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The effect of ultrasound treatment on the particle-size of Wyoming bentonite in aqueous suspensions

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Aqueous suspensions of Na-montmorillonite (1.80 and 0.60 wt%) were aged for three months and were sonicated for different times. Statistical parameters related to volume were determined from particle-size distributions obtained by Laser Shadow Analysis. After a long ultrasound treatment the concentrated suspension showed a gradual increase of the largest particles and of the mean and median particles, as well as a decrease of the percentage of particles with diameters under 1.5 μm. The dilute suspension, on the other hand, showed a decrease of the largest particles and of the mean and median particles, and an increase of the percentage of particles with diameters under 1.5 μm. In the dilute suspension sonication resulted in the formation of small swollen particles by the delamination of large swollen tactoids. In the concentrated suspension the swelling of the clay was not complete during the three months aging period. As a result of the ultrasound treatment tactoids were delaminated and the swelling of the clay particles became more efficient.

1. Introduction

It is well known that an ultrasound treatment of aqueous clay suspensions stabilizes the colloidal system. In general this treatment is carried out in a water bath with a very low sonication energy and for very short periods. To the best of our knowledge, no systematic study has been carried out until recently to understand the processes which lead to the stabilization of the suspensions. In aqueous smectite suspensions, the following three types of species exist simultaneously: (1) kinetically independent primary particles, which are platelets of single unit-layers or tactoids, (2) spherical flocs composed of associated platelets or tactoids with card-house or book-house structures, respectively, and (3) aggregates composed of associated flocs with a block-house structure [1]. With increasing rate of shear the average size of the flocs and aggregates decreases until at a sufficiently large shear rate the suspensions contain either dispersed primary particles (single platelets or tactoids), or small stable flocs, or both types of units.

Pérez-Maqueda *et al.* [2, 3] and Pérez-Rodriguez *et al.* [4] showed that sonication of layer silicates such as vermiculite, muscovite and biotite, resulted in nano-particles. The effect of sonication on particle-size was studied by recording particle-size distribution curves using a Low Angle Laser Light Scattering system. The nano-particles were obtained from the delamination of the silicate crystals and the breakdown of the layers. Grauer *et al.* [5] showed that a short treatment in a "supersonic cleaner" (with weak ultrasound energy) of flocs of a dye-clay complex, resulted in their deflocculating.

Recently [6] we determined particle–size distribution curves of Na-, Ca- and Mn-montmorillonite, in aqueous suspensions in different clay concentrations. by using a Laser Shadow Analysis. In this system, particle-size determination is carried out by the 'time-of-transition' (TOT) technique, in which measurements are made on single particles so that the resolution is relatively high. The TOT method for particle-size determination was described by Wiener *et al.* [7]. The particle-size is determined by the pulse width and to first order the results are not dependent on the optical properties of the particles. Dilution or shaking had almost no effect on the particle-size distribution curves of Ca- and Mn-montmorillonite but the curves of Na-montmorillonite were affected by these treatments. The presence of large particles of Na-montmorillonite was attributed to the extensive swelling of this clay. In the present paper, the effect of ultrasound on size-distribution of Na-montmorillonite determined by Laser Shadow Analysis is described. This study might give information on the swelling of sonicated dispersed clay particles.

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2. Experimental

2.1. Materials

Na-montmorillonite (Wyoming-bentonite) was supplied by Ward's Natural Science Establishment, Inc. The aqueous suspension was prepared by dispersing the manually ground and sieved (80 mesh) clay. The non-clay fraction was separated by sedimentation. Aqueous clay suspensions were prepared in concentrations 1.80 and 0.60 wt% and were aged for three months before used. It is assumed that in this period association-dissociation reactions of platelets-tactoids-flocs-aggregates have reached equilibrium.

2.2. Methods

A "Vibra-Cell" VCX750 (20 kHz) digital ultrasonic processor equipped with 13 mm probe (supplied by MEDENT, Ltd.) was used for the ultrasound treatment. A beaker with 150 ml suspension was placed in an ice bath. Cycles of five seconds ultrasound action followed by five seconds of pause were continuously employed. The ultrasound treatments were carried out during different lengths of time, resulting in accumulated applied energies. The suspension temperature during the treatment was 15–18°C.

The particle-size distributions in the different suspensions before and after ultrasonic treatment, in the range 0.5 to 100 μm were measured by a particle-size analyzer Galai CIS 1 (Computerized Inspection System), equipped with a 3 ml plastic cell for holding the analyzed suspension. The instrument and the working procedure have been described [6]. Portions of the suspensions after ultrasound treatments with different energies were diluted directly in the measurement cell. The final concentration was 0.1 wt%. Since the particle-size distributions were determined immediately after the dilution, it is assumed that they are very similar to the distributions in the non-diluted suspensions.

3. Results of the particle-size distribution study

In the present study statistical parameters were determined from particle-size distributions related to volume, which is equivalent to clay mass, described in percentages as a function of particle-size.

3.1. An aqueous clay suspension of 1.8 wt%

3.1.1. The effect of clay concentration and shaking on the size distribution of the suspended particles

In order to obtain readable and reliable measurements of the particle-size distribution in the 1.8% suspension it was necessary to dilute the suspension. In our previous work [6], we showed that in a 0.3 wt% suspension (obtained by diluting the 1.8 wt% suspension) 90% of the volume of the particulate matter was composed of particles with diameters under 1.0 μm and 10% of particles with diameters between 1.0 and 1.5 μm (in other words volume% of particles under 1.0 and 1.5 μm were 90 and 100, respectively). Larger particles were not detected. In the present study similar particle-size distribution was obtained when the aged suspension was diluted to 0.1 wt%. The particle-size distributions were recorded promptly after dilution, and we assume that the distribution which was determined in the 0.1 wt% is equal, or almost equal, to that occurring in the concentrated suspension. In more dilute suspensions larger particles were detected.

Shaking of the diluted suspension in the measurement plastic cell a short time before recording the particle-size distribution affects the particle diameters and reproducibility of the measurements. For example, in the case of the 0.1 wt% suspension, in one shaking treatment the particle-size increased whereas in another it slightly decreased. The particle-size distributions in the more dilute suspensions also show changes due to shaking [6].

3.1.2. The effect of ultrasound treatment on the size distribution of the suspended particles

Table I summarizes statistical parameters determined from particle-size distributions obtained after different ultrasound energies. Since the reproducibility of the measurements is not perfect, data for each set are given for non-shaken (A), and shaken (B), suspensions. However, the trend in the presented results is significant. The table shows a gradual increase of the largest particles as well as of the mean and median particles, and a decrease of the percentage of particles with diameters under 1.5 μm with the ultrasound energy.

In Fig. 1a the concentrations (in volume%) of particles with different sizes are plotted against the energy of

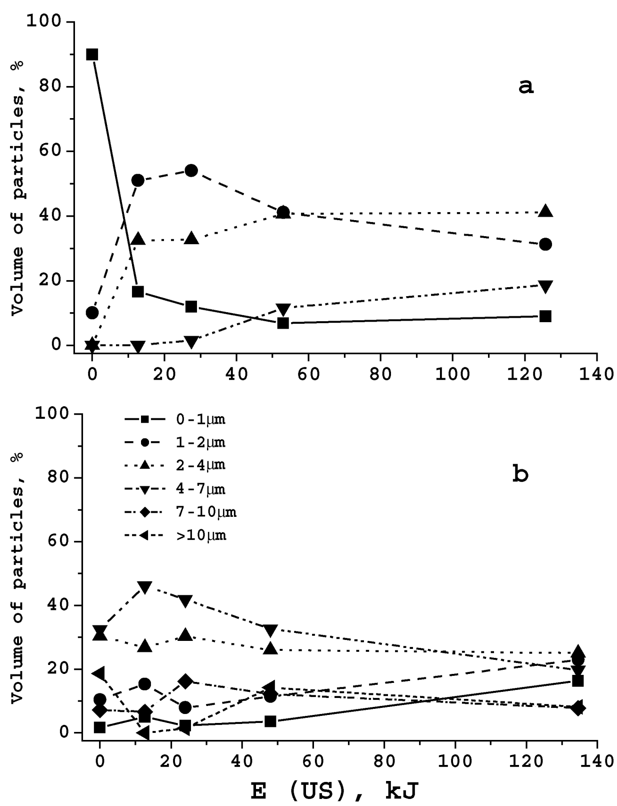

Figure 1 Concentrations (in volume%) of particles with different sizes against the accumulated energy of the ultrasound treatment, (a) 1.8 wt%, (b) 0.6 wt%.

TABLE I The effect of the ultrasound energy on the diameters of the largest, the median and the mean particle (in μm), and the volume% of particles with diameters under 1.5 μm. Concentrations of the mother aqueous clay suspensions were 1.8 and 0.6 wt%. All data are obtained from volume distributions

Ultrasound energy (kJ)		Diameter of largest particle (μm)		volume% of particles with diameters under 1.5 μm		Median particle-size from volume distribution (μm)		Mean particle-size from volume distribution (μm)	
Clay concentration		1.8%	0.6%	1.8%	0.6%	1.8%	0.6%	1.8%	0.6%
A	0	1.5	9.5	100	15	0.79	3.63	0.82	3.60
B	0	1.5	n.d.	100	n.d.	0.78	n.d.	0.80	n.d.
A	13	3.5	10.0	45	13	1.60	4.24	1.74	4.11
B	13	5.0	13.0	31	10	1.88	4.12	2.00	4.80
A	25	5.0	11.0	40	7	1.69	4.31	1.78	4.77
B	25	5.0	13.5	30	6	1.91	4.16	2.14	4.70
A	50	5.0	17.5	26	10	1.98	4.52	2.24	5.92
B	50	6.0	11.5	31	14	1.84	4.07	2.12	4.23
A	130	6.5	12.0	23	32	2.34	3.02	2.65	3.82
B	130	4.5	7.0	26	33	2.17	3.02	2.25	2.94

A: immediately after dilution; B: immediately after shaking. n.d.: not determined.

the ultrasound treatment. Comparing the particle-size distribution before and after the ultrasound treatment of 130 kJ reveals that small particles disappeared and large particles appeared. The figure shows that before the treatment 90 percent of the volume is composed of particles under 1 μm. After this treatment 73 percent became particles with diameters between 1 and 4 μm and only 9 percent remained as particles with a diameter smaller than 1 μm. There are practically no particles with a diameter larger than 7 μm. The figure supports the suggestion that the very small particles (<1 μm) initially present in the suspension are transformed to larger particles.

3.2. An aqueous clay suspension of 0.6 wt%; The effect of ultrasound treatment on the size distribution of the suspended particles

According to Table I there was an increase in the diameter of the largest particle up to ultrasound energy of 50 kJ but at 130 kJ this diameter became smaller. Similar dependencies of particle-sizes on the ultrasound energies were observed in the median and mean diameters. The content of particles below 1.5 μm decreased up to 25 kJ. There was a small increase with ultrasound energy of 50 kJ and a significant increase with 130 kJ.

By comparing the particle-size distribution before the ultrasound treatment and after a treatment of 130 kJ shown in Fig. 1b, it is obvious that the size of many of the particles decreased. The figure shows that before the treatment the percent of particles with diameters below 2 μm was 12, but at the end of this treatment more than 40 vol% are particles with diameters under 2 μm. Particles with diameters in the range 2–4 μm decreased only slightly from 30 to 25 vol%, but particles with diameters larger than 4 μm decreased considerably from 60 to 26 vol%. From this figure it is obvious that the small particles (<2 μm) were obtained during ultrasound treatment on the 0.6% suspension by the mechanism of dissociation of large particles. The irregular increase in the volume% of the 4–7 and 7–10 μm particles with ultrasound energies of 13 and 25 kJ, respectively, may indicate gradual dissociation and swelling reactions.

4. Discussion

According to Peters [8] cavitational collapse sonication on solids leading to microjet and shock-wave impacts on the surface, together with interparticle collisions, can result in particle-size reduction. The effect of mechanochemical treatments on clay minerals was recently reviewed [9]. Grinding results in breakdown of the layers, diffusion of atoms inside the clay crystal, disaggregation of aggregates and delamination of tactoids. When the grinding is carried out in the presence of water, disaggregation and delamination are the principal mechanochemical processes.

In the present study we showed that small particles were obtained by sonication in the dilute suspension but larger particles were obtained in the concentrated suspension. In the dilute suspension water was swollen by the Na-clay during the aging period of three months. Sonication resulted in the formation of small swollen particles by the delamination of larger swollen tactoids. In the concentrated suspension the Na-clay was slightly aggregated and its swelling was not complete during the aging period of three months. As a result of the ultrasound treatment aggregates were disaggregated and clay tactoids were delaminated. As a consequence, swelling of Na-montmorillonite particles became more efficient.

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