

Aggregate Hierarchy in Soils

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Abstract

An Alfisol, a Mollisol and an Oxisol were fractionated into different particle sizes after a range of disaggregating treatments from gentle to vigorous.

The Alfisol and the Mollisol appeared to break down in steps; macroaggregates $>250\ \mu\text{m}$ diameter breaking down to microaggregates $20\text{--}250\ \mu\text{m}$ diameter before particles $<20\ \mu\text{m}$ were released. Vigorous disruption led to particle size distributions similar to those obtained by classical methods used to determine particle size distributions.

The Oxisol was stable to rapid wetting treatments but when aggregate disruption was initiated by vigorous treatments particles $<20\ \mu\text{m}$ diameter were released and there was no evidence of aggregate hierarchy.

Scanning electron microscopy of particles of different sizes showed distinctly single grain particles and aggregates. The microscopic studies indicated the potential role of roots and hyphae in the stabilization of larger aggregates, and for fragments of roots as nuclei for smaller aggregates. Plant debris was not visible in aggregates $<20\ \mu\text{m}$ but clay microstructure was evident.

It is suggested that aggregate hierarchy occurs in Alfisols and Mollisols because organic materials are the dominant stabilizing agents in larger aggregates but in the Oxisol oxides are dominant stabilizing agents and prevent the expression of aggregate hierarchy caused by organic materials.

Keywords: aggregate hierarchy, soil organic matter, scanning electron microscopy.

Introduction

All soils which contain colloidal materials exhibit aggregation where clusters of particles of different sizes remain associated when the soil is stressed mechanically by tillage, raindrop impact or drying and wetting. Aggregation is very important in tilled soils as it controls a range of soil properties which control crop establishment, water infiltration, aeration and drainage and creates a habitat for soil biota.

This paper focuses on the tilled layer of soils and examines the concept of aggregate hierarchy and its implications for soil management. Aggregate hierarchy infers a range of aggregates of different sizes and it is essential in any discussion of soil structure to describe precisely the scale at which structure or aggregation is being studied. This was stressed by Waters and Oades (1991) who pointed out that the range of sizes involved covers up to nine orders of magnitude. It is necessary to define whether one is dealing with the structure of clays in the nanometre-micrometre range, the structure

of microaggregates perhaps a hundred microns in diameter, macroaggregates a few millimetres in diameter, or aggregation at a larger scale.

The concept of aggregate hierarchy has been discussed theoretically by Dexter (1988) with particular emphasis on implications for the bulk density and mechanical strength of aggregates. Dexter outlined the 'porosity exclusion principle' which will apply if aggregate hierarchy exists. The concept is that larger aggregates will have a greater porosity than smaller aggregates because they will contain the pores between the smaller more dense aggregates. The larger aggregates will be weaker and have a lower tensile strength because they contain larger pores which are planes of weakness and failure planes when the aggregate is stressed. Utomo and Dexter (1981) showed that these tenets hold in friable soils based on dry tensile strength of aggregates with a range of sizes measured in tens of millimetres. The concept may not apply to clods formed from strongly sodic clays in which planes of weakness are rare.

Based on the principle that disaggregation will occur due to planes of weakness between stable aggregates it should be possible to determine whether an hierarchical order of aggregates exists or not by systematic studies of disaggregation as disruptive energy is increased. If aggregates breakdown in a stepwise fashion then hierarchy exists. If aggregates breakdown to release silt and clay size materials directly then there is not an hierarchical order.

The experimental work to be described examines potential hierarchy in three very different soils in terms of aggregate stability in water. The baseline is represented by water stable aggregation assessed by wet sieving and sedimentation after slow wetting (Slow), the most gentle disaggregation procedure available. Three methods of increasing intensity of disruption were used (a) wet sieving and sedimentation after fast wetting (Fast); (b) 16 hours end-over-end shaking (16 h) which has been used as a measure of mechanically dispersible clay (Rengasamy *et al.* 1984a; Shanmuganathan and Oades 1982); (c) ultrasonic dispersion (Son) which disrupts larger aggregates and releases most clay sized material. These results are compared with classical particle size distribution to determine clay contents using H_2O_2 and Calgon which destroy some organic colloids.

Experimental

Soil Samples

The surface horizons (0–10 cm) of each of three tilled soils were chosen for study. These are described in Table 1.

Fractionation Procedures

Air dried soils (1000 g, <4 mm) were separated into size fractions by the following procedure. Whole soil (50×20 g) was subjected to slow wetting on filter paper on a washed sand bed at 10 cm suction, and fast wetting by immersing soil in distilled water before mechanically wet sieving (Tisdall and Oades 1979) to obtain the size fractions 2.0–4.0 mm, 1.0–2.0 mm, 0.5–1.0 mm and 250–500 μ m. The <250 μ m fraction was wet sieved manually to obtain the size fractions 150–250 μ m, 90–150 μ m and 53–90 μ m. The <53 μ m fraction was then separated into 20–53 μ m, 2–20 μ m and <2 μ m fractions by sedimentation.

16 hours end-over-end shaking or ultrasonic dispersion (Turchenek and Oades 1978) of 20 g soil/50 ml H_2O suspensions was followed by manual wet sieving and sedimentation to obtain the size fractions >250 μ m, 53–250 μ m, 20–53 μ m, 2–20 μ m and <2 μ m.

Light fractions were obtained using a $ZnBr_2$ solution of density 2.0 $Mg\ m^{-3}$.

Table 1. Soils used in the study

	Alfisol-uncropped ^A	Alfisol-cropped ^B	Mollisol	Oxisol
Great Soil Group	Red-brown earth	Red-brown earth	Chernozem	Krasnozern
Soil Type and Location	Urrbrae fine sandy loam, South Australia	Urrbrae fine sandy loam, South Australia	Melfort clay, Saskatchewan, Canada	Malanda clay, Queensland
pH H ₂ O	5.2	5.4	6.0	4.6
Organic C %	2.8	1.2	5.4	5.6
Particle Size Distribution %				
>250 μ m	3	5	1	1
3-250 μ m	18	21	9	2
20-53 μ m	30	31	30	14
2-20 μ m	30	28	20	13
<2 μ m	19	15	40	70
Clay Type	illite-kaolinite	illite-kaolinite	smectite	kaolinite-oxide

^A Pasture for 40 years.^B Wheat-fallow rotation for 45 years.

Analytical Methods

Particle size distribution was determined using the pipette method (Gee and Bauder 1986). Total carbon was determined by dry combustion using a Leco CR-12 Carbon System with a furnace temperature of 1200°C. Total nitrogen was determined by the Kjeldahl digestion method. Specific surface areas were determined by the classical gas adsorption method.

Scanning Electron Microscopy

Samples were gold-coated and examined with a Cambridge Stereoscan X250 electron microscope.

Results

Bar graphs illustrating the data for the fractionation of the soils (Fig. 1) show from 25 to 85% of soil materials present in aggregates >250 μ m after slow wetting. Fast wetting caused substantial breakdown of these larger aggregates with little if any release of particles <20 μ m. The Oxisol was virtually stable to this treatment. The breakdown of aggregates >250 μ m in the Alfisol and Mollisol thus occurred in a stepwise fashion with larger aggregates producing smaller aggregates but not clay.

Further disaggregation caused by 16 hours shaking also indicated stepwise disaggregation rather than a complete disruption to single grain particles for the Alfisol and Mollisol but not the Oxisol. After shaking the Alfisol retained less than 10% of the soil material in particles >250 μ m but only one half of the clay was released; the other half remaining in aggregates 2-250 μ m in diameter.

In the intensively cropped Alfisol less than 10% of soil materials were present in macroaggregates after rapid wetting but the breakdown was only to particles 20-250 μ m; no further material <20 μ m was produced. However, shaking led to comprehensive disaggregation and released most of the clay present in the soil.

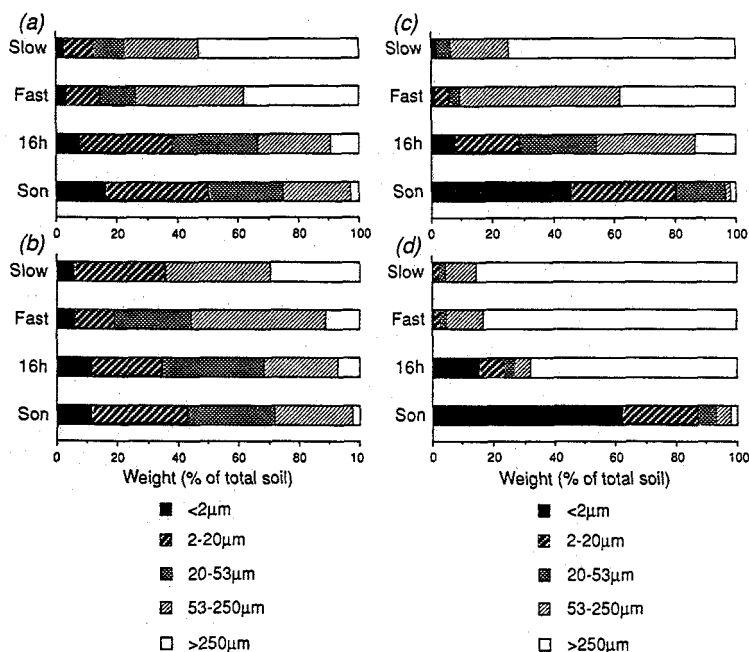


Fig. 1. Particle size distribution with increasing intensity of disruption (a) Alfisol-uncropped, (b) Alfisol-cropped, (c) Mollisol and (d) Oxisol.

In the Mollisol there was significant aggregate disruption by rapid wetting probably reflecting the substantial shrink-swell capacity of the dominantly smectite clay fraction, but even after 16 hours shaking three-quarters of the soil materials were present in aggregates $>20\ \mu\text{m}$ including about 80% of the clay fraction.

The Oxisol was quite different from the Alfisol and Mollisol. It was virtually stable to rapid wetting and released no clay. The breakdown of aggregates $>250\ \mu\text{m}$ caused by both shaking and ultrasonic treatment suggested no aggregate hierarchy as disruption led to predominantly particles $<20\ \mu\text{m}$ with no intermediate steps. The Oxisol thus behaved as a very stable soil with 85% of the soil material stabilized in aggregates $>250\ \mu\text{m}$ but when breakdown occurred it led immediately to the release of clay size particles.

Increased stability of smaller aggregates is shown graphically for the four soils in Fig. 2. The graphs represent the particle size distributions for aggregates destroyed between slow and fast wetting, between fast wetting and 16 hours shaking and between 16 hours shaking and ultrasonic dispersion.

From these curves we can derive mean aggregate sizes disrupted by increasingly vigorous treatments. The data show clearly that greater energy is required to disaggregate smaller aggregates and indicate 3 hierarchical orders of aggregation.

The curves show distinct peaks for the Alfisol and Mollisol illustrating hierarchy but this is not evident for the Oxisol.

Fast wetting disrupted aggregates $>250\ \mu\text{m}$, 16 hours shaking disrupted aggregates about $100\ \mu\text{m}$ in diameter and ultrasonic treatment disrupted

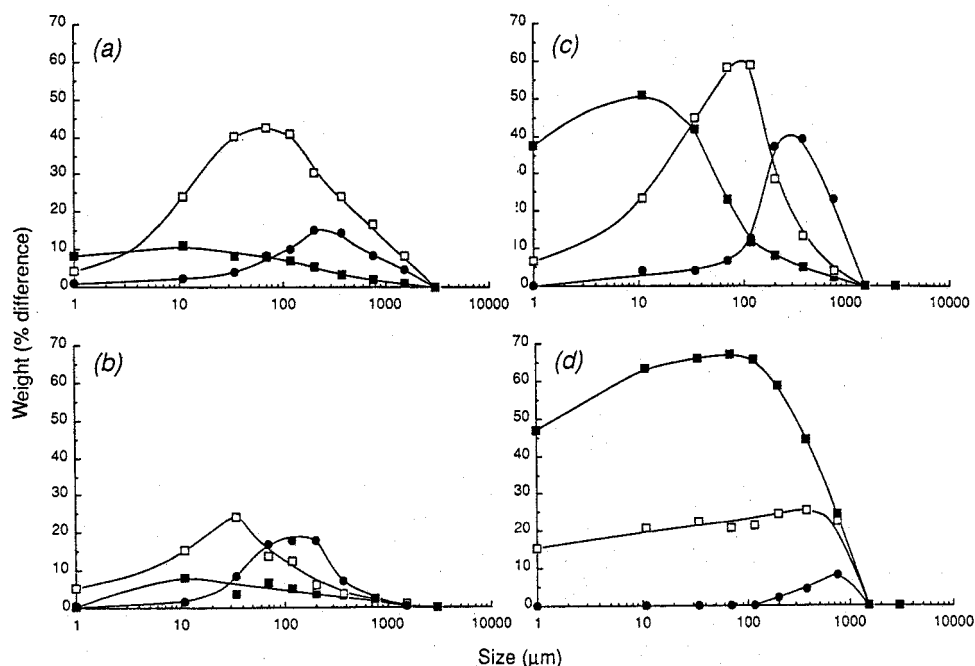


Fig. 2. Dimensions of aggregates destroyed with increasing intensity of disruption ■ Son-16 h, □ 16 h-fast, ● fast-slow, (a) Alfisol-uncropped, (b) Alfisol-cropped, (c) Mollisol and (d) Oxisol.

aggregates 2–20 μm in diameter releasing similar amounts of clay to that released by chemical treatments. The aggregates disrupted by ultrasonics cover a range of particle sizes and there is no evidence of hierarchy below about 20 μm. Aggregation in this size range is very stable and very dependent on organic materials. This is illustrated by measurements of specific surface areas before and after oxidation of organic matter using H₂O₂ (Table 2, see also Turchenek and Oades 1978). Specific surface areas are inversely related to the carbon contents of the fine fractions especially those <2 μm. From the limited data available we cannot see evidence for hierarchy within the clay fraction and cannot suggest any separation between clay aggregates as described for pure clay systems and the 2–20 μm aggregates in soils which contain a mixture of inorganic and organic colloids.

Carbon and Nitrogen in Particle Size Fractions

Carbon and nitrogen contents of all the various particle size fractions were determined and the following conclusions were drawn. For the Alfisol and Mollisol aggregates >250 μm contain a greater concentration of C and N than the soil as a whole. Fractions in the microaggregate range 20–250 μm were lower in organic matter probably due to the presence of substantial quantities of single grain particles in this size range. The largest aggregate sizes tended to be richer in organic matter and this trend was more obvious with the more vigorous disaggregation treatments.

Ultrasonic treatment destroyed all larger aggregates and any particles remaining were either quartz grains or pieces of plant debris which had been freed from inorganic components. This plant debris was concentrated in light fractions which were richest in carbon and had high C:N ratios. Scanning electron microscopy showed the vascular and structural components of plants (Plate 1a). High magnification (Plate 1b) shows clearly what are probably the recalcitrant ligno-cellulose components of plants colonized by fungal hyphae. The hyphae show as collapsed structures due either to the effects of the heavy liquid used to float the fragments or the vacuum in the electron microscope.

Clay fractions were relatively rich in organic matter with C:N ratios below 10 in most cases.

The results for the Oxisol showed uniformity of organic matter content and C:N ratios across the range of particle sizes. C:N ratios were higher than for the other soils.

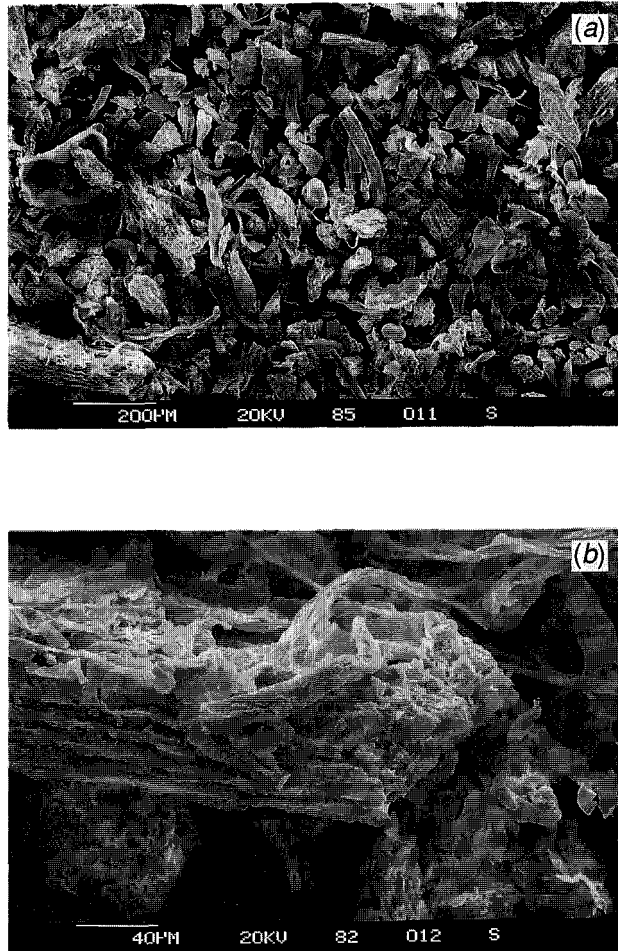


Plate 1. Light fraction from Alfisol-uncropped 53–250 µm fraction (a) plant debris, (b) structural components in plant debris.

Table 2. Specific surface areas and organic carbon contents of Alfisol particle size fractions obtained after slow wetting and wet sieving, and ultrasonic dispersions.a. = specific surface area in m^2g^{-1} . C = organic carbon content in percent

Size (μm)	Alfisol-uncropped				Alfisol-cropped			
	untreated		H_2O_2 treated		untreated		H_2O_2 treated	
	s.a.	C	s.a.	C	s.a.	C	s.a.	C
<i>Slow wetting and wet sieving</i>								
<2	12	4.6	32	1.1	27	2.9	48	0.7
2-20	14	3.5	19	0.4	13	1.5	12	0.3
20-53	5	1.3	8	0.2	4	0.2	3	0.1
53-90	6	1.6			5	0.4		
90-150	7	2.4			8	0.7		
150-250	9	3.0			11	1.0		
250-500	9	2.9			11	1.2		
<i>Ultrasonic dispersion</i>								
<2	39	4.9	70	0.7	54	3.3	87	0.5
2-20	9	2.6	11	0.3	11	1.3	10	0.1

Discussion

The Concept of Micro- and Macroaggregates

The results presented confirm the utility of the concept of micro- and macroaggregates for the Alfisol and Mollisol but not for the Oxisol. The original suggestion to use $250\ \mu\text{m}$ to separate micro- and macroaggregates was made by Edwards and Bremner (1967), consolidated by Tisdall and Oades (1982) and subsequently supported by Elliott (1986) and Miller and Jastrow (1990). The soils involved were Mollisols and Alfisols. The concept of micro- and macroaggregates fits well with the Australian view of slaking and dispersion as described by Emerson (1977). The disruption of macroaggregates is caused by slaking. The result in non-sodic soils is breakdown to microaggregates but not dispersion of finer colloidal materials. The microaggregates are quite stable but can be disrupted by mechanical energy, e.g. raindrop impact or sodicity/electrolyte effects releasing fine particles which will disperse in the aqueous phase.

The Stability of Macroaggregates

The stability of macroaggregates in these soils appears to be controlled by roots and hyphae from vesicular arbuscular mycorrhizae (VAM). There are available in the literature a range of electron micrographs which illustrate elegantly the interaction of ramifying root and hyphal systems with clay and silt particles via mucilages on the surfaces of roots and particularly hyphae (e.g. Tisdall and Oades 1979; Waters and Oades 1991; Foster *et al.* 1983). The scanning electron micrographs (Plate 2) indicate the attachment of inorganic particles to a fragment of root several millimetres long. The fragments were obtained after slow wetting and wet sieving and illustrate clearly the 'sticky string bag' effect of roots as well as hyphae. The encrustation of roots and hyphae by inorganic components protects them from decomposition by microorganisms. However, once the root and the associated hyphae die they are

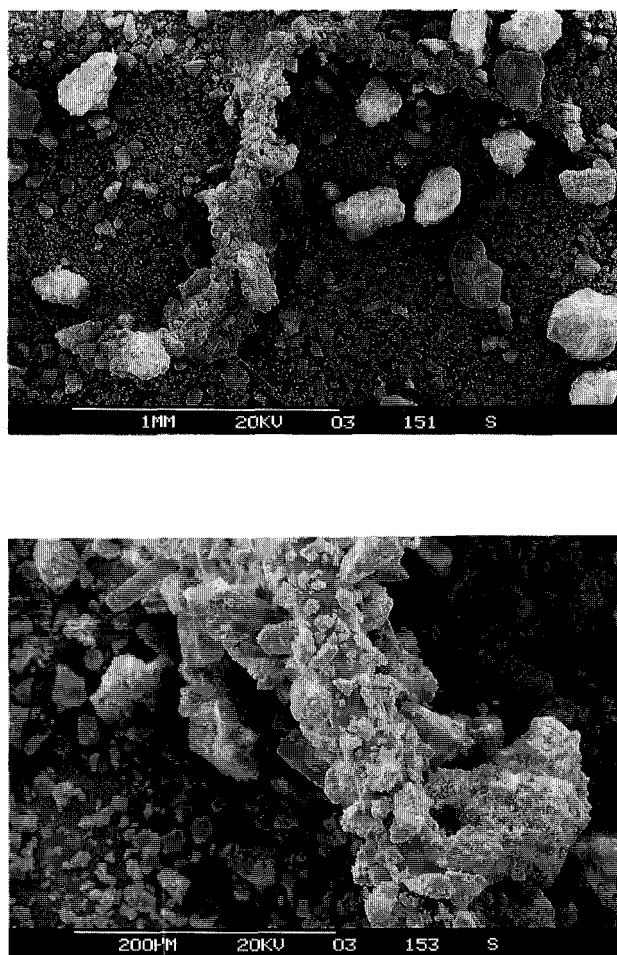


Plate 2. Fine wheat root encrusted with clay and silt particles.

fragmented, probably by fauna and are no longer of sufficient length to perform the role of tying together macroaggregates by physical entanglement. The binding effects for the macroaggregates will disappear at the same rate as the decomposition of the root system and result in the relation between aggregation and organic carbon as controlled by cereal-pasture rotation presented by Tisdall and Oades (1979).

We know that as the decomposition proceeds about one third of the carbon will remain in the soil after one year (Mediterranean climate) and thereafter the decomposition becomes very slow (Ladd *et al.* 1985). At this stage the organic matter concerned is located in microaggregates which we can define as 20–250 μm . Systematic studies of aggregates within this size range has shown that the stability of the microaggregates is still much influenced by cores of plant debris which will serve as a source of energy for soil microorganisms but rapid decomposition is limited by the interaction with the inorganic matrix (Waters and Oades 1991).

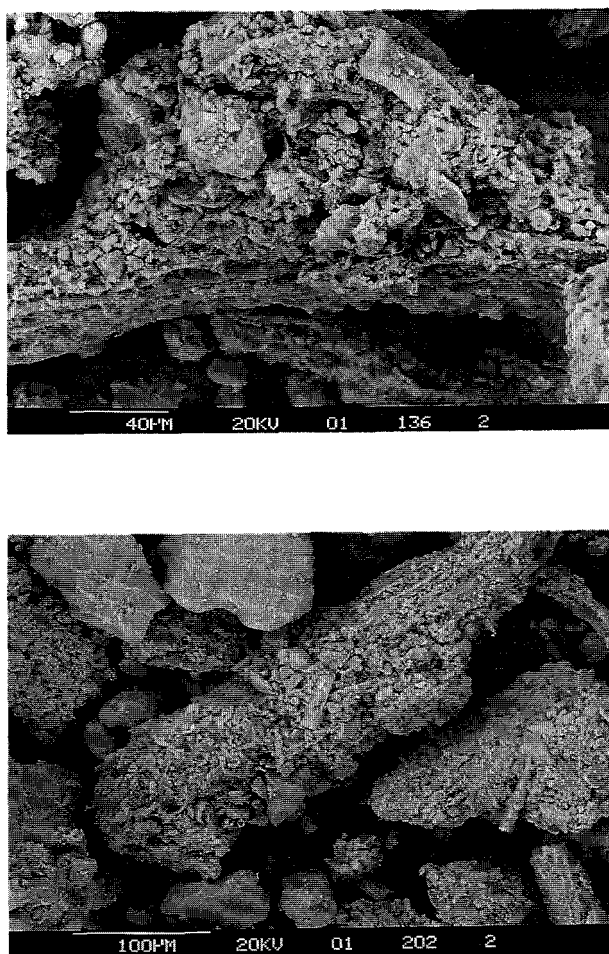


Plate 3. 90–250 μm sized particles from Alfisol-uncropped showing encrusted plant fragments.

The Stability of Microaggregates

For aggregates $<250 \mu\text{m}$ electron microscopy indicates a major role for plant debris in water stability. Many aggregates about 100–200 μm in diameter have cores of plant debris (Plate 3). Some are completely coated with inorganic material but retain an elongate shape with a length to width ratio greater than 2. When these materials are disrupted using ultrasonic energy plant debris as illustrated in Plate 1 is released. Such aggregates are quite stable but intensive cultivation (Alfisol-cropped) leads to a situation where 16 hours shaking caused complete breakdown. This correlated well with the behaviour of this soil in the field which slakes, slumps and disperses under rainfall to form crusts.

In aggregates $<90 \mu\text{m}$ in diameter plant fragments become more difficult to recognize. The aggregates tend to be more equidimensional with length

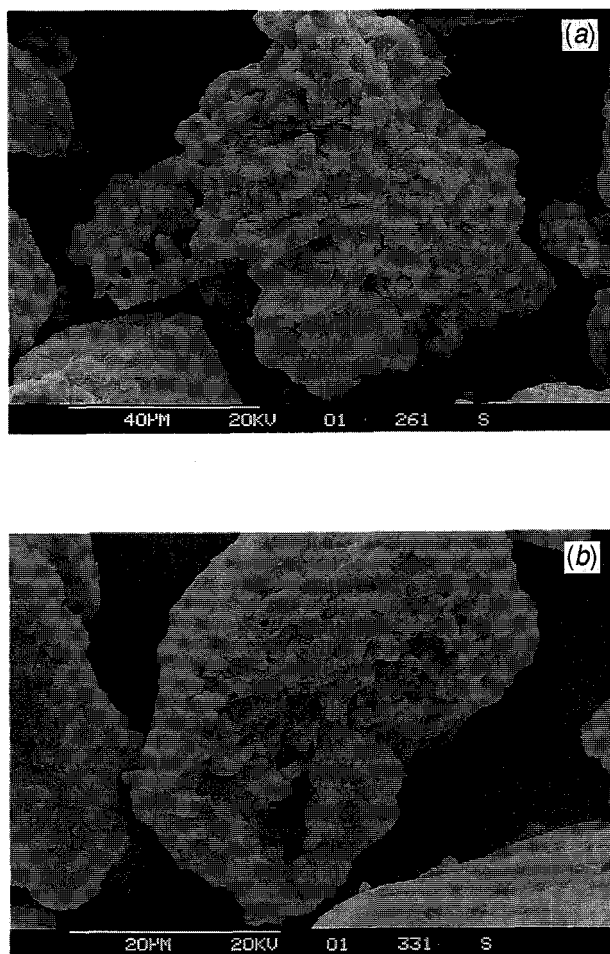


Plate 4. 20–90 μm sized particles from Alfisol-uncropped (a) with lignin and (b) with cavity.

to width ratios up to about 2. The core of such aggregates may contain the last remnants of vascular plant debris such as particles of lignin (Plate 4a) or in fact an empty core (Plate 4b) which is thought to be due to the complete degradation of plant debris leaving a cavity. Thus decomposition products of the organic core were responsible for stable aggregation which has remained long after the organic debris has gone.

Examination of aggregates less than 20 μm diameter indicates that little if any plant debris exists in particles this small. Changes in C/N ratios and the composition of the organic materials as seen by modern spectroscopic techniques suggests that even lignin has been degraded in particles <20 μm diameter. The organic matter in fine fractions is thus dominated by microbial products. At this scale scanning electron microscopy of real soils becomes difficult with respect to identification of biological entities because diagnostic shapes are lacking. What is evident is the microstructure of clays. The three

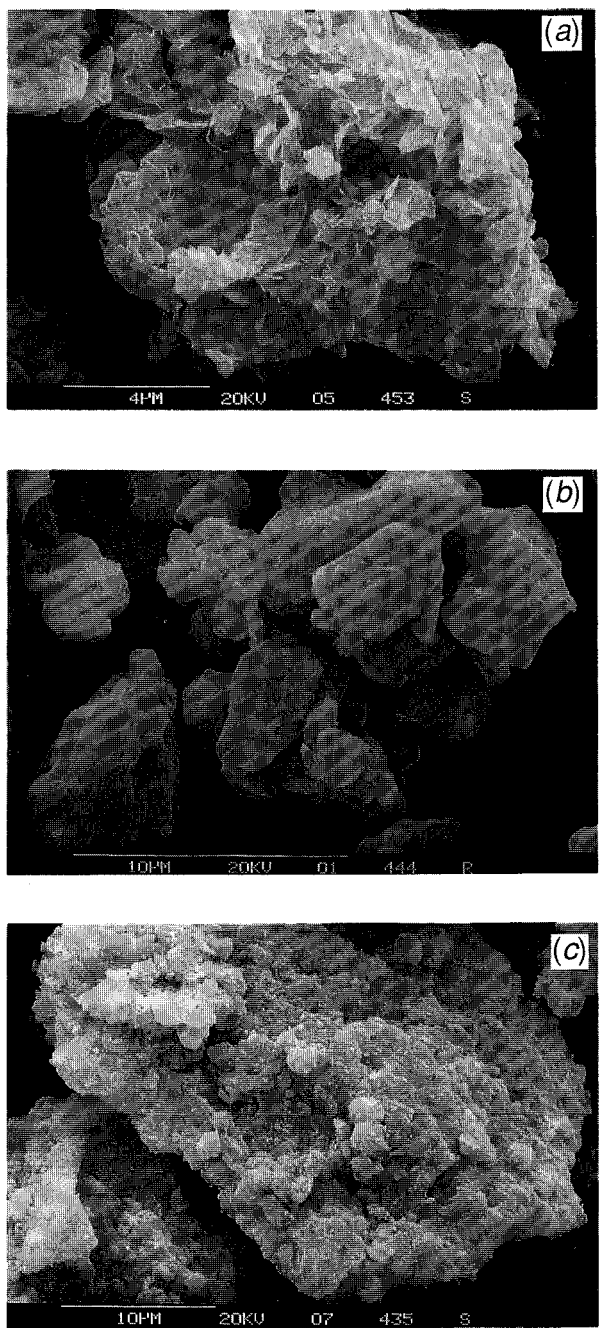


Plate 5. Clay microstructure (a) Mollisol-quasicrystals, (b) Alfisol-domains, (c) Oxisol-assemblages of blocky plates.

soils have different clay mineralogy (Table 1) with the Alfisol dominated by illitic minerals, the Mollisol by smectites and the Oxisol by kaolin and iron oxides. Each of these clay minerals exhibits distinct morphologies which lead to different packing characteristics easily recognisable by SEM (Plate 5). Good examples of quasicrystals of smectite were seen in the Mollisol (Plate 5a), domains of platy illite particles with kaolinite in the Alfisol (Plate 5b) and clusters of fine kaolin crystals, presumably held together by oxides, in the Oxisol (Plate 5c). Quasicrystals based on smectites can be observed in numerous micrographs obtained by TEM of thin sections (Foster *et al.* 1983). Further information on the structure of clays, based on interactions which lead to associations of plates or crystals has been discussed by Rengasamy *et al.* (1984b) and Murray and Quirk (1990) and simplified to schematic diagrams (Fig. 3) by Oades (1987).

The organization of the basic units of aluminosilicate is controlled by particle morphology. The degree of ordering of particles within the first stage clay aggregates ranges from minimal, with large blocky rigid particles with low width to thickness ratios as found in many kaolins, through some degree of parallel stacking of platy particles with width to thickness ratios of less than 10, as in illites, to something approaching perfect parallel alignment of thin flexible plates with width to thickness ratios exceeding 100,

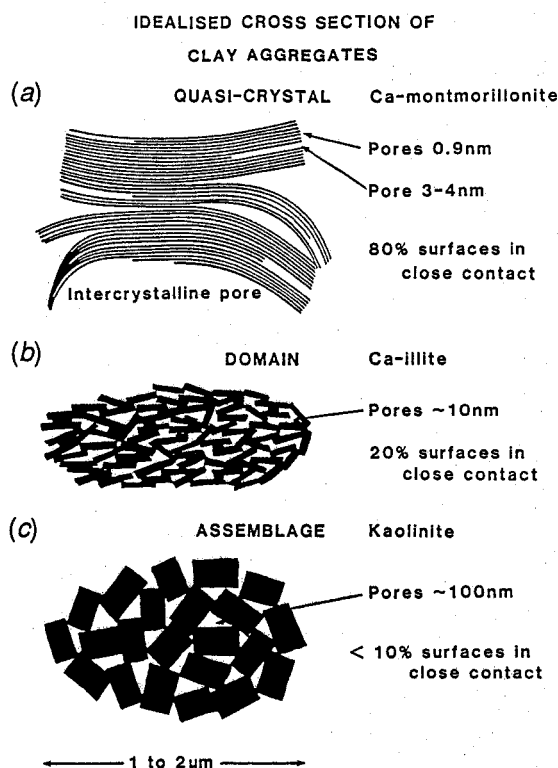


Fig. 3. Clay aggregates (a) quasicrystals, (b) domain and (c) assemblage of kaolinite crystals.

as shown by smectites. The term quasi-crystal is used to describe a stable but expandable arrangement of a number of basic units of elementary lamellae of Ca-montmorillonite (Aylmore and Quirk 1971).

While it is quite clear that clay aggregates exist in the $<20\ \mu\text{m}$ range of the soils studied it is also clear from surface areas before and after oxidation of organic matter using H_2O_2 that organic materials play a major role in stabilizing clay microstructures in soils. However, the data presented here indicates no evidence of stepwise degradation of aggregates smaller than $20\ \mu\text{m}$ diameter where there are close associations of microorganisms, biopolymers and clay structures with no evidence of hierarchy. More detailed studies in the μm size range may reveal some order in the system in the future.

Conclusions

Aggregate hierarchy exists in soils where aggregate stability is controlled by organic materials. It was not evident in an Oxisol where oxides are the putative dominant aggregate stabilizing agents.

The hierarchy is created by the physical entanglements of particles by living roots and hyphae into macroaggregates $>250\ \mu\text{m}$ diameter. On the death of roots and hyphae the stability of macroaggregates will decline at about the same rate at which plant material decomposes in soils, i.e. initially quickly but then more slowly.

The degradation of macroaggregates creates aggregates from about 20 to $250\ \mu\text{m}$ diameter which are considerably more stable than the larger aggregates. Little material $<20\ \mu\text{m}$ is released during this first step in disaggregation. Fragments of plant material serve as cores for many microaggregates and are protected from rapid decomposition by encrustation with inorganic materials. Eventually the plant debris is oxidized leaving a cavity surrounded by aggregations of particles stabilized by the microorganisms, which utilized the plant debris as a substrate, and their metabolic products.

For aggregates $<20\ \mu\text{m}$ diameter there appears to be a random mixture of clay microstructures, biopolymers and microorganisms.

It is suggested that these three steps in aggregation may apply to those surface soils in which structural stability is dominated by organic material.

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