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Diagenesis 3. Aspects of the Diagenesis of Coarse-Grained Siliciclastic Rocks

Ian Hutcheon

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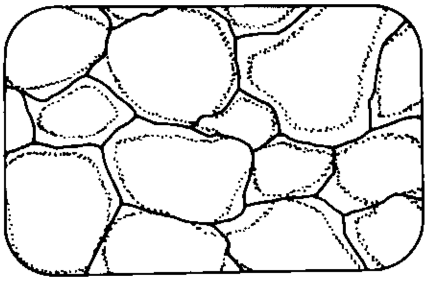
Article abstract

The process of diagenesis, by which sediments become rock, has many physical and chemical aspects. Even a cursory examination of the scientific literature shows that mineral reactions, original depositional environment, diagenetic environment, the flow of fluid, the evolution of organic matter, the compaction of sediment and the precipitation, replacement and destruction of cements all play a role in sandstone diagenesis. The importance of sandstone diagenesis is evidenced by the rapid increase of pure and applied research in this area. The oil and gas industry has greatly accelerated research into sandstone diagenesis, primarily because the nature and distribution of porosity and permeability ultimately governs the extent of oil and gas recovery, and diagenesis is a controlling factor in the formation and destruction of porosity. In addition, as our demand for oil and gas grows, and the relatively easily extracted oil is exploited, there is an increasing emphasis on secondary and tertiary recovery of conventional oil and recovery of heavy oil and tar sands. Enhanced recovery agents, whether physical (injected water or steam) or chemical (surfactants, acids, polymers, caustic andclay stabilising agents), are in direct contact with the pore space of the rock. It is important to understand the present state of diagenesis of any rock, and how that state will be affected by an agent used to stimulate recovery. While it is tempting to divide the subject of diagenesis into those processes which are physical and those which are chemical, the interdependence of physical and chemical processes makes this impossible. The reader also should be warned that even sandstone diagenesis itself cannot be separated from the interplay of differing sedimentary environments, lithologie variation within basins, and setting and evolution of sedimentary basins. All of these factors are ultimately governed by the tectonic setting.

Articles



Diagenesis 3. Aspects of the Diagenesis of Coarse-Grained Siliciclastic Rocks

Ian Hutcheon
Geology and Geophysics
The University of Calgary
Calgary, Alberta T2N 1N4

Summary

The process of diagenesis, by which sediments become rock, has many physical and chemical aspects. Even a cursory examination of the scientific literature shows that mineral reactions, original depositional environment, diagenetic environment, the flow of fluid, the evolution of organic matter, the compaction of sediment and the precipitation, replacement and destruction of cements all play a role in sandstone diagenesis. The importance of sandstone diagenesis is evidenced by the rapid increase of pure and applied research in this area. The oil and gas industry has greatly accelerated research into sandstone diagenesis, primarily because the nature and distribution of porosity and permeability ultimately governs the extent of oil and gas recovery, and diagenesis is a controlling factor in the formation and destruction of porosity. In addition, as our demand for oil and gas grows, and the relatively easily extracted oil is exploited, there is an increasing emphasis on secondary and tertiary recovery of conventional oil and recovery of heavy oil and tar sands. Enhanced recovery agents, whether physical (injected water or steam) or chemical (surfactants, acids, polymers, caustic and clay stabilising agents), are in direct contact with the pore space of the rock. It is important to understand the present state of diagenesis of any rock, and how that state will be affected by an agent used to stimulate recovery.

While it is tempting to divide the subject of diagenesis into those processes which are physical and those which are chemical, the interdependence of physical and chemical processes makes this impossible. The reader also should be warned that even sandstone diagenesis itself cannot be separated from the interplay of differing sedimentary environments, lithologic variation within basins, and setting and evolution of sedimentary basins. All of these factors are ultimately governed by the tectonic setting.

Introduction

Sandstone diagenesis is a subject which has been investigated by many different methods, and this reflects the nature of the processes governing diagenesis. In this review paper, the chemical aspects of the diagenesis of coarse-grained siliciclastic rocks are emphasized. In the broadest sense, diagenesis includes all the changes which affect a sediment, from the time it is physically deposited, until all the constituent grains come to chemical equilibrium during metamorphism. Diagenesis lies between sedimentation and metamorphism, both in the sequence of events in the rock cycle and in the methods used to study diagenesis. Physical changes, such as compaction, fluid migration and pressure solution, are accompanied by chemical changes caused by mineral reactions, mineral replacement, cementation and dissolution.

As rocks are buried, they evolve water, hydrocarbons and various gases. As these fluids move through a sedimentary sequence they will generally follow fractures or the most permeable units. In siliciclastic dominated sequences the most permeable units generally are the sandstones, which become the conduit for the evolving fluids. Sandstone diagenesis is controlled by a maze of interdependent processes, including burial rate, sediment composition and texture, sedimentary and tectonic environment, chemical reaction rates, degree of lithologic variation, hydrodynamic and geothermal gradients and a host of other factors. We will enter this maze by starting with the observed effects of sandstone diagenesis, and continue by examining pressure solution, mineral authigenesis, cementation, and development of secondary porosity and fluid movement. By this route we hope to arrive at some understanding of how all these processes contribute to turning sand into sandstone. Finally, we will briefly examine the practical application of diagenesis to the exploitation of hydrocarbon reservoirs.

Methods of Study

The petrologist studying diagenesis relies on transmitted light microscopy, much like metamorphic or igneous petrologists. However, because many of the materials he is attempting to observe are extremely fine-grained (1-20 μm), he relies heavily on the scanning electron microscope (SEM) and special X-ray diffraction techniques to assist in mineral identification and petrography. In many diagenetic studies extensive use is made of SEM petrography. With an energy dispersive spectrometer, the SEM can provide qualitative mineral analyses suitable, when combined with X-ray diffraction data and morphology, to assist in the identification of clay minerals and other authigenic phases. The scanning transmission electron microscope (STEM) offers the possibility of determining the exact nature of very fine intergrowths of authigenic minerals and the potential for accurate chemical analyses of very small particles.

Many other methods may be applied during the study of clastic diagenesis. Resin pore casts and mercury injection tests can provide detailed information about the size and distribution of pores and are especially useful in studies relating to the recovery of hydrocarbons. Fluid inclu-

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sion studies of clastic rocks are not common (see Currie and Nwachukwu, 1974), but the success of fluid inclusion studies of carbonates implies that they will become more common. Stable isotopes and methods of bulk chemical analysis (X-ray fluorescence) have been applied to the study of diagenesis, but separating authigenic minerals from allogenic phases is always difficult and may complicate interpretation of the results of such studies. Some very successful studies combine organic geochemistry with detailed study of the diagenetic events, either by characterizing the organic material (Foscolos and Powell, 1980), or by employing properties such as the vitrinite reflectance of coal (Hutcheon *et al.* 1980), which is related to time and temperature of burial.

Diagenesis and Porosity

The most obvious physical change in siliciclastic sediments during burial is the loss of porosity, which was the subject of an early study by Taylor (1950). Figure 1 incorporates data from the Gulf Coast of Texas (Stuart, 1970), Northeast Pacific (Galloway, 1974) and the Mackenzie River Delta (Schmidt and McDonald, 1979a). It is obvious that porosity tends to decrease with increasing depth, but less obvious that there may be zones in which porosity decrease is not as rapid, or in which porosity actually increases. The porosity loss, in general, results from compaction of grains, pressure solution of grains and growth of authigenic minerals (cements). It can be seen in Figure 1 that there is no single relationship between porosity and depth. Variations occur between basins and between units within basins, reflecting the many interacting processes which contribute to diagenesis. Maxwell (1964) attempted to quantify by experiment the relationships between porosity and other factors, and concluded that increasing temperature, age, pressure, amount of fluid flow and amount of matrix all tend to lead to more rapidly decreasing porosity. These conclusions are reinforced by Selly's study of porosity evolution in the North Sea (1978). A number of studies indicate that porosity evolution and the general path of diagenesis are greatly affected by the presence of hydrocarbons, early hydrocarbon migration tending to preserve porosity and immature diagenetic assemblages (Lowry, 1956; Wilson, 1977).

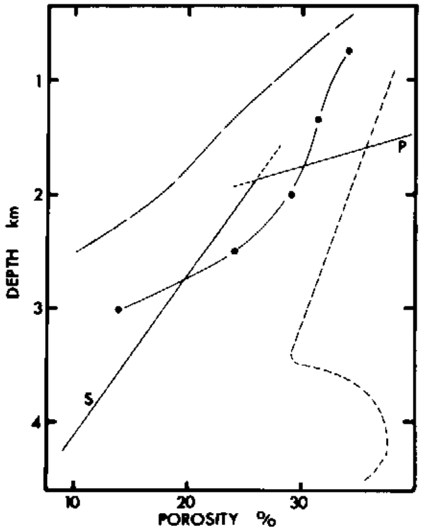

Figure 1 *Porosity vs. depth curves for sandstones can be generalized for a number of sedimentary basins. Data for the Northeast Pacific are from Galloway (1974), and are shown by the long-dashed (geothermal gradient = 39 C/km) and dot-dash (geothermal gradient = 29 C/km) lines. Relationships also are shown for the Gulf Coast (Stuart, 1970, short-dash curve) and the Mackenzie Delta (Schmidt and McDonald, 1979a, solid curves). The "P" and "S" on the data from the Mackenzie Delta indicate primary and secondary porosity, respectively.*

Galloway (1974) notes that within the Queen Charlotte basin sandstone cementation is accelerated in areas of higher geothermal gradient (Figure 1), which implies that the rate of diagenetic alteration is temperature dependent. Sandstones from the Northeast Pacific seem to lose porosity more rapidly than their counterparts in more stable tectonic regimes, such as the Gulf Coast of the United States and the Mackenzie Delta. The sands in the Northeast Pacific basins, in addition to being in an area of higher heat flow, have higher contents of volcanic rock fragments, feldspars and other immature or chemically less stable constituents than sands in the Gulf Coast or Mackenzie Delta. It would seem that chemical instability (or compositional "immaturity") of sandstones also plays a role during diagenesis.

The data from the Mackenzie Delta are interpreted by Schmidt and McDonald (1979a) to show two different trends of porosity loss with depth. Porosity which these authors refer to as "primary," or porosity represented by the original interstices in the sediment, is lost more rapidly than "secondary" porosity, which results from fractures, shrinkage and dissolution of preexisting cements or grains. On Figure 1, the porosity-depth curve for the Gulf Coast sands shows a rapid increase in porosity with depth below 3500 m. The Gulf Coast basin contains significant volumes of "overpressured" or "geopressured" rocks. This means that rather than the normal depth-pressure curve expected for an open column of water, pore fluids in the Gulf Coast sands are isolated from communication with the surface and can develop and sustain abnormally high water pressures. These "overpressures" affect the evolution of diagenesis in the Gulf Coast.

In summary, we see that porosity is occluded with depth, but the rate of porosity destruction (and presumably attendant diagenetic processes) is strongly affected by geothermal gradients, initial sediment composition, tectonic setting, presence of secondary porosity and presence of overpressure. We shall see that other factors also are important during diagenesis. Many of our ideas about diagenesis are based on the Gulf Coast because of the large amount of data from that area. It would be incorrect to assume that all basins will show the same diagenetic trends as the Gulf Coast.

Pressure Solution and Quartz Overgrowths

Pressure solution is a subject worthy of an entire article, but since this process contributes to sandstone cementation a very brief review is presented here. Two types of petrographic phenomena are attributed to pressure solution, stylolites and dissolution of grains at grain-to-grain contacts. Dissolution at grain-to-grain contacts is more commonly observed in sandstones than are stylolites, which are more common in carbonate rocks. In 1863, H.C. Sorby described convincing petrographic evidence to support the concept of pressure solution, and since that time there has been an abundance of petrographic work to bolster Sorby's original observations. A satisfactory theoretical explanation for pressure solution has been far more elusive, beginning with Reicke's (1895) work on solids under homogeneous stress in a fluid and continuing to the present day experimental work of Sprunt and Nur (1977), DeBoer *et al.* (1977) and Robin (1978). Robin (1978) gives a brief but excellent summary of the development of theories regarding pressure solution. Figure 2 summarizes a theory proposed by Robin. Pressure along the interface between grains in not constant, nor is it equal to the pressure exerted by the fluid in the pore space. The difference in thermodynamic chemical potential produced by this pressure difference is sufficient to put quartz into solution, allowing it to migrate along the grain boundary and to be precipitated in the pore space. Robin's analysis verifies the conclusion that force across a grain contact is a function of burial depth, grain size, grain shape and grain packing, and all these factors must play a role in the pressure solution process.

Figure 3 shows various petrographic features often attributed to pressure solution. In sandstones, interpenetrating grain-to-grain contacts, sutured contacts, concavo-convex contacts and quartz overgrowths in many cases are taken as evidence of pressure solution. Spang *et al.* (1979) have demonstrated that clay seams, stylolitic in part, may cause solution of adjacent rock. The subsequent collapse will produce a texture very much like a pressure solution stylolite. In the strict sense, this texture actually results from chemical reactions among minerals, which may proceed in the complete absence of directed stress, and thus is not truly "pressure" solution. Also, the work of Foscolos and Powell (1980) shows the presence of considerable amounts of inorganic amorphous silica at depths up to 2000 m in the Sverdrup and Beaufort-Mackenzie basins. The solubility of amorphous silica, often in the form of animal skeletons, is considerably higher than that of quartz, and dissolution of amorphous silica and reprecipitation as quartz can lead to the formation of quartz overgrowths, a feature often ascribed to pressure solution but which, in this case, is not related to directed pressure.

Quartz overgrowths are commonly the most significant porosity-reducing feature in the diagenesis of quarz-rich sandstones. Reactions between clay minerals and replacement of silica-bearing minerals by other minerals, such as carbonates, can provide sources of silica. The variability in the development of quartz overgrowths is governed by the nature of the host grain, with monocrystalline quartz grains producing different forms of overgrowths when compared to polycrystalline quartz (Waugh, 1970). The "dust lines" associated with quartz overgrowths (Figure 3a) mark the interface between the overgrowth and the host grain and are often voids (Pittman, 1972). The voids seem to form because nucleation is initiated at a number of sites on the surface of the host grain and, as competitive crystal growth proceeds, a small number of the original nuclei will grow to form the overgrowth. The voids represent the interstices between these original nuclei.

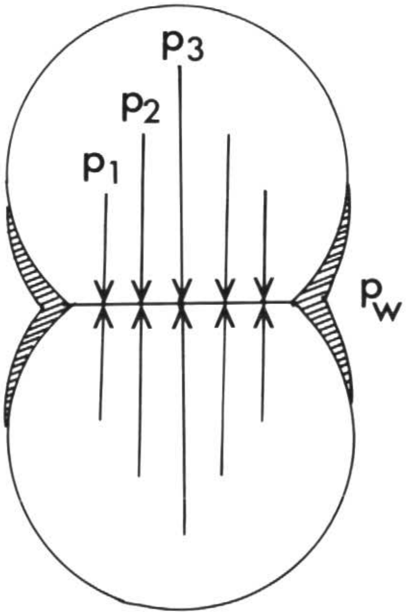

Figure 2 *An idealized interface between two spherical quartz grains is shown with the pressure distribution as postulated by Robin (1978). The pressures p1 to p3, at the grain contact, are not equal, nor are they equal to the fluid pressure, pw. An overgrowth is shown on the grains (shaded area). The difference in pressures supplies the chemical potential gradient, which leads to the diffusion of silica from the grain contact to the pore space.*

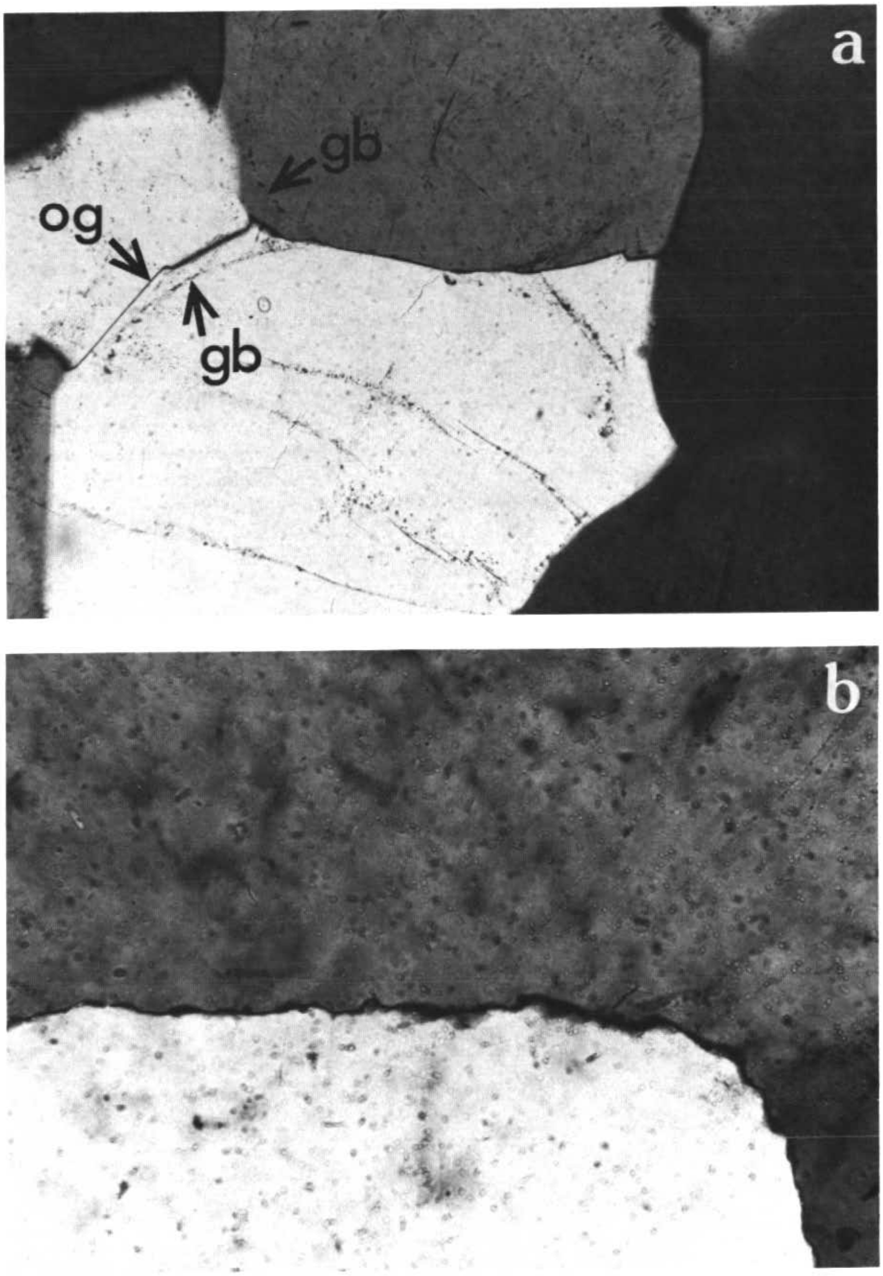

Figure 3 *These photomicrographs show textures commonly associated with pressure solution. In 3a (60 X) quartz grains show "dust rings" where the original grain boundary (gb) has been overgrown by euhedral quartz (og). The concavo-convex contact between the two grains cuts the dust rings and the overgrowths, indicating solution of material at the interface. In 3b (400 X) a highly sutured contact has developed between two grains in a quartz sandstone.*

Authigenic Cementation

Wilson and Pittman (1977) emphasize the necessity and textural criteria for distinguishing between allogenic (introduced during deposition) and authigenic (growth *in situ* during diagenesis) clay minerals. Figure 4 shows typical morphologies of kaolinite, chlorite, smectite and illite when found as authigenic minerals in sandstone. Most workers accept the authigenic nature of carbonate or other types of pore-filling cements (Figure 5). In many instances there is not a sufficient local source within the sediment to produce all the observed cementing material, suggesting that material for cement formation must be scavenged from the pore water. The material in the pore water may be derived from connate saline water (seawater), dissolution of soluble rocks (i.e. evaporites) by percolating groundwater or expulsion from shales and other rocks undergoing mineral or organic reactions. Curtis (1978), Boles (1979) and Boles and Franks (1979) have recognized the probability that material for cementation may be transferred from shales, via pore waters, to sandstones.

The paths of cementation in sandstones are extremely complex. For example, Galloway (1974) recognizes relatively early calcite pore-fill formed near the surface, and a phase of calcite cementation displaying replacement textures which occurs only at depths greater than 1200 m. Numerous authors have documented paragenetic sequences for authigenic minerals in sandstones in which appearances and disappearances of minerals overlap (see, for example, Galloway, 1974; Surdam and Boles, 1979; Mankiewicz and Steidtmann, 1979; Tillman and Almon, 1979). This implies that, rather than the appearance and disappearance of a single mineral, there are reactions taking place, via the pore water, among the minerals in a sandstone.

Boles (1982) has recognized active albitization in Frio Sandstones from the Gulf Coast. Using scanning electron microscopy, the reaction

$$\underset{\text{quartz}}{2\,SiO_2} + \tfrac{1}{2}H_2O + H^+ + Na^+ + \underset{\text{plagioclase}}{CaAl_2Si_2O_8} \quad (1)$$
$$= \underset{\text{plagioclase}}{NaAlSi_3O_8} + \underset{\text{kaolinite}}{\tfrac{1}{2}Al_2Si_2O_5(OH)_4} + Ca^{++}$$

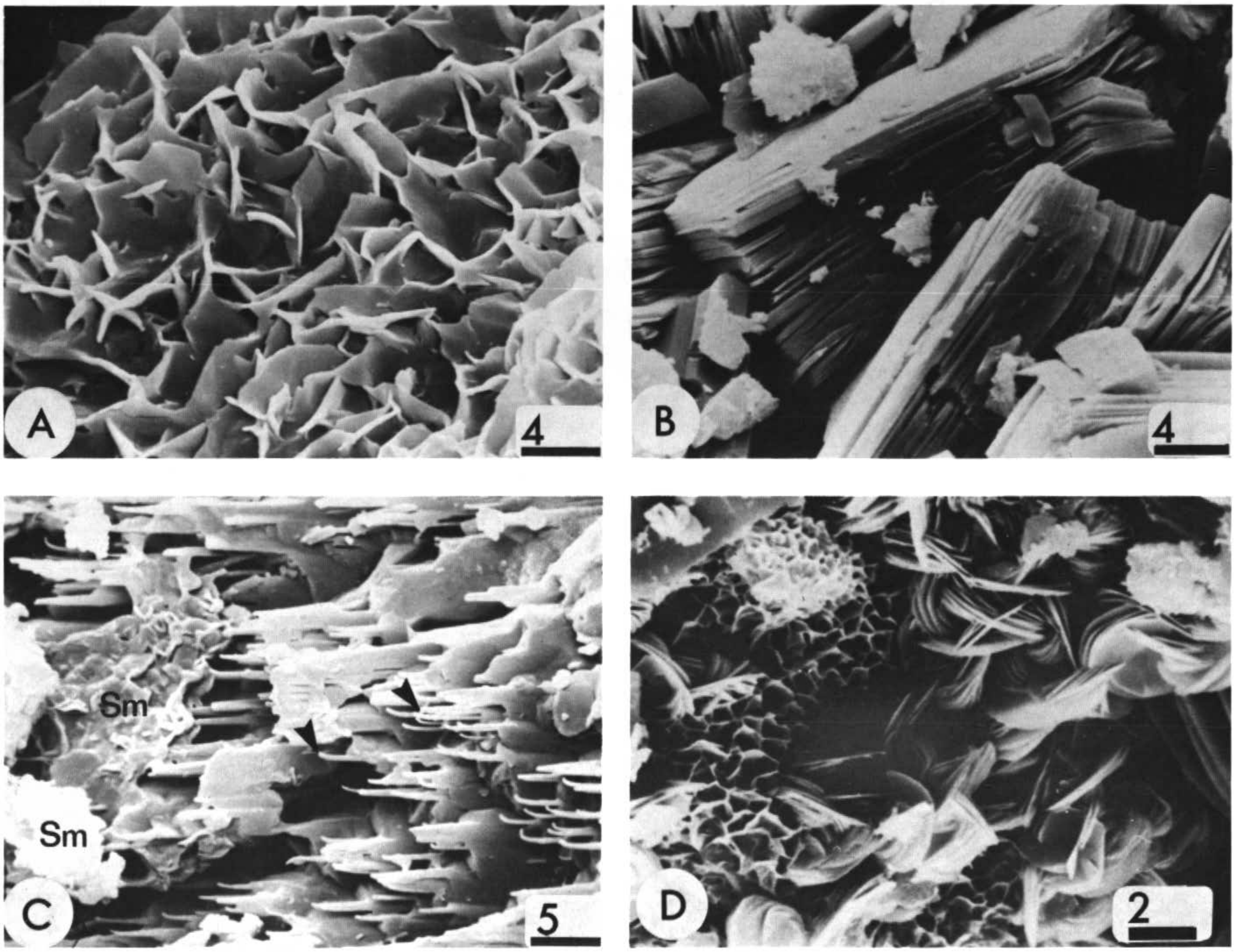

Figure 4 *Scanning electron micrographs of (a) smectite honeycombs, (b) euhedral kaolinite booklets, (c) illite fibres (marked by black arrows) growing on altered plagioclase feldspar and (d) rosettes of iron-rich chlorite. The bar scale on each photograph is in um. All micrographs were taken by Alan Oldershaw.*

can be documented. By plotting the activity ratio of Ca^{++}/Na^{+} vs. pH, contoured for 100, 125 and 150°C, and also plotting the compositions of waters (in these same variables), Boles convincingly demonstrates that albitization is occurring, probably at the present time, in the subsurface of the Gulf Coast. The reaction shown provides a source of calcium for the production of carbonate cements. Many diagenetic mineral reactions are interrelated in a similar way, and to obtain a complete picture of diagenetic evolution a complex sequence of textures, indicative of mineral reactions, must be interpreted. The studies of Merino (1975) and Nesbitt (1980) demonstrate that if we consider the minerals within the pore space as a chemical system (Figure 6), we can treat many of the chemical problems in sandstone diagenesis using equilibrium thermodynamics. We will see that chemical kinetics, mass transfer and fluid movement also may have a major impact on sandstone diagenesis.

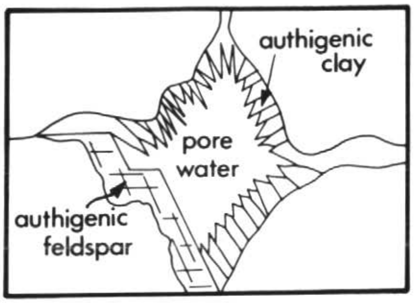

Figure 6 *The minerals in the pore space, which are in contact with the pore fluids, often can be considered as a system which may come to thermodynamic equilibrium.*

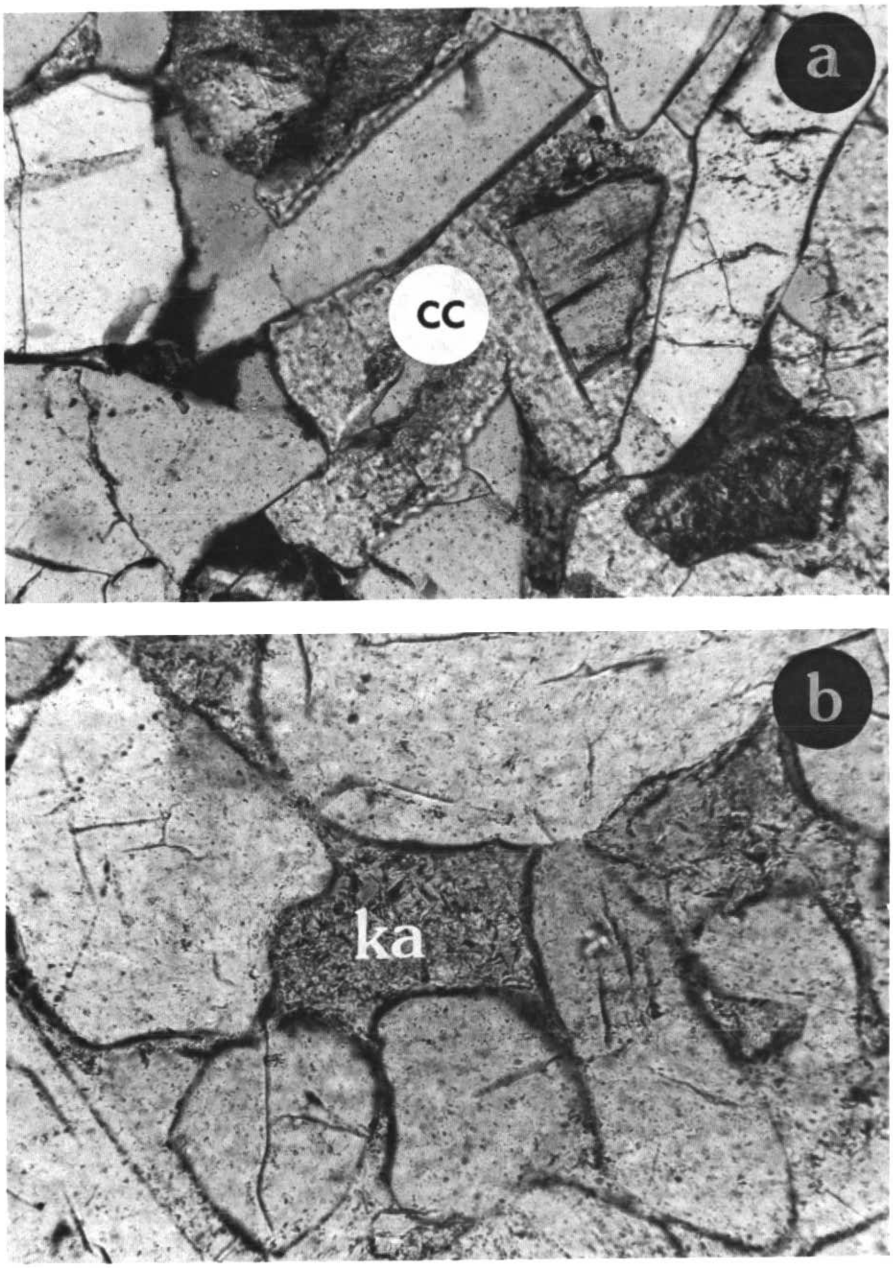

Figure 5 *Authigenic, pore-filling cements are shown in both photomicrographs. In 5a (160 X) calcite is shown enclosing angular quartz grains in a sandstone. The calcite is well crystallized (cc) and shows some euhedral crystal faces. In 5b a pore in a sandstone with well-rounded quartz grains is filled by authigenic kaolinite (160 X).*

In examining the published work on chemical aspects of sandstone diagenesis, it would be easy to conclude that minerals and aqueous solutions are the only substances we need deal with in diagenesis. From fluid inclusion studies of sediments Burruss (1981) cites examples of hydrocarbon, methane or carbon dioxide-bearing fluid inclusions associated with diagenetic events. The importance of carbon dioxide in controlling carbonate dissolution and precipitation has long been recognized. Many authors have noted the production of methane and carbon dioxide during the diagenesis of organic matter. While this may be the case, how much of the vapour is carbon dioxide and how much is present as other species is a moot point. In 1942, Germann and Ayers noted the production of carbon dioxide from limestones with trace amounts of silica, aluminum, iron and magnesium at temperatures as low as 98°C. More recently, Muffler and White (1969), in a study of the Salton Sea geothermal field, postulated a carbon dioxide-producing mineral reaction between clay minerals and carbonates. Hutcheon *et al.* (1980) have documented the mineral reaction

$$5\,\underset{\text{dolomite}}{CaMg(CO_3)_2} + \underset{\text{kaolinite}}{Al_2Si_2O_5(OH)_4} + \underset{\text{quartz}}{SiO_2} + 2\,H_2O \qquad (2)$$

$$= \underset{\text{chlorite}}{Mg_5Al_2Si_3O_{10}(OH)_8} + 5\,\underset{\text{calcite}}{CaCO_3} + 5\,CO_2$$

which is observed in the Kootenay Formation, at approximately 5000 m (stratigraphic plus structural) burial. Figure 7 shows the topology of reaction (2) as a function of temperature at constant pressure in a mixture of carbon dioxide and water. As temperature increases, reaction (2) proceeds to the carbon dioxide-bearing side, and this carbon dioxide is added to the vapour phase, increasing the mole fraction of carbon dioxide in the vapour and making higher temperatures necessary for the reaction to proceed. The mineral reaction will follow along the curve in Figure 7 until the miscibility surface between carbon dioxide and water is reached and carbon-dioxide will no longer dissolve in an

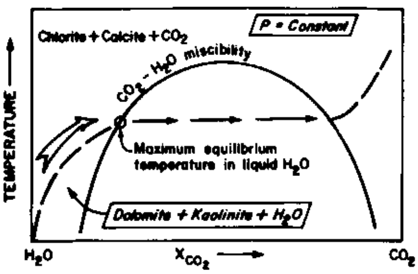

Figure 7 *Mineral reactions similar to the one shown will attempt to control the carbon dioxide content of a vapour phase during burial. As the carbon dioxide content in the vapour increases, the reaction is stable at higher temperatures. Once the miscibility surface (solid curve) between carbon dioxide and water is reached, carbon dioxide-rich vapour is produced.*

aqueous fluid. At that point, carbon dioxide is produced as a vapour. Most fluids observed in sedimentary rocks are aqueous liquids; it is reasonable to expect that the intersection of the mineral reaction and the carbon dioxide – water miscibility surface represents the maximum equilibrium temperature for the mineral reaction in liquid water. Hutcheon *et al.* (1980) describe this process in more detail, and show calculated phase diagrams which indicate the maximum temperature for reaction (2) in the Kootenay Formation is about 180°C, in agreement with coal maturation data for the Kootenay Formation.

A number of interesting observations can be made as a result of chemical studies of diagenesis. Boles and his coworkers have shown that many diagenetic processes appear to be active. In other words, we can observe from drill core that ankerite cementation and albitization are taking place as the sediment is buried. By using mineral chemistry and equilibrium thermodynamics, it has been possible to show that material has moved between shales and sandstones during diagenesis. This result is directly comparable to the study by Hower *et al.* (1976) in which the smectite-illite transformation in mudrocks is interpreted as an active process.

The research of many authors demonstrates that, rather than single minerals appearing and disappearing, there are complex mineral reactions with pore fluid in the subsurface. To properly understand diagenesis, it is necessary to document these reactions with careful petrography, combining X-ray, thin section and electron microbeam techniques.

The results of Muffler and White (1969) and Hutcheon *et al.* (1980) show that mineral reactions may produce carbon dioxide. This is an alternate, additional source to carbon dioxide produced by organic reactions. Carbon dioxide is an interesting constituent of diagenetic processes in that some authors have attributed carbonate cementation to carbon dioxide production (Curtis, 1978), while other authors attribute dissolution of carbonate (and the formation of "secondary porosity") to the effect of carbon dioxide (Schmidt and McDonald, 1979a). Although these authors are referring to processes which take place at different burial depths, the effects of carbon dioxide production on carbonate stability require clarification.

Secondary Porosity

Many authors have examined the importance of secondary porosity in the development of sandstone reservoirs (Hayes, 1979; Loucks *et al.* 1, 1979). A comprehensive account of the role and recognition of secondary porosity is given by Schmidt and McDonald (1979a, 1979b). They recognize porosity produced by fracturing, shrinkage, dissolution of sedimentary material and dissolution of authigenic material, but consider the dissolution of carbonates to be a major contributor to secondary porosity in sandstones. The data of Schmidt and McDonald show a marked reduction in the rate of porosity loss with burial depth (Figure 1, line labelled "S") over depth ranges in which secondary porosity is being destroyed. As stated previously, Schmidt and McDonald ascribe dissolution of carbonates to carbon dioxide produced by organic diagenesis, whereas Curtis (1978) blames the identical process for the carbonate precipitation. Which interpretation is correct? As is usual in geological problems, both are, maybe! *

Using calcium carbonate as an example, the dissolution of carbonates is controlled by a reaction similar to (3):

$$\underset{\text{calcite}}{CaCO_3} + H^{+} = HCO_3^{-} + Ca^{++} \qquad (3)$$

assuming, as is often the case, that the pH range is such that HCO_3^- is the dominant acqueous carbonate species in solution. If the pH (activity of hydrogen ion) is controlled by mineral reactions with an aqueous phase, or by organic materials, such as acetic acid, the addition of carbon dioxide will increase the amount of HCO_3^- by dissociation of H_2CO_3. This will cause an increase in the solubility product for reaction (3) and lead to carbonate precipitation. However, if the pH is not fixed by some organic matter or minerals in the rock something different will happen. The addition of carbon dioxide will produce H_2CO_3 which, being a weak acid, will dissociate, producing hydrogen ions and lowering the pH. This will decrease the solubility product for reaction (3), leading to carbonate dissolution. Present knowledge of water-rock systems does not allow unequivocal determination of pH from mineral assemblage data; therefore recourse lies in obtaining reliable, accurate water chemistry from subsurface waters.

Geological Controls of Diagenesis

Siever (1979) has summarized plate tectonic controls on diagenesis. He notes source area, burial rate, heat flow and residence time as generally important factors. While it is obvious that burial environment and original sediment composition and texture, which are determined by depositional environment, are two important geological controls, there is a paucity of studies which relate these two important variables. A study by Foscolos *et al.* (1982) marks the beginning of a project devised to relate diagenetic alteration to burial and sedimentary environment. Concentration of effort in this area is badly needed.

Fluid Flow and Diagenesis

It is clear that as sediments are buried they lose water and other volatile constituents. Many of our concepts of sandstone diagenesis are founded on studies of the Gulf Coast, an area of low topographic relief and active burial. Movement of fluid in the Gulf Coast is upward and laterally outward along permeable units (Magara, 1976; Neglia, 1979). In addition, the fluids are largely connate marine water and water from the dissolution of evaporites, modified by diagenetic mineral reactions (Carpenter, 1978; Hanor, 1979). Most ancient sedimentary basins are not found in this particular setting. For example, the Alberta basin is in close proximity to a mountain range which provides sufficient hydraulic potential that meteoric water circulates through permeable units to depths on the order of 3000 m below ground surface (Toth, 1980). This origin for waters has a strong influence on water composition and therefore influences the nature of mineral reactions taking place between pore fluids and the enclosing rocks. Concentrations of dissolved solids in formation waters in Alberta are commonly 50,000 mg/l, but can range up to 200,000 in Devonian rocks (Hitchon *et al.* 1971; Hitchon, 1980). Since some of these waters can circulate to a great range of depths, fluid movement is a process which can transport large amounts of material, even in the time scale of the Holocene (Toth, 1980).

The presence of overpressured zones in many basins implies that porous and permeable units may be isolated from communication with other permeable rocks by impermeable bodies. These permeable sands still show evidence of diagenetic features associated with other diagenetic settings, and it appears that fluid movement must have occurred. Also, spatial patterns of mineral distribution may be developed, as, for example, in the alteration of volcanic rocks (Surdam and Boles, 1979). Wood and Surdam (1979), Cassan *et al.* (1981) and Wood and Hewett (1982) all consider convective fluid movement to be an important process. Cassan *et al.* (1981) surmised that convective fluid movement controls the dissolution of feldspar and cementation by quartz and calcite in the Gabon Basin by imposing temperature changes as a result of convection cells.

Although convective flow is an hypothesis which requires further verification by careful study of diagenetic mineral assemblages, there is little doubt that fluid flow, whether by upward moving compaction water, circulating groundwater cells in hydrodynamically complex basins or convective flow, is a controlling process for mass transport in porous sandstones. Fluid flow probably dominates over diffusion controlled mass transfer for mass movement on the large scale. Nevertheless, diffusion is an important process in pressure solution (Robin, 1978) and may play a greater role in sandstone diagenesis than is presently recognized. Studies such as those by Aagaard and Helgeson (1982) mark the beginning of the use of kinetics as a means of understanding sandstone diagenesis.

Diagenesis and Hydrocarbon Reservoirs
Intergranular, dissolution, fracture and micro-porosity are important in sandstone reservoirs. The authigenic clay content of sandstone reservoirs can create microporosity, which is largely ineffective, and low permeabilities by decreasing the size of pore apertures. The high surface area to volume ratio of most clay minerals and the tendency for expansion of the smectites or migration of kaolinite when exposed to fresh water are limiting factors in oil recovery. Many of these factors are affected by diagenesis (Pittman, 1979). Consequently, much effort is being expended to understand the role of diagenesis in hydrocarbon recovery, encouraging an overall increase of interest in sandstone diagenesis.

The recovery efficiency of sandstones has strong positive correlations with high porosity, small pore-to-throat size ratios,

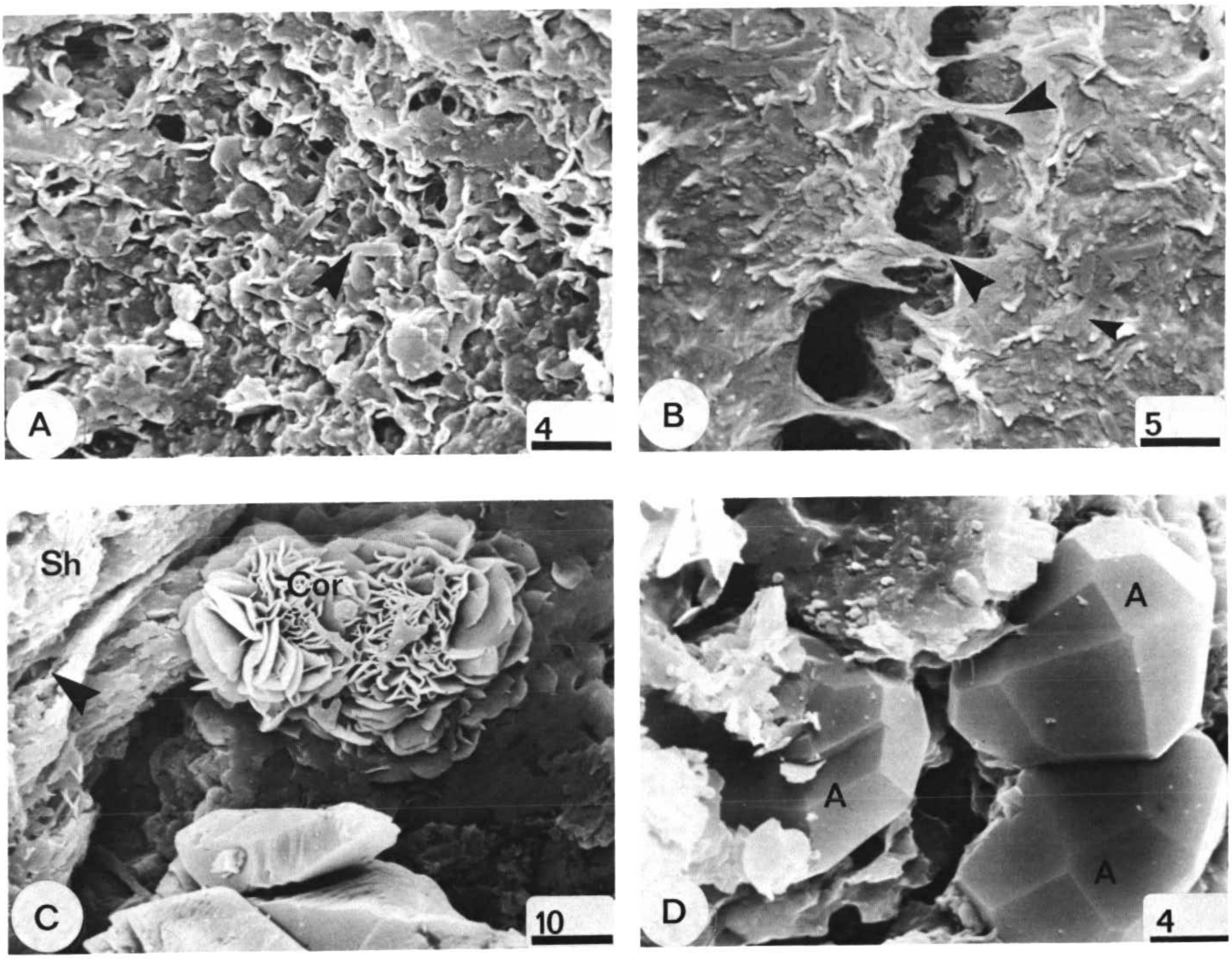

Figure 8 *During steam stimulation, fine smectite and illite (8a and 8b) are reacted to produce coarse grained corrensite (Cor, a smectite-chlorite intergrowth which in this case is not identified by X-ray diffraction), and coarse grained, euhedral analcime (8c and 8d). The scale on each photograph is in µm. All micrographs were taken by Alan Oldershaw.*

small mean particle size and low percentage of carbonate (Wardlaw and Cassan, 1979). Of these properties, pore-to-throat size ratio and carbonate percentage can be markedly affected by diagenesis. For example, in studies of reservoir sandstones it is common to see illite bridging pore throats (Figure 8) and effectively doubling or tripling pore-to-throat size ratios. Kaolinite (Figure 4b) commonly forms aggregates and pore-fillings which are larger than the pore throats. When interstitial flow velocities become high, during recovery, these aggregates and individual booklets may become detached from their substrate and lodge in the pore throats, shutting off flow in that part of the rock. The smectite group of clay minerals tends to swell when in contact with water due to osmotic swelling and the adsorption of water molecules in the interlayer positions. This swelling can be up to two orders of magnitude of the solid volume occupied by smectite with a corresponding decrease in porosity and permeability. Further, all clay minerals have high surface area to volume ratios, and tend to have electrically charged surfaces, making them effective at binding water and causing high irreducible water saturations. The differing cation exchange capacities for groups of clay minerals indicate different surface charge distributions. Clays with higher cation exchange capacities, such as smectite, tend to be more effective at binding water than such clays as kaolinite, which has a low cation exchange capacity. Van Elsberg (1978) has noted that many amorphous hydroxides have similar properties to clay minerals, and are very difficult to detect. In addition, the role of amorphous material in sandstone diagenesis is poorly understood, with very few workers (Foscolos and Powell, 1980, being notable exceptions) recognizing the importance of amorphous material. Almon and Davies (1981) and Thomas (1981) outline possible problems created by the presence of clay minerals, and offer general methods for minimizing their effect on recovery. Almon (1981) also outlines the necessity for using diagenetic trends as a method for porosity prediction in oil exploration. Table 1 summarizes some of the more general ways in which clay minerals can affect recovery and common methods of treating these problems.

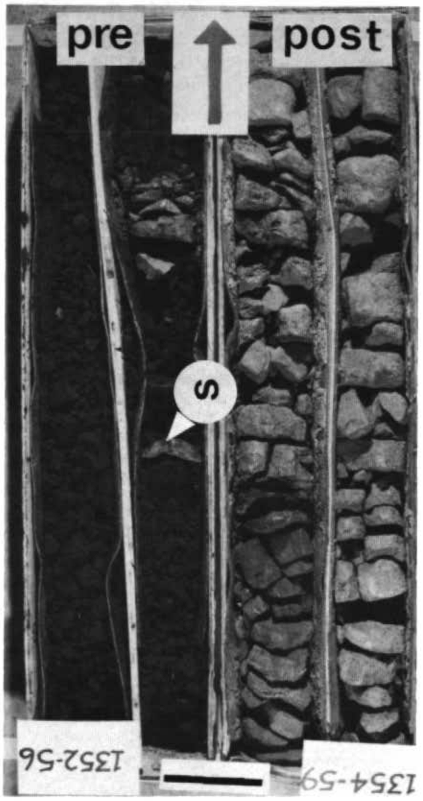

Figure 9 *A pre-steam core from the Cold Lake tar sand is a pasty mixture of bitumen and mineral grains. Two years of cyclic steam injection and steam flooding have removed most of the oil and lithified the sand. The minerals in Figures 8c and 8d cement the individual grains together.*

Table 1 *Problems and treatments for authigenic minerals in reservoirs*

Mineral	Problem	Incompatible with:	Compatible with:	Remedy
Smectite	swelling	fresh water	KC1, hydrocarbon	HC1, HF
Mixed-layer Smectite-Illite	swelling	fresh water	KC1, hydrocarbon	HC1, HF
Illite	microporosity	fresh water	KC1, hydrocarbon	HC1, HF
Kaolinite	mobile fines	high flow rate	low flow rate	clay stabilizers
Chlorite	Iron precipitate	0_2 - rich, pH 3.5 +	HC1, organic acid	HC1, organic acid
Calcite, dolomite	CaF_2 precipitate	HF		HC1

After Almon and Davies (1981)

Heavy Oil and Tar Sands (artificial diagenesis)

The tar sands and heavy oil deposits in Alberta and Saskatchewan represent a huge resource, but oil viscosities are so high in the tar sands that there is little or no conventional production. Schemes to mobilise the vast amounts of oil that are too deep to be mined involve heating, either by steam injection or by in-situ combustion. Mineralogical alteration, especially of pore space minerals, is to be expected and may be observed both in laboratory tests (Perry and Gillott, 1979, 1982) and in actual field results (Sedimentology Research Group, 1981). The impact of steam on volcaniclastic tar sands from Cold Lake is seen in Figure 9. When compared to the well-lithified, oil-free portion of a core cut after two years of steam injection, a pre-steam injection core is a pasty, unconsolidated mixture of mineral grains and tar. Detailed petrographic work demonstrates that an early, fine-grained diagenetic assemblage (less than 4 μm) of illite, smectite, zeolites and chlorite is replaced by coarse (4-10 μm) smectite and large euhedral analcime crystals (20 μm, see Figure 8). Figure 10 shows that decreases in porosity correspond to decreased kaolinite and increased smectite content in the less than 2 μm size fraction. The reaction postulated for these mineral alterations is

$$\underset{\text{kaolinite}}{7\ Al_2Si_2O_5(OH)_4} + \underset{\text{quartz}}{8\ SiO_2} + 2\ Na^+ \qquad (4)$$
$$= \underset{\text{smectite}}{2\ NaAl_7Si_{11}O_{30}(OH)_6} + 7\ H_2O + 2\ H^+$$

(Sedimentology Research Group, 1981) and in conjunction with albite- and analcime-bearing reactions can be used to construct a phase diagram to outline the stability fields for analcime, kaolinite, smectite and albite. The phase diagram constructed (Figure 11) verifies the mineralogical and textural observations that smectite is a high temperature phase compared to kaolinite. The diagram also explains the textural observation that analcime is the last-formed phase in these rocks. When injection stops, more saline formation water will invade the rocks and the phase diagram predicts that analcime will be stable at higher salinities than kaolinite and smectite at the same temperature. All the studies mentioned demonstrate that "artificial" diagenesis does occur during steam injection or in-situ combustion. The studies also indicate that artificial diagenesis affects the petrophysical parameters of the rock and, presumably, the efficiency of oil recovery.

Summary and Conclusions

The most obvious feature of sandstone diagenesis is the modification and general reduction of porosity. Compaction, pressure solution and authigenic cementation by mineral reactions are the principle processes contributing to this porosity modification. The rate and pathway of sandstone diagenesis is strongly influenced by sediment composition, which is in turn dependent on sedimentary and tectonic environments. Temperature also is a regulating factor, most easily recognized by the different paths of diagenesis in areas of varying geothermal gradients. Upon burial, sandstones display evidence of "active" diagenesis which can be modelled by mineral-solution equilibria. Much of the material required for sandstone diagenesis (especially in very quartz-rich sands) is transferred from other rock by fluid movement. Fluid movement may result from upward and outward flow of compaction water, circulation of ground water in hydrodynamically complex basins or by convective flow within basinal units.

The properties of hydrocarbon reservoirs are affected by diagenesis, and can be dramatically altered by drilling fluids or enhanced recovery techniques. Very detailed understanding of possible interaction between minerals, aqueous fluids, hydrocarbons and gases is required in order to maximize hydrocarbon recovery.

The emphasis on more sophisticated ways of modelling fluid and mass transfer (Wood and Hewett, 1982) and dealing with the overall kinetics of processes which are governed by reactions at the mineral-solution interface (Aagaard and Helgeson, 1982) indicate the direction of physical and chemical research on sandstone diagenesis. The application of scanning transmission electron microscopy (STEM) offers the hope that our ability to deal with the composition and structure of very small mineral grains will keep pace with the theoretical data. The chemistry of mineral surfaces, the kinetics of mineral reactions and the thermodynamic properties of clay minerals and complex aqueous solutions at elevated temperatures and pressures are not well enough understood to solve the scientific and practical problems of diagenesis.

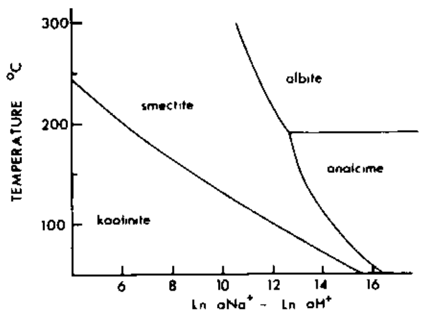

Figure 11 *Phase relations at constant pressure and silica activity for kaolinite, smectite, albite and analcime show that for a given fluid composition smectite is a higher temperature phase than is kaolinite. In addition, at constant temperature analcime requires higher values of the equilibrium constant (at constant pH, this could be interpreted as increased salinity) than do kaolinite or smectite.*

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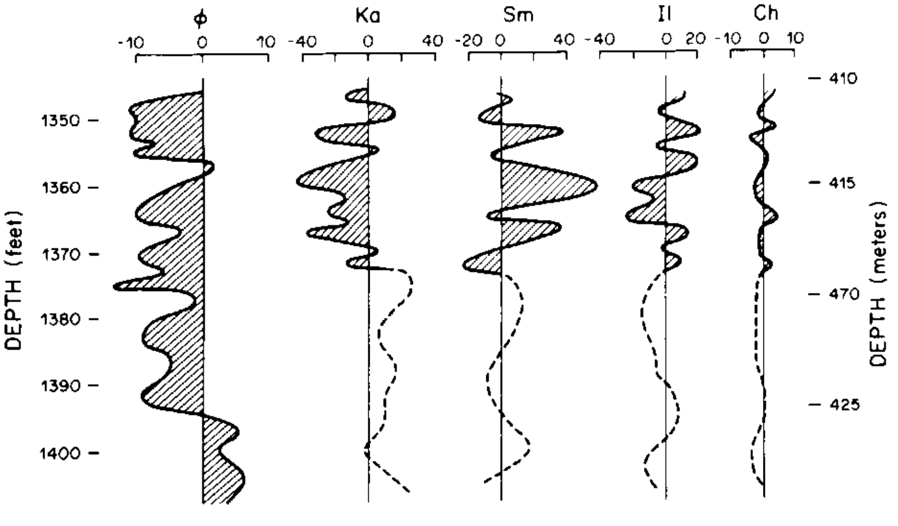

Figure 10 *This figure shows changes in porosity (Φ), kaolinite (ka), smectite (sm), illite (il) and chlorite (ch) contents in the tar sand samples examined. When going from the pre-steam to post-steam samples, a negative change means a decrease, and a positive change means an increase. The antipathetic relationship between smectite content and kaolinite content is the basis for the mineral reactions postulated. Note that increases in smectite content correspond to decreases in porosity.*

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