



# A model for predicting solid particle behavior in petroleum sludge during centrifugation



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## HIGHLIGHTS

- Developed a model to predict solid transference in centrifuged petroleum sludge.
- The model classified solid particles into three groups during centrifuging.
- Experiment results multistage centrifugation agreed well with model predictions.

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## ABSTRACT

An efficient model is proposed for predicting the behavior of solid particles in petroleum sludge during centrifugation, from the particle size distribution and petroleum viscosity. Petroleum sewage sludge (PSS) and oil-tank bottom sludge (OBS), respectively collected from a sewage reservoir containing petroleum waste water and a petroleum oil storage tank, were studied. The solid particle removal and oil recovery rates were measured to assess the efficiency of the model. The model predicted that solid particles could be separated into three groups during centrifugation, with critical separation sizes of 69.2 and 80.3  $\mu\text{m}$  for the PSS and 91.2 and 110.3  $\mu\text{m}$  for the OBS, and that particles larger than the smaller critical separation size could be removed from each sludge efficiently at room temperature without pretreatment. The predicted cumulative particle removal rates using a three-stage centrifugation process were 50%, 61%, and 70%, for the three PSS-particle size ranges (in ascending order), which agreed well with experimental measurements (54%, 65%, and 75%, respectively). The experimental water/oil emulsion recovery rates were 86%, 81%, and 75%, respectively. Preheating enhanced the solid particle removal rate by 29% for PSS whose viscosity decreasing quickly with increasing temperature. Adding a solvent increased the solid particle removal rates to 90% for the PSS and 82% for the OBS, but caused the water/oil emulsion recovery rates to decrease to 69% for the PSS and 67% for the OBS. The model and experimental results suggest that the optimum centrifugation time should be determined after assessing the petroleum viscosity and the solid particle size distribution.

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## 1. Introduction

Rapid economic growth has made China the second largest energy and raw material consumer in the world, and China generates extremely large amounts of waste, including petroleum sludge, every year. The terms “petroleum sludge” and “oily sludge” refer to two types of waste, waste generated by refinery plants and waste products from crude oil transportation facilities and storage tanks, respectively [1]. It is estimated that more than 6 million tons of petroleum sludge have been produced each year since 2010 in China [2]. Petroleum sludge is recognized as a hazardous waste because of its high concentrations of various toxic and dangerous components, which include bacteria, heavy metals, and organic

pollutants such as polycyclic aromatic hydrocarbons [3,4]. However, petroleum sludge can also be regarded as a resource because it contains valuable hydrocarbons that can be recovered and re-used as fuel or in chemical products [5].

Petroleum sludge is often a complex mixture of water/oil emulsion and solid particles, typically containing approximately 30% oil, 40% water, and 30% solids, by mass. A number of methods have been developed to recover hydrocarbons from petroleum sludge, including catalytic thermal cracking, microbial treatment, chemical extraction, and mechanical centrifugation [6]. Catalytic cracking is widely used, but it is costly and requires complex facilities, the reaction conditions need to be precisely controlled to maintain safety, and the used catalysts may cause secondary pollution [7]. Microbial treatment is an environmentally friendly solution but gives a very low energy recovery rate [8]. Chemical extraction allows the hydrocarbons to be recovered efficiently but consumes

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large amounts of chemical reagents and water resources [9]. Microwave treatment has recently been used to extract bitumen, upgrading the heavy oils and decreasing the viscosity, but it has not been used commercially because of its uncertain potential and high setup costs [10].

Centrifugation provides a basic mechanical separation approach for removing solids from petroleum sludge, and it has advantages in its simplicity, low industrial implementation costs, and low environmental impact [11]. Centrifuging petroleum sludge separates it into an upper and a lower layer, which are predominantly a water/oil emulsion and the solid particles, respectively. Hydrocarbon liquids can be recovered from the upper layer but the lower layer has to be subjected to further treatment. The solid particles in the lower layer mainly comprise silicon sand [12,13] and other minerals, iron fragments, and heavy metal species, making petroleum sludge unsuitable for bio-treatment [14].

According to the principles of sedimentation, during centrifugation, a particle will be dragged, by centrifugal force, in the radial direction through the sludge, toward the bottom of the container. The distance that the particle moves is defined as the settling distance, and this depends on the viscosity of the water/oil emulsion, the centrifugation speed, and the particle size. The longer the settling distance is, the more easily the particle can be separated. Petroleum sludge usually contains solid particles ranging in size from several micrometers to millimeters [15]. Small particles may be retained in the upper layer after centrifugation, having a negative effect on the quality of the recovered hydrocarbon resources. Particle transfer behavior needs to be studied so that we can understand how to increase the particle settling distance during centrifugation, to allow the particle removal rate to be improved and a purer upper layer to be obtained. However there has been only a very limited amount of research on this topic [16].

As a mixture of water/oil emulsion and particles, the viscosity of the emulsion will affect the settling distance significantly. Kanti et al. [17] developed a dynamic liquid viscosity law for predicting the viscosity of crude oil, and Werner et al. [18] further developed that theory, proposing a new viscosity model based on the mass fractions of the “SARA” (saturates, aromatics, resins, and asphalt- enes) components present. Ukwuoma and Ademodi [19] measured the apparent viscosities of Nigerian oil sand at different temperatures, showing that the viscosity decreased with temperature. Kendall and Monroe [20] proposed a mixing rule for Bingham fluid that also proved to be applicable to heavy oil. A number of mixing rules for use in crude petroleum oil viscosity calculations have been reported, and Centeno et al. [21] tested the applicability of seventeen mixing rules to four crude oils with different American Petroleum Institute (API) gravity values, showing that most of the rules were unsuitable for a high viscosity petroleum sludge with a low API gravity value.

In the study presented here, a new model was developed for predicting the behavior of solid particles during the centrifugation of petroleum sludge, based on the particle densities and size distributions and the fluid viscosity based on the SARA components ratio. The settling of solid particles during the centrifugation of two types of petroleum oil sludge (from a petroleum storage tank and a petroleum wastewater reservoir) was studied. Experiments were conducted to evaluate the validity of the model predictions and to assess the effects of pretreating the sludge and using multistage centrifugation on the removal of solid particles and the water/oil emulsion recovery rate.

## 2. Solid particle transference model

To predict the transference of solid particles, the viscosity of the water/oil emulsion must first be obtained. The viscosity of the petro-

leum sludge can be measured using a rheometer, but this parameter cannot be directly used to predict the transference because the particle properties will significantly affect the results. The viscosity of the water/oil mixture in the sludge mainly depends on the SARA components, the viscosities of which are proportional to their API gravity values. In this paper, the SARA components are classified as light or heavy. The light components, which include saturated compounds and aromatics, have API gravity values of between 10° and 70°, and the heavy components (including resins and asphalt- enes) have API gravity values of below 10°. We used the two phase mixing rule developed by Kendall and Monroe [20]:

$$\mu = x_A \mu_A^{1/3} + x_B \mu_B^{1/3}, \quad (1)$$

where  $\mu$  is the viscosity of the mixture,  $\mu_A$  and  $\mu_B$  are the viscosities of the light and heavy components in the mixture, respectively, and  $x_A$  and  $x_B$  are the corresponding mass ratios.

The sedimentation velocity of particles in the water/oil emul- sion during centrifugation can be described by Stokes' Law:

$$v_c = \frac{d_e^2 (\rho - \rho_0) g}{18\mu} \frac{\omega^2 R}{g}, \quad (2)$$

where  $v_c$  is the sedimentation velocity,  $d_e$  is the solid particle diam- eter equivalent,  $\rho$  and  $\rho_0$  are the densities of the solids and the water/oil emulsion, respectively,  $\omega$  is the centrifuge rotation speed, and  $R$  is the radius of the centrifuge.

The sedimentation velocity of a particle during centrifugation will be affected by other particles in its vicinity, and a correction factor,  $\eta_\phi$ , is introduced, as suggested by Hawksley [22], to take the mutual interference between particles into account:

$$v'_c = \eta_\phi \cdot v_c \quad (3)$$

and

$$\eta_\phi = (1 - \phi)^2 \exp \left( -\frac{5.53\phi}{1.64 - \phi} \right), \quad (4)$$

where  $v'_c$  is the modified sedimentation velocity taking mutual interference between particles into account and  $\phi$  is the volume fraction of the particles.

For particles with the same density, the larger the particle is, the longer the settling distance will be. Solid particles of different sizes will move to different positions along the radial direction of the container according to their settling distances. After centrifugation, large particles will have aggregated in the bottom of the container, and the sludge can be separated into upper and lower layers. Fig. 1 shows a schematic diagram of the centrifugation process, with the dividing line between the upper and lower layers marked. Using Eq. (3), the settling distance,  $l$ , for a particle, after being centrifuged for  $t$  seconds, can be calculated using the equation:

$$l = v'_c \cdot t = \eta_\phi \frac{d_l^2 (\rho - \rho_0) g}{18\mu} \frac{\omega^2 R}{g} \cdot t \quad \begin{array}{ll} l < L_{upper} & \text{group 1} \\ L_{lower} > l > L_{upper} & \text{group 2,} \\ l > L_{lower} & \text{group 3} \end{array} \quad (5)$$

where  $L_{upper}$  and  $L_{lower}$  are the dividing lines for the settling dis- tances for the upper and lower layers, respectively. The dividing line between the upper and lower layers was approximately 3 mm from the container bottom for the petroleum sludge used in this study, i.e.,  $L_{upper} = 7$  mm and  $L_{lower} = 10$  mm. If the centrifugation time  $t$  and dividing line position  $l_d$  are known, a critical size  $d_l$  can be defined for determining whether a particle can move to the lower layer during centrifugation:

$$d_l = \sqrt{\frac{18\mu \cdot L_{upper}}{\eta_\phi \omega^2 R (\rho - \rho_0) t}} \quad (6)$$

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