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Coagulation of oil in water using sawdust, bentonite and calcium hydroxide to form floating sheets

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Abstract

The coagulation of oil in water is valuable for the removal of oil from water. The use of a mixture of bentonite and sawdust, with sawdust being the vast majority, is highly effective for the coagulation of oil in water, giving coagulation efficiency 92% or above. A minor amount of calcium hydroxide may be optionally added to the mixture to increase the coagulation efficiency to 94%. The use of organobentonite in place of unmodified bentonite increases the coagulation efficiency further to 95%, but increases cost. Sawdust by itself sinks in water. However, the coagulated aggregates float on water when sawdust is used with the bentonite. These aggregates are sheet-like, with the oil–bentonite–sawdust serving as a continuous matrix and apparently no upper limit to the aggregate sheet size. They contain, for example, 81 vol.% oil, 15 vol.% sawdust, 3 vol.% bentonite (with basal spacing 14.4 Å) and 1 vol.% calcium hydroxide. Upon compression, 73% of the oil in the aggregate is removed. Without sawdust, the aggregates sink in water and the coagulation efficiency is only 37%. The sawdust functions as a fibrous framework for the attachment of the coagulating oil and bentonite, thus facilitating floating aggregate formation and the subsequent removal of the aggregates. In the presence of unmodified bentonite, sawdust enhances the coagulation efficiency significantly, while calcium hydroxide enhances the coagulation efficiency by a much lower degree.

1. Introduction

Coagulation refers to the transformation of a liquid to a soft, semisolid, or solid mass. Coagulation requires the use of a small amount of a coagulant, which, through adsorption, absorption and flocculation, causes coagulation.

The amount of coagulant required for coagulation is much smaller than the amount of adsorbent that is required for capturing the same amount of liquid. Activated carbons are adsorbents that have been used for the adsorption of phenolic compounds in wastewater (Richard et al., 2009) and of methylene blue in aqueous solutions (Karagoz et al., 2008). Sawdust is a low-cost adsorbent that has been used for the adsorption of oil (Tiwari et al., 2009) and methylene blue (Bello et al., 2010) in water. Organobentonite has been used as an adsorbent of oily liquids (Gitipour et al., 1997). Clay aerogel made by freeze-drying is a sorbent that has been used for the sorption of aqueous liquids (Gawryla and Schiraldi, 2009). Sorbents are affected by fouling, but coagulants are not. This is because coagulation takes place outside the coagulant particles, whereas sorption takes place inside the pores of the sorbent. In addition, because adsorption involves adhesion to a surface, it suffers from the relatively small amount of material that can be adsorbed.

After coagulation, the coagulated material may be removed by skimming, filtration, flotation or sedimentation. The buoyancy of the coagulated material is attractive, as it facilitates the removal of the coagulated material by scooping or other mechanical methods. Furthermore, the sedimentation of the coagulated material may negatively impact benthic organisms (Niu et al., 2010).

The coagulation of oil is relevant to the removal of oil from water, as needed for the clean-up of oil spills (Ajjisolsys et al., 2007). The formation of the coagulated material promotes the dispersion of stranded oil, as relevant to the recovery of oiled shorelines (Owens and Lee, 2003; Stoffyn-Egli and Lee, 2002). Oil coagulation is also useful for the treatment of industrial wastewater (Abdelaal, 2004; Almaliky, 2010). Oil is found in numerous types of wastewater, with the types of oil including insoluble hydraulic fluids, crude oils, bunker oils and lubricant oils.

Due to their low cost, natural coagulants, such as minerals, are attractive for large-scale environmental applications. When minerals are used, the coagulated material is known as oil–mineral aggregates (OMA), which are classified as droplet, solid and flake aggregates (Stoffyn-Egli and Lee, 2002). The size of the aggregates increases with increasing degree of hydrophobicity of the minerals (Zhang et al., 2010).
Due to their high abundance and fine microstructure, clay minerals (particularly montmorillonite) are used as coagulants (Tansel and Sevimoglu, 2006; Wincele et al., 2004). Montmorillonite constitutes 90% of the composition of an industrial grade bentonite, which is commonly used as a coagulant (Abdelaal, 2004; Almilky, 2010; Laurent and Laurent, 1993).

There are two main classes of bentonite, based on the dominant exchangeable ion that is weakly bound in the double layer of montmorillonite. They are sodium bentonite and calcium bentonite. Sodium bentonite swells more in water than calcium bentonite and has excellent colloidal properties. Organically modified bentonite (called organobentonite) is manufactured by modifying bentonite with quaternary ammonium cations via a cation exchange process. Organobentonite is particularly effective as an oil coagulant (Atther, 1999, 2008).

In relation to surface charge neutralization, metal salts are used as coagulants for oil. Metal salts that have been used for this purpose include calcium sulfate (CaSO₄·2H₂O) (Cambiella et al., 2008), aluminum hydroxide (Canizares et al., 2008), aluminum chloride hydrate (Albu and Beauchamp, 2005) and ferric chloride (Badawy and Ali, 2006). Due to the negative charge associated with the silicate layers of montmorillonite and the positive charge associated with the ions in a metal salt, the combined use of bentonite and aluminum magnesium hydroxide (Abend et al., 1998) enhances the efficiency of coagulation. The pH also affects the coagulation process (Canizares et al., 2008; Hempoonsert et al., 2010).

This paper is primarily aimed at advancing the technology of oil coagulation in water by the combined use of bentonite and sawdust. The use of sawdust to facilitate oil coagulation using bentonite is innovative. Sawdust has been previously used as a filter medium and sorbent in the treatment of oil-in-water emulsions (Cambiella et al., 2008), but it has not been previously considered for helping the process of coagulation.

A second objective aims at advancing the technology of oil coagulation in water by the combined use of bentonite, sawdust and calcium hydroxide. Calcium hydroxide is expected to be more attractive than the previously used aluminum hydroxide (Canizares et al., 2008) for interaction with bentonite, due to the higher charge of the Al³⁺ ions compared to the Ca²⁺ ions making it relatively difficult for the Al³⁺ ions to reside between the montmorillonite layers.

A third objective is to compare the effectiveness of organobentonite and unmodified bentonite for oil coagulation in the presence of calcium hydroxide and sawdust. This comparison is practically important, since organobentonite is much more expensive than unmodified bentonite.

A fourth objective is to investigate the effect of calcium hydroxide on the coagulation in the presence of unmodified bentonite and sawdust, and the effect of sawdust on the coagulation in the presence of unmodified bentonite and calcium hydroxide.

A fifth objective is to evaluate the effectiveness of removal of oil from the coagulated material by compression. This objective relates to the feasibility of obtaining liquid oil by squeezing the coagulated material.

2. Experimental methods

2.1. Materials

Tap water was used in the experiments. The oil is HE-175 vacuum pump oil (a hydraulic fluid), which is a highly distilled pure hydrocarbon oil (solvent refined neutral paraffinic oil) from Leybold (Export, PA). The chemical formula is (CH₂)n, where 20 ≤ n ≤ 40. It is an amber viscous liquid, insoluble in water, with density 0.88 g/cm³, vapor pressure less than 1 kPa at 20 °C, and boiling point above 200 °C.

The calcium hydroxide is a white powder from J.T. Baker, Phillipsburg, NJ (Product 1372-01). It contains 97.0% Ca(OH)₂, 2.3% CaO, 0.9% magnesium and alkaline salts (as SO₄), 0.03% Fe and 0.02% Cl⁻. Its density is 2.24 g/cm³ and it is slightly soluble in water (0.185 g/100 cm³ at 0 °C). Its melting point is 580 °C. Its particle size is such that 99% passes U.S. 325 mesh (44 μm).

The sawdust was obtained from a carpenter shop. Its density is 1.65 g/cm³. It has fibrous morphology, with the fiber diameter ca. 10 μm.

The sodium bentonite powder (M325, provided by Ashbury Graphite Mills, Inc., Ashbury, NJ) contains 2–6% free SiO₂ and has less than 10% moisture. It has a cation exchange capacity (CEC) 92 cmol/kg, density 2 g/cm³ and negligible solubility in water. 98.65% of the powder passes through U.S. 325 mesh (44 μm).

The organobentonite is Cloisite 10A, as provided by Southern Clay Products, Inc., Gonzales, TX. It consists of montmorilloniteintercalated with a salt of dimethyl benzyl hydrogenated tallow with quaternary ammonium cations and chloride anions and basal spacing d0019.2 Å. The CEC is 125 cmol/kg. Its particle size distribution is: 10% less than 2 μm, 50% less than 6 μm and 90% less than 13 μm. The density is 1.90 g/cm³. Montmorillonite is hydrophilic, but ion exchange involving the ammonium salt renders the clay more hydrophobic. The organobentonite has a reduced surface energy, which is well-suited for use with oil.

In order to reach the research objectives, four coagulant formulations were prepared, namely (i) organobentonite, calcium hydroxide and sawdust; (ii) unmodified bentonite, calcium hydroxide and sawdust; (iii) unmodified bentonite and sawdust, and (iv) unmodified bentonite and calcium hydroxide. The proportions of the ingredients are given below. In this paper, unmodified bentonite is also referred to as bentonite.

In formulations (i) and (ii), the mass ratios of bentonite (either unmodified bentonite or organobentonite), sawdust and calcium hydroxide are 0.375:1.000:0.125, and the volume ratios are 0.198:1.00:0.059. In formulation (iii), the bentonite proportion increased, so that the mass ratio bentonite:sawdust was 0.500:1.000, and the volume ratio was 0.26:1:00.

In formulation (iv), the mass ratio bentonite:calcium hydroxide was 0.375:0.125, and the volume ratio was 0.19:0.056.

2.2. Coagulation evaluation experiments

Coagulation evaluation is conducted using a small-scale experiment and a medium-scale experiment. The medium-scale testing involved an area of 2700 cm² for the surface of the liquid (water with oil), whereas the small-scale testing involved a corresponding area of 54 cm². The small-scale method was used for initial evaluation of all the formulations, with three tests performed for each formulation. Subsequently, the medium-scale method was used for further evaluation of formulation (ii), which was found to be the most effective by small-scale testing in terms of performance and cost. The medium-scale method involved only one test.

2.2.1. Small-scale experiments

In 250 cm² of tap water (pH = 7) placed in a 500 ml glass beaker with internal diameter 83 mm, 5 g of oil (5.7 cm³, i.e., 2.2 vol.%) were added. The liquid mixture was stirred with a magnetic stirrer at 70 rpm for 30 min and then it was allowed to stand for 1 min, whereby the oil droplets floated on the water surface. Then the various coagulant formulations (bentonite, organobentonite, calcium hydroxide and/or sawdust, as applicable), formed by manual mixing, was sprinkled gradually on the surface of the liquid in the beaker by a plastic spoon. Then over a period of 75 min, the oil was allowed to coagulate and form oil–solid aggregates.

In the formulations involving sawdust, the aggregates floated on water whereas in the formulation without sawdust, the aggregates sank. The floated aggregates on water were collected with a spoon.
Aggregates that sank in water were collected from the bottom of the beaker with a spoon after decantation of the supernatant water. All the aggregates that had been removed manually from the beaker were filtered for 45 min using previously weighed filter paper (Whatman Grade No. 2). After filtration, the aggregates and the filter paper were dried in a vacuum drying oven at 110 °C for 30 min. Subsequently they were allowed to cool at room temperature and equilibrate with air for 24 h and finally they were weighed.

The liquid was poured out of the beakers and the oil which was retained in the inner surface of the beaker and the surface of the spoon was cleaned with filter paper. After vacuum drying at 110 °C, the soaked filter paper was weighed and the weight of the retained oil was recorded and was subtracted from the original oil weight. The coagulation efficiency \( E \) refers to the fraction of oil that is removed by coagulation and was calculated from the equation

\[
E = 1 - \left( \frac{5.00 \times R + B + S + F}{G} \right) / (5.00 - R),
\]

where \( R \) is the weight of the retained oil, \( B \) is the bentonite (or organobentonite) weight, \( C \) is the \( \text{Ca(OH)}_2 \) weight, \( S \) is the sawdust weight, \( F \) is the filter paper weight, and \( G \) is the weight of the coagulated material together with the filter paper after drying.

### 2.2.2. Medium-scale experiments

The medium-scale experiments involved a relatively large area over which oil coagulation occurred. The method used is basically the same as in the small-scale experiments except for the container area and the amounts of materials.

Only formulation (ii) was used. A rectangular black plastic container 600 × 450 × 150 mm was used. The materials used were water (10 kg), oil (150 g), bentonite (11.25 g), calcium hydroxide (3.75 g) and sawdust (30 g). Relative to the mass of the oil, the proportions (by mass) of bentonite, calcium hydroxide and sawdust were the same as in the small-scale experiments (i.e., bentonite/oil ratio = 0.075, calcium hydroxide/oil ratio = 0.025, and sawdust/oil ratio = 0.2). The oil/water mass ratio was slightly lower in the medium-scale experiments compared to the small-scale experiments (0.015 and 0.02 respectively). The concentration of oil in the liquid was 1.7 vol.% in the medium-scale experiments and 2.2 vol.% in the small-scale experiments.

The filter paper (3.086 g) was a stack of three coffee filters (CVS, basket style) with basal diameter 82.5 mm made from paper. In contrast to the small-scale experiments, the mixture of oil and water was stirred manually with a plastic spoon for 30 min and the coagulation time was 24 h instead of 75 min.

### 2.3. Microscopic examination of the coagulated material

The coagulated material corresponding to each of the four formulations was examined by scanning electron microscopy (SEM) coupled with elemental analysis by X-ray spectroscopy (EDS), using a Hitachi S270 system. The purpose was to study the microstructure of each type of aggregate. Prior to SEM work, the specimens were carbon coated.

Unless stated otherwise, the coagulated material examined was the oil-containing aggregate after drying to remove the water retained after filtration (Section 2.2.1). Moreover, the coagulated material was also examined after drying and subsequent exposure to room temperature and vacuum drying. After drying of the oil, the previously sticky and agglomerated aggregates were disaggregated.

### 2.4. X-ray diffraction analysis

Powder X-ray diffraction (XRD) of the original bentonite and the coagulated material (formulation (ii)) was conducted with CuKα radiation (40 kV, 30 mA) using a Siemens Kristalloflex diffractometer equipped with a diffracted-beam graphite monochromator.

### 2.5. Evaluation of oil removal from coagulated material by compression

The extent of oil removal from the coagulated material by compression was evaluated. The coagulated material was placed between two pre-weighed paper towels. The coagulated material and the filter papers ("sandwich") was weighed and the mass of the coagulated material used in the evaluation was obtained. Then the sandwich was compressed for 15 min perpendicular to its main area with a force of 400 lb, which corresponded to a pressure of 3.3 ± 2.1 MPa. The variation in pressure resulted from the variation in the area of the tested material. During compression part of the oil was expelled from the coagulated material and was soaked by the paper towel. Subsequently, the coagulated material was removed from the paper towels and the latter were then weighed.

The difference between the mass of the paper towels before and after oil soaking yielded the mass of the oil that was removed from the coagulated material during this compression process. The mass of the removed oil divided by the mass of the coagulated material prior to compression yielded the mass fraction of oil removed from the coagulated material. This mass fraction divided by the mass fraction of oil in the coagulated material prior to compression (as determined in the coagulation evaluation described in Section 2.2.1) gave the fraction of oil in the coagulated material that was removed by compression. The test was conducted in triplicate corresponding to formulation (ii).

### 3. Results and discussion

#### 3.1. Small-scale experiments

For all four formulations, solid aggregates were used, which are defined as mixed oil and solid materials of variable morphology which depended on whether sawdust was present or not.

For all three formulations involving sawdust, the aggregates floated on the water, with the coagulated material occurring as a single sheet-like aggregate of diameter limited by the diameter of the beaker and thickness ca. 5 mm (Fig. 1). This size was much larger than that of conventional OMA in the form of solid aggregates (0.01 mm after Stoffyn-Egli and Lee, 2002). The sheet-like floating aggregates differed from conventional OMA in that they contained sawdust at much higher concentration than the bentonite content. Although sawdust sank in water, the aggregates with bentonite floated on water because the sawdust functioned as a fibrous framework for the attachment of the coagulated oil and bentonite and the oil had lower density than water. Thus, the sawdust facilitated the formation and subsequent removal of large sheet-like aggregates.

In formulation (iv), which did not contain sawdust, the aggregates sank in the water and formed agglomerates with particle size of the order of 1 mm. Although the particles were interconnected to a limited degree, they did not form a complete sheet.

Although the proportion of bentonite was much lower than that of the sawdust, bentonite was the main coagulant. In a simple experiment in which bentonite was mixed with either oil or water to form a clay ball, we found that bentonite adsorbed both oil and water. The affinity of bentonite for oil assisted bentonite to serve as a coagulant. In addition, bentonite acted as emulsifier, with the opposite charges on the oil droplets and montmorillonite causing attraction.

The oil–sawdust–bentonite aggregates obtained in this work for the first time are valuable not only for the separation of oil from water, but are also valuable as a handleable fuel. This is because both oil and sawdust in an aggregate are fuels and they constitute the majority of the aggregates. For example, in case of formulation (ii), the aggregates contained 75.6 wt.% oil, 16.3 wt.% sawdust, 6.1 wt.% bentonite and 2.0 wt.% calcium hydroxide, i.e., 81.5 vol.% oil, 14.7 vol.% sawdust, 2.9 vol.% bentonite and 0.9 vol.% calcium hydroxide. The large size of

#### 2.9 vol.% bentonite and 0.9 vol.% calcium hydroxide. The large size of
these aggregates and the mechanical integrity provided by the sawdust, which acts as a reinforcement, make it convenient to use these aggregates as a fuel. In addition, the aggregates may be squeezed to remove the oil from the aggregates, thus providing liquid oil fuel.

Table 1 shows the coagulation results for formulations (i), (ii), (iii) and (iv). Formulation (i) gave the highest coagulation efficiency. Comparison of the coagulation efficiency for formulations (i) and (ii) showed that organobentonite was more effective than unmodified bentonite. Formulation (ii) was more effective than formulation (iii), indicating that Ca(OH)₂ enhanced the coagulation efficiency in the presence of bentonite and sawdust. Formulation (iii) was much more effective than formulation (iv), indicating that sawdust was much more important than Ca(OH)₂ in enhancing the coagulation efficiency in the presence of bentonite.

Although formulation (i) is the most effective among the four formulations studied, it is expensive, due to the presence of organobentonite. Thus, formulation (ii) is more attractive when both cost and performance are taken into consideration.

The four formulations involve either organobentonite or unmodified bentonite as the base coagulant, which functions as coagulant, adsorbent and emulsifier (Gianotti et al., 2008; Giese and van Oss, 2002). Due to the strong thixotropic behavior of bentonite or organobentonite, Ca(OH)₂ (a coagulation aid) and/or sawdust (an agent that provides framework and buoyancy and serves as an oil adsorbent) is used to reduce the thixotropy. Calcium hydroxide strengthens the coagulating function of bentonite, as it provides Ca²⁺ ions, which strongly adsorb on both bentonite and oil drops (Liu et al., 2004). The sawdust may be substituted by stalk powder, paper powder or other natural, manmade fiber powder.
3.2. Medium-scale experiments

The coagulated material formed a sheet with size restricted by the container walls (Fig. 2). Thus, the upper limit of the aggregate size probably exceeded 600 mm. In fact, probably there was no upper limit, provided that the coagulant mixture was uniformly applied to the surface of the liquid containing the oil, because the sawdust provides a continuous framework to the resulting aggregate sheet. The aggregate after drying weighed 186.908 g. The retained oil weighed 2.901 g. Hence, according to Eq. (1), the coagulation efficiency was 0.924, i.e., slightly lower than the corresponding value of 0.942 obtained in small-scale experiments. The fraction of oil that was retained was 1.9%, compared to the corresponding value of 4.4% for small-scale testing. This difference is attributed to the greater difficulty in recovering all of the retained oil in the medium-scale experiments. The accuracy of the coagulation efficiency was thus lower for the medium-scale experiments than the small-scale experiments.

3.3. Microscopic examination

3.3.1. Aggregates containing oil

Figs. 3–6 show SEM photographs of the aggregates obtained by using formulations (i), (ii), (iii) and (iv) respectively. In spite of the presence of sawdust, fibrous textures are not present in Figs. 3–5, suggesting that the sawdust and the bentonite formed a rather uniform solid aggregate. Figs. 3 and 4 show particles that were essentially encased in a continuous oil–bentonite–sawdust matrix. In contrast, in Fig. 5 individual particles were not observed, partly due to the absence of calcium hydroxide, but the oil–bentonite–sawdust matrix was continuous. In the absence of sawdust, the microstructure was less smooth, with solid particles (about 5–10 μm in size) that protruded partly from an oil–bentonite matrix (Fig. 6). The particles in Figs. 3 and 4 appeared smaller, because they were essentially embedded in the matrix.
3.3.2. Aggregates with the oil dried

The aggregates with the dried oil were examined for formulations (ii) and (iv). Formulation (ii) contained sawdust, whereas formulation (iv) did not.

Although a continuous span of solid was observed for the oil-containing aggregates corresponding to formulation (ii) and the sawdust essentially was not discerned (Fig. 4), the sawdust skeleton and the particles clung to it were observed after drying the oil (Figs. 7 and 8). The relatively large particles, which resembled to patches of particle agglomerates, are attributed mainly to bentonite, as shown by the EDS spectra (Fig. 7). The relatively small particles were rich in calcium, so they are attributed to calcium hydroxide (Fig. 8).

For formulation (iv), the sawdust skeleton was absent, as expected. However, particles were observed (Fig. 9a). The particles protruded more clearly compared to the materials prior to drying the oil (Fig. 6). They belong to bentonite and calcium hydroxide, as shown by the EDS spectra (Fig. 9b).

3.4. X-ray diffraction determination of the bentonite interlayer distance

Fig. 10 shows X-ray diffraction patterns of bentonite in the as-received state and that in the oil-coagulated state. The montmorillonite basal spacing ($d_{001}$) increased from 12.0 Å in the as-received state to 14.4 Å in the coagulated material (with sawdust and calcium hydroxide, formulation (ii)). These values are consistent with those previously reported for untreated bentonite and organic-treated bentonites respectively (Gitipour et al., 1997). This suggests that oil is intercalated between the clay layers in bentonite in the coagulated state. However, it is also possible that the increase in $d_{001}$ is due to an increase in the moisture content. Furthermore, it is possible that the increase in $d_{001}$ is due to the Ca from Ca(OH)$_2$ and the tap water exchanging Na from the montmorillonite, as $d_{001}$ of Ca-montmorillonite is 15 Å (Ferrage et al., 2005). At any rate, the consequence is a volume increase of 20% for the bentonite. As mentioned previously in relation to formulation (ii), the aggregate contained 81.5 vol.% oil, 14.7 vol.% sawdust, 2.9 vol.% bentonite and 0.9 vol.% calcium hydroxide. If intercalation of oil had occurred in the aggregate, the observed 20% increase in the bentonite volume would be due to oil and the aggregate composition would be 80.9 vol.% free oil, 14.7 vol.% sawdust, 3.5 vol.% oil-intercalated bentonite and 0.9 vol.% calcium hydroxide. The vast majority of the oil in the coagulated material was not intercalated in the montmorillonite interlayer space.

Fig. 10a also shows the presence of a small proportion of hydrated bentonite in the as-received bentonite. The impurity peak is probably due to mica (Tettenhorst and Corbato, 1993). Fig. 10b shows the presence of an intense and broad hump, which is probably due to turbostratically disordered Ca-montmorillonite (Lutterotti et al., 2010). Superposed on this hump are a peak attributed to Ca(OH)$_2$ (Rao et al., 2009) and a peak attributed to montmorillonite (Gitipour et al., 1997).

3.5. Removal of oil from coagulated material by compression

Based on the masses of the ingredients, the mass fraction of oil in the coagulated material was 0.756. The results of three tests involving formulation (ii) show that the removed oil amounted to 55 ± 4% of the aggregate mass prior to compression. This corresponded to 73 ± 5% of
the oil in the aggregate prior to compression. The scatter of the data was mainly due to the variation in the specimen area.

4. Conclusion

The use of a mixture of bentonite and sawdust, with sawdust being the vast majority, is highly effective and cost-efficient for the coagulation of oil in water, with coagulation efficiency greater than 92%. A minor amount of calcium hydroxide may be optionally added to the mixture to increase the coagulation efficiency in excess of 94%. Sawdust (79.6 vol%), unmodified bentonite (15.8 vol%) and calcium hydroxide (4.7 vol%) used as a mixture gave coagulation efficiency 94% for an oil concentration of 2.2 vol.% in water. This formulation is recommended for use in the cleaning up of oil spills.

Sawdust by itself sank in water. However, the coagulated aggregates floated on water when sawdust was used with the bentonite. Large sheet-like aggregates formed, with the oil-bentonite–sawdust serving as a continuous matrix. The upper limit of the aggregate size exceeded 600 mm and was facilitated by sawdust. The aggregates contained 81 vol.% oil, 15 vol.% sawdust, 3 vol.% unmodified bentonite (with interlayer distance 14.4 Å) and 1 vol.% calcium hydroxide. Upon compression, 73% of the oil in the aggregate was removed. In contrast, previously reported OMAs were much smaller and did not contain sawdust. The sawdust-free aggregates had small size and were not smooth in morphology; they sank in water and had low coagulation efficiency (37%). The sawdust functioned as a fibrous framework for the adsorption of oil droplets and bentonite particles, thus facilitating the formation and the subsequent removal of large aggregates.

In the presence of unmodified bentonite, sawdust enhanced the coagulation efficiency significantly, while calcium hydroxide enhanced the coagulation efficiency to a considerably lower degree. The use of organobentonite (with dimethyl benzyl hydrogenated tallow as modifier) instead of unmodified bentonite in the formulation with calcium hydroxide and sawdust slightly increased the coagulation efficiency to 95%. However, organobentonite is much more expensive than unmodified bentonite.

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