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# Geochemistry of Sedimentary Ore Deposits

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With 149 Figures

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*On the front cover: Pyrite-cemented breccia in shale-hosted  
barite deposit of Arkansas.*

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# Preface

This book is an outgrowth of my interest in the chemistry of sedimentary rocks. In teaching geochemistry, I realized that the best examples for many chemical processes are drawn from the study of ore deposits. Consequently, we initiated a course at The University of Cincinnati entitled "Sedimentary Ore Deposits," which serves as the final quarter course for both our sedimentary petrology and our ore deposits sequence, and this book is based on that teaching experience. Because of my orientation, the treatment given is perhaps more sedimentological than is usually found in books on ore deposits, but I hope that this proves to be an advantage. It will also be obvious that I have drawn heavily on the ideas and techniques of Robert Garrels.

A number of people have helped with the creation of this book. I am especially grateful to my students and colleagues at Cincinnati and The Memorial University of Newfoundland for suffering through preliminary versions in my courses. I particularly thank Bill Jenks, Malcolm Annis, and Dave Strong.

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January 1983

J. Barry Maynard

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## CHAPTER 1.

# Introduction

"I should see the garden far better," said Alice to herself, "if I could get to the top of that hill: and here's a path that leads straight to it—at least, no, it doesn't do *that*—" (after going a few yards along the path, and turning several sharp corners), "but I suppose it will at last. But how curiously it twists! It's more like a cork-screw than a path! Well *this* turn goes to the hill, I suppose—no, it doesn't! This goes straight back to the house! Well then, I'll try it the other way."

Alice in *The Looking Glass*  
From Lewis Carroll, *The Complete Works of Lewis Carroll*. (New York, Vintage Books, 1976, page 156.)  
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Ore deposits come in a bewildering range of types, and their study involves almost all subdisciplines of geology. Accordingly, it has been difficult to cover the subject matter of economic geology satisfactorily in one book or in one course. In practice, some sort of restriction is made. For North American geologists, there has been a tendency, now fading, to regard economic geology as synonymous with the study of hydrothermal deposits. At one time this was carried to the extreme of insisting that almost all deposits were, in fact, hydrothermal. More recently, the importance of sedimentary processes in ore genesis has begun to be appreciated, but the sedimentology necessary for the understanding of these processes is hard to integrate with the standard economic geology curriculum. It seems to me that a workable solution to this problem is to make divisions along the lines of the traditional areas of geology. Thus, one can approach volcanic ores from the viewpoint of a volcanologist, sedimentary ores from that of a sedimentologist. The ores are then treated within the context of the surrounding rocks, rather than as isolated entities. This book attempts to fill this purpose for sedimentary ores.

Most sedimentary ores are formed by chemical processes, and these processes are the subject of this book. Accordingly, I have largely excluded placers. Ores are defined to be economic accumulations of metals, and I have not discussed non-metallics. Most non-metallics have an evaporite origin, a style of deposition well covered elsewhere (e.g., Braitsch 1971). The remaining metallic deposits



form a more or less coherent group whose precipitation is mostly controlled by Eh gradients. That is, a particular metal dissolves at, say, low Eh, then migrates along the gradient to be precipitated at high Eh, or vice-versa.

What, then, is meant by "sedimentary ores"? How do they differ from deposits classed as "syngenetic" or "strata-bound"? I use sedimentary in the sense of "formed by sedimentary processes." Almost all such ores are also strata-bound, but many strata-bound ores seem to have been formed by hydrothermal processes (see Chapter 7). Similarly, many sedimentary ores are syngenetic, that is, formed at the same time as the surrounding sediment. But there is also an important class of syngenetic ores that is volcanic in origin (see Chapter 8), and some sedimentary ores are epigenetic. It is useful to make the following distinctions (based on Tourtelot and Vine 1976):

*Sedimentary*—Formed by sedimentary processes, at low temperatures and pressures near the Earth's surface, hosted by sedimentary rocks or soils.

*Strata-bound*—Enclosed by essentially parallel layers of sedimentary rocks. May be cross-cutting within a layer.

*Syngenetic*—Formed at the same time the sediment was deposited. Sedimentary examples are iron and manganese ores, volcanic examples are the Kuroko-type Cu-Pb-Zn ores.

*Early diagenetic (or syndiagenetic)*—Formed after deposition of the sediment, but while still in contact with overlying water via diffusion through pore fluids. Pyrite in modern sediments obtains its sulfur in this way.

*Late Diagenetic*—Formed after the sediment is closed to the overlying water, but from metals derived from within the same stratigraphic sequence. Some U deposits in tuffaceous rocks may have this origin (Chapter 6).

*Epigenetic*—Formed after consolidation of the sediment and usually cutting across the strata; metals supplied from outside the system. Two varieties may be distinguished:

*Ground water-epigenetic*—Formed by cold surface waters. An example is roll-front U.

*Hydrothermal-epigenetic*—Formed by hotter ( $>100^{\circ}\text{C}$ ) solutions, usually from depth. Carbonate-hosted Pb-Zn deposits are mostly of this type.

Two tools for the study of the geochemistry of ores that are used extensively in this book need some introduction: stable isotopes and equilibrium diagrams. The stable isotopes of S, C, and O are proving to be invaluable in the study of sediments, and I have tried to include the data on these isotopes that apply to sedimentary ores. For a general introduction to stable isotopes in geology, see Hoefs (1980); here I will briefly review some of the principles.

Stable isotope analyses are presented on a relative scale of so-called delta values. That is

$$\delta = 1000(R_{\text{sample}}/R_{\text{standard}} - 1),$$

where  $R$  is the ratio of the amount of the heavy to that of the light isotope. Thus, for sulfur,

$$\delta^{34}\text{S}/1000 = (^{34}\text{S}/^{32}\text{S}_{\text{sample}} / ^{34}\text{S}/^{32}\text{S}_{\text{standard}}) - 1,$$

from which it can be seen that a sample with a ratio identical to the standard will have a delta value of 0 permil, a sample with more of the heavy isotope will have a positive delta, and one with more of the light, a negative delta. The terms light and negative or heavy and positive tend to be used interchangeably. For each element of interest, an arbitrary standard has been chosen. For S the choice was troilite of the Canyon Diablo meteorite, for O "standard mean ocean water," for C a fossil belemnite. Except for seawater, these primary standards are practically exhausted and measurements are made with secondary standards, but all results are reported in terms of the original standards.

In sedimentary systems, wide ranges of delta values are encountered, reflecting large fractionations. For S and C, biologic activity is the most important cause of fractionation; for O it is evaporation and precipitation of water. Fig. 1-1 shows that S in seawater, which is mostly in the form of  $\text{SO}_4^{2-}$ , has a narrow range of values around +20, but S in sedimentary rocks, which is mostly present in  $\text{FeS}_2$ , is much more variable and can be considerably more negative, a result of bacterial sulfate reduction. C (Fig. 1-2) shows a similar biological effect: marine carbonates are around 0 permil (as might be expected from the use of carbonate C from a marine fossil as a standard), but organic matter, both living and fossil, is appreciably lighter. Thus, carbonates whose carbon comes from oxidized organic matter can be detected by their light C. For O (Fig. 1-3), meteoric waters are noticeably lighter than seawater, which is caused by a preference for the lighter isotope during evaporation of water (see Hoefs 1980, fig. 31). O isotopes are also affected by temperature. As diagenesis proceeds and temperature in-

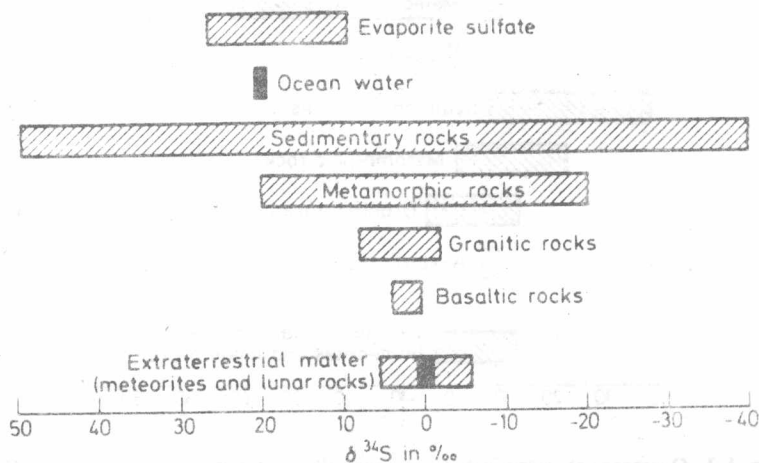


Fig. 1-1. Range of S isotopes in geologic systems. Note the difference between mantle-derived S and sedimentary sulfides (Hoefs 1980, fig. 12).

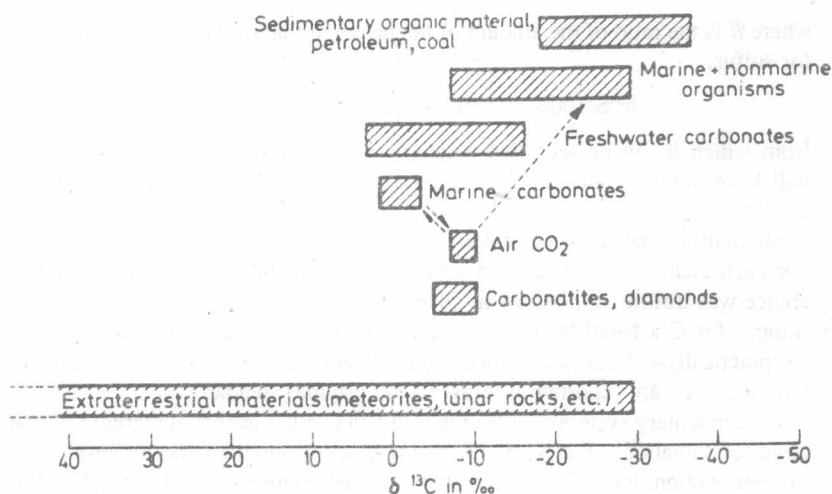


Fig. 1-2. C isotopes in geologic systems. Carbonate carbon derived from seawater is much heavier than organic carbon, and hence than carbonate formed by oxidation of organic matter (Hoefs 1980, fig. 9).

creases, O in minerals tends to become lighter. Thus it is vital to distinguish the timing of mineral formation when studying O isotopes.

The isotope ratios of these elements in seawater and sediments have changed with time, which has an effect on the values found in ore deposits. Changes in  $\delta^{34}\text{S}$  of sulfate in evaporites, which is almost identical to that in coeval seawater, are particularly well documented (Holser and Kaplan 1966, Schidlowski and

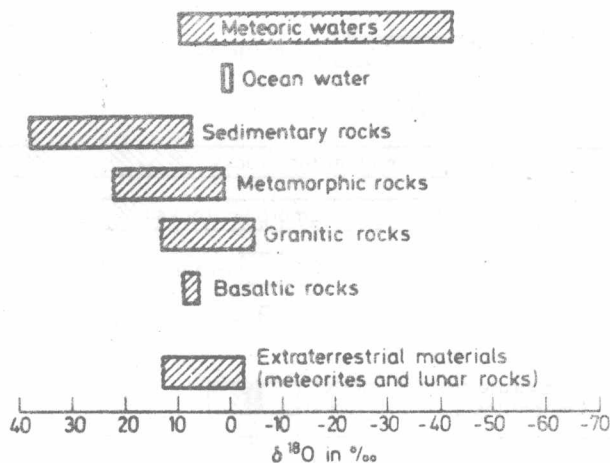
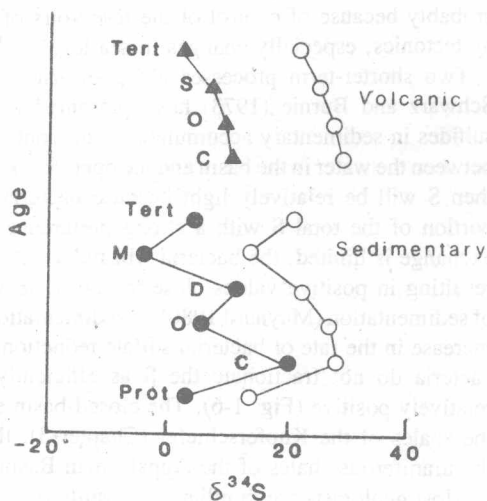


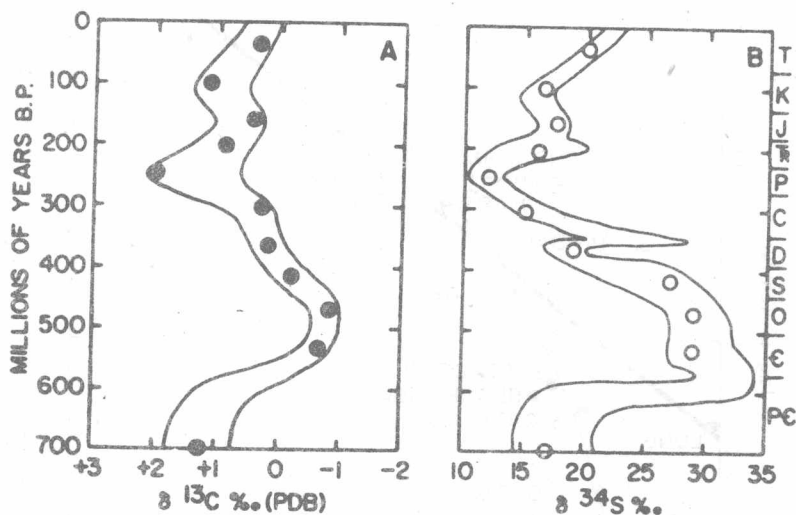
Fig. 1-3. O isotopes of meteoric water are generally lighter than those of seawater (Hoefs 1980, fig. 10).

**Fig. 1-4.**

S isotopes of evaporites and massive sulfide deposits show a parallel trend with time, suggesting that much of the S in the sulfides comes from seawater (after Sangster 1968, fig. 8).



others 1977, Claypool and others 1980). Strata-bound sulfide deposits show a striking parallelism to this seawater curve (Fig. 1-4), providing strong evidence for the participation of seawater in their genesis. Veizer and Hoefs (1976) documented similar but smaller changes in C and O isotopes with time, and Veizer and others (1980) have shown that the S and C curves are correlated (Fig. 1-5),



**Fig. 1-5.** Variation of C and S isotopes of carbonates and sulfates with time. Lower Paleozoic rocks have relatively light C and heavy S, indicating that a high proportion of the available C was deposited as carbonate, of the S as pyrite. Since the Carboniferous, the trend has been towards more organic C and sulfate S (after MacKenzie and Pigott 1981, fig. 3).

probably because of control of the reservoirs of reduced and oxidized S and C by tectonics, especially changes in sea level (Mackenzie and Pigott 1981).

Two shorter-term processes also can affect the S isotopes of an orebody. Schwarz and Burnie (1973) have presented a model in which S isotopes of sulfides in sedimentary accumulations are controlled by the degree of exchange between the water in the basin and the open sea. If exchange is rapid and complete, then S will be relatively light because bacterial reduction will only involve a portion of the total S with a strong preference for  $^{32}\text{S}$ . If, on the other hand, exchange is limited, the bacteria will reduce all of the S coming into the basin, resulting in positive values close to coeval seawater. The other control is rate of sedimentation (Maynard 1980). If sedimentation is rapid, there is a concomitant increase in the rate of bacterial sulfate reduction. At high rates of reduction, the bacteria do not fractionate the S as efficiently, so the resulting sulfides are relatively positive (Fig. 1-6). The closed-basin effect seems to be important for the shales of the Kupferschiefer (Chapter 3), the sedimentation-rate effect for the uraniferous shales of the Appalachian Basin (Chapter 6).

Most geologists are familiar with equilibrium diagrams showing relationships between mineral phases and variables such as Eh, pH, temperature, or composition of the fluid. The standard treatment of this subject, which contains several applications to sedimentary ores, is Garrels and Christ (1965). Another more recent reference with a good short summary of these principles is Froese

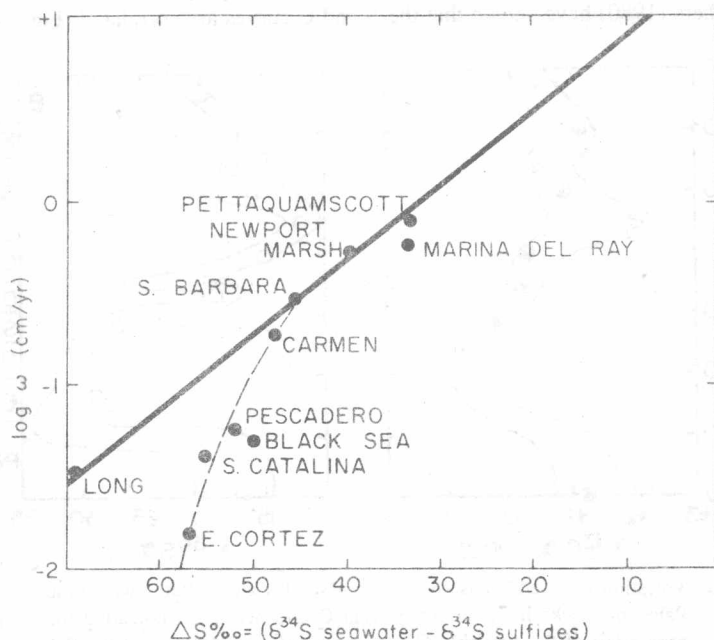


Fig. 1-6. Fractionation of S between seawater and sulfides increases as the rate of deposition ( $\omega$ ) decreases, up to a spread of approximately 50 permil (Maynard 1980, fig. 2).

(1981). Here I would like to make a few points about the use of such diagrams and sources of data for their construction. Any thermodynamic relationship can only apply when the system under investigation is at equilibrium, a situation notoriously rare in sediments. Why, therefore, are equilibrium diagrams used so often in this book? Such relationships tell us what *should* happen in a system, and thus provide a first approximation of its geochemical behavior. Then, by comparing this first model with observation, we can decide what other factors need to be considered. Usually these are reaction rates, an aspect of geochemistry that is, as yet, poorly understood. I have presented a few examples of kinetic studies for Mn and U, and have tried to identify other areas that could be studied in the same way.

Thermodynamic data for minerals are still in a state of flux, particularly for Al-bearing phases. I have, wherever possible, used the compilation of Robie and others (1978). Other sources, along with the values selected for use in the diagrams, are given in the Appendix. Please note that in this book the new international convention for expressing energy in joules (or kilojoules) rather than calories is followed. To convert, use the factor

$$1 \text{ joule} = 0.239 \text{ calories.}$$

Similarly, the equation relating the equilibrium constant for a reaction ( $K$ ) to the change in free energy ( $\Delta G_r$ ),

$$\Delta G_r = -RT \ln K,$$

becomes, at 25°C,

$$\Delta G_r = -5.707 \log K.$$

If the reaction involves oxidation and reduction, the standard electrode potential ( $E^\circ$ ) is also used:

$$\Delta G_r = 96.487 n E^\circ$$

where  $n$  is the number of electrons, and  $\Delta G_r$  is expressed in kilojoules. These relations can then be used with the standard equations for free energy change

$$\Delta G_r = \Delta G_f(\text{products}) - \Delta G_f(\text{reactants})$$

where  $\Delta G_f$  represents the free energy of formation of the substance from the elements, and the Nernst equation

$$E_h = E^\circ + (0.059/n) \log(a_{\text{oxid}}/a_{\text{red}})$$

where  $a_{\text{oxid}}$  and  $a_{\text{red}}$  are the activities of the oxidized and reduced species, to construct the type of diagrams used in this book. To use the Nernst equation in the above form, write the reactions so that the oxidized species are products. For instance,



Note that the electron should always appear on the right-hand side.

In cases where no thermodynamic data are available, there are several schemes

**Table 1-1.** Free energies of silication of various oxides to be used in calculating free energies of formation of layer silicates by the method of Tardy and Garrels (1974).

	Kcal	Kj
K <sub>2</sub> O	-184.32	- 771.21
Na <sub>2</sub> O	-162.8	- 681.2
MgO	-151.51	- 633.9
CaO	-171.0	- 715.5
FeO	- 64.6	- 270.3
Fe <sub>2</sub> O <sub>3</sub>	-177.50	- 742.68
Al <sub>2</sub> O <sub>3</sub>	-383.2	-1603.3
SiO <sub>2</sub>	-204.6	- 856.1
H <sub>2</sub> O	- 57.8	- 241.8
Mg(OH) <sub>2</sub>	-201.7	- 843.9
K <sub>2</sub> O exchange	-184.32	- 771.21

for making approximations. I have followed that proposed by Tardy and Garrels (1974), but see Lippmann (1977) for a critical treatment of these approximations. Basically, Tardy and Garrels' approach uses known values of free energy for sheet silicates to estimate the free energy of Al<sub>2</sub>O<sub>3</sub>, SiO<sub>2</sub>, etc., within lattices of layer silicates, then uses these values to calculate free energies for unknown cases by simple summation. In order to use this approach in conjunction with the other free energy values in this book, it is necessary to correct their numbers slightly to account for different accepted values for their "known" minerals. To do so, use the numbers in Table 1-1. For example, a pure iron chamosite might have the composition Fe<sub>2</sub>Al<sub>1.43</sub>Si<sub>1.43</sub>O<sub>5</sub>(OH)<sub>4</sub>. Its free energy of formation would then be estimated by first writing the formula as the sum of the oxides



The free energy is then

$$2(-64.6) + 0.72(-383.2) + 1.43(-204.6) + 2(-57.8) = -813.28 \text{ Kcal,} \\ \text{or } 3402.85 \text{ Kj.}$$

If a mineral contains Mg, first assign as much as possible to Mg(OH)<sub>2</sub>, then the remainder to MgO.

Such calculations can give only a crude approximation of free energy of formation of a mineral, but reasonable values usually result, keeping in mind the uncertainties in the "well-known" values. Further, the numbers so obtained can be used with some confidence in establishing the topology of equilibrium diagrams even if absolute values for the parameters plotted on the axes remain somewhat uncertain. Thus, the relationships among minerals can be explored, but there are limitations in comparing observed mineral assemblages with measured or inferred water chemistry.

## CHAPTER 2.

# Iron

"Would you tell me, please, which way I ought to go from here?"  
"That depends a good deal on where you want to get to," said the Cat. "I don't much care where—" said Alice. "Then it doesn't matter which way you go," said the Cat. "—so long as I get somewhere," Alice added as an explanation. "Oh, you're sure to do that," said the Cat, "if you only walk long enough."

Alice and the Cheshire Cat in *Alice in Wonderland*  
From Lewis Carroll, *The Complete Works of Lewis Carroll*. (New York: Vintage Books, 1976, pages 71-72.)  
Reprinted with permission of the publisher.

Iron has been mined from a variety of kinds of deposits, but production is now almost entirely from two types: banded iron-formations and oolitic ironstones. Banded iron-formations are rocks of mostly Precambrian age that are composed of interlaminated quartz and iron minerals. They can be subdivided into two varieties (Gross 1980): Algoma deposits, which are relatively small with an obvious volcanic association, and Lake Superior, which are much larger and have a shallow-shelf, orthoquartzite-carbonate association. The Algoma type is abundant in the Archean, but an Ordovician example is found at Bathurst, New Brunswick (see Chapter 8), and there is a possible analogue associated with the Carboniferous Pb-Zn deposit at Tynagh, Ireland (see Chapter 7). Lake Superior-type ores, by contrast, are confined to a particular time interval at around 2 billion years before the present. Oolitic ironstones have a more clastic association than the iron-formations, and are higher in Al. Instead of banding, their most prominent sedimentary structure is oolites made up of hematite or chamosite. Chert is rare, and they are found in rocks of a variety of ages from Proterozoic to Pliocene.

As of 1970, these two types constituted more than 90 percent of the world's iron production, with Lake Superior ores accounting for about 65 percent (Gross 1970, table 3). At that time, ironstones made up about 20 percent of the production, but this proportion has been decreasing: the last iron mines in Great Britain, which are in Jurassic oolitic ironstones, were closing in 1980. The extensive Lake Superior-type deposits of Australia are now coming into production, displacing smaller deposits, so that Lake Superior-type ores probably



make up 75 percent or more of production today. One reason for their predominance is a change in the nature of the feed for blast furnaces. Formerly, only ores that had been naturally enriched by the leaching of most of their  $\text{SiO}_2$  (the "direct shipping ores") could be used. Since the introduction of pelletized feeds in 1955, there has been a shift to artificially concentrated cherty taconites because of lower shipping costs and lower energy costs for blast furnace operation (Kakela 1978).

Because of the differences in their chemistry and depositional setting, we will discuss iron-formations and ironstones separately. First, however, it is useful to examine some aspects of the distribution of iron in the Earth's crust.

Iron is the third most abundant metal, after Si and Al. Therefore, it is a major constituent of most rocks (Table 2-1), and its ores are actually rock types in their own right. The formation of these iron rocks depends on changes in oxidation state: Fe is mobilized under reducing conditions, precipitated under oxidizing. Thus, it will migrate from areas low in oxygen towards areas high in it, provided a pathway exists. Note, in Table 2-1, that igneous rocks, although varying in total iron, have about the same proportion of  $\text{Fe}^{3+}$  to  $\text{Fe}^{2+}$ . Sedimentary rocks, in contrast, show a wide range of oxidation states, indicative of the presence of environments of differing oxidation potential. Therefore, it is in sediments that the greatest potential for iron enrichment exists, and almost all iron ores are sedimentary. We will see that most of the other sedimentary ore metals also depend on redox gradients for their enrichment.

**Table 2-1.** Abundance of iron in common rock types:

	FeO (%)	Fe <sub>2</sub> O <sub>3</sub> (%)	$\frac{\text{Fe}_2\text{O}_3}{\text{FeO} + \text{Fe}_2\text{O}_3}$
<b>Igneous Rocks</b>			
alkali-olivine basalts	7.9	4.2	0.35
tholeiitic basalts	9.5	3.2	0.25
granodiorites	2.6	1.3	0.33
granites	1.5	0.8	0.35
<b>Sedimentary Rocks</b>			
Sandstones:			
quartz arenite	0.2	0.4	0.67
lithic arenite	1.4	3.8	0.73
graywacke	3.5	1.6	0.31
arkose	0.7	1.5	0.68
Shales and slates			
red	1.26	5.36	0.81
green	1.42	3.48	0.71
black	4.88	0.52	0.10
Seawater (as Fe)		0.007 ppm	

Source: Wedepohl 1969, tables 26-K-2, 26-K-3; Wedepohl 1971a, tables 6.2, 8.1, 12.2.