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ABSTRACT

The biological clogging of natural porous media, often in conjunction with physical or chemical clogging, is encountered under a wide range of conditions. Wastewater disposal, artificial groundwater recharge, in situ bioremediation of contaminated aquifers, construction of water reservoirs, or secondary oil recovery are all affected by this process. The present review provides an overview of the techniques that are used to study clogging in the laboratory, or to monitor it in field applications. After a brief survey of the clogging patterns most commonly observed in practice, and of a number of physical and chemical causes of clogging, the various mechanisms by which microorganisms clog soils and other natural porous media are analyzed in detail. A critical assessment is also provided of the few mathematical models that have been developed in the last few years to describe the biological clogging process. The overall conclusion of the review is that although information is available on several aspects of the biological clogging of natural porous media, further research is required to predict its extent quantitatively in a given situation. This is particularly true in cases that involve complicating factors such as predation or competition among organisms.

KEY WORDS: bioremediation, groundwater, soil, hydraulic conductivity, artificial recharge, bacteria, microorganisms, subsurface environment.

I. INTRODUCTION

In studies on water transport in saturated soils, sediments, and, more generally, porous media, the saturated hydraulic conductivity at any given location is usually assumed to be constant through time. Experimentalists have, however, known for many decades that this parameter, indicative of the medium's ability to transmit water, can, and often does, vary considerably with time. As early as 1905, Slichter reported that

> previous experimenters on the flow of water through sands and gravels experienced much difficulty on account of the progressive reduction in flow of water through the sand when an experiment extended over a considerable length of time. No means had been found for avoiding this difficulty; even the use of distilled water was not entirely effective

In his laboratory experiments on sand tanks, Slichter (1905) used formaldehyde to prevent variations in flow rate and, although he made no measurements of microbial numbers or metabolic activity, he attributed the effect of formaldehyde to the inhibition of the growth of organisms within the sand. A few years later, Green and Ampt (1911) encountered what may be interpreted as a time decrease in saturated hydraulic conductivity in soil cores under laboratory conditions. These authors made water flow downward through the columns and observed in some of them an 80-fold reduction in flow rate during the first 2 weeks of the experiment, with the flow virtually stopping at the end of 3 months. By reversing the direction of water movement, they partially eliminated the decrease, which they attributed to silting action within the column as a result of downward percolation. In the early 1920s, Winterer (1922, 1923) carried out an extensive series of percolation experiments and pointed out a striking similarity between the curves of percolation rate vs. time for the different soils considered, all exhibiting three successive and apparently distinct phases. A decrease in flow rate immediately after the onset of an experiment was typically followed by a brief period of rather sharp increase after which the percolation rate decreased slowly but continuously over an extended period of time.

These early laboratory studies, and the flurry of investigations that followed, attest to the practical importance of this time dependence of the saturated hydraulic conductivity in a wide variety of practical situations. In a number of cases, reductions of the saturated hydraulic conductivity of soils or aquifer materials have beneficial consequences and are actively promoted. Talsma and van der Lelij (1976), for example, report a decrease over time of the hydraulic conductivity near the soil surface in a rice field under protracted ponding conditions. This decrease might in principle prevent excessive losses of water by deep drainage and thereby improve the typically very poor (e.g., Tuong et al., 1994) water use efficiency of irrigated lowland rice. Mirtskhulava et al. (1972) mention that an antipercolation technique was tested, apparently with success, and was routinely used in the USSR from 1951 to 1968 to artificially diminish the saturated hydraulic conductivity of sediments at the bottom of man-made lakes, pools, ponds, and other reservoirs through the application on the soil surface of a screen composed of a cellulose-rich material. The authors state that decomposition of the screen by anaerobic microorganisms induced gleyfication of the soil and resulted in decreased hydraulic conductivity. Ahmad et al. (1996) used compaction and a “bio-plastic sandwich” composed of manure, vegetative material, and soil to reduce seepage from water ponds in Pakistan by a factor of three.

In another practical situation, namely, the water flooding of petroleum reservoirs in secondary oil recovery projects (Updegraff, 1983a,b; Jack et al., 1983), the clogging of portions of the reservoir has beneficial consequences and may be profitably exploited. In a water flooding process, regions of high permeability in the reservoir rock, often called “thief” zones (Latil, 1980), constitute preferential pathways for the flood water, causing the bypassing of pockets of residual petroleum in the low permeability regions. Selective clogging of the thief zones, a process sometimes referred to as “stratification rectification” (Crawford, 1983), reduces fingering in the water flood and results in a higher percentage of the petroleum-bearing formation being swept by water, leading to higher oil recovery. Among plugging agents in current use, many are dispersed solids or semisolids in suspension (e.g., powders such as talk, zinc oxide, cement, colloids, resin, and emulsions soluble in caustic soda) (Latil, 1980). Lappin-Scott et al. (1988) suggest, as clogging agents, the use of starved cells that are later resuscitated in situ.

Aside from these various situations where one might have interest in promoting the biological clogging of porous media, there are other cases where biological clogging has deleterious consequences. Numerous authors report practical difficulties in the field disposal of wastes of various origins because of marked reductions of hydraulic conductivity over time. This problem particularly concerns industries that generate large quantities of organic wastes, such as dairy (Davis et al., 1973) and paper industries (Marchand, 1971). However, it also affects the disposal of domestic waste waters (e.g., Laak, 1970; McIntyre and Riha, 1991), sewage sludge (Thomas et al., 1968), and septic tank effluents (Bouma, 1971, 1975a,b; Daniel and Bouma, 1974; Jawson, 1976; Kristiansen, 1981a,b,c). Certain techniques, such as trickling or percolating filters, that are commonly used in the pretreatment of these wastes suffer to some extent from the same problem (e.g., Bruce, 1969; Gray, 1981) and different methods, such as the addition of microbicides to the percolating fluid, have been developed to alleviate it.

Surface waters used for artificial groundwater recharge often contain appreciable quantities of suspended sediments and microorganisms. In many instances (e.g., Rebhun and Schwarz, 1968; Behnke, 1969; Ripley and Saleem, 1973; Wood and Signor, 1975; Wood and Bassett, 1975; Vigneswaran and Suazo, 1987; Warner et al., 1994), accumulation of this suspended particulate matter causes the progressive clogging of the soil in the pits, ponds, or trenches used for aquifer recharge. A similar situation arises in flood water spreading operations (e.g., Berend, 1967). Parameters such as suspended solids content and biodegradable organic carbon, and column flow tests, used to compare clogging potentials of recharge waters, are not useful predictors of plugging in injection wells (Bouwer, 1996).

In the production of drinking water, the clogging of extraction or “discharge” wells is often a serious problem, particularly in cases where water is extracted from anaerobic zones in aquifers (Van Beek and Kooper, 1980). In the river region in the Netherlands, Van Beek and Van der Kooij (1982) estimate that the majority of the wells are clogged, leading to severely reduced water production. This clogging can be partially alleviated through back-flushing, subsurface pasteurization, chlorination, or regeneration with hydrochloric acid or sodium hypochlorite (Van Beek and Van der Kooij, 1982). However, the effect of these various remedial processes is only temporary, and they need to be repeated on a regular basis.

The experimental evidence that has accumulated in the last few decades thus reveals that the time dependence of the saturated hydraulic conductivity is a common occurrence in a wide range of situations, and that it can be controlled to a certain extent in cases where it has deleterious consequences. In other instances, it can even be used to advantage. Therefore, to more efficiently control or promote reductions in saturated hydraulic conductivity, the various mechanisms responsible for clogging need to be determined and sufficiently understood.

Strictly speaking, the time dependence of the saturated hydraulic conductivity of a porous medium is a purely physical phenomenon, a progressive variation in the medium’s physical resistance to water flow resulting from the reduction in size of the pore space or from changed friction coefficients or fluid viscosity. However, the causes of these structural and rheological changes, and consequently the mechanisms truly responsible for the time dependence, are usually not only of a physical nature. For convenience, they may be organized into three classes: physical, chemical, and microbial (Baveye and Jacquet, 1987; Vandevivere and Baveye, 1992a). It is clear that in particular situations, factors from each of these three classes interact and occur simultaneously (e.g., McGauhey and Krone, 1967; Jawson, 1976; Kreissl, 1978; Metzger et al., 1983; Warner et al., 1994) and that, in practice, it may not always be straightforward to determine which class of mechanism is prevalent. Nevertheless, from a reductionist viewpoint, the above classification appears to serve a useful purpose as a framework for research.

Among the three classes of mechanisms responsible for the clogging of soils and aquifer materials, the microbial one is at the moment of particular interest. Indeed, with the rapid emergence of groundwater and soil contamination as the primary environmental issues, the notion has evolved that microbial activity plays a major role in the development and decay of contaminant plumes in the subsurface (Molz et al., 1986; Ghiorse and Wilson, 1988). Much research effort is currently being devoted to designing efficient ways to stimulate the in situ biodegradation of organic contaminants in aquifers (e.g., Lee et al., 1988). Some of the techniques investigated in this respect involve the injection of growth substrates (C sources, electron acceptors) in- or downstream of contaminant plumes to create biologically active zones (BAZs). If, as a result of stimulating microbial activity, local clogging occurs to any significant extent in the vicinity of injection wells, it may result in increased injection pressures or, alternatively, in decreased injection rates. Even if clogging is successfully prevented, for example, by chlorination or by substrate pulsing (Roberts et al., 1990), in the vicinity of the injection wells, significant reductions of the saturated hydraulic conductivity may still occur in the BAZs as a result of the growth and metabolism of microorganisms, causing the BAZs to be bypassed by the injected nutrient solution and jeopardizing the chances of success of the bioreclamation efforts.

To facilitate future work on the design of efficient bioreclamation procedures and to understand more completely the role of microorganisms as clogging agents in the situations briefly reviewed above (rice fields, secondary oil recovery, artificial groundwater recharge, groundwater extraction), it would appear useful to assess what is currently known about the effects of microorganisms on the transport properties of porous media.

In this general context, the objective of the present review is to analyze the existing literature dealing with the influence of the growth and metabolism of microorganisms on the time dependence of the saturated hydraulic conductivity of porous media. This review is organized as follows. In the first section, following this introduction, we describe the experimental equipment most commonly used in studies on the temporal variation of the saturated hydraulic conductivity. Also addressed in this section are the methodologies used to assess the extent of clogging under field conditions. In the next section, typical spatial and temporal clogging patterns commonly observed in practice are described. The next section outlines physical and chemical mechanisms that may give rise to variations of the saturated hydraulic conductivity over time. It is necessary to be aware of the nature of these physical and chemical mechanisms and of the extent to which they may partially mask microbial effects and interact with them. The next section reviews in detail, from a number of angles, the available experimental evidence concerning the role of microorganisms in the clogging of soils and aquifer materials. Finally, the last section analyzes, from a conceptual standpoint, the few attempts that have been made to date to model mathematically the clogging process in aquifer materials and compares model predictions with recently published experimental data.

II. LABORATORY AND FIELD MEASUREMENT OF THE SATURATED HYDRAULIC CONDUCTIVITY

A. LABORATORY MEASUREMENT

The measurement of the saturated hydraulic conductivity, K_s, in laboratory columns is relatively easy to perform. If the flow rate and the hydraulic head gradient are measured simultaneously, K_s can be calculated by solving the following form of Darcy's law:

$$Q = A \cdot K_s \cdot \Delta H / z \quad (1)$$

where Q is the flow rate ($L^3\ T^{-1}$), A is the cross section area of the column (L^2), K_s denotes the saturated hydraulic conductivity (LT^{-1}), H is the hydraulic head, (water potential per unit weight, L), $\Delta H \equiv H_{in} - H_{out}$, and z is the distance between the two points where H is measured (L). The hydraulic head, H, is the sum of the pressure potential and the gravitational potential. The gravitational potential is the elevation relative to an arbitrary reference plane and the pressure potential, under saturated conditions, is proportional to the hydrostatic pressure in the porous medium. The pressure potential can be measured with pressure transducers (e.g., Raiders et al., 1986; Torbati et al., 1986) or with water manometers, often termed piezometers. A piezometer is simply a tube connected to the porous medium at one end, so as to permit the passage of the liquid phase, and open to the atmosphere at the other end. When the absolute pressure inside the column is greater than 110 kPa (1.1 bar), it becomes more practical to use mercury manometers. The key advantage of using pressure transducers vs. the more traditional piezometers is that they permit the measurement of the pressure potential to be fully automated. They also tend to be more sensitive than piezometers to small changes in hydraulic head.

As defined in Eq. 1, the saturated hydraulic conductivity K_s is a global property (e.g., Baveye and Sposito, 1984), characteristic of the soil column as a whole, like the porosity of the soil column or its bulk density. In studies on the time dependence of the saturated hydraulic conductivity, it is of particular interest to determine where in the soil column the clogging or unclogging occurs. To this end, piezometers are typically inserted in the column to evaluate the hydraulic head at various locations between the inlet and outlet. For each of the layers that these piezometers delineate in the porous medium, the saturated hydraulic conductivity K_{si} can be calculated using Eq. 1, recast as follows (e.g., Cunningham et al., 1990, p. 699; Jury et al., 1991):

$$K_i = Q / A \cdot z_i / (\Delta H)_i \quad (2)$$

where $(\Delta H)_i$ corresponds to the drop in hydraulic head across the ith layer in the column and z_i is the thickness of this layer.

The column (often termed permeameter column or simply permeameter) containing the porous medium whose K_s value is of interest can be operated at a constant flow rate or under a constant hydraulic head difference. The choice between these two options usually depends on the type of field situation that is being simulated in the laboratory. Both have been used more or less equally frequently. When it is desired to keep the flow rate constant, a peristaltic pump is usually used to maintain a constant flow of liquid to the column. On the other hand, when one desires to keep the hydraulic head loss constant, the column can be ponded with a volume of water held constant by means of an overflow port (e.g., Ripley and Saleem, 1973; Rice, 1974; Hills, 1976; Vigneswaran and Suazo, 1987) or, alternatively, a so-called Mariotte bottle is sometimes used (e.g., Orlob and Radhakrishna, 1958; Gupta and Swatzendruber, 1962; Lance and Whisler, 1972; McIntyre and Loveday, 1974; Vandevivere and Baveye, 1992a; Ragusa et al., 1994; Seki et al., 1996). It is likely that the time dependence of K_s would be different under conditions of constant flow or constant head loss. Indeed, the column held at constant flux will become increasingly pressurized as clogging proceeds, whereas the effect of clogging on the column held at constant ΔH will be a proportional reduction of the flow rate. Whereas an increase in pressure could lead to the dislodging of local plugs, the decrease in flow rate would slow down the supply of nutrients to the microflora present in the column.

There are still two other ways by which permeameter columns can be operated. Neither one has been used much in studies on clogging. In the falling head method (Hilton and Whitehall, 1979), neither Q nor ΔH are held constant. In the oscillating head method (Poulovassilis, 1972), the same volume of liquid flows back and forth through the permeameter in a batch mode. A more detailed account of laboratory methods to measure K_s can be found in McIntyre and Loveday (1974) and in Klute and Dirksen (1986).

Various artifacts can interfere with the measurement of K_s. Even though the porous medium is initially fully saturated with the liquid, partial desaturation can occur in the course of an experiment, even in the absence of gas production by microorganisms. Partial desaturation may occur if three conditions are fulfilled simultaneously: the liquid flows downward through the column, the effluent tubing opens to the atmosphere at a level underneath that of the top of the column, and a layer of lesser hydraulic conductivity forms at the column inlet as a result of slaking, clay dispersion, or biological clogging. These conditions lead to negative pressure potentials in the soil immediately underneath the surface crust and may therefore cause the soil to desaturate in this part of the column (McIntyre and Loveday, 1974; Daniel and Bouma, 1974). It is very likely that this phenomenon occured in a large number of studies on clogging for which no precautions were taken to prevent the development of negative pressure potentials (e.g., McCalla, 1945 and 1950; Allison, 1947; Jones and Taylor, 1965; De Vries, 1972; Frankenberger, 1979; Frankenberger et al., 1979; Okubo and Matsumoto, 1979 and 1983; Tollner et al., 1983). Even when precautions are taken to preclude the establishment of subatmospheric pressures in the permeameter, another purely physical phenomenon can lead to partial desaturation. In cases where the injected fluid is at a significantly higher pressure than atmospheric, such as when the flow is driven by a pump through a relatively impermeable porous medium, dissolved gasses can come out of solution as the pressure drops to atmospheric level at the outlet end of the permeameter (Raiders et al., 1986).

Another possible artifact is related to wall effects, in particular to the preferential flow of liquid along the interface between the porous medium and the solid container. For example, De Kimpe and Laverdière (1982) observed that 30 to 80% of the flow through paraffin-embedded soil cores occurred at the boundary between the soil and the paraffin. The importance of this effect, which can be decreased by increasing the diameter of the permeameter, still remains to be assessed. It is possible that preferential flow along the walls might be of lesser importance when the columns are filled with a noncompressible material, such as sand.

Another artifact concerns the motility of bacteria. Motile bacteria are able to swim in free water by up to 10 cm in less than 1 h (Stanier et al., 1987) and therefore rapidly spread from the soil or sand column to the entire apparatus, including retaining meshes or porous plates, the inlet chamber, or the nutrient medium reservoir. This will result, among other things, in a modification of the chemical composition of the injected liquid with time, especially when nutrients have been added. This phenomenon complicates the analysis of experiments in which the effect of the nutrient medium composition on clogging is investigated. Whenever significant bacterial growth occurs in the inlet portion of the flow apparatus, upstream of the porous medium itself, there is also a risk that the observed, overall reduction of the saturated hydraulic conductivity of the system be due to processes that occur almost entirely outside the soil or sediment sample. A similar situation may arise when inoculation is carried out in the inlet tubing, directly in the percolating solution. The injected organisms may colonize the internal surfaces of the tubing, as well as be filtered by meshes or porous plates in contact with the inlet surface of the porous medium. This seems to have been the case in the experiments of Lappin-Scott et al. (1988) and Vandevivere and Baveye (1992a), who observed the development of a filter cake on the inlet surface in their laboratory columns. In the experiments by Seki et al. (1996), the hydraulic conductivity of a glass filter at the inlet end of the column decreased by three orders of magnitude as a result of bacterial and fungal growth.

B. HOW EVIDENCE OF CLOGGING IS OBTAINED IN THE FIELD

The vast majority of field studies on clogging do not include actual measurements of K_s probably because of the difficulty associated with this measurement when the depth of saturation is unknown, the soil properties nonuniform, and the flow geometry ill defined, as it often is the case in field trials. One exception is the work of Schiff and Johnson (1958). Their field trial consisted of laboratory columns that were partly driven into the soil. Because the flow geometry was artificially controlled, it was possible to calcuate K_s in the same manner as it is done in the laboratory. Another approach to circumvent the difficulties associated with K_s measurement in field trials was adopted by Chang et al. (1974). These authors buried laboratory columns in pond sediments, where they became clogged, and later retrieved them to measure K_s in the laboratory.

Similarly, Howsam and Tyrrel (1989) described a sand-filled flow-through cell designed to measure biofouling in water production wells.

In field experiments involving land disposal of wastewater, the development of clogging of the ponded soil is usually monitored by measuring the water infiltration rate, whereas maintaining the depth of ponding at a constant level by way of an overflow port or a motorized valve activated by a water level float (Johnson, 1958; Wood and Bassett, 1975; Siegrist, 1987). It is not possible, however, with this type of set-up, to find out where in the soil profile clogging processes take place.

In field studies related to water-producing wells, the occurrence of clogging is characterized by a decrease in a well's specific capacity, defined as the discharge flow rate per meter of drawdown, where drawdown is the difference between the water level in the well during operation and inaction (van Beek, 1984). Clogging of water-injection wells, on the other hand, can be detected by decreased injection rate at a given injection pressure or by an increased injection pressure, at a fixed pumping rate. The installation of observation wells in the vicinity of the water-injection wells allows the measurement of hydraulic head gradients and therefore the identification of the region of the aquifer subjected to clogging (Oberdorfer and Peterson, 1985). A complicating factor in this analysis is the fact that part of the observed changes in pumping rate, injection pressure, or hydraulic head gradients over time may be due to the rise of the water table in the vicinity of the well. Proper account of this rise must be made in evaluating the intensity of the clogging.

III. CLOGGING PATTERNS

Various descriptions have been made of the evolution in time of the clogging of porous media (e.g., Otis, 1985; Vigneswaran and Suazo, 1987), both under ponded and intermittently ponded conditions. Even though there is a wide variety of clogging behaviors reported in the literature, it is useful at this point to describe in some detail the patterns observed by Allison (1947) and Okubo and Matsumoto (1979, 1983), both of which apply to situations of protracted water submergence.

Allison (1947) characterizes as "S-shaped" the time changes in permeability he observed when applying river water to soils (Figure 1a). These S-shaped curves consisted of three successive phases. Especially in soils with a low hydraulic conductivity, the first phase was an initial decrease in K_s, lasting for up to 20 d. The second phase led to an increase in K_s, and was noticeably more pronounced when the initial wetting of the soil took place from top to bottom, that is, when the soil was suddenly submerged. The third phase began, according to Allison (1947), after the maximum K_s value was reached. It consisted of a progressive but very marked decrease in K_s. The decrease was precipitous at first and then eventually leveled off after 2 to 4 weeks, at which point the saturated hydraulic conductivity had reached a very low value. The minimum reached by K_s between the first two phases could be due to a balance between the dominating processes in each of the two phases (Allison, 1947). A similar balance between opposite mechanisms was thought to be responsible for the occurrence of a maximum between the second and third phases.

A different pattern was observed by Okubo and Matsumoto (1979, 1983) (Figure 1b). These authors used a physically and chemically stable sand as the porous medium in their laboratory experiments. They percolated through columns packed with this sand various solutions of glucose, starch, peptone, and skim milk, all with carbon to nitrogen ratios of 1.44 and with total concentrations in organic carbon similar to those found in secondary treated wastewater. The pattern emerging from these experiments also consisted of three successive phases (Okubo and Matsumoto, 1983). At first, the infiltration rate decreased rapidly. Okubo and Matsumoto (1983) observed that effluent contained dissolved oxygen during this phase. After approximately 10 d, the infiltration rate stopped decreasing or even started to increase slightly for a period of up to 10 d, during which the dissolved oxygen content of the effluent was low. After this stationary or transition phase, the infiltration rate started again to decrease, and leveled off after about 40 d. During this time the effluent did not contain any measurable amount of dissolved oxygen.

The time evolutions of clogging that have been reported in the literature very often lack one or both of the first two phases reported by Allison (1947) and Okubo and Matsumoto (1979, 1983). The reasons for the existence of such a variability in the observed behaviors is not surprising, in view of the range of methods that have been used to measure K_s, and in view also of the various physical, chemical, and biological processes that, alone or in combination, can cause K_s reductions in porous media. These processes are analyzed in detail in later sections of this review.

In contrast with the variability of the temporal behaviors, the spatial patterns of clogging reported in the literature are characterized by a much greater uniformity. Indeed, in most clogged soils or aquifer materials subjected to prolonged submersion, the observed reduction in K_s takes place predominantly in a region close to the inlet (Otis, 1985; Vigneswaran and Suazo, 1987). Gupta and Swartzendruber (1962), for example, report that, in their sand columns, the saturated hydraulic conductivity dropped to 0.15% of its initial value within the first 1.5 cm, and that little change in K_s was observed at deeper depths. Rice (1974) notes that for water with a high concentration of suspended solids (SS), clogging proceeded fast but did not extend deeper than 4 cm from the surface, whereas for water low in SS, biological clogging was considered very significant, developing slower but extending somewhat deeper than 4 cm. For the non-motile bacterium used in their experiments, Vandevivere and Baveye (1992a) show that bacterial cells induce very locaized clogging because their rapid metabolism results in steep nutrient gradients, which preclude growth downstream. With non-motile bacteria, the exact location of this zone of pronounced clogging depends on where the bacteria are inoculated in the porous medium. In the experiments described by Vandevivere and Baveye (1992a), when the bacteria were introduced in the sand columns by inoculation in the feed line, K_s decreased only in a very thin layer (<3 mm) at the inlet end of the columns. When the bacteria were introduced at a certain depth into the columns, clogging proceeded at a slower pace and in a thicker layer located downstream of the point of inoculation. Of course, under field conditions, microbial populations present in aquifer materials or reservoir rocks contain motile strains, which are expected to determine the evolution of the spatial clogging pattern.

In one of the only studies of clogging under field conditions carried out to date, Oberdorfer and Peterson (1985) worked with injection wells at two sites in different sedimentary formations in Hawaii. These authors repeatedly observed the same type of pattern, slightly departing from the "typical" cases described above. For the first few days after the beginning of their experiments, clogging in the sedimentary materials took place predominantly within 50 cm of the injection wells. Oberdorfer and Peterson (1985) attributed this clogging to the entrapment of the organic matter suspended in the recharge water. After a few weeks, however, the zone of most intense clogging moved to a region located between 70 cm and 130 cm from the injection wells. Clogging in this region was interpreted as the result of dinitrogen gas production.

IV. PHYSICAL AND CHEMICAL CAUSES OF CLOGGING

As we mentioned in the introduction of this section, the time dependence of the saturated hydraulic conductivity of a porous medium is a physical phenomenon, brought about by a change in the capacity of the pores to conduct water. This decreased capacity may be the result of a decrease in the size of pores and/or of their interconnections, or even of a complete filling of pores. In the present section, the physical and chemical processes that can induce these modifications of the pore space are briefly reviewed.

A. PHYSICAL PROCESSES

According to McGauhey and Winneberger (1964, 1965) and McGauhey and Krone (1967), compaction by superimposed loads, smearing of the surface, and migration of fines are among the physical factors that can potentially lead to appreciable K_s reductions. Of these, the latter process is probably the most relevant once the porous media have been flooded. Under saturated flow conditions, percolating waters with a high concentration of suspended solids have been observed frequently to lead to severe clogging through a process of filtration (e.g., Rebhun and Schwarz, 1968; Rice, 1974; Okubo and Matsumoto, 1983, Ragusa et al., 1994). Three mechanisms limiting particle migration and causing their accumulation in porous media are reviewed in detail and summarized by McDowell-Boyer et al. (1986).

When the influent water contains suspended solids of a size commensurate with that of the particles of the porous medium, suspended solids penetration does not occur to any significant extent, and accumulation at the surface leads to the formation of a filter cake, which reduces the overall hydraulic conductivity of the medium. Smaller solids that may be suspended in the percolating water are then retained in the filter cake, enhancing further the resistance to flow. For suspended solids smaller than the particles of the porous medium, penetration inside the pore space is possible. Under these conditions, rather than the formation of an inlet filtercake, one commonly observes straining of the suspended solids inside some of the pores, which, as a result, become increasingly clogged (e.g., Fehr et al., 1992; Goldenberg et al., 1993). Large pores, however, may retain their full water carrying capacity, so that the overall K_s of the medium may not always be noticeably affected by this process. If the suspended solids are very small (that is, of colloidal size), thus possessing large specific surface areas, and if the conditions for flocculation are met, deposits thicker than one monolayer may form on the pore walls. The thickness of such deposits depends on the balance between hydrodynamic forces and strength of aggregation. In practical situations, suspended solids are likely to exhibit a wide range of sizes. Under these conditions, the observed outcome is expected to involve to some extent all three of the mechanisms briefly described above.

Vegetation may also result in declines in saturated hydraulic conductivity in wetland environments. In an experiment designed to simulate an artificial wetland constructed using sand, McIntyre and Riha (1991) observed that the hydraulic conductivity in both control and vegetated boxes was approximately halved in 150 d. An additional 14% decrease in the vegetated boxes compared with the control was attributed to soil compaction due to root growth and to production of root exudates.

Submergence of a soil may give rise to the disintegration of aggregate structure. Allison (1947) hypothesizes that dispersion and swelling accounted for the first phase in the time evolution of the saturated hydraulic conductivity of soil samples, during which K_s decreased markedly. He also considered that further dispersion under prolonged water saturation, as might occur to some extent in the third phase, could result from attack of microorganisms on organic binding agents of the aggregates. Among the factors that may contribute to structure destabilization (e.g., Ponnamperuma, 1984), two are strictly physical. Saturation of a soil causes a reduction in the cohesive forces between particles, whereas the entrapment of air within the aggregates can lead to significant pressure build-up. Depending on the mechanical properties of the particles constituting the soil and on the nature of the binding agents (polysaccharides, humic acids, sesquioxides), both of these processes may, under certain conditions, lead to the breakage of the aggregates and to the dispersion of their constituents in the surrounding pore space.

If a porous medium is not perfectly saturated at the onset of a percolation experiment, the progressive dissolution or the release of entrapped gas bubbles into the flow stream can significantly alter the measured K_s value (e.g., Faibishenko, 1985). Simply ponding a soil column does not completely eliminate the air from the pores (e.g., Ahmad, 1963). Although less air may remain entrapped, progressively wetting a soil column from beneath does not lead to complete saturation either (Allison, 1947; Pillsbury and Appleman, 1945; Christiansen, 1944). Allison (1947) hypothesized that air entrapment was responsible for the second phase he identified in the evolution of K_s over time. This conjecture was verified later by other researchers; wetting porous media under vacuum (Gupta and Swatzendruber, 1962; Shaw et al., 1985), under an atmosphere of CO_2 (Christiansen et al., 1946; Ripley and Saleem, 1973), or with a combination of these two methods (Raiders et al., 1986; Jenneman et al., 1983) indeed essentially eliminate this phase of K_s increase. The water could also be degassed beforehand, for example, by boiling (Gupta and Swatzendruber, 1962), to dissolve bubbles faster. Quantitatively, the process of gas entrapment can have significant consequences; Orlob and Radhakrishna (1958) estimated experimentally that, in their system, a 10% increase in the volume of entrapped air led to a 35% reduction in K_s.

B. CHEMICAL PROCESSES

The geometry of the pore space is linked with the chemical properties of the solid particles of soils or aquifer materials, and of the percolating aqueous solution. Electrolyte concentration, fraction of organic compounds in the aqueous phase, pH, redox potential, as well as the mineralogical composition of the solid phase, its surface characteristics, and the chemical reactions (precipitation/dissolution) it is involved in, all affect the shape and stability of the pores and therefore determine the value of the saturated hydraulic conductivity of the medium.

In terms of clogging, the processes of dispersion and swelling seem to be of particular significance (e.g., McGauhey and Winneberger, 1964, 1965, and McGauhey and Krone, 1967), even though their exact role in the clogging is still the object of controversy. Several investigators concluded that decreasing the electrolyte concentration and/or increasing the sodium adsorption ratio of the percolating solution causes clay-sized particles to disperse and lodge in soil pores. The latter is hypothesized to induce the observed reductions in the K_s of clay-containing soils, especially if the exchangeable sodium percentage is high (Felhendler et al., 1974; Frenkel et al., 1978; Pupisky and Shainberg, 1979; Shainberg, 1984; Das and Datta, 1987; Shainberg and Levy, 1992; Rengasamy et al., 1996).

This conclusion is based on the observation, made in a number of studies, that dispersed clay appeared in the effluent solutions at the same time K_s was experiencing a sharp drop. Abu-Sharar et al. (1987) note, however, that in these percolation experiments, the concentrations of dispersed clay in the effluent solutions peaked when approximately one pore volume of the infiltrating solution had passed through the soil columns. The concentration of dispersed clay then declined with further leaching, whereas the saturated hydraulic conductivity continued to decrease. On the basis of these observations and of independent experiments on three Californian soils, Abu-Sharar et al. (1987) argue that the appearance of clay in the effluents indicates that dispersed clay was mobile in the soil columns and thus cannot be plugging a major part of the soil cores. According to these authors, the major cause of K_s reduction is the blockage of large conducting pores as a result of aggregate failure (slaking). However, Vinten and Nye (1985) and Abu-Sharar and Salameh (1995) observed clogging due to clay particles present in the infiltrating water. Experimental results obtained by Vandevivere and Baveye (1992d) show that highly mobile particles (in this case, cells of Arthrobacter sp. strain AK19) can cause appreciable K_s reductions.

In some cases, high concentrations of organic compounds may result in increased hydraulic conductivity in clays. Quigley and Fernandez (1989) observed that hydraulic conductivity of water-compacted clay increased by as much as three orders of magnitude in the presence of aqueous leachate solutions containing greater than 70% hydrocarbon. Below 70% hydrocarbon the hydraulic conductivity was decreased by approximately a factor of two. The authors hypothesize that the high concentrations of organic compounds caused the electrical double layer to contract, resulting in increased hydraulic conductivity.

V. BIOLOGICAL CAUSES

A. EVIDENCE OF BIOLOGICAL CLOGGING

The significance of biological vs. nonbiological clogging can be assessed by the use of control treatments that suppress biological activity. A rapid K_s decline that was observed in columns packed with sand operated with a constant flux of deionized water was eliminated when phenol (0.1% final concentration) was dissolved in the solution or when the experiment was run at low temperature (1.5°C) (Gupta and Swartzendruber, 1962). Raising the temperature induced rapid clogging. Addition of NaClO has also been shown to be effective in suppressing clogging in granular porous media such as glass beads (Shaw et al., 1985) or reservoir sand (Geesey et al., 1987). In soils, sodium azide (NaN_3) has been used by Seki et al. (1996) to alleviate clogging (see also Miyazaki, 1993). A 2% formaldehyde solution was used with similar success by Ragusa et al. (1994). However, in general, treatments with microbicides such as phenol (0.1%), clorox (1%), Cl_2 (60 ppm), or formaldehyde additions do not seem as effective at maintaining high infiltration rates in soils as they are in mineral porous media (Allison, 1947; Johnson, 1958; Ripley and Saleem, 1973). The reason for this observation may be that in general these disinfectants are less effective in the complex environment of a soil or that the oxidant nature of some of these disinfectants brings about a degradation of organic cements and a breakdown of the soil structure (Johnson, 1958). Sterilization, on the other hand, occasionally has been shown to completely eliminate clogging in soil systems. Allison (1947) ran percolation experiments in soil columns with and without prior sterilization with ethylene oxide. Whereas, after an initial rise, the saturated hydraulic conductivity declined rapidly in the non-sterile soil, it remained constant throughout the duration of the experiment in the sterile soil. Moreover, after seeding the sterile soil with fresh soil inoculum, rapid clogging developed. Similar results have been obtained with steam sterilization of soil columns (Frankenberger et al., 1979) or by maintaining the soil columns at a low temperature (McCalla, 1950).

If clogging is induced by the activity of microorganisms, one would expect that the addition of growth substrates should accelerate the process. This, indeed, has often been observed. For example, additions of carbon and energy sources such as plant residues, monosaccharides, disaccharides, or alcohols both accelerate and enhance soil clogging (Allison, 1947; McCalla, 1950; Frankenberger et al., 1979). Further addition of mineral N resulted in even greater clogging (Frankenberger et al., 1979).

Finally, were the principal cause of clogging linked in some way to the activity of microorganisms, one would expect to find a correlation between the K_s decline and various indicators of microbial activity. Indeed, various investigators have found that the extent of clogging is correlated with the number of colony forming units (CFU) in soil profiles (McCalla, 1950; Johnson, 1958; Gupta and Swartzendruber, 1962; Frankenberger et al., 1979). An exception to that rule is described by Frankenberger et al. (1979), who found fewer CFUs, both aerobic and anaerobic, in soil samples taken from the column in which the greatest reduction in K_s had been observed. As this particular column had been continuously infiltrated with a solution rich in glucose (500 mg L^{-1}) and KNO_3 (50 mg L^{-1}), it is possible that the important stimulation of the microbial processes that should have ensued generated a toxic effect that inactivated most microorganisms at the sampling time. Be that as it may, indicators of microbial activity such as phosphatase activity (Frankenberger et al., 1979), substrate disappearance, or product accumulation (Thomas et al., 1966; Oberdorfer and Peterson, 1985; Ehlinger et al., 1987) usually correlate well with the extent of clogging.

B. TYPES OF ORGANISMS FOUND IN CLOGGED SOILS

Once the key clogging agent is identified as being biological, the next question one may ask relates to the type of organisms involved in the process. Bacteria are the predominant microorganisms in flooded rice soils (Yoshida, 1975, 1978; Sethunatthan et al., 1983) and in aquifers. As noted by several investigators, the predominance of prokaryotic forms seems to be characteristic of most waterlogged systems (Mitchell and Alexander, 1962; Laak, 1970; Jawson, 1976; Kristiansen, 1981c; Starkey and Karr, 1984; Shaw et al., 1985; Raiders et al., 1986). Identification of particular physiological groups of microorganisms by microscopic observation of clogged systems has been limited in the past to groups possessing distinct morphological features such as iron bacteria (Kuntze, 1982; Updegraff, 1983a; van Beek, 1984; Cullimore and Mansuy, 1987), methanogenic bacteria (Ehlinger et al., 1987), or algae (Bubela, 1985). On the other hand, isolation of microorganisms from samples taken in clogged systems allows further characterization of their specific metabolism. Adopting this approach, a number of investigators identified various strains of aerobic bacteria (Laak, 1970; Updegraff, 1983a), facultative anaerobic bacteria (Mitchell and Nevo, 1964; Laak, 1970; Jack et al., 1983), and strict anaerobic bacteria such as sulfate-reducing bacteria (Davis, 1967; Wood and Bassett, 1975; van Beek and van der Kooij, 1982). Examining bacterial populations recovered from sand-filter columns treated with either a high- or low-carbon loading, Pell and Ljunggren (1996) used principal component analysis to distinguish several groups of bacteria with different carbon substrate and supplemental nutrient requirements. It should be pointed out that the isolation of particular organisms from clogged soils does not indicate whether these isolates were active in the environment from which they were obtained. Furthermore, the microorganisms that are responsible for the observed K_s reductions may not be culturable on standard laboratory media. Therefore, it may be more meaningful to monitor in situ the activity of different groups by measuring the consumption/production of chemical species that are characteristic of different types of metabolism. This particular approach has linked K_s reductions to the activity of aerobic bacteria (Okubo and Matsumoto, 1979), denitrifying bacteria (Lance and Whisler, 1972; Jawson, 1976; Oberdorfer and Peterson, 1985; Ronen et al., 1989), acetogenic bacteria (Okubo and Matsumoto, 1979; Ehlinger et al., 1987), sulfatereducing bacteria (Wood and Bassett, 1975; Jawson, 1976; Siegrist, 1987), methanogenic bacteria (Jawson, 1976; Kristiansen, 1981a; Tollner et al., 1983), and photosynthetic sulfur bacteria (Tollner et al., 1983).

Although the anaerobic environment that develops under prolonged submergence can lead to large reductions in the number of eukaryotic organisms (Mitchell and Alexander, 1962), various types of eukaryotes have nevertheless been observed in clogged porous material. Fungi have been found associated with clogged porous material on numerous occasions (Ripley and Saleem, 1973; Okubo and Matsumoto, 1983; Seki et al., 1996). Ragusa et al. (1994) obtained a reduction of the hydraulic conductivity of soil samples to less than 22% of its original value after inoculation with benthic green algae Chlorococcum sp. Clogging appeared to be a result of the development of an algal/bacterial mat on the soil surface. Protozoans are also occasionally involved in the clogging process. Ciliates have been detected in severely clogged sand columns (Okubo and Matsumoto, 1983). Hilton and Whitehall (1979) identified a variety of ciliated protozoa in the fouled surface layer of rapid sand filters. Likewise, Calaway (1957) found protozoans, especially ciliates, and metazoans to be important members in the ecology of intermittent sand filters. The exact function of eukaryotes in porous media with respect to K_s is poorly understood. Their occurrence in clogged layers suggests that they contribute to reductions in K_s. However, it is possible that they were simply injected with the influent feed water or that they were feeding on the microorganisms responsible for the clogging. In column experiments, DeLeo and Baveye (1997) found that periodic additions of amoebae at the column inlet did not appreciably reduce hydraulic conductivity. Via predation of bacteria, amoebae instead merely delayed the occurrence of clogging. In field situations, there is circumstantial evidence suggesting that protozoa may inhibit reductions in K_s in a eutrophic aquifer (Sinclair et al., 1993).

A question that remained unsettled until recently concerns the relative impact of aerobic and anaerobic metabolisms on K_s. Recurrent statements can be found in the literature that refer to the need for an anaerobic environment for large K_s reductions to occur. Such statements are based on the frequent observation that an accelerated phase of clogging is coincident with a decline in the redox potential or the appearance of black FeS deposits, which are signs of a reduced environment (Thomas et al., 1996; Nevo and Mitchell, 1967; Wood and Bassett, 1975; Okubo and Matsumoto, 1983; Siegrist, 1987). This association of K_s reductions with anaerobic conditions is usually explained by the slow degradation of organic matter in anaerobic environments, leading to a rapid accumulation of organic residues that plug the pores (Thomas et al., 1966; Nevo and Mitchell, 1967; Kristiansen, 1981a). Allowing a soil profile to drain, thereby promoting the mineralization of the accumulated organic material, usually restores the original infiltration capacity (Thomas et al., 1966; Chang et al., 1974).

Conceptually, however, the accumulation of plugging organic substances (which may or may not include biomass) is the net result of the balance between their production and their decay via mineralization. Because both processes appear to proceed faster under aerobic conditions (e.g., Nevo and Mitchell, 1967; Okubo and Matsumoto, 1983), one cannot a priori decide that anaerobic conditions will lead to greater accumulations. At least under certain conditions, a greater accumulation of organic matter can occur under aerobic than under anaerobic conditions (Winneberger et al., 1960). In fact, a close analysis of published data casts doubts on the complete dismissal of aerobic metabolism as a significant part of the clogging process. Various types of porous media containing no or very little organic matter became clogged when supplied with deionized water (Updegraff, 1983a; Shaw et al., 1985). Under those conditions, it is very unlikely that the flowing suspension would ever become anoxic. It is nonetheless possible that once the attached biomass starts decaying, sufficient amounts of nutrients are released to deplete the solution of its O_2. This was, however, not observed by Taylor and Jaffé (1990a), who supplied sand columns with a mineral medium containing up to 7.2 mg l^{-1} of methanol as the sole carbon source. During the 9-month-long experiment, K_s decreased by 3 orders of magnitude, although the column effluent was never found to contain less than 2.6 mg O_2 l^{-1} in trial runs. It is conceivable that organic matter turnover within the attached biofilms caused a local depletion of O_2, thereby inducing anaerobic metabolism. In other studies, there was an initial phase of clogging occurring in an aerobic environment followed by a second phase during which anaerobic conditions prevailed (Thomas et al., 1966; Okubo and Matsumoto, 1979, 1983). During the aerobic phase, K_s was seen to decline 10- to 100-fold, and, during the anaerobic phase K_s further declined 10-fold. Some investigators compared the extent of clogging in columns supplied with either aerobic or anaerobic solutions. Loading soil columns with aerobic effluent always resulted in higher clogging rates and greater extents of clogging compared with anaerobic effluents (Kropf et al., 1975; Okubo and Matsumoto, 1983, Raiders et al., 1986; Cullimore and Mansuy, 1987).

Another piece of evidence in support of the role of aerobic organisms is the fact, mentioned previously, that the region where clogging is most severe often consists of a very thin layer immediately underneath the infiltrative surface (e.g., Gupta and Swartzendruber, 1962; Rice, 1974; Okubo and Matsumoto, 1979; Raiders et al., 1986). Independent observations suggest that aerobic conditions are likely to prevail in this portion of the soil profile. For example, according to Ponnamperuma (1984), molecular diffusion suffices to maintain an oxidized zone, a few millimeters thick, in paddy soils. Therefore, it becomes apparent that aerobic metabolism cannot be entirely dismissed as part of the clogging process.

In a recent study, Vandevivere and Baveye (1992a) provide conclusive evidence that bacterial reductions of the K_s of natural porous media do not require the development of anaerobic or microaerophilic conditions in these media. Their results show that a strictly aerobic bacterial strain, Arthrobacter sp. strain AK19, is able to reduce K_s in sand columns by three to four orders of magnitude in a matter of a week.

C. SUCCESSION OF TYPES OF BACTERIAL METABOLISM IN SUBMERGED SOILS

Because clogging caused by microorganisms evolves in time and space, it is informative to characterize the spatio-temporal successions characterizing microbial communities. Such knowledge could result in the identification of the microorganisms and mechanisms responsible for the decrease in K_s. According to Parr and Papendick (1971), it is essential to be aware of the redox condition, the nature of microbial metabolism, and the degree of soil anaerobiosis when resolving problems of poor internal drainage. In this section, the spatial and temporal successions of different microbial metabolisms in submerged soils that are not subjected to water flow are examined, followed by a discussion of cases in which the liquid phase moves through the soil matrix.

In a static waterlogged soil, enzymatically mediated reactions result in a lowering of the redox potential of the system (Koyama, 1955; Takai et al., 1956; Bell, 1969; Whisler et al., 1974; Yoshida, 1975). A convenient parameter for the redox intensity is the pE, defined as $pE = -\log(e) = Eh/0.059$, where (e) is the electron activity and Eh the redox potential. In the same way that the pH is proportional to the relative tendency of a solution to accept or transfer protons, the pE measures the relative tendency of a system to accept or transfer electrons (Stumm and Morgan, 1981). Thus, the higher the redox potential of a system, the lower the activity of the electrons, the greater the tendency to accept or transfer electrons, the greater the tendency of an oxidant to be reduced, and the more free energy will be released during the redox reaction (Stumm and Morgan, 1981).

Bacteria harvest energy by enzymatically catalyzing redox reactions that would otherwise proceed spontaneously, although usually at an exceedingly slow pace (Champ et al., 1979; Stumm and Morgan, 1981). For ecological studies, a useful classification of bacteria is based on the electron acceptor used for their energy-yielding reactions. Chemoorganotrophic bacteria oxidize organic molecules to obtain energy. They can use a wide array of oxidants to oxidize the organic matter, among which the most important are O_2, NO^-_3, Mn^{4+}, Fe^{3+}, and SO^{2-}_4 (cf. Table 1). According to thermodynamic theory, a given reductant (or electron donor) will reduce the stronger oxidant available in the system to maximize the release of free energy. Thus, more energy-yielding reactions, e.g., aerobic respiration or denitrification (Table 1), should precede processes that are less energy-yielding, such as sulfate reduction or methanogenesis. Enzymatically catalyzed reactions seem to follow that rule to a large extent. Indeed, the initiation of NO^-_3, Mn^{4+}, Fe^{3+}, SO^{2-}_4, and CO_2 reduction in soil systems follows the thermodynamic rule (Patrick and Delaune, 1972; Patrick and Reddy, 1978; Jakobsen et al., 1981).

Measurements of the redox potential, Eh, in soils should be interpreted with caution and, in any case, such measurements provide only qualitative information on the stage of reduction of the system. According to Champ et al. (1979), it is nevertheless possible to identify three redox zones in waterlogged soils and sediments with the use of an electrode. A zone with a redox potential greater than + 0.3 V, where O_2 and NO^-_3 are the dominant oxidants, a zone with a redox potential between + 0.3 V and + 0.1 V, where Mn^{4+} and Fe^{3+} are the dominant oxidants, and finally a zone with redox potential lower than + 0.1 V, where SO^{2-}_4 reduction occurs.

Fermentative microorganisms use dissolved organic molecules both as electron donors and as electron acceptors. Hydrogen is a common product of fermentation pathways (Brock and Madigan, 1988, p. 622). The production of H_2 enables CO_2 to be used as an electron acceptor by methanogens (Bryant, 1979). Hydrogen can also serve as an electron donor for sulfate-reducing bacteria. The H_2/H^+ couple maintains the redox potential at -300 mV (Bell, 1969). As indicated by H_2 evolution, fermentative pathways occur when the O_2 and NO^-_3 are depleted (Koyama, 1955; Takai et al., 1956; Bell, 1969). Thus, there is a temporal succession of physiological groups in which high energy-yielding reactions precede processes that are less energy-yielding. Aside from a purely thermodynamic explanation, this observed succession probably also results from enzyme inhibitions, toxicity, and competition mechanisms (Jakobsen et al., 1981). Moreover, as is typically the case for methanogens, certain biochemical pathways rely on the presence of a particular substrate that occurs as a byproduct of the metabolism of other groups. Some overlap between reductions of different electron acceptors are regularly observed (Takai et al., 1956; Patrick and Reddy, 1978; Jakobsen et al., 1981, Ehlinger et al., 1987). At least in one case (Jakobsen et al., 1981), this overlap could not be explained by the existence of microsites because the system was well mixed.

The reduced solutes present in submerged soils diffuse toward the oxic boundary at the soil-water interface where they are reoxidized spontaneously or enzymatically, whichever happens first. Among the specialized microorganisms that thrive in this narrow region, characterized by a steep redox gradient, are the iron bacteria that catalyze the oxidation of Fe^{2+}, the methylotrophs that oxidize the methane, and the sulfur bacteria that oxidize the sulfides. Patrick and Delaune (1972) show that these various reductants are oxidized sequentially in space according to thermodynamic principles. Thus S^{2-} is oxidized first, then Fe^{2+}, then Mn^{2+}.

In open systems characterized by a continuous supply of fresh medium with dissolved organic carbon and electron acceptors, all successive steps of redox reactions that occur sequentially in closed systems, could theoretically proceed indefinitely in successive and spatially contiguous zones along the direction of flow (Champ et al., 1979). The ensuing profiles of redox potential and oxidant-reductant couples, illustrated in Figure 2, have indeed been observed in ponded soils (Wood and

Bassett, 1975) and in aquifers (Champ et al., 1979; Mariotti, 1986; Barcelona et al., 1989; Bennett et al., 1993; Eganhouse et al., 1993; Baedecker et al., 1993). However, if the flow rate decreases as a result of clogging under gravity flow, for example, the rate of oxidant depletion at a particular depth eventually becomes greater than the rate of oxidant import. Eh then decreases or, in other words, the Eh gradient becomes progressively steeper (Wood and Bassett, 1975). Under these conditions, the bulk of the system successively undergoes the different metabolic stages described for closed systems, ending with CH_4 production. Only in a narrow zone, close to the point of injection of the substrates, do these various stages exist concurrently.

D. INFLUENCE OF GRAIN-SIZE DISTRIBUTION OF POROUS MEDIA

Because the microbial clogging of a porous medium amounts to a reduction of its ability to transmit water, one would expect that the rate and the extent of the clogging would be strongly influenced by the initial transport characteristics of the medium. In typical aquifer materials and in structureless soils, these characteristics are correlated with the grain-size distribution of the medium. A coarse-textured material is expected to have a high saturated hydraulic conductivity, whereas the opposite is true for fine-textured materials.

Only a few studies have addressed this issue of the effect of the physical environment on the microbial clogging process. Kalish et al. (1964) show that the rate of K_s decline is significantly higher in a low-permeability sandstone than in a high-permeability one. Raiders et al. (1986) conducted continuous flow experiments with two cores of Berea sandstone, one of higher permeability than the other, connected in parallel without cross flow. When a solution containing a Bacillus strain and nutrients was injected into this dual-core system, the higher permeability core, which initially transported 76% of the total flow, was preferentially plugged, so that it transmitted only 10% of the fluid. Torbati et al. (1986) incubated unsterilized cores of Berea sandstone with nutrient solutions. After incubation, the permeabilities had decreased and the pore entrance size distributions shifted toward smaller sizes, suggesting that the larger pores within the cores were partially plugged. The results of Cunningham et al. (1991) and Vandevivere and Baveye (1992d) are arguably the most conclusive to date, concerning the relation between clogging intensity and initial saturated hydraulic conductivity (cf. Figure 3). Up to an α value of approximately 0.14, the experimental data of K_s/K_{so} vs. α exhibit a very consistent pattern: fine grain sizes lead to a much more significant clogging than coarse ones. Some of this pattern disappears at higher α values, where the behavior of the three intermediate curves (for the 0.7 mm, 0.54, mm and 0.12 mm sands) changes appreciably. Nevertheless, the two extreme data sets, for the 1-mm glass beads and for the 0.09-mm sand, respectively, remain distinctly apart over the whole range of α values where data are available (Vandevivere et al., 1995).

The behavior exhibited in Figure 3 cannot be satisfactorily explained in terms of porosity differences. Indeed, unstructured, fine-textured materials generally have a higher porosity than coarse-textured ones, so that a given biomass would occupy a lesser portion of the total porosity in the former case than in the latter. Another parameter, which is strongly linked with the granulometry, may explain some of the observations; the specific surface area decreases sharply with an increase in average particle size. In coarse-textured porous media, therefore, the specific surface area available for colonization by microorganisms is significantly less than that in fine-textured materials. A given biomass may have a tendency to accumulate in the form of a biofilm on the internal surfaces of a coarse-textured porous medium. On the other hand, as shown clearly on scanning electron micrographs (cf. e.g., Figures 3 and

7 in Vandevivere and Baveye (1992a) or Figure 5b in Vandevivere and Baveye [1992b]) the bacterial coverage of solid surfaces in fine-textured porous media is often sparse and heterogeneous.

Rittmann (1993) argues that the grain-size distribution does not, in and of itself, determine the distribution of microorganisms in the pore space. He proposes a new parameter, called normalized surface loading, to explain observed patterns of colonization of porous media. This normalized surface loading depends on a number of variables, such as the bulk liquid flow rate in the porous medium, the substrate concentration entering the system and the substrate concentration in the effluent, and a “reference” flux, defined as the minimum flux producing a deep, steady-state biofilm in the system. Introduction of this reference flux is largely heuristic, because it cannot be measured in practice. The normalized surface loading is inversely related to the volume of the porous medium in which the clogging occurs. This volume introduces a high level of arbitrariness in the use of the normalized surface loading concept, because to obtain a high value for this parameter (supposedly associated with the presence of biofilms), one only needs to reduce the volume of the system under consideration. This feature makes the normalized surface loading of little use in natural porous media, such as soils or aquifers, which can be sampled at a multitude of scales.

E. MECHANISMS OF BACTERIAL CLOGGING

1. ACCUMULATION OF CELL BODIES

The saturated hydraulic conductivity of a porous medium is a function of the physical properties, such as the viscosity and gravimetric density, of the fluid it contains, and of the physical properties of the porous medium itself, such as the volume, size and interconnectedness of water-conducting pores, and the “roughness” of the pore walls. Theoretically, the presence of bacterial cells in soils could alter all these properties and thereby affect K_s.

The effect on the fluid properties can easily be discounted on the basis of numerous studies (e.g., Shaw et al., 1985) involving the injection of bacterial suspensions in model core systems. In these studies, the K_s decline is progressive and is correlated with the gradual accumulation of bacteria retained in the column. Had K_s been affected by a change of the physical properties of the fluid, the effect should probably have been noticeable from the onset of the experiments.

The importance of the increased friction caused by the development of a biofilm has been studied mostly in cylindrical pipes, for which it has been shown that the occlusion of the pipe lumen due to the existence of a biofilm could not account for the observed loss of pressure head (Picologlou et al., 1980). The finding that the head loss was proportional to the abundance and length of attached filamentous microorganisms prompted the hypothesis that pressure head losses in the pipes were caused by the increased frictional resistance associated with these filaments in a manner that is analogous to increased drag in streams due to bottom vegetation (Picologlou et al., 1980; McCoy et al., 1981). In porous media, however, the increased roughness or frictional resistance of the porous medium particles resulting from bacterial growth at surfaces seems insignificant, as suggested by the work of Characklis et al. (1987). In their experiment involving simulated porous media inoculated with a Pseudomonas aeruginosa strain, the permeability (the component of K_s that is independent of the fluid properties) was a linear function of the equivalent pore diameter. This equivalent pore diameter is the ratio of fluid volume over wetted surfaces and as such quantifies the size of the fluid-conducting pores. On the basis of these results, Characklis et al. (1987) conclude that increased roughness (frictional resistance) of the pore walls is inconsequential and that bacterial cells affect K_s mainly by altering the pore geometry, that is, by reducing their cross-sectional area.

There is a large body of literature, mainly relevant to oil production, that describes the plugging of consolidated material during the injection of bacterial suspensions (e.g., Kalish et al., 1964; Raleigh and Flock, 1965; Davis, 1967; Updegraff, 1983a and 1983b). As these studies involved killed cells and porous media with most of their pores in the same size range as bacteria, the mechanism of plugging is straightforward, mainly the consequence of mechanical sieving at pore throats. For that reason, these papers are not reviewed here.

Several authors have attempted to correlate the extent of clogging with bacterial numbers. Gupta and Swartzendruber (1962) found that K_s in sand columns was unaffected at bacterial densities below 0.4×10^6 CFU g^{-1}, whereas 100-fold K_s reductions were observed for bacterial densities higher than 1.3×10^6 CFU g^{-1} (Figure 4). Elementary calculations showed that, at the critical threshold where K_s drops precipitously, the bacterial cells would occupy only 0.03% of the total void volume of the sand. It should be pointed out, however, that the analysis of Gupta and Swartzendruber (1962) rested on the questionable assumptions that the extraction of the cells from the sand was 100% effective, and that the subsequent preparation of the bacterial suspension was able to perfectly dissociate aggregated cells (Vandevivere and Baveye, 1992a). Moreover, the plating technique used by Gupta and Swartzendruber (1962) does not allow the detection of either nonviable cells or cells (in particular, all the strict anaerobes) that are unable to multiply on the substrate used for plate counting (Thornston's standardized agar medium). In some cases, plate counts can underestimate the total counts in clogged sand by three orders of magnitude (Kristiansen, 1981c), a finding that decreases considerably the meaningfulness of the microbiological data presented by Gupta and Swartzendruber (1962). The same limitation applies to the data reported by Frankenberger et al. (1979), who also found low plate counts, both aerobic and anaerobic, that never exceeded 10^7 CFU g^{-1} in soil columns whose K_s values had declined by up to three orders of magnitude. Total counts, in addition to plate counts, were carried out by Kristiansen (1981c) in clogged sand supplied with wastewater. The top few centimeters, which corresponded to the clogged layer, contained up to 3×10^{10} cells g^{-1} of sand (direct counts), which was equivalent to 1.4% of the total void volume of the sand.

More recently, Vandevivere and Baveye (1992a), in experiments in sand columns inoculated with Arthrobacter sp. and using a phospholipid assay method, determined that for biomass densities less than 4 mg wet weight cm^{-3}, no significant reduction in K_s was detected. Above that value, large reductions in K_s occurred. The K_s was reduced by one, two, and three orders of magnitude as the biomass density reached 10, 20, and 35 mg wet weight cm^{-3}, respectively. When expressed as the percentage of the pore volume occupied by bacterial cells, these densities amounted to 2.4%, 4.8%, and 8.5%, respectively.

In a subsequent study, using a mass balance approach, Vandevivere and Baveye (1992d) computed biomass densities at various stages during the clogging of sand columns, under conditions in which neither exopolymers nor bacterial mats at the inlet end of the permeameters were produced. These authors found that biomass densities occupying 3.8% and 6.3% of the pore space caused 10- and 100-fold K_s reductions, respectively.

As the above experimental results clearly indicate, bacterial cells can dramatically reduce the K_s of porous media, even though in most cases they only occupy a tiny fraction of the pore space. This contradiction has led a number of authors to rule out a direct effect of bacterial bodies on hydraulic (e.g., Seki et al., 1996). However, bacterial cells could cause significant clogging if they were strategically located at grain contacts or at pore necks throughout the sand. Vandevivere and Baveye (1992a,b,d) suggest that this strategic localization may be related to the propensity of some bacteria to form aggregates of various sizes. In particular, Arthrobacter AK19 (Vandevivere and Baveye, 1992a) does not form biofilms on sand grain surfaces but rather appears to form three-dimensional aggregates that occupy the lumen of pores. The cells seem able to withstand convective transport by remaining attached after cell division, thereby forming large, loosely packed assemblages. Peripheral cells of these assemblages adhere to pore walls by means of exopolymeric linkages or via physical adsorption. Some loose aggregates of cells seem, however, to be simply trapped at pore constrictions: they might have been carried across a certain distance by the percolating liquid before being filtered out. Similar observations were made with other microorganisms (Vandevivere and Baveye, 1992b,d).

2. PRODUCTION OF BACTERIAL EXTRACELLULAR POLYMERS

Most prokaryotic organisms excrete slimy or gummy materials on their surfaces (Brock and Madigan, 1991). Although the vast majority of these excretions identified so far consists essentially of polysaccharides (Marshall, 1985), they will be referred to in the following as exopolymers instead of exopolysaccharides because in the vast majority of studies dealing with soil clogging, their polysaccharidic nature was not ascertained.

Exopolymers are not recalcitrant molecules because they do not accumulate indefinitely in soils. Indeed, traditional enrichment techniques usually yield isolates having the ability to use the polymers as sole source of carbon and energy (Mitchell, 1965). If exopolymers are to play a significant role in clogging processes, they should resist rapid decomposition or at least undergo mineralization at a slower pace than their rate of production. Both exopolymer production and degradation depend on a variety of environmental parameters such as soil type (Chang et al., 1974), moisture conditions in the soil or sediment (Roberson and Firestone, 1992), the presence of surfaces (Vandevivere and Kirchman, 1993), temperature (Geesey et al., 1987), redox potential (Nevo and Mitchell, 1967), the availability and nature of organic substrate (Avnimelech and Nevo, 1964; Tollner et al., 1983; Linton et al., 1987), nitrogen availability (Dudman, 1964; Avnimelech and Nevo, 1964; Williams and Wimpenny, 1977 and 1980), O_2 concentration (Eagon, 1956; Dudman, 1960 and 1964; Sutherland, 1977; Starkey and Karr, 1984), and the physiological status of microorganisms (Uhlinger and White, 1983; Lappin-Scott et al., 1988). Thus, there may exist some combination(s) of these parameters that favor(s) the production of polysaccharides over their mineralization, which would explain their accumulation in clogged soil. Another possibility to account for slowed mineralization of microbial exopolymers may involve certain chemical reactions. Polyuronides (polymers of uronic acids) appear more resistant to degradation than polysaccharides (Mitchell and Nevo, 1964; Tollner et al., 1983), possibly because of the formation of salts with metals (Martin and Richards, 1969) or complex compounds with other materials (Avnimelech and Nevo, 1964).

Allison (1947) was apparently the first to suggest that microbial waste products such as slimes might play a significant role in the clogging process. Exopolymers, as suggested by the term “slime” used originally, are usually highly hydrated owing to their hydrophilic nature and contain about 99% water (Rittman and McCarty, 1980). This high degree of hydration accounts for the difficulty associated with the observation of bacterial exopolymers via electron microscopy (Vandevivere and Baveye, 1992e). Exopolymers can form either a gel structure or a highly viscous sol (Gardner, 1972). It is therefore apparent that exopolymers could affect K_s either by increasing the viscosity of the fluid or by decreasing the volume and size of fluid-conducting pores. Moreover, bacterial exopolymers can cause high frictional resistance (Characklis, 1971).

The work of Raiders et al. (1986) seems to indicate that the viscosity effect is negligible. These authors induced clogging in sand columns by percolating them with a solution that was selective for a slime-producing strain and found that the K_s decline was localized, at least during the first stages, at the entrance of the columns. Had the K_s reduction been caused by an increase of the fluid viscosity, clogging would have been expected to occur throughout the column.

It hardly comes as a surprise that gel-like exopolymers would efficiently block the flow through a network of fine pores by reducing the effective porosity, that is, the volume of effectively conducting pores. An underlying assumption to that hypothesis, however, is that the water molecules associated with the exopolymer are immobile or at least do not participate in the macroscopic transport of the fluid (Taylor and Jaffé, 1990a). In other words, the wetted exopolymers must be impermeable. Several studies lend credence to this assumption. For example, Ehlinger et al. (1987) observed a twofold decrease of the liquid residence time in a packed reactor inoculated with a slime-producing consortium. Because the flow rate was kept constant, this result indicates that the pore volume available for flow had declined by half. Another piece of evidence is provided by Cullimore and Mansuy (1987). These authors found that after inoculation of a gravel pack with a microbial consortium rich in slime-producing iron bacteria and susained flow of ironcontaining nutrient medium, the extensive clogging that resulted was paralleled by a reduction of the volume of mobile water to nondetectable levels. In another column study, inoculation of sand with two slime-producing Pseudomonas strains resulted in a four-order magnitude decrease in hydraulic conductivity at the column inlet (Hoyle, 1994). The cause of the clogging was determined to be a layer of gelatinous slime filling the column inlet reservoir.

Slime-producing bacteria have often been detected in clogged media (Mitchell and Nevo, 1964; Shaw et al., 1985; Raiders et al., 1986). However, these organisms seem ubiquitous in natural systems (Costerton et al., 1981) and therefore the detection of slime producers in clogged media does not imply causality but only points out the possibility that they are involved in clogging processes. A number of arguments have been presented in favor of this possibility. After inoculation of various porous media with exopolymer-producing organisms, rapid K_s declines are usually observed (Jack et al., 1983; Shaw et al., 1985; Cullimore and Mansuy, 1987; Cunningham et al., 1991). In some of these experiments, microscopic analysis of clogged materials reveals the presence of large amounts of amorphous deposits in the pore space (Shaw et al., 1985; Ehlinger et al., 1987). When sand columns were supplied with a solution containing purified exopolymers, K_s declined (Mitchell and Nevo, 1964). The production of exopolymers can be controlled in situ by altering the composition of the nutrient medium (Avnimelech and Nevo, 1964; Tollner et al., 1983, Jack et al., 1983; Okubo and Matsumoto, 1983; Raiders et al., 1986; Ehlinger et al., 1987). When the production of exopolymers was stimulated in situ in that manner, accelerated clogging usually ensued. For example, Vandevivere and Baveye (1992a) observed a 20-fold K_s reduction in a sand column colonized by an Arthrobacter strain. When the C/N ratio of the percolating fluid was increased from 39 to 77, the bacteria started producing large amounts of exopolymers and K_s further decreased 2-fold (40-fold total).

In a subsequent study, the same authors demonstrated the dramatic impact that exopolymers can have on K_s (Vandevivere and Baveye, 1992b). They inoculated sand columns with two bacterial strains that produced exopolymers and others with mutants of these strains that had been selected for their inability to produce exopolymers. Among the two exopolymer-producing strains, one produced a capsule and the other loose slime layers. Whereas all four strains colonized the sand columns more or less equally successfully, only the slime producer affected significantly the K_s of the sand column, leading to more than a 100-fold decrease. The other three strains had only a negligible effect on K_s. These results were obtained for bacterial colonization occurring within the sand. Different results were obtained when growth also proceeded at the inlet boundary of the sand columns, that is, when inoculation was done in the feed line. In this case, all strains caused large K_s reductions through building up of a superfical mat, or crust, coating the inlet end of the sand columns. The exopolymer-producing strains had a lesser effect under these conditions than their respective non-exopolymer-producing mutant. It was suggested that the presence of exopolymers in such crusts makes the cell packing less dense, and thereby more permeable.

These data indicate that exopolymers indeed may serve as key clogging agents in porous media under some conditions. However, they do not indicate how relevant exopolymers are to clogging processes existing in field applications. This latter issue has mainly been addressed by carrying out chemical assays and by analyzing the correlation between polysaccharides and the degree of K_s reduction. In a number of cases, the K_s decrease was correlated to an increase of the polysaccharide content (Avnimelech and Nevo, 1964; Mitchell and Nevo, 1964; Chang et al., 1974; Tollner et al., 1983; Ragusa et al., 1994). Usually, a higher correlation was found between K_s and polyuronide content. At times, however, no significant relationship was found between K_s and the polysaccharide content (Thomas et al., 1966; Jawson, 1976; Hilton and Whitehall, 1979). An inverse relationship, that is, a K_s decrease associated with a decrease of the polysaccharide content, was found by Siegrist (1987). Complicating the analysis of these results is the fact that the polysaccharide content has in the past usually been assayed in unfractionated samples, therefore including the polysaccharides of the cell protoplasm or those in plant residues, when the latter are added to the soils.

3. ENTRAPMENT OF GASEOUS END-PRODUCTS

Numerous studies have shown the presence of a gaseous phase in submerged soil profiles either in the field (Ahmad, 1963; Chen et al., 1972; Wood and Bassett, 1975; Hills, 1976) or in laboratory columns (McCalla, 1950; Poulovassilis, 1972; Lance and Whisler, 1972; Bubela, 1980; Kristiansen, 1981a; Tollner et al., 1983; Ronen et al., 1989). As was discussed in a previous section ("Physical processes"), air bubbles can become entrapped in submerged soils as a result of purely physical mechanisms. Various indications of the biological origin of bubbles include the fact that organic matter additions enhance the process (Tollner et al., 1983; Ronen et al., 1989) or that they are rich in typical products of anaerobic metabolism such as N_2, H_2S, and CH_4 (Jawson, 1976; Tollner et al., 1983; Kristiansen, 1981a; Bubela, 1985). A powerful means to assess the biogenic origin of a gas (N_2) is to compare its isotope ratio with that of air. Biogenic gases typically show an enrichment in the lighter isotope (Vogel et al., 1981). This technique does not seem to have been used yet in studies on clogging.

The presence in a porous medium of gas bubbles, if sufficiently large as to become trapped in the network of pores, causes a reduction of K_s because the bubbles decrease the size of the water-conducting pores, in the same manner as solid particles would (Orlob and Radhakrishna, 1958). Orlob and Radhakrishna (1958) measured a reduction of K_s by 35% after increasing the air content by 10% in a uniform sand. When the sand contained both fine and large grains, the desaturation had a still greater effect on K_s. In fact, air bubbles need not to be larger than pore diameters to accumulate in the pores. They can adhere to surfaces, possibly at hydrophobic sites, as shown in Figure 5, or they can become entrapped in the matrix of exopolymers that often surrounds sessile bacteria (Battersby et al., 1985). The latter phenomenon can be important enough to cause the detachment of attached colonies (Harremoës et al., 1980).

Although microorganisms produce a wide range of poorly soluble gases, for example, CO_2, N_2, H_2, and CH_4, it is mostly N_2 production that has been studied in relation to K_s reduction. This is probably related to the importance of the N cycle and also to the fact that O_2 and H_2 have rarely been reported to accumulate in the soil atmosphere. Indeed, the O_2 produced in algal mats at the water-sediment interface (Tollner et al., 1983; Bubela, 1985) is probably rapidly reduced, and H_2 is readily used by various microorganisms such as sulfatereducing bacteria (Jakobsen et al., 1981), methanogenic bacteria (Bryant, 1979), denitrifying bacteria (Yoshida, 1975), or fermentative bacteria (Gottschalk, 1986). For these reasons, and also to reflect the availability of information, the production of gas by denitrifiers and methanogens are analyzed further.

A. DENITRIFICATION

Denitrification, or dissimilatory nitrate reduction with N_2 as a major product, involves the respiration of organic molecules with NO^-_3 as a terminal electron acceptor. This reaction can be carried out by a large and heterogeneous group of aerobic bacteria (Gottschalk, 1986, p. 122) and tends to maintain the soil redox potential at approximately 0.2 V (Rowell, 1981). Certain bacteria can carry out the reaction with alternate electron donors such as Fe^{2+} ((Mariotti, 1986), H_2, or S^{2-} (Yoshida, 1975). The widely reported stimulation of denitrification after organic matter additions (e.g., Bowman and Focht, 1974) does not rule out the potential significance of these parallel pathways because the availability of alternate electron donors can increase after organic matter additions. Excess carbohydrate can retard the denitrification process (Bowman and Focht, 1974). Dissimilatory nitrate reduction is not the only source of N_2. Nitrifying bacteria can, under certain conditions, evolve N_2 (Yoshida, 1975). The dissimilatory reduction of NO^-_3 is also carried out by certain fermentative bacteria that couple the oxidation of NADH with the reduction of NO^-_3 to NH^+_4 in a process referred to as fermentative nitrate reduction (Gottschalk, 1986). According to Stanier et al. (1987, p. 554), this reaction is highly competitive with denitrification, the majority of NO^-_3 added to most soils being reduced to NH^+_4 rather than to N_2.

Denitrification is often considered to occur only in the absence of O_2. The enzymes involved in denitrification are not formed under conditions of high O_2 tension and are strongly inhibited by O_2 (Gottschalk, 1986, p. 122). Therefore, the simultaneous occurrence of denitrification and aerobic respiration that is at times observed in soils or sediments (Oberdorfer and Peterson, 1985; Mariotti, 1986; Trudell et al, 1986) traditionally has been explained by the concept of anaerobic microsites (e.g., Brock and Madigan, 1991). More recent data (Lloyd et al., 1987) suggest that, even in a well-mixed aerobic system, denitrification persists, although NO^-_3 tends not to be reduced all the way to N_2 and N_2O accumulates instead. Such is also the case when the electron donor/NO^-_3 ratio is small (Yoshida, 1975). Thermodynamic considerations seem to provide a firm grounding for these observations because the oxidation of carbohydrates coupled with the reduction of NO^-_3 to N_2 theoretically yields, at pH 7, 95% of the free energy released by the coupling with the reduction of O_2 (Champ et al., 1979).

Evidence that microbial production of N_2 in the field causes K_s declines does not abound. Oberdorfer and Peterson (1985), in their field study of wastewater injection into an aquifer, observed a progressive supersaturation of N_2 simultaneously with a depletion of other N species and a reduction of K_s, which led them to suggest the presence of gas locks. Several investigators observed a relative accumulation of N_2 in the gas phase of clogged soil profiles, both in the field (Wood and Bassett, 1975) and in laboratory columns (Lance and Whisler, 1972; Kristiansen, 1981a; Tollner et al., 1983). These latter observations do not, however, demonstrate unequivocally the production of N_2 gas because a ponded soil profile whose surface layer has a reduced K_s as a result of clogging typically exhibits a zone of negative pressure (< P atm) at some depth (Daniel and Bouma, 1974). Therefore, a purely physical explanation can account for the occurrence of a gas phase, and microbial respiration can account for the enrichment in N_2. However, in another set of studies using experimental apparatuses that precluded the establishment of subatmospheric pressures, several authors have shown the potentially deleterious effect of denitrification on K_s when the percolating liquid contained large amounts of nitrates (Harremoës et al., 1980; Raiders et al., 1986; Ronen et al., 1989). Under these circumstances, complete recovery of K_s can be obtained by applying a vacuum or flushing with CO_2. Both procedures effectively eliminate the gas locks (Poulovassilis, 1972; Lance and Whisler, 1972; Ronen et al., 1989).

B. METHANOGENESIS

Methanogens, or methane-producing archaebacteria, carry out the final steps in the sequence of redox reactions taking place in anaerobic environments, typically becoming active when the sulfate reduction process has come to a halt and the redox potential has reached -0.2 V (Jakobsen et al., 1981). At times, however, the two reactions are simultaneous (Takai et al., 1956; Takai, 1970). Several reasons seem to limit the activity of methanogens to the tail end of the successive redox reactions. First of all, they rely on the end-products of various types of fermentations for their supply of growth substrates, such as acetate, formate, methanol, CO, CO_2, and H_2 (Zeikus, 1977; Blaut et al., 1985; Gottschalk, 1986, p. 252; Lobo and Zinder, 1988). Moreover, they are inhibited by O_2, NO_3^-, and SO_4^{2-} (Jakobsen et al., 1981).

Swartzendruber and Gupta (1964) tested the hypothesis that crystalline water surrounding dissolved methane molecules would increase the viscosity of the fluid to the point of reducing the K_s of a fine quartz sand. After dissolving CH_4 in water and running the solution aseptically through columns packed with the sand, K_s was not found to decline more than in control columns supplied with pure water. Swartzendruber and Gupta (1964) concluded that the production of CH_4 by microorganisms could not cause clogging. This statement appears, however, unwarranted in view of the fact that no methanogens were involved in the experiments and that the solutions percolated through the sand were undersaturated with respect to methane, making the formation of bubbles in the pore space very unlikely. Theoretically, in a system with a moving liquid phase, methane bubbles could form and decrease K_s only if the rate of production exceeds the removal rate (equal to the product of the flow rate with the methane concentration). For example, Takai et al. (1956) measured rates of methane production up to 8 ml d^{-1} g^{-1} of soil in submerged paddy soil maintained under static (no-flow) conditions. Assuming a CH_4 concentration of 3% (vol/vol) at saturation (Swartzendruber and Gupta, 1964), such a rate would lead to supersaturation of CH_4 and possible bubble formation when the infiltration rate into the soil becomes less than approximately 5 m d^{-1}. Therefore, it is not surprising that the gas phase frequently detected in ponded soil profiles often contains significant amounts of methane (Jawson, 1976; Kristiansen, 1981a; Tollner et al., 1983). Nevertheless, one needs to interpret such observations with caution because, as discussed previously, the occurrence of a gas phase may not be directly caused by the microbial production of gases.

There seems to be only two studies having demonstrated via direct measurement the adverse effect of methanogens on soil K_s. Reynolds et al. (1992) monitored the movement of de-gassed, temperature-equilibrated water through repacked laboratory columns of catotelm peat. In unsterilized columns, the saturated hydraulic conductivity decreased sharply over a period of 78 d, while concomittantly, the volumetric gas content and the gaseous methane concentration in the peat increased significantly. These observations suggest that in some cases there may be a strong correlation between in situ accumulation of anaerobe-generated methane and severe decreases in hydraulic conductivity. However, because Reynolds et al. (1992) did not investigate other possible mechanisms (e.g., biomass accumulation, extracellular polymer production) by which methanogens could clog the porous media, one cannot deduce from the existence of this correlation that there is a causal link between methane production and decreases in hydraulic conductivity.

In a recent research effort, Sanchez de Lozada et al. (1994) analyzed the extent to which a methanogen can clog sand columns. Two permeameters packed with clean quartz sand were sterilized, saturated with water, inoculated with Methanosarcina barkeri and percolated under upward flow conditions. After approximately 5 months, the K_s of the sand had decreased to 3% and 25% of the highest values measured earlier in the two permeameters. At that point, gas-filled regions in the sand were clearly visible through the transparent walls of the permeameters, and methane bubbles were continuously released from the columns into the effluent. Scanning electron microscopy observations and biomass assays indicated that cell mass accumulation did not contribue significantly to the observed decrease of K_s. This decrease was therefore attributed to pore blocking due to the entrapment of methane bubbles.

4. ACCUMULATION OF INSOLUBLE SULFIDE SALTS

The hypothesis that FeS precipitates, which are commonly found in anaerobic environments rich in organic matter, cause clogging was formulated in the 1940s in the petroleum industry (O'Bryan and Ling, 1949) and in the early 1960s in studies investigating reasons for the failure of septic tank effluent disposal systems (Winneberger et al., 1960). Since then, strong evidence has been obtained showing that FeS precipitates have a negligible effect on K_s at least under the conditions of the investigations. The formation of FeS precipitates and its effect on K_s will, however, be reviewed here in some detail because of the abundance of studies on the subject and because it is still possible that certain specific systems are subject to clogging by FeS colloids.

The formation of sulfide is generally microbially catalyzed (Jakobsen et al., 1981). Sulfide is the end-product of the dissimilatory sulfate reduction in which sulfate acts as the electron acceptor in an energy-yielding oxidation reaction (Gottschalk, 1986). Sulfate reduction has been claimed to be the most common and extensive microbiological process on Earth (Iverson and Olson, 1984). Sulfate-reducing bacteria are widely distributed in nature (Ogata and Bower, 1965) and occur in deep and shallow groundwater worldwide (Iverson and Olson, 1984). In accordance with thermodynamic predictions, enzymatic sulfate reduction was found to occur at redox potentials between -175 mV and -350 mV with an optimum around -300 mV (Jakobsen et al., 1981). In the sequence of redox reactions taking place in waterlogged environments, it corresponds to the third redox zone, as defined by Champ et al. (1979), thus taking place only after the reduction of O_2, NO^-_3, Mn^{4+}, and Fe^{3+}. There is a narrow pH optimum around 6.7. For a long time, sulfate-reducing bacteria were only known to oxidize a number of organic acids and alcohols incompletely, with acetate as an end-product (Gottschalk, 1986, p. 260):

$$2CH_3CHOHCOOH + SO^{2-}_4 \text{-->} 2CH_3COOH + 2CO_2 + S^{2-}$$

Only recently have isolates been obtained that complete the oxidation to CO_2 (Gottschalk, 1986). Most sulfate-reducing bacteria are able to reduce sulfate with H_2 and many can utilize CO_2 as a carbon source, thus growing autotrophically (Gottschalk, 1986). According to Gottschalk (1986), sulfate-reducing bacteria are strict anaerobes, but Iverson and Olson (1984) point out that most of them are O_2 tolerant.

Because, as indicated above, Fe^{3+} reduction precedes SO^{2-}_4 reduction in the sequence of redox reactions, the Fe^{2+} species is usually abundant where sulfides are being produced. Ferrous iron readily associates with sulfide to form FeS, which has a very low solubility product and, therefore, a strong tendency to precipitate as black colloids (Ogata and Bower, 1965). FeS colloids bind most strongly to humus and organic matter and do not seem to accumulate on mineral soil particles (Ford et al., 1968). As a result, FeS appears to be much more effective as a plugging agent when it can combine with organic residues, bacteria, or oil (Davis, 1967). The consistency of FeS precipitates in soil profiles supplied with organic-rich effluents has been characterized as a jellylike gelatinous mass (McGauhey and Krone, 1967; Ford et al., 1968; Wood and Bassett, 1975). Calcite ($CaCO_3$) is also a common precipitate associated with the formation of sulfides (Ogata and Bower, 1965).

In the early 1960s, a group of engineers at the University of California (Winneberger et al., 1960; McGauhey and Winneberger, 1964, 1965; McGauhey and Krone, 1967) claimed that FeS was a major factor in soil clogging during percolation with sewage or septic tank effluents. However, that conclusion was based entirely on the observation that, in the soil profile, a black layer had developed that was much thicker than the superficial biocrust. Many other studies have shown that a thick zone of FeS accumulation often develops in clogged soils submerged with wastewater (Mitchell and Nevo, 1964; Thomas et al., 1966; Ford and Beville, 1968; Ford et al., 1968; Wood and Bassett, 1975; Kristiansen, 1981a). The possible role of FeS as a plugging agent of water-production wells has been studied in the delta region in the Netherlands (van Beek and Kooper, 1980; van Beek and van der Kooij, 1982; van Beek, 1984). The progressive clogging of the aquifer sediments surrounding wells is usually accompanied by an increasing content of FeS, organic matter, and $CaCO_3$ in the produced water (van Beek, 1984). The injection of NaClO in the aquifer is sometimes used to regenerate water-production wells that become clogged (van Beek and Kooper, 1980). This procedure usually liberates SO^{2-}_4, presumably a product of the oxidation of sulfides with HClO. Moreover, Van Beek's (1984) results show that the water obtained from clogged wells contains significantly more sulfate-reducing bacteria than non-clogged wells. As pointed out by the author, these observations do not prove that the precipitation of FeS catalyzed by sulfate-reducing bacteria is the cause of the clogging, but may simply indicate that the presence of S^{2-} is an indicator of conditions that are conducive to clogging.

In fact, there is strong evidence that FeS does not significantly affect K_s. During the artificial recharge of groundwater, Oberdorfer and Peterson (1985) noticed a progressive reduction of K_s in the aquifer sediments at a distance of approximately 0.5 m from the injection well. Chemical analysis indicated that FeS had accumulated in the sediments adjacent to the well but not in the region where K_s declined. In his studies of wastewater percolation through sand columns, Kristiansen (1981a) also found that the region of FeS accumulation did not correspond to the region of K_s decline. Jawson (1976) observed a steady increase of the FeS content during 14 months of wastewater application to soil columns, but K_s decreased only during the first half of that period, remaining constant afterwards. By inducing nonenzymatic precipitation of colloidal FeS in sand columns, no change of K_s could be obtained by Mitchell and Nevo (1964). However, this may not simulate adequately the biological mechanism because, as indicated above, FeS appears to be a more effective plugging agent when it can combine with microbial cells and their waste products. Mitchell and Nevo (1964) also induced FeS precipitation by adding sulfur together with casein in the percolation solution. The clogging was most severe in the sand columns treated with casein alone, in which FeS precipitation was not observed. Laak (1970) established that an FeS content of 0.4% was necessary to cause clogging of sand columns. Because the clogging material analyzed in their experiments always contained less than 0.4% of FeS, they discounted the role of FeS as a plugging agent.

5. EFFECT OF IRON BACTERIA

The deposition of ferric oxihydroxides in water supply systems is a worldwide problem (Davis, 1967; Ford, 1979a; Kuntze, 1982; van Beek, 1984; Driscoll, 1986; Applin and Zhao, 1989). This process can also cause failure of drainage systems in agricultural fields (Ford and Beville, 1968; Ford et al., 1968; Kuntze, 1982; Houot and Berthelin, 1992) and of enhanced oil recovery schemes (Davis, 1967). Although the relative importance of the enzymatic vs. spontaneous process can vary, there is no doubt that ferric oxihydroxides deposition causes major clogging in a variety of situations. Particular conditions, which are reasonably well understood and are described below, are nonetheless required for ferric oxihydroxide deposition to occur.

Soils contain large amounts of iron (Kuntze, 1982). Aside from typical iron-containing minerals such as ferrihydrite, lepidocrocite, goethite, or hematite, clay minerals can contain up to 30% Fe (Kuntze, 1982). A very large number of soil microorganisms can reduce Fe^{3+} to Fe^{2+} (Parr and Papendick, 1971; Fischer, 1986). According to Fischer (1986), microbial reduction of Fe^{3+} may include all Fe^{3+} oxides of a given soil compartment, although reduction of amorphous forms, such as lepidocrocite, are thermodynamically more favorable than crystalline forms such as goethite. In the sequence of redox reactions taking place in a submerged soil containing organic matter, Fe^{3+} reduction takes place slightly after the Mn^{4+} reduction and before the SO^{2-}_4 reduction (Jakobsen et al., 1981). The dissolution of Fe^{3+} is accelerated by the production of iron-chelating molecules (siderophores) by soil microorganisms that increase the solubility of Fe^{3+} by several orders of magnitude (Fischer, 1986). Because these chelators bind Fe^{2+} as well as Fe^{3+}, they render the Fe^{3+} reduction thermodynamically more favorable by lowering the activity of Fe^{2+} ions (Fischer, 1986). The stability of Fe^{2+} depends on the pH, Eh, Fe^{2+} activity, and on temperature (Fischer, 1986). Thus, an increase of the pH, the Eh, or the Fe^{2+} activity will cause a rapid spontaneous precipitation of Fe^{3+}.

In most cases, microbial oxidation of Fe^{2+} is associated with the activity of acidophilic iron-oxidizing bacteria such as Thiobacillus ferrooxidans (Kuntze, 1982). A number of iron bacteria catalyze the oxidation of Fe^{2+}, but only the acidophilic ones have been shown to unequivocally derive energy from the process (Fischer, 1986). Neutrophilic iron bacteria typically produce exopolymers that actively bind Fe^{3+} (Fischer, 1986). These bacteria require O_2 as an electron acceptor and therefore their activity relies on the introduction of O_2 to the system (Kuntze, 1982). Ralph and Stevenson (1995) observed that faster oxidation of Fe^{2+} occurred in the presence of an inoculum consisting of well-bore sludge than in a sterile system. Bacteria can oxidize Fe^{2+} either directly (enzymatically) or indirectly (non-enzymatically). Non-enzymatic oxidation includes processes such as rises in pH or Eh and breakdown of Fe^{3+}-chelators such as oxalate, citrate, humic acids, or tannins (Driscoll, 1986).

There are two major ways by which O_2 is introduced in an anaerobic solution flowing through soils or sediments. The first is the desaturation, or aeration, of the system in response to artificial drainage. The second involves the mixing of waters with different chemical compositions as is the case in groundwater pumping operations.

Pipe drainage is the most widespread method of soil reclamation (Kuntze, 1982). A commonly encountered hazard in those operations is the clogging of drains and surrounding filter material caused by iron sludge deposition (Kuntze, 1982). The iron sludge usually contains high amounts of organic matter, 20% to 50%, which suggests that microorganisms participate in the precipitation (Kuntze, 1982). Indeed, iron bacteria, such as Gallionella, Leptothrix, and Thiothrix, have been found repeatedly and abundantly in clogged drainage systems (Kuntze, 1982; Houot and Berthelin, 1992). According to Ford (1982), the iron sludge is a red to tan gelatinous deposit containing extensive amounts of iron in association with bacterial slimes. These slimes probably play an important role in the clogging process because nonenzymatically oxidized Fe^{2+} is a rather poor clogging agent unless sorbed onto bacteria (Ford, 1979b). Because these bacteria are either aerophilic or microaerophilic, preventing air from entering the drainage pipe, for example, by maintaining it submerged below the water table, is a simple means of preventing iron clogging (Kuntze, 1982). Another simple measure that alleviates the clogging is the use of a backfill material that contains no organic matter, to prevent the development of the anaerobic environment that promotes iron reduction (Ford et al., 1968).

Shallow aerobic and deep anaerobic groundwaters often mix in and around extraction wells that draw water from unconfined aquifers (van Beek, 1984). Under these conditions, iron bacteria multiply profusely because the flow velocity and, therefore, the supply of nutrients is greatly increased by pumping. These bacteria can thrive at very low concentrations of O_2 and Fe^{2+}, such as 0.2 mg 1^{-1} (Davis, 1967; Ford, 1982; van Beek, 1984). Moreover, their excellent adhesive properties enable them to colonize surfaces exposed to very high flow velocities (van Beek, 1984). In the absence of preventive measures, severe well clogging inevitably follows (van Beek, 1984; Driscoll, 1986). A common preventive measure consists of the injection of aerated water to precipitate Fe^{3+} in the aquifer instead of around the well (Driscoll, 1986).

F. PORE COLONIZATION PATTERN AND SEVERITY OF BIOLOGICAL CLOGGING

The rate and extent of the clogging of porous media by bacteria has been shown to be closely related to their ability to colonize the pore space. For example, MacLeod et al. (1988) show that ultramino bacteria, that is, dwarf cells resulting from prolonged starvation, move through porous media to a much larger extent than their non-starved (normal-sized) counterparts and, as a result, have a much lesser tendency to plug the porous medium. Correlation between bacterial transport and clogging was also evinced in the experiments of Vandevivere and Baveye (1992d). A bacterial strain that did not move easily through a sand column, presumably because of its tendency to coalesce into large flocs, induced greater clogging than another strain that coated sand grains with a monolayer of individual cells. Other strategies employed by bacteria to withstand transport by convection may include the formation of strings of cells. In some cases, the ability to withstand transport, or the development of a specific pore colonization pattern, may result from environmental constraints. For example, DeLeo and Baveye (1997) observe that a Bacillus strain, which occurs as individual cells when grown in pure culture, has an increased tendency to form aggregates when subjected to high grazing pressure by an Acanthamoeba.

It is possible that many bacterial strains, used in pure-culture studies, may be unable to clog porous media to any significant extent because of their poor ability to colonize pore spaces (Vandevivere and Baveye, 1992b). However, even poor pore colonizers induce severe clogging when the cells colonize the inlet boundary of sandstone cores (Lappin-Scott et al., 1988) or sand columns (Vandevivere and Baveye, 1992a,b). In these cases, all strains tested rapidly built up a blanket-like crust or “skin” plug (Lappin-Scott et al., 1988), with a much reduced permeability.

G. CLOGGING PREVENTION AND SPONTANEOUS UNCLOGGING

In principle, any of the biocides mentioned earlier (in Section V.A) could be used to prevent clogging or at least to maintain bacterial numbers below the level at which they begin to have significant effects on the saturated hydraulic conductivity of the host porous medium. The same objective may be reached by controlling the concentration of the growth-limiting substrate injected in the subsurface (e.g., an electron acceptor in bioremediation strategies). Another approach, which is meant to prevent microbial clogging in the immediate vicinity of injection wells, when more than one substrate is necessary to support microbial growth, is to rely on “staggered” injection (Figure 6). The primary substrate, needed for the biodegradation of the organic contaminant (secondary substrate) via cometabolism, and an electron acceptor are injected as separate pulses. This method, which has been used in the field by Roberts et al. (1990) (see also Cookson, 1995), should prevent growth-promoting conditions from developing close to the injection wells, yet should allow growth to occur at some distance from the well. At that point, clogging may still result and may have deleterious effects on aquifer bioremediation, in spite of nutrient pulsing. This point has not been addressed by Roberts et al. (1990).

Once a soil or sediment is clogged by microorganisms, it may be of interest to restaure the medium’s original hydraulic conductivity. Various treatments have been shown to be partially and temporarily effective at removing organic plugs resulting from the activity of bacteria. These include chemical treatments with H_2O_2 (Jawson, 1976; Mickelson et al., 1989), Cl_2 (Johnson, 1958), various acids (Driscoll, 1986), or sodium hypochlorite (Shaw et al., 1985; cf. Figure 7), and mechanical treatments such as pressurization (Shaw et al., 1985) or reverse flow (Gupta and Swartzendruber, 1964; Raiders et al., 1986).

At times, however, partial and temporary removal of the organic plug occurs spontaneously without any change in the flow rate or quality of the applied solution. These sloughing events have been detected by visual observation (Honda and Matsumoto, 1983; Cullimore and Mansuy, 1987; Harremoës et al., 1980), by abrupt change of the nutrient utilization pattern (Okubo and Matsumoto, 1979 and 1983), or by sudden K_s increases (Mitchell and Nevo, 1964; Kropf et al., 1975; Okubo and Matsumoto, 1979 and 1983; Pell and Ljunggren, 1984; Cullimore and Mansuy, 1987).

Sloughing has been attributed to the increased shear stress brought about by the reduced cross-sectional area available for flow (Taylor and Jaffé, 1990a; Characklis et al., 1987), to the disruptive forces of growing N_2 bubbles entrapped in actively-denitrifying biofilms (Harremoës et al., 1980), to the accumulation of toxic metabolites (Kropf et al., 1975), to anaerobic decomposition or grazing that weaken the forces of adhesion of sessile colonies (Honda and Matsumoto, 1983; Cullimore and Mansuy, 1987), or to shifts of microbial flora (Okubo and Matsumoto, 1983; Pell and Ljunggren, 1984). The latter two explanations, which may in fact describe the same process, were best characterized by Okubo and Matsumoto (1983). In their studies of synthetic wastewater injection through sand columns, they observed a partial recovery of K_s that was simultaneous to a peak of biomass in the column effluent and the establishment of anaerobic conditions. Their interpretation was that obligate aerobes had died off as a result of O_2 depletion and were removed with the column effluent as a result of anaerobic decomposition. Some of the data presented by Tollner et al. (1983) lend credence to that explanation. These authors noticed a partial recovery of K_s after addition of organic waste to ponded soil columns, an addition that was expected to induce a shift of the microbial community structure.

A peculiar aspect about the concept of sloughing, probably owing to the poorly understood nature of this complex phenomenon, is that it has been used to account both for K_s increases and K_s decreases. The former probably are intuitively easier to grasp, but the latter have been advocated by a number of researchers. For example, Characklis et al. (1987) interpreted a phase of accelerated clogging by a mechanism of biofilm detachment under the action of shear stress and subsequent filtration of drifting clumps at strategic points such as pore constrictions. Taylor and Jaffé (1990a) used the same interpretation to account for biomass accumulation and K_s reduction in deeper regions of the columns that lacked essential growth nutrients. In the unsaturated conditions prevailing in trickling filters used for water purification, sloughing has been recognized as a major limiting factor for the system performance (Howell and Atkinson, 1976).

H. INTERACTIONS BETWEEN BIOTIC AND ABIOTIC EFFECTS

In his experiments involving septic tank effluent application onto soil, Siegrist (1987) obtained the best fit ($R^2 = 0.95$) for the decreasing infiltration rate by expressing it as a function of both the BOD and the suspended solids, the latter comprising organic and inorganic material. This suggests that both the microbial activity taking place at the expense of the BOD and the physical filtration of suspended particles played a role in the clogging process. Other investigations showed that physical clogging, attributed to the trapping of suspended and colloidal material in the pores, and biological clogging, attributed to the accumulation of cells and waste products in the pores, can occur sequentially or simultaneously at different depths in a soil or sand profile (e.g., Ripley and Saleem, 1973; Chang et al., 1974; Ragusa et al., 1994).

These two processes are probably not simply additive but rather interactive. As suggested by McGauhey and Winneberger (1965) in their extensive study on land disposal of septic tank effluents, an initial reduction of K_s is induced by the physical filtration of suspended solids. Part of these solids being organic, this initial phase is followed by a rapid proliferation of bacteria that causes further clogging. As the porosity of the surface cake decreases, finer and finer particulates are trapped that, of course, decreases K_s further. This interactive effect is illustrated by Shaw et al. (1985), who injected suspensions containing bacteria, inert solid particles, or both, in columns of sintered glass beads. The largest reduction of K_s occurred when the suspension contained both bacteria and inert solids. Bacterial exopolymers seem particularly effective in trapping the suspended solids (Pell and Ljunggren, 1984; Shaw et al., 1985).

VI. MODELING BIOLOGICAL CLOGGING

A complete mathematical model of biological clogging should be able to predict the temporal and spatial variation of K_s on the basis of input data relative to the physical and chemical characteristics of the porous matrix (porous medium) and of the liquid phase, the flow characteristics (pressure gradient or flow rate), and to specific features of the microbial population. Conceptually, it is convenient to separate such a model into two main components. The first one would include a set of equations describing the accumulation of the microorganisms and of their metabolic end-products that interfere with the flow. The second part would predict the reduction of K_s as a function of the accumulation of microorganisms and end-products. Adopting this division of labor, we shall first describe a number of approaches that have been adopted in the past to model the accumulation of biomass. Subsequently, an outline and a critical evaluation will be provided of the various computational attempts that have been made to relate clogging to biomass density increases. As convenient as this division may seem, one should, however, always keep in mind that most processes modeled in the first component are affected by the output of the second component, principally through a modification of the pore water velocity, and therefore that both components are in fact intimately coupled (Characklis et al., 1987).

A. ACCUMULATION OF BIOMASS

One possible approach is to establish an empirical relationship between the accumulation of biomass and the supply of liquid. For example, Okubo and Matsumoto (1979) assumed a linear relationship between the volume of the biomass accumulated inside the column and the cumulated volume of synthetic wastewater added to sand columns. This model can be somewhat refined by including a saturation coefficient, as follows:

$$dX_{att.} / dt = kC(1 - X_{att.} / X_{att.max}) \quad (3)$$

where $X_{att.}$ is the amount of attached biomass per unit total volume (ML^{-3}), $X_{att.max}$ is the maximum value for $X_{att.}$, and k is a fitted parameter. The variable C can either represent the rate of supply of a nutrient, in which case Eq. 3 may be used to model microbial growth, or, as in Characklis et al. (1987), it can represent the concentration of unattached cells, in which case Eq. 3 models the process of cell attachment to solid surfaces. In writing Eq. 3, no attempt is made to describe separately and in detail the various processes controlling the fate of the attached biomass at the pore scale. These processes are, instead, lumped together. In consequence, the parameter k in Eq. 3 can only be evaluated by fitting and thus lacks physical or biological significance.

Mechanistic models, on the other hand, should attempt to involve explicitly all the individual processes that significantly affect the amount of attached biomass. According to Characklis et al. (1987), these processes should at least include sorption, desorption, detachment, attachment, and growth. Detachment refers to sloughing of biomass flocs by shear forces whilst attachment refers to the subsequent filtration of these flocs at pore constrictions. In support of the Characklis et al. (1987) viewpoint, the experimental data of Taylor and Jaffé (1990a) indicate that both growth and biomass transport affect the rate of bacterial clogging. Therefore, mechanistic models of clogging need to account for the transport of unattached bacteria as well as for the growth of both attached and unattached bacteria. To model bacterial growth, the transport of the limiting nutrient(s) also ought to be modeled.

At present, various models describe bacterial transport without incorporating a growth term (Yates and Yates, 1991; Harvey and Garabedian, 1991; Tan et al., 1994). Other models describe bacterial growth and decay, generally in conjunction with biodegradable solute transport, but without considering biomass transport (e.g., Molz et al., 1986; Baveye and Valocchi, 1989, 1991; Wood et al., 1994). Combining mathematically these two types of models is not very difficult. However, the key problem resides in the independent estimation of the many parameters included in a "complete" model describing both growth and transport.

B. REDUCTION OF THE SATURATED HYDRAULIC CONDUCTIVITY

Different empirical relationships have been obtained that relate K_s to the accumulation of biomass (Taylor and Jaffé, 1990a; Vandevivere and Baveye, 1992a) or to various indexes of biological activity (Frankenberger et al., 1979; Frankenberger and Troeh, 1982). Empirical relationships were also produced by Cunningham et al. (1991), although only in the form of diagrams. Siegrist (1987) correlated the infiltration rate directly to the BOD and TSS (total suspended solids) of the applied liquid. Such empirical relationships, however, are only applicable to systems that in many ways are similar to those for which the empirical relationships were obtained.

To obtain mathematical expressions of more general applicability, several investigators have attempted to develop mechanistic models of biological clogging processes. A rigorous analysis requires solving the Navier-Stokes equation for appropriate boundary conditions. However, the structure of the Navier-Stokes equation and the boundary conditions make the analytical solution virtually intractable (Scheidegger, 1974). Because an exact solution can be obtained for the flow of liquids through straight circular tubes (the Hagen-Poiseuille equation), most investigators have made the simplifying assumption that the pore space in porous media could be represented by bundles of parallel capillary tubes of uniform radius in which the biomass forms an impermeable biofilm of uniform thickness covering the walls of the tubes. Using this approach, Okubo and Matsumoto (1979) derived a model that showed good agreement with their experimental data. However, the first part of their model, describing the accumulation of the biomass, was empirical and included parameters that could not be determined independently.

An alternative to the Hagen-Poiseuille equation is the Kozeny-Carman equation, which describes a porous medium as if it were a conduit with a complicated shape but a constant cross-sectional area. The tube radius is replaced by the concept of hydraulic radius, defined as the ratio of the volume of liquid to the wetted surface area. The Kozeny-Carman equation yields the following expression for the saturated hydraulic conductivity, K_s (LT^{-1}), of a porous medium (Scheidegger, 1974; Vandevivere et al., 1995):

$$K_s = \rho g / c_0 \eta \; f^3 / S^2 \quad (4)$$

where ρ denotes the fluid specific mass density (ML^{-3}), g is the acceleration of gravity (LT^{-2}), f is the porosity of the (unclogged) porous medium (L^3L^{-3}), c_0 is a constant, η is the fluid viscosity ($ML^{-1}T^{-1}$), and S is the internal specific surface area per unit volume of the porous medium (L^2L^{-3}). In a porous medium that, for some reason, becomes clogged, the K_s ratio, according to the Kozeny-Carman equation, is given by

$$K_s / K_{s0} = f^3 / f^3_o \; S^2_o / S^2 \quad (5)$$

where the subscript "o" denotes an initial value, associated with the unclogged porous medium.

This approach was followed by Ives and Pienvichitr (1965) to predict the clogging of sand during the deep filtration of micrometer-size inert beads (Ives and Pienvichitr, 1965). To calculate the change of porosity and surface area caused by the entrapment of the beads, Ives and Pienvichitr (1965) made the assumption that the beads coated the walls of cylindrical capillaries with a film of uniform thickness. Their resulting equation can be written as follows (Vandevivere et al., 1995):

$$K_s / K_{s0} = (1 - \alpha)^{3-2p} \quad (6)$$

In this equation, p is an adjustable parameter reflecting the tortuosity of the actual pore space and the "biovolume ratio" (L^3L^{-3}), the bulk volume of colloidal particles per unit pore volume of "clean", unclogged porous medium (cf. Figure 3), is defined as $\alpha \equiv (f_0 - f)/f_0 \equiv \beta\sigma/f_0$, with β denoting the ratio of the total volume of aggregates of cells to the solid volume of the cells (L^3L^{-3}). In this definition, f denotes as before the water-filled porosity of the porous medium and the subscript "o" denotes an initial value, associated with the unclogged porous medium.

The model of Ives and Pienvichitr (1965) is more versatile than that of Okubo and Matsumoto (1979). Indeed, it is easy to show that the latter is a special case of Eq. 6, obtained by setting p equal to 0.5 (Vandevivere et al., 1995). Nevertheless, despite its greater versatility, Eq. 6 has an intrinsic limitation in the extent of the clogging that it can predict (Vandevivere et al., 1995). Indeed, because physically the parameter p cannot be smaller than zero, the value $(1 - a)^3$ represents, at any given a, the lowest value of the K_s ratio that can be predicted by the model of Ives and Pienvichitr (1965).

Taylor et al. (1990) derive a number of models on the basis of the Kozeny-Carman equation. For "clean" porous media, values for the initial porosity f_0 and internal surface S_0 are obtained by assuming that the solid phase can be represented by regular packings of uniform spheres. These geometrical arrangements are termed cubic (f_0 = 47.64%), orthorombic (f_0 = 39.54%), tetragonalspheroidal (f_0 = 30.19%), and rhomboedral (f_0 = 25.96%). Taylor et al. (1990) further assume that, in clogged systems, a biofilm develops in such a way that all the spheres constituting the porous medium are coated with an impermeable film of biomass of constant thickness L_f. Thus, biofilm growth effectively increases the volume, and decreases the surface area, of the solid phase in a given volume of the porous medium. A comparison of the relationship between K_s/K_{so} and the porosity ratio f/f_0 ($\equiv 1 - \alpha$) obtained in these 4 cases (cf. Figure 3 in Taylor et al. [1990]) reveals that this relationship is largely insensitive to the assumed packing arrangement.

A second type of model derived by Taylor et al. (1990) is based on the "cut-and-random-rejoin" approach introduced by Childs and Collis-George (1950) and later also adopted by Marshall (1958), Millington and Quirk (1959), and Mualem (1976). This approach assumes that a porous medium contains pores of various radii that are randomly distributed in space and that, when adjacent planes of the medium are brought into contact, the overall hydraulic conductance across the plane depends statistically upon the number of pairs of interconnected pores and geometrically after their configuration (Taylor et al., 1990). The additional assumption that biofilms uniformly coat the internal surface of the pores makes it possible to describe mathematically the relationship between biomass accumulation and K_s reduction. Two of the models developed by Taylor et al. (1990) using this approach, one based on the Kozeny-Carman equation and another based on the permeability model of Millington and Quirk (1959), lead to unphysical predictions in systems with a wide distribution of pore sizes. Indeed, under such conditions, these two models predict that clogging of the pores results in an increase of the medium's saturated hydraulic conductivity. The two remaining models, based on the permeability reduction models of Childs and Collis-George (1950) and Mualem (1976), lead to results that are physically plausible and very similar in the case of porous media with a narrow range of pore sizes. The Mualem-based model predicts a slightly more severe reduction of K_s, at a given value of α, than does the Childs and Collis-George permeability reduction model. Unlike the models of Ives and Pienvichitr (1965) or the models based on the Kozeny-Carman equation, the Mualem-based model has the advantage of containing an adjustable parameter λ, which reflects the homogeneity of the pore size distribution of the porous medium.

When the models briefly described above are tested against the experimental data of Cunningham et al. (1991), it becomes apparent (cf. Figure 8) that all three mathematical models slightly overestimate the extent of the clogging in the coarse-textured glass bead system. At the same time, they vastly underestimate the clogging in all other cases for which data are available (compare Figure 3 and Figure 8). By varying the value of 1 in the Mualem model, it is possible to bring its predictions into better agreement with the experimental data. For example, setting 1 = 0.8 provides a very good fit to the data for the 1-mm glass beads. However, to obtain a similarly good concordance for the sands for which clogging data were presented in Figure 3, one would have to use uncharacteristically high values for 1.

Two main reasons can be invoked to account for these observations. The first is related to the description of the geometry of the pore space. In different ways, all three models idealize this geometry, as explained above. It is possible that the assumptions made by each model are somehow less appropriate in fine-textured materials than they are in coarse-textured ones. One has to bear in mind, however, that the range of mean particle diameters covered in the experiments of Cunningham et al. (1991) and Vandevivere and Baveye (1992d) spans barely more than one order of magnitude and that there is no complicating factor like particle aggregation. Under these conditions, one would expect the assumptions embodied in the models to be equally valid (or invalid) for all the textures used in the experiments.

The second possible reason for the disparities between observations and model predictions in Figure 8 is related to the assumption, made in all three models, that the microorganisms responsible for the clogging form biofilms of constant thickness that uniformly coat the grain surfaces. This assumption of continuous biofilm coverage may be appropriate when one deals with smooth, macroscopic surfaces in contact with a liquid phase (Costerton et al., 1987), with cylindrical conduits (Block et al., 1993), or with relatively coarse-textured porous media at specific flow rates, and it may be possible in these cases to visualize the biofilms or to estimate their thickness (Bakke and Olsson, 1986). However, even under those conditions, there has been a definite trend in the literature in recent years to associate the term “biofilm” with structures that are far from uniformly coating surfaces. They consist instead of heterogeneous assemblages of cell clusters, pores, and conduits, all of which are characterized by a complicated geometry (e.g., de Beer et al., 1994). In clogged soils and aquifer materials, as we mentioned earlier (Section V.D), experimental evidence obtained (e.g., by Shaw et al. [1985] and Vandevivere and Baveye [1992a,b,c,d]) suggests that a continuous biofilm may not always be an accurate representation of the way microorganisms are distributed (see also Baveye and Valocchi [1989] and Baveye et al. [1992]). These observations indicate that mathematical models assuming the microorganisms to be present only in biofilms uniformly coating the solid surfaces oversimplify the pore-scale geometrical configuration of clogged soils or aquifer materials, and in that sense are not sufficiently mechanistic. A possible alternative is to assume that instead of (or in addition to) forming biofilms, microorganisms form plugs in the interstitial space of porous media.

To check if this assumption could in principle account for the clogging patterns observed in Figure 8, Vandevivere et al. (1995) propose a simplistic mathematical model, in which cylindrical “pores” are obstructed by plugs occupying a fraction α of the volume of each pore. To evaluate the saturated hydraulic conductivity of the plugged portion of the porous medium, it is assumed that the K_s value of the plugs, denoted by K_{splug}, is so low compared with K_{so} that the head loss occurring in the unplugged portion of each capillary is negligible. Under this condition and with the added assumption that the flow of water through the plugs can be described by Darcy’s equation, the K_s of the plugged porous medium is given by:

$$K_s\ (\alpha) = f_0 K_{splug}\ /\ \alpha \quad (7)$$

In the case of the 0.09 mm sand (with $f_0 = 0.38$), the most severe clogging is observed at $\alpha = 0.178$, where the ratio K_s/K_{so} is equal to 0.0023. This observation would be compatible with equation (7), provided K_{splug}/K_s is set equal to 0.0018. Experimental data relative to K_{splug} in the experiments of Vandevivere and Baveye (1992d) unfortunately are unavailable. Nevertheless, this estimate of K_{splug}/K_s appears reasonably plausible. Indeed, the Arthrobacter sp. strain used by Vandevivere and Baveye (1992d) has cells that are clay-sized (<2 mm) to very-fine-silt-sized (2 to 5 µm). Therefore, one would expect that the dense bacterial aggregates observed on the micrograph presented by Vandevivere and Baveye (1992d, Figure 10(B)) would have saturated hydraulic conductivities similar to those of clay or fine-silt media, that is, of the order of 100 to 1000 times lower than the hydraulic conductivity of sand (Marshall and Holmes, 1988). The value of the saturated hydraulic conductivity of these aggregates may, in specific cases, depend on a number of parameters. In a different, yet perhaps related system, measurements and calculations for activated sludge flocs indicate that age, particle size, shape, and fluid velocity all can affect floc permeability (Li and Ganczarczyk, 1992). In particular, as the size of the particles composing the flocs decreases to 1 to 2 μm, the percentage of permeable flocs decreases rapidly.

Whereas the cylindrical pore model described above presents a simplified method to estimate bulk plugged hydraulic conductivities, more sophisticated approaches are also being developed. For example, Imdakm and Sahimi (1987) proposed a Monte Carlo method to model particle transport by a random walk technique. Subsequently, Sahimi and Imdakm (1991) formulated a mechanistic model describing a network of pores in which particles are subject to physical and chemical forces during transport. These and other similar models (e.g., Loehle and Johnson, 1994) might be used to simulate the rate and extent of biological clogging in soils and sediments.

The results of Figure 8 and the discussion in the preceding paragraphs have particular significance with regard to the practice of using computer simulations to assess the usefulness of in situ bioreclamation as a strategy to clean up polluted aquifers. In at least one case (Taylor and Jaffé, 1991), some of the models mentioned above were used to obtain an estimate of the level of clogging that could be expected in a given field situation. Not surprisingly, it was concluded that alleviating clogging problems should be relatively easy in practice, especially when an in situ bioremediation strategy involves the injection only of an electron acceptor. Taylor and Jaffé (1991) predict that under these conditions, clogging in the biologically active zone may be largely prevented by modifying the electron acceptor concentration in the injection water, by increasing the well pumping rate or by introducing the electron acceptor in aquifer materials through multiple injection wells. However, in view of the poor performance of the biofilm-based clogging models in Figure 8, it is likely that the conclusions reached in simulation efforts using these models would be relatively unreliable, except perhaps in coarse-textured aquifers.

VII. CONCLUSIONS AND PROSPECTS FOR FUTURE RESEARCH

The clogging of porous media is intrinsically a physical phenomenon, resulting from a decrease in the capacity of the system to conduct water. For convenience, however, the processes responsible for this clogging may be organized in three classes: physical, chemical, and biological (microbial). Under each one of these categories fall a large number of processes. For example, the microbial clogging may be brought about by the accumulation of cells in the pore space, by the production of extracellular polymers, by the release in the pore space of gaseous byproducts, by the microbially mediated accumulation of insoluble precipitates.

Information on these individual processes has been accumulating steadily in the last few decades, making feasible their identification and, to some extent, their control under laboratory and field conditions. Various complicating factors, still largely unstudied to this date, should, however, make one wary of extrapolating too quickly to field situations some of the observations made under controlled laboratory conditions. Competition between microorganisms may lead to the selection of an organism or of a group of organisms with an extreme behavior, leading to much more or much less clogging than one might have expected on the basis of experiments involving pure cultures only. Similarly, protozoan predation or the propagation of bacteriophages may have a significant effect on the rate and the extent of clogging in natural porous media. With advances in our knowledge in these areas may come an increased ability to take advantage of microbial clogging in situations in which it has already proven beneficial, and to control it whenever it has deleterious consequences.

ADDED MATERIAL

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TABLE 1 Standard Free Energies, ΔG^0, Associated with Microbially Mediated Redox Reactions Involved in the Degradation of Organic Matter, Represented by the Model Compound CH_2O (data from Champ et al., 1979; Christiansen et al., 1994)

Reaction	Equation	ΔG^0 (w)(FNa) KJ
Aerobic respiration	$CH_2O + O_2$ --> $CO_2 + H_2O$	-502.3
Denitrification	$CH_2O + (4/5)NO^{-}3 + (4/5)H^+$ --> $CO_2 + (2/5)N_2$ $(7/5)H_2O$	-476.8
Mn(IV) reduction	$CH_2O + 2MnO_2 + 4H^+$ --> $2Mn^{2+} + 3H_2O + CO_2$	-340.3
Fe(III) reduction	$CH_2O + 8H^+ + 4Fe(OH)_3$ --> $4Fe^{2+} + 11H_2O + CO_2$	-115.0
Sulfate reduction	$CH_2O + (1/2)SO^{2-}4 + (1/2)H^+$ --> $(1/2)HS^- + H_2O + CO_2$	-104.7
Methane fermentation	CH_2O --> $(1/2)CH_4 + (1/2)CO_2$	-92.9

FOOTNOTE

a ΔG^0 (w) = ΔG^0 RT ln $[H^+]^p$ where $[H^+] = 1.0 \times 10^{-7}$ mol L^{-1} and p is the stoichiometric coefficient for H^+.

Note: The values of ΔG^0 are corrected for ph = 7.

FIGURE 1. Clogging patterns observed by (a) Allison (1947) and (b) Okubo and Matsumoto (1979). The vertical lines and the number correspond to an arbitrary division of the clogging pattern into a sequence of phases with distinct behaviors.

FIGURE 1B

FIGURE 2. Typical distribution of terminal electron-accepting processes in (top_ deep pristine aquifers (adapted from Lovley, 1991) and (bottom) shallow aquifers contaminated with organic compounds (adapted from Lovley et al., 1994).

FIGURE 4. Relationship between saturated hydraulic conductivity ratio and number of bacteria per g of sand in a sand column. (Data from Gupta and Swartzendruber, 1962.)

FIGURE 5. Schematic representation of a porous medium with biochemically produced gas bubbles and entrapped air bubbles. Some small bubbles clog pore conduits without significantly reducing the volumetric water content of the porous medium. (From Ronen et al., 1989. Reprinted by permission of Kluwer Academic Publishers.)

FIGURE 6. Schematic illustration of a clogging prevention method by pulse injection of electron acceptor and primary substrate. (After Cookson, 1995).

FIGURE 7. Illustration of the ability of sodium hypochlorite to partially counter the influence of microbial activity on the saturated hydraulic conductivity of a model glass bead core. (Data from Shaw et al., 1985.)

FIGURE 8. Predictions of the mathematical models of Ives, Mualem and Kozeny-Carman compared with experimental data of Cunningham et al. (1991). The symbols are identical to those in Figure 3. The predictions of the Mualem model are based on a value of 3.98 for the adjustable parameter λ.

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