorus, underneath carbon, and above germanium, and at the center of an X formed by boron, nitrogen, gallium, and arsenic. All of these are fairly abundant elements, as is typical for the lighter end of the elemental spectrum. But silicon has most of them beat; it's the eighth most common element in the universe.

Silicon is even more common here on Earth, especially in the crust that we live on, where it is second in abundance only to oxygen. There's also no escaping

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silicon on Mercury, Venus, and Mars, on the Moon and the moons of Jupiter, or in roughly 96% of meteorites, either. You walk on silicon, breath it in, drink it up, and work with it all the time. In short, you've never spent a day of your life without being in contact with nontrivial amounts of it in some form or other.

## 1.2 Silica

But, despite its abundance, you've probably rarely encountered silicon on its own. Unless you are prone to making silicon wafers or computer chips (or smashing open electronic devices or solar cells), you could go your whole life without seeing silicon in its elemental form, which is a strangely lightweight, approximately silver-colored metal. In nature, such free, atomic silicon exists as the tinted darkness within smoky quartz and possibly not much else. Silicon prefers to be bound to oxygen, something there is no shortage of here on Earth.

Alone together, silicon and oxygen are known as silica. Under most surface Earth circumstances, the form they take is four oxygen atoms defining the corners of a triangular pyramid at whose heart sits a single atom of silicon, as sketched in Fig. 1.1 (where the dark lines represent the chemical bonds between silicon and the oxygen atoms and the dashed lines outline the pyramid). This silica tetrahedron forms the basis for most silicate minerals and it exists for two reasons. First, silicon, like carbon but unlike most other elements, has four bonds that it needs to fill with the help of other atoms. But, second, the four oxygen atoms that volunteer for the cause repel each other. The pyramid shape of the silica tetrahedron is the balance struck by the repulsive forces of the four oxygen atoms bound to the central silicon.

But a silica tetrahedron is more than just a stable chemical pyramid. It, if you can stomach the anthropomorphism, aches for more for even the four oxygen atoms leave a silica tetrahedron incomplete. Just as each silicon atom needs to engage

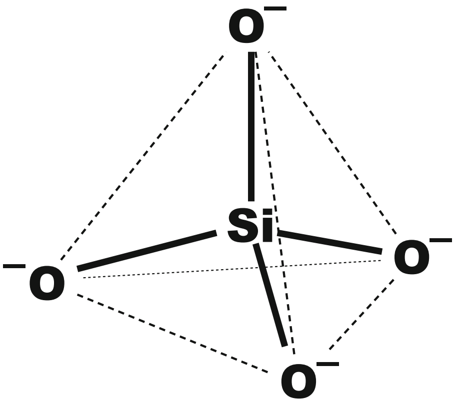

**Fig. 1.1** The silica tetrahedron. The *solid lines* indicate bonds between atoms. The *dashed lines* outline the tetrahedral shape of the ensemble

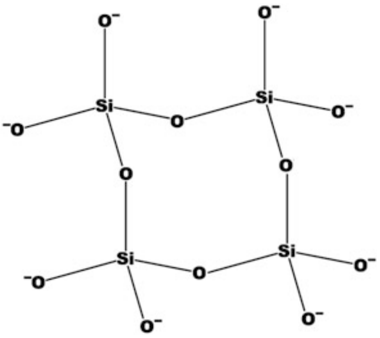

**Fig. 1.2** Silica tetrahedra joined to form silica

itself in four bonds, each oxygen atom needs to engage itself in two. This means each oxygen atom in a solitary silica tetrahedron is unsatisfied; none of the oxygen atoms in a solitary silica tetrahedron have more than their single bond with the central silicon. One solution to this is for a number of silica tetrahedra to come together so that each oxygen atom becomes shared by two silicon atoms, as in Fig. 1.2. Extraneous oxygen atoms are dispensed with until there are, on average, only two oxygen atoms for every one silicon atom. This yields a bigger, more complex silica, also known as silicon dioxide, also known as $SiO_2$.

"But!" you are saying and we hear you. No matter how many silica tetrahedra join together, unfulfilled oxygen atoms will remain at the periphery, as can also be seen in Fig. 1.2. In the presence of water, many of these terminal oxygen atoms pick up a hydrogen atom as their second bond, making for a number of OH groups (called hydroxyl groups) at the edges. That, finally, is enough to satisfy them.

## 1.3 Silicic Acid

Silica completely dissolved in water becomes a bunch of liberated silica tetrahedra whose oxygen atoms have each hooked up with a hydrogen atom (Fig. 1.3). This dissolved silica has the most fearsome name of silicic acid and its basic chemical formulation can be written as $Si(OH)_4$.

But silicic acid is not really fearsome. You drink it all the time and you've got it running through your veins (Yawn).

As with many acids, what makes silicic acid an acid isn't that it bores holes through things á la alien ectoplasm but that it can lose one or more of its hydrogen atoms to exist as a charged species (also known as an ion). In an alkaline solution, which is one that has a high pH and therefore a low concentration of hydrogen ions ($H^+$ in chemical notation), some of the $Si(OH)_4$ molecules let go of one hydrogen to become the silicate ion $SiO(OH)_3^-$ plus a free $H^+$. This addition of $H^+$ to the solution acidifies the solution by increasing its hydrogen ion concentration, lowering its pH. Thus is silicic acid an acid.

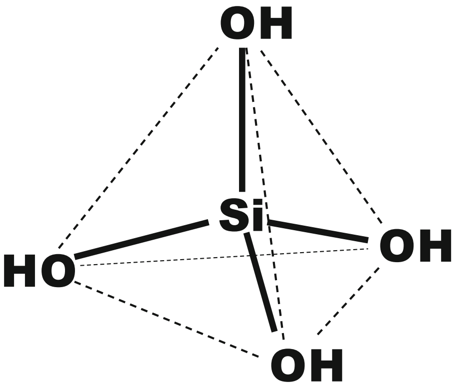

**Fig. 1.3** Silicic acid, aka dissolved silica

In the book, we'll tend not to call silicic acid *silicic acid* but instead *dissolved silica*. Because we can. But also because it underscores the fact that dissolved silica is not a solid, but a solute.

## 1.4 Silicate

The silicates are where things start to get complex and we go a little bit nerdy on you. Bear with us!

Because of its four lonely, negatively charged oxygen atoms, the silica tetrahedron forms the basis for the wide variety of silicate minerals that make up the silicate rocks that constitute almost the entirety of the Earth's crust.

As pointed out in the brief rundown at the beginning of this chapter, silicates are materials that have silicon-containing units of negative charge. Anything that contains a silica tetrahedron fits this bill. This includes not only fairly pure silica like quartz and significantly more hydrated silica like opal, but also clays, serpentines, micas, feldspars, zeolites, garnets, zircons, pyroxenes, and amphiboles, just to name several mere broad categories of minerals.

These rock-forming silicate minerals can be incredibly different from one another in terms of their physical properties, chemical behavior, and appearance. Think of a wet clay, kaolinite, for instance. It has a smell, it can absorb water, it can dissolve in water, it is moldable, at least until it has been fired into a rigid form. Now think of a quartz crystal. It is transparent, hard, odorless, crystalline, and it takes not quite literally forever to dissolve in water. Both of these two minerals have silica tetrahedra as their basic basis. The difference between them is possible because of the chemistry of the silica tetrahedron, in large part due to the extra bond each corner oxygen desires. Depending on what ions are around to fulfill the oxygen atoms and on other factors, like the temperature and pressure at the time the mineral is forming, incredibly different interlocking (or not interlocking) mineral frameworks are possible. Silica tetrahedra may be linked to silica tetrahedra in chains, rings, or sheets, and these chains, rings, or sheets may be anchored together by regularly, repeatedly placed ions of other elements.

Without getting seriously into mineralogy, we'd like to give you a taste for this. While you could make it to the end of your days without knowing anything about this and suffer no ill effects, it is cool to pick up a rock and be able to imagine what lies within in terms of the crazy three-dimensional organization of its atoms. Plus, the underlying silicate structure of minerals is why they are what they are and they do what they do.

The simplest silicate structure consists of silica tetrahedra that are isolated from one another and thus not bonded together into a rigidly connected framework. To give one example, this is the case for the mineral olivine, which, when gemstone quality, is called peridot. In olivine, the disconnected silica tetrahedra are arranged in an orderly fashion within a sea of magnesium and/or iron atoms, depending on the type of olivine. A pure magnesium olivine, $Mg_2SiO_4$ is forsterite. A pure iron olivine, $Fe_2SiO_4$ is fayalite. And there are plenty of mixtures in between.

Believe it or not, the framework of olivine has global implications. Its loose, open structure makes it easy to dissolve and this dissolution, as with the dissolution of all silicate minerals, consumes carbon dioxide. Thus olivine's ease of dissolution makes it an ideal candidate for removing some of the excess carbon dioxide we've spewed into the atmosphere (a topic which is explored at the end of the final chapter of this book). This could help us avoid some of the global warming and climate change that is bearing down upon us, our descendants, and all other organisms on the planet.

The next step up is when silica tetrahedra bind together to form a regular, repeating, and predictable (that is, crystal) framework, such as in the case of quartz. But, visually speaking, the chains, double chains, rings, sheets of rings, and three-dimensional frameworks of rings of silica tetrahedra interbedded regularly with other ions are where things get interesting geometrically speaking.

You could go to any old mineralogy textbook or to any of dozens of websites to see the detailed molecular structures of specific silicate minerals (and you should; it's interesting). Plus the one thing this silica book isn't about is silicate mineralogy. So we decided to go the artistic route. Here's our pitch for what cool quilts, coloring books, or paintings silicate structures could inspire.

Take the cyclosilicates (Fig. 1.4). This family of minerals includes the beautifully blue, exceedingly rare, and correspondingly pricey benitoite (the very beautiful official gemstone of the state of California) and the beryl you may know as emeralds. In these cyclosilicate minerals, a regular number of silica tetrahedra link together to form rings (three tetrahedra in the case of benitoite; four, five, or even six in the case of other minerals, as you can see clearly in the geometric representation of beryl in Fig. 1.4). These rings of linked silica tetrahedra are isolated from the other rings of silica tetrahedra in the minerals and held in place within the crystal structure of the mineral by their bonds to other elements, hence the lovely regularity of the spacing between what looks like six-pointed stars in Fig. 1.4.

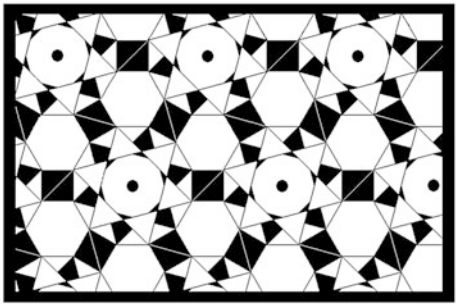

**Fig. 1.4** An artistic representation of the structure of the cyclosilicate mineral beryl

Why does this happen? Those needy oxygen atoms. Because no matter how many silica tetrahedra you link together, there will always be some oxygen atoms that still need their second bond and this is where the other elements come in. In beryl, each six-tetrahedra ring consists of six silicon atoms and 18 oxygen atoms. This leaves 12 of the oxygen atoms in the ring lacking their desired second bond. The shortfall is filled by three beryllium atoms (which each seek to make two bonds) and two aluminum atoms (which each seek to make three).

This is all represented in Fig. 1.4. The aluminum atoms sit at the center of the rings of silica tetrahedra, as represented by the small, dark circles. The beryllium atoms sit between adjacent rings and, illustrated two-dimensionally, the bonds they have with neighboring rings are the diagonally bisected black squares anchoring the rings. The rings of six smaller sized white triangles at the surface of the image as it faces you are the rings of silica tetrahedra, as are the rings of six black triangles just beneath them. Unseen are all the rings beneath them. Who knew such complex tranquility could lurk within a crystal gemstone?

Sheet silicates, where rings of silica tetrahedra link directly together to form sheets of silica, are also pretty cool, although slightly less bedazzling (Fig. 1.5). In serpentine minerals (including the half-dozen referred to as asbestos), clay minerals (such as kaolinite, the main constituent of porcelain; vermiculite, friend to gardeners; and talc, friend to babies' bottoms but enemy to women's ovaries), and micas (including biotite and the sheety medieval Russian windowpane material muscovite), numerous flat sheets of networked rings of silica tetrahedra are

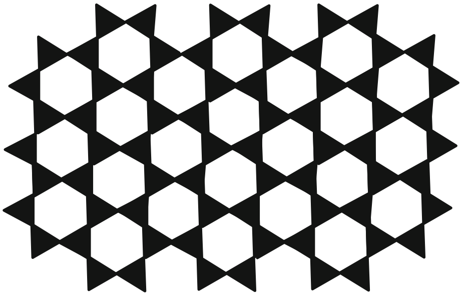

**Fig. 1.5** A stylized representation of a sheet of linked rings of silica tetrahedra

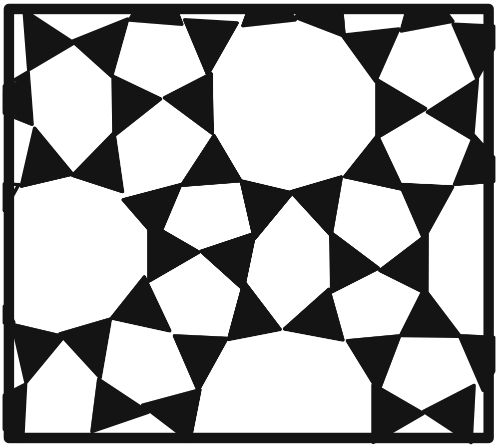

**Fig. 1.6** A two-dimensional, suitable-for-quilting representation of the silica tetrahedra framework of a simple zeolite mineral

sandwiched between layers of things like aluminum and oxygen and are anchored by strategically located hydrogen atoms. What our simplified artistic rendering (Fig. 1.5) shows is a face on view of one sheet of networked rings of silica tetrahedra.

Zeolite minerals are also quite quiltable, although two-dimensional representations of zeolites, such as Fig. 1.6, fail to do them justice. Zeolite minerals, both naturally occurring and designed, consist of three-dimensional frameworks based on silica ring structures with connections on several layers. Their most noteworthy feature is spiraling stacks of rings of silica tetrahedra. These stacks are like microscopic tunnels through the mineral and can act like filters or traps for ions. They are what make zeolites so good at catalyzing chemical reactions and adsorbing pollutants. Industrially, zeolites are indispensable. But you probably know zeolites best as non-clumping cat litter; all those tunnels also make them great at absorbing liquid and entrapping malodorous entities like ammonia.

These and so many more are the ways that silica tetrahedra may be linked together with themselves and other ions to form all the different possible silicate minerals. But as this is a silica story book, not a silica coloring book, we will leave it here and hope two things: that you've picked up some appreciation for the behavior of the silica tetrahedron and that someone reading this will pick up the ball and run with it and start the art of silicate quilting.

## 1.5 Silicone

Before this chapter ends, we need to mention silicone. As a term, silicone covers a variety of polymers based on repeating units of Si–O–Si mixed in with atoms like carbon and hydrogen. Silicone crops up a lot in modern life, for instance, as Silly Putty and silicone caulk, grease, oven mitts, bakeware, and breast and testicle implants. There are silicone hydrogel contact lenses, as well, and silicone is common in shampoo and hair conditioner. With a list like this, silicone obviously has tales to tell. But silicone is not silica (meaning it is not made up of silica tetrahedra), so aside from a brief mention of what happens when silicones get into sewage, we won’t be sharing its stories here.

# Chapter 2
# The Origin of Life Was Brought to You in Part by Silicate Rocks

Beginning at the beginning sounds like a good call. Or, if not at *the* beginning, at least at a big beginning. Will the origin of life on Earth do? Silica was more than there. Together silicate minerals, seawater, and hydrothermal heat set in motion a surprisingly simple sequence of chemical reactions that resulted in Earth's first metabolism, the first major hurdle overcome in the development of life.

Metabolism is a word that can make you think of the need to exercise and of those people, damn them, who can eat whatever they want to without piling on the pounds. But metabolism means much more than the rate at which calories are burned. Yes, the catabolic biochemical reactions of metabolism generate energy by oxidizing organic matter (made up of carbohydrates, lipids, and proteins). But metabolism also consists of anabolic biochemical reactions that consume energy during their construction of organic matter (namely the organic compounds that make up living organisms).

The initial invention of a metabolism capable of building things up as well as burning things down was a pretty big deal. Not only did it tick the first box of the instruction sheet *Basic Requirements for Life*, metabolic pathways tend to be long, convoluted chains of chemical reactions that start from a specific point and end at specific point, along the way producing specific products from specific ingredients.

Many metabolic pathways are complex enough to be the chemical equivalent of circular assembly lines. They take up a small compound and incorporate it into a larger molecule. They then either produce or consume energy while rearranging the larger molecule multiple times, as if solving a Rubik's cube. Eventually they break up the now rather rearranged larger molecule, eject the desired product, and rearrange the remaining materials to resurrect the enzymes and/or other molecules they started with. Then the cycle repeats.

As an example, take Fig. 2.1. That revolving wheel of chemical reactions, which is spinning continuously inside pretty much each and every one of your living cells, is the sort of thing a typical metabolic cycle entails. It is a complex masterpiece that managed to get itself invented out of chaos and disorder.

It would seem to be a miracle. But when you sit down and ponder the simple chemical reactions that would have been going on between water and silicate rocks

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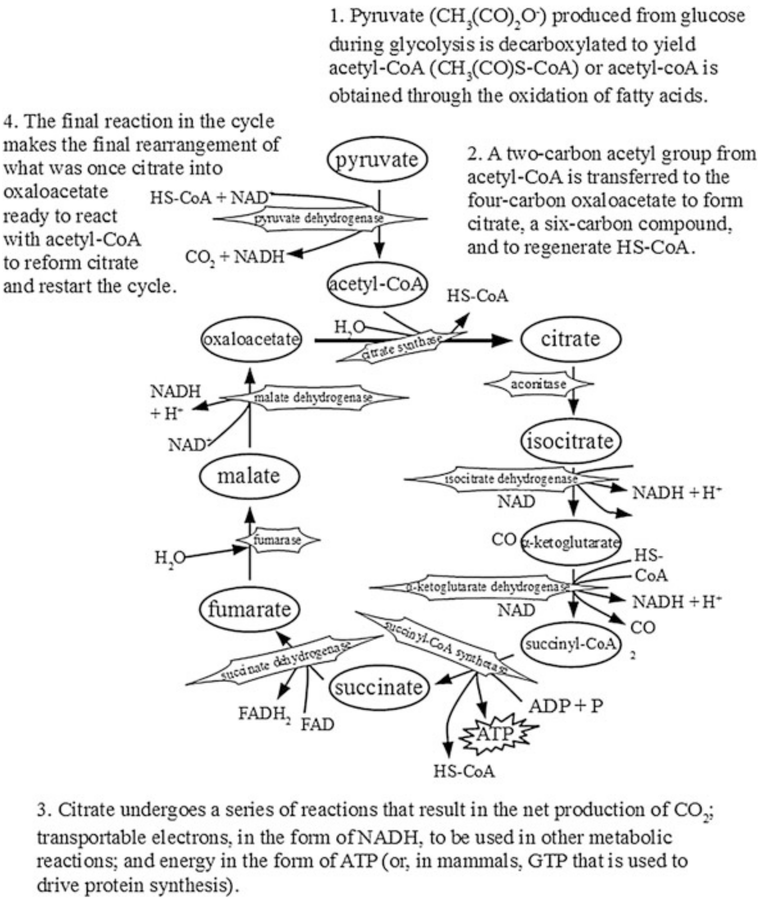

1. Pyruvate ($CH_3(CO)_2O^-$) produced from glucose during glycolysis is decarboxylated to yield acetyl-CoA ($CH_3(CO)S$-CoA) or acetyl-coA is obtained through the oxidation of fatty acids.

2. A two-carbon acetyl group from acetyl-CoA is transferred to the four-carbon oxaloacetate to form citrate, a six-carbon compound, and to regenerate HS-CoA.

3. Citrate undergoes a series of reactions that result in the net production of $CO_2$; transportable electrons, in the form of NADH, to be used in other metabolic reactions; and energy in the form of ATP (or, in mammals, GTP that is used to drive protein synthesis).

4. The final reaction in the cycle makes the final rearrangement of what was once citrate into oxaloacetate ready to react with acetyl-CoA to reform citrate and restart the cycle.

**Fig. 2.1** All aerobic life on earth uses the citric acid cycle to generate energy by oxidizing acetate

on the flanks of undersea mountains approximately four billion years ago, the invention of metabolism starts to seem unavoidable.

And so we shall begin with this big beginning, or rather, a little bit before it because metabolism didn't invent itself in a vacuum. Metabolism came about on Earth shortly after the Earth's formation. The conditions that prevailed at that time set both requirements and constraints upon how life could have initially unfolded.

## 2.1 Setting the Stage

In the early days of our Solar System, there was no Earth nor any other planets, just a large, thin disk of matter rotating about the developing Sun. That disk consisted of gases and dust bumping, crashing, and crunching into each other. Sometimes when things collided, they, or at least bits of them, got stuck together. Because bigger bodies are harder to smash to pieces than smaller ones, bigger bodies began to accumulate until the inner Solar System came to be a mess of smash-ups of perhaps half a thousand planetesimals on conflicting paths. Ka-blam! Ka-blam! Ka-blam!

Most meetings resulted in the larger subsuming the smaller. Each crash added to a to-be-planet's mass and continued to reduce the overall number of objects in the inner Solar System (sometimes 1 + 1 = 1). Eventually, by this process, the crowded chaos of tiny bodies within the inner Solar System tidied itself up into the four terrestrial planets that are still there today (Mercury, Venus, Earth, and Mars) plus an asteroid belt.

The final smash for the almost-but-not-quite-Earth was with Theia. Theia, a proto-planet the size of Mars, was the smaller of the two. They may have been otherwise kindred, as surmised from the similarity in the composition of the present-day Earth and present-day Moon, which may be mostly made of Theia. If this is the case, it is likely that for millions of years, the proto-planet siblings had shared an orbit around the Sun. Accreting from the material that had accumulated at this same distance from the Sun would have meant the two protoplanets were made from materials of similar elemental and isotopic composition.

Locked in by the opposing pulls of the gravity of the Sun and the gravity of the proto-Earth, Theia either led or followed the proto-Earth by 60° as they orbited about the Sun.[1] The geometric side effect of this was that Theia circled the Earth (and vice versa) exactly one time for each full run around the Sun. So stable was this co-orbitation, Theia should still be there today, more than four and a half billion years down the line. (That would make for a very different story, one of life emerged on two similarly habitable planets and by now either learning to get along or fighting each other to death.)

But Theia is no longer there, just ahead of or just behind the Earth in orbit about the Sun. Thank the gravity of some passing planet, probably Jupiter, whose massive, wandering mass wreaked havoc in the Solar System in its early days. Tugged out of whack, Theia's orbit relative to the proto-Earth went from loopy but sustainable to simply dangerous. On one wobbly turn around the proto-Earth, Theia struck.

It's safe to say that none of us can really, fully grasp it, not even the modelers who study it using computer models and the laws of physics. It was no game of billiards. When one planetesimal struck the other, they didn't just briefly deform and ricochet. They demolished, shattered, melted, vaporized, fused, and ejected. These were the repercussions for the materials that made up Theia and the proto-Earth.

One long-standing theory for what then happened goes like this: The iron-rich core of Theia, too heavy to escape the gravity of the proto-Earth after impact, was trapped and sank to merge with the iron-rich core of the proto-Earth. To this day,

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[1] If the Earth was at 6 o'clock on an almost perfectly circular orbit whose center was the Sun, Theia would have been at either 4 o'clock or 8 o'clock.