



Research paper

Can hydrocarbon contamination influence clay soil grain size composition?

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ABSTRACT

Microstructural (fabric, forces and composition) differences due to hydrocarbon contamination in a clayey soil (glacial till) were studied using scanning electron microscopy (microfabric analysis) and sedimentation bench test (PSD). Uncontaminated and contaminated glacial till from NE Poland (in the area of a fuel terminal) were used for the study. Because the contaminated samples exhibited lower electric charge, the electrostatic repulsive forces in the contaminated samples were much lower than in uncontaminated samples. All sedimentary-based particle size test results gave similar results and were inconclusive to find difference between contaminated and uncontaminated samples. Mastersizer PSD data show smaller aggregates in contaminated in comparison to the uncontaminated sample. The present test's finding may support earlier microstructure investigation results, where diesel-contaminated clay soil shows more open porosity, consisting consequently of looser aggregates, which were easier dispersed in water during Mastersizer measurements than the more coherent uncontaminated clay sample. This in effect increased the percentage of finer than 0.1 mm grain fraction within the broad diameter range in contaminated samples results, in comparison with the uncontaminated sample results.

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1. Introduction

Particle space arrangement within a soil, known as a texture, is a very important factor that determines its physicochemical properties. These properties play a major role in a range of technologies and manipulation of colloidal stability as well as in engineering projects dealing with main slurry dewatering, dams, footings and slope stability. Despite the fact that many microstructural studies have been carried out around the world, this subject is still poorly understood. One of the key structural elements in soil study is grain size composition, known also as particle size distribution (PSD), which holds very important information that characterises soil and its basic physical properties. PSD depends on many factors like soil mineral composition, chemistry, content of salt and moisture, exchangeable cation composition and perhaps much more. The complex dependencies between PSD and many physicochemical environmental states make this parameter the fingerprint-like characterisation tool, enabling the prediction of major physical properties and comparison between different samples for engineering evaluation.

Similar to the soil texture, simple and cheap PSD examinations may encode more complex information about tested samples. Such information may enable researchers to bypass numerous sophisticated and expensive investigations by monitoring soil system. An example of this

is to detect hydrocarbon-contaminated soil and monitoring the effectiveness of cleaning processes.

Contamination of a soil by petroleum substances, characterised by physicochemical properties different from water, was found to have a deteriorating effect on its physical, mechanical and filtration parameters (Bowders and Daniel, 1987; Fernandez and Quigley, 1988; Izdebska-Mucha, 2005; Khamehchiyan et al., 2007; Korzeniowska-Rejmer and Izdebska-Mucha, 2006; Singh et al., 2009; Uppot and Stephenson, 1989). Hydrocarbon pollution is common, especially in heavily industrial places where careless operators do not particularly look after the environment. Such places include fuel depots and army polygons. In work by Izdebska-Mucha et al. (2011), a microstructural examination of clean and diesel oil-contaminated clay-rich soil was conducted. Pollution was present at former Soviet army polygon sites, and investigations were needed before the proper cleaning method was chosen. The investigation found that uncontaminated soil has a comparatively medium packed matrix type of microstructure dominated by face-to-face (FF) and edge-to-edge (EE) contacts with small, elongated voids between compact aggregates. By contrast, in the contaminated soil, the clayey matrix is more loosely packed, and disintegration of natural aggregates was observed. These structural results may suggest that compact aggregates in an uncontaminated soil sample may be richer in highly dispersed clay particles than in contaminated ones. This hypothetical prediction was tested by performing simple PSD investigations. For this purpose, our present investigation focuses on PSD measurements for the same samples

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that were investigated in Izdebska-Mucha et al. to validate previously obtained microstructural results. Different grain size distribution methods were validated to find out if any of them may explain previously described microstructural changes.

2. Materials and methods

The natural clayey soil used for this study was a glacial till from NE Poland from a 2 m deep exploration pit located in an area unaffected by pollution; the other sample came from a contaminated area. Samples were collected from two glacial till horizons that have been found to occur in the vertical section of these artificial exposures. These tills were formed during the upper stadia of the Wisła Glaciation.

The soil samples were collected from the area of a fuel terminal where an accidental spillage of diesel oil occurred from underground fuel tanks. In the area where down flow and stagnation of diesel oil took place, petroleum-derived substances migrated upwards in the soil, causing pollution up to the groundwater level. The hydrocarbon content at a depth of 2 m was 5395 (mg/kg dry weight), which was the sum of shorter chain C₆-C₁₂ (1457 mg/kg dry weight) and longer chain >C₁₂ (3938 mg/kg dry weight) hydrocarbons. Soil samples were taken from two pits located, respectively, in contaminated and unaffected areas. The distance between the pits was about 50 m, and the samples were collected ten months after the pollution incident. The near surface glacial till soil samples have been named: GL NS_uncontaminated soil and GL ON_contaminated soil.

Force measurements were conducted using a Nanoscope III, AFM (Digital Instruments) in force mode, utilizing a J scan head. A standard fluid cell with a scan rate of 1 Hz was used for the measurements. The AFM cantilever was triangular, tipless silicon nitride with a spherical colloidal probe (2.5 μm in diameter) purchased from NOVASCAN. The spring constant was a nominal 0.12 N/m.

The electrokinetic potential (zeta potential or ζ) in the clay samples was measured using Zetasizer (NanoSeries), manufactured by Malvern Ltd., United Kingdom. PSD was investigated using three methods. One method was based on the laser diffraction and was done on Malvern 3000 Mastersizer equipment, United Kingdom, and two other, hydrometer and pipette methods which were standard sedimentary methods based on the Stocks law.

3. Results and discussion

3.1. General characterisation

The mineral composition results displayed in the X-Ray Diffraction (XRD) patterns (Fig. 1) show the presence of clay minerals like illite, kaolinite and smectite. Of the non-clay minerals, most abundant were quartz and accessory minerals. Both contaminated and uncontaminated samples were indifferent from XRD results.

Transmission electron microscopy (TEM) results from the uncontaminated sample shown in Fig. 2A display aggregates about 1.5 μm long. This aggregate consists of larger and slightly curved sheets, foiled with extremely dispersed platy in morphology particles of diameters well below 100 nm. These nano-clays look firmly connected to larger clay sheets forming stronger aggregates. Contaminated sample morphology shown in the TEM micrograph (Fig. 2B) displays a lot of clay nanoparticles aggregated, blanketed by organic film envelopes.

Clay platy particles usually carry high electric potential as the zeta potential measurements show values measured for the uncontaminated sample NS in DI water was −31 mV (electric conductivity 0.0021 mS/cm) and increased to −44 mV in 0.01M NaCl (electric conductivity 1.38 mS/cm) solution. Interestingly, the zeta potential values measured for the contaminated (ON) sample varied and show positive value in DI water from +5.94 to +3.5 mV (electric conductivity 0.0035 mS/cm). After ultrasonic treatment in 0.01M NaCl solution, this sample's (ON) zeta potential recovered and reached −38.2 mV (electric conductivity 1.45 mS/cm). The smectite flakes therefore experienced strong electric forces that hold aggregates firmly, but after particle dispersion in aqueous solution, they experienced strong repulsion forces when approaching each other. The contaminated sample showed very low and even positive zeta potential to make electrostatic forces weaker. Therefore, the van der Waals attraction forces prevail and edge-to-face (EF) contacts between smectite thin flakes become favourable in this colloidal system, building within aqueous environments weak coagulated aggregates and looser, easily dispersive aggregates in the sample dry state.

Differences in physical properties can be illustrated in SEM micrographs in Fig. 3. For this demonstration, samples were dispersed in DI water, then deposited onto the hydrophilic surface of silicon wafer and dried in air. Significant differences can clearly be seen: the uncontaminated sample (NS) forms a uniform film consisting of submicron

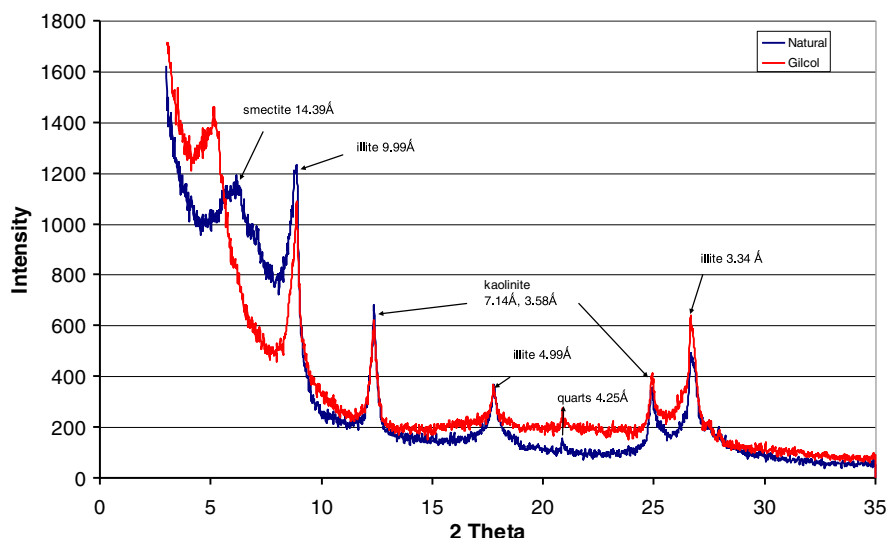


Fig. 1. XRD pattern from oriented clay fraction natural and ethylene glycol treated sample shows presence of kaolinite, illite and smectite.

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