Understanding the effects of high temperature processes on the engineering properties of soils

Comprendre les effets des procédés a haute température sur les propriétés des sols

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ABSTRACT: High temperature processes such as in situ smouldering and thermal remediation techniques can achieve rapid removal of organic contaminants from soils in much shorter time periods than traditional remediation technologies. Thermal remediation processes use heat or heated water to volatilise the contaminant within the soil to enable its extraction. High temperatures affect the particle size distribution, mass loss, mineralogy and permeability of the soil. In sandy soils, the particle size decreases with increasing temperature due to a mobilisation of fines, which is likely due to the bond of fines to the sand grains being affected by temperature. In clayey soils, the overall particle size increases with increasing temperature due to aggregation and cementation of the clay fraction. Permeability seems to be affected by treatment type rather than temperature alone, comparing heat treated and smouldered samples showed an increase of sand permeability by approximately two magnitudes. This study illustrates the effects of high temperature and smouldering processes on soil characteristics and dynamic behaviour. Monitoring during and after aggressive remediation is advisable so that rehabilitation measures can be implemented before site redevelopment.

RÉSUMÉ : Des procédés a haute température tels que la combustion lente in situ et des techniques de traitement thermique peuvent achever une élimination rapide des contaminants organiques des sols en beaucoup moins de temps que les technologies de traitement traditionnelles. Les procédés de traitement thermique utilisent la chaleur ou de l'eau chauffée pour vaporiser les contaminants dans le sol pour permettre leur extraction. Des températures élevées affectent la distribution granulométrique, la perte de masse, la minéralogie et la perméabilité du sol. Dans les sols sablonneux, la taille des particules décroît avec l'augmentation de température due à une mobilisation des particules les plus fines, probablement dû à la liaison de ces particules aux grains de sable, affectée par la température. Dans les sols argileux, la taille des particules augmente avec l'augmentation de température due à l'agrégation et la cimentation de la fraction argileuse. La perméabilité semble être affectée par le type de traitement plutôt que par la température uniquement, des échantillons traités par la chaleur ont montré une augmentation de la perméabilité du sable d'environ deux ordres de grandeur par rapport à ceux traités par combustion lente.Cette étude montre les effets des températures élevées et des procédés de combustion lente sur les caractéristiques du sol et sur son comportement dynamique. Il est conseillé d'utiliser un système de surveillance pendant et après traitement agressif afin que les mesures de réhabilitation puissent être appliquées avant le réaménagement du site.

KEYWORDS: Thermal behaviour of soils, smouldering remediation, high temperature

1. INTRODUCTION

Soils can be exposed to elevated temperatures naturally through wild, forest or peat fires or through thermal remediation processes designed to mitigate contamination by hazardous organic chemicals. Most research on soil properties and their heat dependency is based on forest fires and therefore concentrates on erosion rates, ground stability and nutrients affected by fire severity. The effects of exposure to temperatures up to 500°C have been studied widely (Are et al., 2009; Certini, 2005; Rein, 2009; Rein et al., 2008). Literature published on heat treatments of clay evaluates the effects of temperatures up to 1000°C (Tan et al., 2004). Exposures of 200 – 850°C have been observed in soils during wildfires (Certini, 2005; DeBano, 2000; Mataix-Solera and Doerr, 2004; Rein et al., 2008). Moderate (300-400°C) and high (>450°C) temperature processes, such as hot water extraction, thermal desorption, soil heated vapour extraction, incineration or smouldering are used widely to treat contaminated soils (Araruna Jr et al., 2004; Chang and Yen, 2006; Gan et al., 2009; Kronholm et al., 2002; Lee et al., 2008; McGowan et al., 1996; Pironi et al., 2011; Pironi et al., 2009; Switzer et al., 2009; Webb and Phelan, 1997). Most research on soil remediation techniques focuses on the remediation result and less on the effects the process has on the soil properties itself. In some cases, the effects on soil properties may be a criterion for selection of the remediation technique (Chang and Yen, 2006; Pironi et al., 2011) or the soil properties may influence the results (Webb and Phelan, 1997). There is little research on the effects of thermal remediation processes on soil properties (Araruna Jr et al., 2004; Pironi et al., 2009). Based on the observations of soil erosion and subsidence after wildfires, further understanding of the effects of high temperature remediation processes must be developed.

The maximum temperatures observed in contaminant remediation vary by the process that is used. With the exception of smouldering remediation, all of these remediation techniques use heat or heated water to volatilise the contaminant within the soil to enable its extraction. Maximum temperatures for these technologies are typically adjacent to the heat source with more moderate target temperatures of 80-100^{o}C achieved within the wider treatment zone. The contaminant must be collected and treated (Chang and Yen, 2006; Gan et al., 2009; Kronholm et al., 2002; Lee et al., 2008; McGowan et al., 1996; Webb and Phelan, 1997). These processes maintain high temperatures in the soil for weeks to months or longer. In contrast, smouldering remediation uses the contaminant itself as fuel for the combustion reaction (Pironi et al., 2011; Pironi et al., 2009; Switzer et al., 2009). In laboratory studies, the soil particles are exposed to high temperatures on the order of 1000°C for coal tars and 600-800°C for oils for up to 60 minutes. Field scale efforts may result in exposure durations on the order of hours or longer.

Elevated temperatures have been shown to alter the mineralogical composition of soil. These effects have been studied extensively in relation to the effects of wildfires on soil properties. Colour change in soils has been observed after wildfire and after smouldering remediation. In most cases it changes from yellowish brown to reddish brown. This is due to the oxidation of soil iron content from goethite to maghemite or hematite (Goforth et al., 2005; Ketterings and Bigham, 2000). Decomposition of soil particles, especially clay minerals, starts at temperatures above 550°C (Certini, 2005). These temperatures are rarely reported for wild and forest fire, but temperatures up to 1200°C can be achieved during smouldering remediation (Pironi et al., 2009; Switzer et al., 2009).

This study aims to characterise the effects of moderate and high temperatures as well as smouldering on soil properties to determine the impact changes will have to the soil and predict possible complications that may arise during or after remediation treatment. Silica sand and kaolin clay are used as constituents of a synthesised simple soil. Clean untreated, heat-treated and contaminated/smouldered materials are evaluated to determine the impacts of the treatment conditions on soil properties.

2. MATERIALS AND METHODS

Coarse silica sand (Leighton Buzzard 8/16, Sibelco, Sandbach, UK) and kaolin clay (Whitchem Ltd, UK) were used as the base soil for all of the experiments. The sand contains 99% silicon-dioxide, has a mean grain size of 1.34 and a bulk density of 1.7g/cm^3 (Switzer et al., 2009). The sand and clay were accepted as received and the sand was subjected to the same pre-treatment. A programmable muffle furnace (Nabertherm L9/11/SKM, Nabertherm GmbH, Lilienthal, Germany) was used for all heating experiments. The sands evaluated after smouldering remediation were prepared in a 3m^3 experiment involving coal tar mixed with coarse sand. The initial concentration of this mixture was 31000 ± 14000 mg/kg total extractable petroleum hydrocarbons before treatment and the average concentration after smouldering remediation across the majority of the vessel was 10 ± 4 mg/kg (Pironi et al., 2009).

Table 1. Heat treatment programs

Sample Name	Pre-heating time (min)	Peak temperature for 60min	cooling down time (min)
Untreated			
105	30	105°C (24h)	0
250	30	250°C	0
500	30	500°C	~ 60
750	60	750°C	~ 180
1000	60	1000°C	~ 240

2.1 Sample Preparation and Heat Treatment

The silica sand was washed and wet sieved using a 425µm screen to eliminate any loose fines and air dried for several days. In case of mixed samples the dried silica sand was mixed with 10% mass kaolin clay and 5% moisture content before being heat treated. For each test, the required amount of samples was heated in the furnace following the heat treatment programmes listed in Table 1. After the required exposure duration, the samples were removed from the muffle furnace and placed in a desiccator to cool. Samples heated to temperatures above 500°C were allowed to cool in the furnace to 200°C before transfer to the desiccator.

2.2 Laboratory Testing

Particle density was measured using the gas-jar method suitable for coarse soils. Minimum density was measured using 1000g of sand in a 1L glass measuring cylinder with 20mL graduation BS1377-2:1990 and BS1377-4:1990). Maximum density was determined using the vibrating hammer method (BS1377-4:199). Particle size distribution for the sand was determined using a sieving method (BS1377-2:1990) using 1.18mm, 600µm, 425µm, 300µm and 212µm sieve sizes. The Atterberg Limits for the clay were determined using the cone penetration and rolling methods as outlined in BS1377-2:1990.

The sand-clay mixtures were prepared by dry-mixing 90% sand and 10% clay (by mass) and then adding distilled water to achieve a 5% moisture content. The sample was then thoroughly kneaded in a plastic bag by hand for 10 minutes and allowed to rest for 2 hours before any heat treatment.

3. RESULTS AND DISCUSSION

3.1. Mineralogy

During the heat treatment testing and after smouldering remediation, a colour change of the silica sand was observed (Figure 1). Exposure of this material to high temperatures results in colour change from yellowish brown to reddish brown with increasing temperature for the silica sand grains and a change from yellow to pinkish red for the crushed silica sand. This colour change is associated with the dehydration reaction of goethite with increasing temperatures to form hematite or maghemite. During the dehydration, the density of the iron-hydroxide increases from 4.3 mg/m^3 for goethite to 5.2 mg/m^3 for hematite (Wenk and Bulakh, 2004). The sand is comprised primarily of silicon dioxide; iron oxides make up a small fraction of its composition. High temperatures may cause additional changes in mineralogy that may be less likely to be detected by visual examination (Goforth et al., 2005; Pomiès et al., 1998). For example, silicon dioxide becomes unstable with high temperatures and forms silica polymorphs such as trydimite or cristobalite (Hand et al., 1998; Wenk and Bulakh, 2004). Thermal treatments (100-1200°C) on fly ash have transformed quartz minerals to cristobalite and smaller particles exhibit a characteristic glassy composition due their faster cooling time (Mollah et al., 1999).

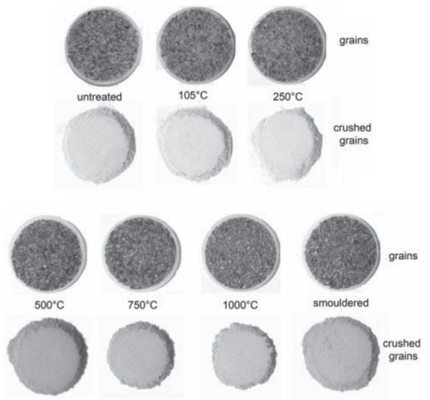

Figure 1. Silica Sand grains and crushed grains after heat treatment.

During testing that required the addition of distilled water, the clay was observed to discolour in the mixed samples, but the distilled water stayed clear (Figure 2). This is very likely associated with the iron oxidation reaction described above. It is possible that this surface reaction enables some of the iron oxides to become mobile and attach themselves onto the clay particles causing this discolouration (Zhang et al., 2011). In the clay-only samples, slight colour changes from white to greyish white were observed. In the smouldered samples for 10% clay and 20% clay mixtures with sand, the colour change was to a darker grey than the heat-only samples. This colour change was likely influenced by staining from the coal tar as well as the inherent colour change of the kaolin.

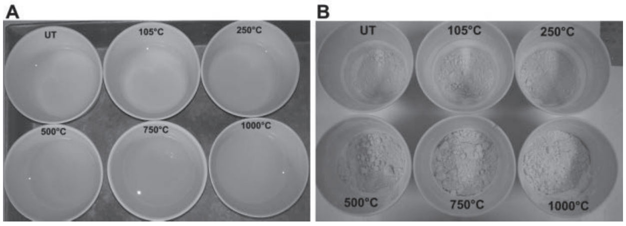

Figure 2. A: Kaolin clay (sand-clay mixture) fraction after heat treatment; B: Kaolin only after heat treatment.

3.2. Particle Size Distribution and Densities

In contrast to mineralogy, elevated temperatures did not seem to affect the particle density or minimum/maximum bulk densities of the silica sand. No real relationship was apparent between treatment temperature and density. For the particle density, the values are consistently near 2.65mg/m^3, which is a value that is widely used in geotechnical engineering calculations. The maximum and minimum densities are equally unaffected by heat treatment or smouldering. These observations are not consistent with the literature on wild and forest fire effects on soil properties, which suggests that bulk density would increase with temperature (Are et al., 2009; Certini, 2005). The lack of organic matter may explain the contrast. The results in this study, which show no significant change in density, suggest that the changes in soil density that are observed after wildfires are associated primarily with effects on organic matter and potentially the smaller silt and clay–sized particles.

Heat treatment has a small but appreciable effect on particle size distribution. As exposure temperature increases from 250 to 1000°C, the sample retained on the 1.18mm sieve increases. The variation in particle size distribution may be linked to the loss of mass beyond the initial moisture content. As temperature increases, mass loss increases. Although there is a dehydration reaction from goethite to hematite in the sand, the fraction of iron oxide relative to the total composition of the sand is too small for this reaction alone to account for the whole additional mass loss. For the silica sand kaolin clay mixture the trend is slightly different (Table 2). The sample retained on the 1.18mm sieve increases very slightly for 250°C, followed by an overall decrease for 250, 500, 750 and 1000°C. For 105 and 250°C the clay coats the sand grains allowing them to be retained on the 1.18mm sieve, for temperatures above 500°C this coating is destroyed resulting in less sample being retained. The coating effect increases the sand fraction >1.18mm by 7 to 16% compared to the higher temperature samples. This is not an increase in the sand fraction but an increase in grains the size of this fraction due to the additional clay coating. This coating could have an impact on the permeability and shear behaviour of these lower temperature samples after heat treatment depending on how easily it can be destroyed or removed by grain interaction or interaction with water.

Table 2. Sieve analysis results for silica sand – 10% kaolin clay mixtures (5% MC) for different heat treatments

Sample	SIEVE ANALYSIS	
	1.18mm	<1.18mm
	% retained	
105	81.8 ± 1.9	18.2 ± 2.1
250	82.7 ± 0.8	17.3 ± 1.0
500	74.5 ± 3.2	25.5 ± 3.6
750	65.6 ± 3.6	34.4 ± 3.7
1000	67.7 ± 0.8	32.3 ± 1.5

3.3. Atterberg Limits for kaolin clay

High temperature processes impact the dynamic properties of soils, particularly liquid and plastic limits at the highest temperatures. This impact on the clay fraction can lead to changes in dynamic behaviour for the clay – sand mixtures. The Atterberg limits for the temperature treatment up to 500°C are similar, especially the liquid limits are all within 64±2%, where the liquid limit for 750°C increases to 81% (Table 3) and this clay has a very high plasticity range compared to the lower temperatures. This is likely due to the increased dehydration of the clay at this temperature. These results are in contrast to Tan et al (2004) (Tan et al., 2004) who recorded an decrease in both liquid and plastic limits with increasing temperature treatment, including non-plastic behaviour for the clays above 400°C. This difference in behaviour can be two-fold. Firstly it can an affect based on the state of the tested sample, especially in regards to initial moisture content. Tan et al (5) uses over consolidated natural clays from Turkey, where this study investigated commercial loose kaolin powder with no moisture content. Secondly, the behaviour can be based on the main mineral contained in the sample, montmorillonite (2:1 clay) for the natural clays from Turkey compared to kaolinite (1:1 clay) for the commercial powder samples. Kaolininte does not swell in the presence of water whereas montmorillonite does swell. Based on this distinction, the responses of montmorillonite and other swelling clays to heat treatment may be different from the responses of kaolinite. Further work is necessary to explore the responses of montmorillonite and other clay minerals during thermal and smouldering remediation processes. The liquid limit test for the sample treated at 1000°C was not possible due to the clay not mixing properly with the water and behaving slightly non-newtonian, which means as the mixing motion stopped the sample liquefied and it was impossible to create a testable sample. Initially, the clay mixed well with the water and it was possible to produce a paste but with increasing water content the behaviour changed and the sample only stayed solid under a constant mixing motion, after stopping the mixing the sample quickly liquefied and dispersed. Storage in a sealed container did not yield different results. In contrast to the other samples (105-750°C treatments), no clay paste was formed. Instead, a stiff clay layer formed at the bottom of the bag with an overlying layer of clean water (Figure 3). This is an unexpected behaviour of the clay and no explanation has been found in the literature. It is likely that the temperature of 1000°C causes de-hydroxylation of the clay minerals, followed by aggregation of the particles and sintering (Fabbri et al.). The net result is that the kaolin particles seem to become hydrophobic. The induced hydrophobicity will affect dynamic properties of the soil such as grain-grain and grain-water interactions. In swelling clays, the effects are expected to be similar to those observed in kaolinite, though based on previous work (Tan et al., 2004), the shift toward hydrophobic particles may occur at lower temperatures. Because other clays are swelling, the effects of the dehydration and melting reactions are expected to have more substantial effects on clay volume as well as grain-grain and grain-water interactions.

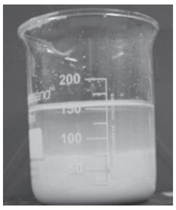

Figure 3. Kaolin clay after 1000°C treatment.

This work has demonstrated that high temperature remediation processes may have significant, long-term effects on soil properties and these effects must be taken into account as part of a holistic approach to aggressive, high-temperature soil remediation.

Table 3. Atterberg Limits and BSCS for kaolin clay for different treatment temperatures

Sample	Liquid Limit w_L	Plastic Limit w_P	Plasticity Index I_p	Plasticity Chart Classification
	%			
105°C	64.4	35.9	28.5	MH: silt, high plasticity
250°C	63.7	30.8	32.9	CH: clay, high plasticity
500°C	65.2	42.7	22.6	MH: silt, high plasticity
750°C	81.6	57.4	24.1	MV: silt, very high plasticity
1000°C	*ND*[1]	*ND*	*ND*	*ND*

[1]: Not Determined

4. CONCLUSIONS

High temperature exposure in the form of thermal treatment and smouldering remediation result in changes to soil properties. These changes are very likely to affect dynamic behaviour such as infiltration, permeability and shear behaviour. The impact appears to be different depending on the sample composition, sand only or sand-clay mixtures. This is due to the mineralogical composition and grain size of these two soil components. This study shows that some results are in contrasts to similar tests (kaolin compared to natural clays from Turkey) and this highlights the complexity of soils and their behaviour. This study gives a good insight into possible changes due to thermal or smouldering treatment. It shows that even lower temperatures (<500°C) can have an impact on the soil, especially on the clay-sand mixture samples. The observed coating of sand particles by clay can impact the infiltration and shear behaviour of the sample. If the coating can be easily removed than this can affect the structure of the sample and in turn weaken the sample or cause collapse after infiltration. This coating can also protect the sand grains from further impact by heat treatment and stabilise the sample. Further analysis is required to fully understand the effect of the clay coating and its stability. The change of Atterberg limits for the kaolin clay with increasing temperature shows that very high temperatures (1000°C) can severely change the behaviour of the soil. Further testing with other clays is necessary to fully understand the relationship between mineralogy and Atterberg Limits.

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