Article
Solidification/Stabilization of Contaminated Soil in a South Station of the Khurmala Oil Field in Kurdistan Region, Iraq

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Abstract: Currently, the primary source of pollution is crude oil production. Crude oil production has dramatic consequences for farmlands, communities, and in terms of the construction materials required for earthworks. The main aims of the present study were to reduce the level of pollution caused by oil production in the Khurmala soil and then reuse it as a construction material. Soil remediation using the solidification/stabilization method was applied in the field using Portland limestone cement (CEM II). The performance of using CEM II in the remediation process was then investigated in the laboratory by taking the natural, contaminated, and treated soils from the Khurmala site. Furthermore, the results of the soils were compared with their corresponding soil samples using ordinary Portland cement (OPC). The comparison was performed by investigating the physical, chemical, and mechanical properties of the soils. The discussion was supported using the scanning electron microscopy (SEM) results. Chemical and SEM results revealed that there were fourfold and tenfold decreases in the percentage of oil and grease using OPC and CEM II, respectively, confirming the higher performance of using CEM II over OPC. The values of the coefficient of permeability, shear strength parameters, and California bearing ratio of the treated soils were significantly improved, compared to those of the contaminated soils.

Keywords: cement; soil contamination; oil field; SEM; treatment

1. Introduction
Kurdistan is an autonomous region rich in natural resources in the northern part of Iraq, including crude oil. In the Republic of Iraq, there are several oil fields, some of which are located in the Kurdistan Region. The area has 13 petroleum fields, one of which is the Khurmala oil field. It is located in the southwest of Erbil City, where 64 crude oil wells are operated. The oil sector is a crucial contributor to the Iraqi Kurdish economy.

The activities and stages of crude oil production in Khurmala, including discovery, loading/unloading stations, and storage facilities, have an adverse effect on all modes of life and ecosystems [1,2]. The environmental impact of these processes cannot be overlooked or disregarded. Among the impacts, the soil pollution which can alter soil engineering properties is considered to be the most worrying, due to its negative impact on civil engineering infrastructure protection [3–5]. Crude oil contaminated soil is possibly the result of oil being released from gas, liquid, or solid components; compounds; or mixtures, leading to changes in the soil’s physical or chemical composition [2]. Crude petroleum is regarded as the most dangerous source of soil pollution. If soil has been contaminated with crude oil, it becomes inappropriate for engineering purposes due to the effect of crude oil on shear strength parameters, resulting in a lack of bearings and immoderate settlement and resulting in the extreme cracking of existing foundations and structures [6]. Nevertheless, it should be noted that the majority of soil pollution occurred in the past, although it continues today through regular industrial and agricultural activity [7,8]. Moreover, soil pollutant outcrops can result from agricultural activity, leaking from aboveground or underground storage tanks and accidental discharges [9,10].
Several researchers, including Akinwumi et al. [11], Wang et al. [12], Oluremi and Osuolale [13], Khamechiyan et al. [9], and Kermani and Ebadi [14], investigated the geotechnical properties of oil-contaminated soils, indicating decreased soil strength and increased plasticity due to oil contamination. The permeability of soils also decreased significantly. Furthermore, quartz sand completely saturated with engine oil can lead to a substantial reduction in soil friction angles and a dramatic increase in soil volumetric strain [15]. In addition, Alfach and Wilkinson [16] reported that the contamination of soil by crude oil had an adverse effect on the base of the pile regarding geotechnical behavior degradation. Moreover, Nasehi et al. [17] and Khosravi et al. [18] investigated the impact of the contamination of gas oil on fine and coarse-grained soil’s geotechnical properties; a decrease in the MDD and the optimum humidity levels was also observed with the increase in Atterberg’s clay and silt limits.

Various methods have been used in recent years to remediate crude-oil-polluted soil. The solvent/surfactant soil-washing technique shows that petroleum pollution soils can cause solubility and extract crude oil soil components [1]. Although biosurfactant solutions have a considerable capacity to extract crude oil from polluted soil by washing conditions, the results showed that the washing-temperature efficiency of crude oil removal from contaminated soil was the most significant factor, compared with the least influential factor which was washing time [2]. In turn, the bioremediation of crude oil polluted soil was achieved by isolating strains of the most efficient biodegradable material in the laboratory; this study demonstrates that many aromatic and saturation hydrocarbons with a chemical composition that is similar to that of crude oil were extracted successfully by the strain [19]. With different remediation approaches, the active degradation of crude oil contaminated saline soil can be achieved by using nitrogen additions, the inoculation of arbuscular mycorrhizas, and the cultivation of Suaeda salsa [20]. In other studies, soil was remediated through pyrolytic treatment. Compared to the reaction time, the pyrolytic efficacy was more affected by the working temperature [21]. Almost all studies in the literature, as mentioned earlier, were focused on agriculture, soil science, and the climate. Hence, it is crucial to analyze these research results for the aim of engineering applications in order to promote practical soil remediation. Thus, oil-contaminated soils must be cured efficiently with methods to enhance the mechanical and geotechnical properties of the soil [22,23]. Furthermore, the solidification/stabilization method, which is accomplished by incorporating cement [24,25], lime [26], fly ash [27], as well as other bonding products into a mixture which is used to impale the contaminants in the polluted medium and ensure long-term safety, is the most effective technique. Solidification describes a process that converts contaminated media into a homogenous solid material with strong structural integrity through its encapsulation in order to change its physical properties. Stabilization describes a process that minimizes contaminated soil’s hazardous potential by limiting its solubility, mobility, or toxicity. Therefore, satisfactory results can be achieved using this technology. For example, Akinwumi et al. [28] and Yu et al. [29] stated that an improvement in crude oil soil achieved with a different proportion of Portland cement increased its strength and reduced its permeability and plasticity, making the soil more suitable after the cement treatment. Similarly, Shah et al. [3] reported better results of soil geotechnical properties with the utilization of various additives, such as cement, lime, and fly ash to stabilize contaminated soils with crude oil.

In further experimental work, Wang et al. [30] indicated that the results of the geotechnical properties presented a notable increase in undrained shear strength, solid content (water content), and Atterberg limit values of the soil, achieved through using different curing times and various doses of cement after stabilizing the mature fine tailings. Additional research carried out by Nasr [31] examined the sand’s strength behavior when contaminated with oil by utilizing the cement kiln dust (CKD) to determine the stabilized soil’s engineering properties for use in rural road construction. Results showed that with the addition of CKD, the unconfined compressive strength and California bearing ratio (CBR) values of the oil-contaminated sand were increased. The stability of polluted sand
decreases with the increase in oil percentage. Consequently, Al-Rawas et al. [32] concluded that oil-contaminated land could be reused when stabilized with cement or cement bypass dust, due to the enhanced geotechnical properties of construction and engineering applications, offering practical, safe, and cost-effective solutions.

This study investigated the effectiveness of using two different types of cements to stabilize oil-contaminated soil. Additionally, the physical, mechanical, and chemical behavior of both polluted crude-oil soil and stabilized soil has been studied in order to enable their reuse as earth construction materials. To date, this kind of research has not been carried out in the oil fields in Kurdistan Region, Iraq.

2. Scope

The Khurmala oil and gas field is spread from 30 km southwest of Erbil City, and is 22 km long and 3 km wide. The crude oil produced in the field wells that are spread across the Khurmala dome was collected at stations (1—North: collection from a network of 29 oil wells; 2—South: collection from a network of 26 oil wells; and 3—Middle: collection from a network of 20 oil wells) through various 150 mm flow lines and then sent to the Central Process Station (CPS-1 and CPS-2) through 500 mm trunk pipelines. After processing, the crude oil was pumped for export.

The historical activities of the Khurmala dome began in 1935 with the drilling of the first well. In 1935–1977, 12 wells were drilled. Development then began in 1988 and the first oil production occurred in 2009.

Crude oil is among the leading causes of terrestrial pollution due to its superior ability to spread, interact, and penetrate the soil in many forms and various means through its dependence on biological, physical, and air variables. There are several different sources of hazards in crude oil contaminated soil in the Khurmala oil field, including exploration and application processes. Therefore, a goal was set to minimize the amount of pollution in the field and remediate the contaminated soil by constructing specific concrete containers for collecting waste crude oil and mixing the contaminated soil with an appropriate cement type, respectively.

The above activities create significant soil pollution due to inappropriate disposal, oil spills, tank leakage, and pipeline breakage [29]. In the Khurmala oil field, the main sources of soil pollution are as follows:

1. Burning pit: This is a pit that is prepared to collect the crude oil that is tested and drained during oil well testing through a special pipe called the burning pipe. A check is necessary to determine the quantity and quality of crude oil, utilizing a test point and a flow meter attached to the burning pipe, as shown in Figure 1a.

2. Random pit: If the pipeline is not accessible for a particular location, the alternative is unloading. The oil in the tankers must be tested. The tested oil then has to be randomly handled. The tested oil is dealt with by disposal in a designated pit called the random pit, as shown in Figure 1b, from which oil can leak into the soil from the older pits. Therefore, these old pits must be remedied. Fortunately, in the Khurmala oil field, a particular separator system is currently used. An oil–water separator system is designed to isolate total quantities of oil and suspended solids from the oil refinery wastewater effluents. This system is based on preventing any leakage into the surrounding and underground soil under Health Safety and Environmental regulations (HSE), as shown in Figure 1c.

3. The absence of a closed drain system in the facilities (including pumps, equipment, pipes, and valves) frequently causes various oil leakage accidents, which can cause severe pollution to the surrounding soil, as shown in Figure 1d. These problems can be controlled through the use of close drain systems linked to a piping system connected to a particular basin for this leakage. Unfortunately, this system is not currently in use at the Khurmala oil station.
4. Flow lines under or above the ground that transport crude oil from the well to stations are subjected to corrosion due to H$_2$S if a corrosion inhibitor is not used, leading to holes in these pipes, causing oil leakage and then soil contamination.

![Flow lines](image1)

Figure 1. Most sources of polluted soils at Khurmala oil field: (a) burning pits, (b) random pits, (c) separator system, and (d) oil leakage from the facilities.

3. Materials and Methods

3.1. Materials

Soil, Portland limestone cement (CEM II), ordinary Portland cement (OPC), and crude-oil were the primary materials used in this work. In this section, the physical, mechanical, and chemical properties of these materials are described as follows:

3.1.1. Soil

This research was carried out on natural, contaminated, and treated soils. All soils were obtained from an oil pit at the south station’s Khurmala oil field treatment area (latitude: 39.0424; longitude: 39.76083).

Figure 2 shows the grain size distribution curve for the natural soil. The soil is classified under the Unified Soil Classification System (USCS) as silty sandy soil (SM). These characteristics designated according to the American Standard of Testing Materials (ASTM). Table 1 shows the geotechnical properties of the soil which were obtained by
performing the tests in the Geotechnical Laboratory, Civil Engineering Department, College of Engineering, Salahaddin University-Erbil, Erbil, Kurdistan Region, Iraq, while Table 2 shows the chemical characteristics of the natural soil that were obtained by performing the tests in the Kurd Central Research Facilities (KCRF) laboratory in the Soran District, Erbil City.

![Grain size distribution curve](image)

**Figure 2.** The grain size distribution curve of the natural soil.

**Table 1.** The geotechnical properties of the natural soil.

| Soil Properties         | Natural Soil | Standard                        |
|-------------------------|--------------|---------------------------------|
| Natural moisture content| w (%)        | 2.4                             | ASTM D2216 [33]                 |
| Specific Gravity        | Gs           | 2.67                            | ASTM D854 [33]                  |
| Gravel (%)              |              | 19.51                           | ASTM D421-85(2007) [33]         |
| Sand (%)                |              | 66.05                           | ASTM D2217-85 R98 [33]          |
| Fines (%)               |              | 14.45                           |                                 |
| Cu                      |              | 3.47                            | ASTM D2487 [33]                 |
| Cc                      |              | 1.06                            | AASHTO A-2-4                    |
| Maximum dry unit weight | $\gamma_d$ (kN/m$^3$) | 17.6                  | ASTM D698 [33]                  |
| Optimum moisture content| (%)          | 12.6                            |                                 |
| Angle of internal friction| $\phi$     | 28.56°                          | ASTM D3080 [33]                 |
| Cohesion                | $C$ (kPa)    | 34.5                            |                                 |
| Coefficient of permeability| $k$ (cm/s) | 3.47 $\times$ 10$^{-5}$        | ASTM D2434 [33]                 |
| CBR                     | Unsoaked CBR % | 41.883            | ASTM D1883 [33]                 |
|                        | Soaked CBR %  | 25.257                          |                                 |

**Table 2.** The chemical characteristics of the natural soil.

| Parameter          | Unit          | Value  |
|--------------------|---------------|--------|
| pH                 |               | 7.7    |
| Electrical conductivity | $\mu$mho/cm | 703    |
| Alkalinity         | mg/L          | 39     |
| Carbonate          | mg/L          | 0      |
| Bicarbonate        | mg/L          | 39     |
| Sulfate            | mg/L          | 104    |
| Chloride           | mg/L          | 132    |
| Total organic carbon| %            | 0.43   |
| Oil and Grease     | %             | 0.52   |
3.1.2. Crude Oil

The petroleum specimen was obtained from the Khurmala Station in Iraqi Kurdistan, run by the Kar Group Petroleum Production Company. A description of the fundamental crude oil properties is shown in Table 3. The Khurmala Block refinery authorities provided the values of the crude oil properties, which have a value of American Petroleum Institute (API) gravity equal to 32.29 at 15.6 °C, and a value of specific gravity equal to 0.8639 at 15.6 °C.

Table 3. The physical properties of the Khurmala crude oil.

| Test | H₂S (ppm) | BS&W * (%) | Total Sulphur (%) | Salt (ptb) ** | Density (kg/m³) | API Gravity (at 15.6 °C) | Viscosity (mm²/s) |
|------|-----------|------------|-------------------|-------------|----------------|--------------------------|------------------|
| Standard Results | UOP 163 | ASTM D4010 [34] | ASTM D4294 [35] | ASTM D3230 [36] | ASTM D1298 [37] | ASTM D1298 [37] | ASTM D7042 [38] |
| Results | 41.3 | 0.6 | 2.22 | 229 | 863.1 | 32.29 | 12.8 |

* Basic sediment and water content of crude oils. ** Ptb = pounds of salt per thousand barrels of crude oil.

3.1.3. Cement

In this study, CEM II is available in the local market and used in the field (according to BS EN 196—Methods of testing cement). Simultaneously, a locally produced OPC is available in the Erbil market and used in the laboratory study. The cements’ chemical composition is presented in Table 4.

Table 4. The physical and chemical properties of the CEM II and OPC.

| Chemical Analysis | CEM II (%) | OPC (%) |
|-------------------|------------|---------|
| SiO₂ | 20.04 | 20.17 |
| CaO | 61.84 | 63.11 |
| Al₂O₃ | 4.37 | 4.22 |
| Fe₂O₃ | 3.71 | 3.78 |
| MgO | 3.48 | 3.82 |
| SO₃ | 2.67 | 2.08 |
| Insoluble Material | 0.32 | 0.59 |
| Loss of Ignition | 3.05 | 1.55 |
| Lime Saturation Factor | 0.87 | 0.96 |
| C₃A | 5.3 | 4.79 |
| C₃S | 42.09 | 63.94 |
| C₂S | 25.72 | 9.79 |
| C₄AF | 11.28 | 11.5 |

3.2. The Solidification/Stabilization Process of Pollutant Soil in the Field

Solidification/stabilization requires the immobilization of the polluted soil constituents through a process of chemical modification into insoluble substances or by encapsulating the solid. Mixing the polluted soil with cement results in this process. The treated soil in the site underwent a solidification/stabilization process at the Khurmala oil field treatment area. The soil was mixed with CEM II (1 ton cement/7 m³ soil) at approximately 8–9% by weight of the soil with a water–cement ratio of 40%, and the treated soil was then left as a construction earth material for two months in order to gain an equilibrium between cement and soil, before being reused in the area. The main goals in this process were the following:

- Improve soil handling and physical characteristics;
- Minimize available surface area for the movement or loss of pollutants and limit fluid movement by the total hard matrix volume;
- Minimize the solubility of the contaminant into the amount of contaminated soil.

The project was initiated on 16 January 2019 and lasted until 19 November 2019. The method of treatment included the following:
1. International drilling fluids and engineering services, a Qmax solutions company, waste management division, provided services of remediating and encapsulating all the pit's wastes, such as oily sludge and contaminated soil waste.

2. The remediation involves remediating the contaminated oil pit, reserve pit (Figure 3a), and overflow pit at the south production station, located at Khurmala Site, in the Erbil City.

3. The equipment and machinery were mobilized to the southern production plant site on 15 January 2019.

4. The contaminated soil in the oil pit was first treated by digging and treating all the soil contaminated with the existing crude oil (Figure 3b). The total volume of treated soil reached 2980 m$^3$.

5. The treated soil was stored beside the oil pit (Figure 3c) to be backfilled, after reconstructing and lining the pit, and prepared as a construction earth material.

6. When the test results of the oil pit’s bottom and sides showed that it was cleaned of contaminant, the pit was reconstructed and lined up with a geosynthetic clay layer with high-density polyethylene liner to be backfilled with remediated soil (Figure 3d).

7. The oil pit was backfilled with treated soil, covered with a GCL liner on the top (Figure 3e), then backfilled to 3.5 m of fresh soil from the area around the pit, leveled, and compacted to the natural ground level.

8. The site underwent general clean-up and restoration. The procedures of the treatment are illustrated in Figure 3.

3.3. The Solidification/Stabilization Process of Pollutant Soil in the Laboratory

The main objectives of the laboratory tests were as follows: (1) to check whether the process of the solidification/stabilization of pollutant soil in the field was performed effectively in the field and (2) to emphasize that the CEM II is a suitable type of cement used in the process. To achieve this, natural, contaminated, and treated soil samples were collected from the Khurmala site. Then, all samples were transported in closed, labeled plastic bags to the Geotechnical Laboratory, Civil Engineering Department, College of Engineering, Salahaddin University-Erbil, Erbil, Kurdistan Region, Iraq, in order to study their physical, mechanical, and chemical properties. In addition, the impact of stabilizing crude oil polluted soil treated by OPC was studied and compared to the treated sample with CEM II from the field.

The treated (CEM II and OPC) soil specimens in the laboratory were prepared by mixing the contaminated specimens (at oil content 14%). The specimens were mixed with 8.7% of ordinary Portland cement by weight with a water–cement ratio of 0.4 in order to match the field conditions. The mixed samples were placed into closed containers for two weeks, allowing possible reactions between the soil and cement.

The clean soils were taken as reference samples. These were obtained from a location that was ensured, through the detection of vision and color, to be uncontaminated. The site had similar geological conditions to the contaminated site. The clean samples were taken 10 cm from the earth’s surface.

3.4. Laboratory Test Program

Laboratory work was designed to obtain parameters, including the specific gravity, compaction, coefficient of permeability, un-soaked CBR and soaked CBR, direct shear, and scanning electron microscopy (SEM) tests for the natural soil, soils polluted with crude oil, and contaminated soil samples stabilized with CEM II and OPC. The laboratory investigation was performed to explore the impact of different types of cements on the geotechnical properties of oil-contaminated soils. On average, three specimens were used to avoid any uncertainty and scattering in data.

The soils’ compaction characteristics were studied by conducting a standard compaction test following ASTM [34]. The MDD and the OMC were obtained from the compaction curve for all the soils.
Figure 3. The treatment process of pollutant soil: (a) the oil pit before treatment; (b,c) the treatment of contaminated soil in the oil pit; (d) reconstruction and lining of the oil pit; and (e) backfilling of the pit with treated soil and leveling.

The shear strength parameters of the soils are essential to consider, as they influence the design of many geotechnical engineering projects, such as embankments, soil slopes stability, and foundations. Direct shear tests were performed according to the method recommended by ASTM [33]. The samples were tested at their MDD and OMC. Soaked and un-soaked CBR tests were performed on oil-contaminated soil samples with and without cement and clean soil samples as described in ASTM [33]. The falling head equipment was used to determine the permeability coefficient. The test was performed on all soils. The
technique, used to assess permeability via the falling head method, was compatible with the work of Head [39].

4. Results and Discussion

The findings show that the values of specific gravity of both the natural and treated CEM II and OPC were 2.67, 2.68, and 2.38, respectively. The untreated soil had the lowest value of specific gravity of 2.35. This could be attributed to the high oil content (which was up to 14%).

4.1. Compaction Test Results

The compaction test results are shown in Figure 4 in the form of dry density versus water content. Generally, the compaction curves of the contaminated soil and both treated soils moved below the natural soil curve. The MDD of the contaminated soil substantially decreased with a 14% oil content to a low value of MDD (1.625 g/cm$^3$) due to the oil content in contaminated soils. This reduction is attributed to the effect of the specific gravity value of crude oil on the soil. Moreover, with silty sandy soils polluted with crude oil, the particles separated as the voids filled with the oil and coated the granules. Therefore, a decline in dry density was observed as the soil transited into a loose material state. Similar results are reported by Al-sanad et al. [40], Meegoda et al. [7], and Nasr [31]. Nevertheless, these findings differ from other studies by Khamechiyan et al. [9], Al-Rawas et al. [32], and Nasehi et al. [17]. At the same time, no discernible change in the OMC was noticed between the natural soils and polluted soil by crude oil.

![Figure 4. Compaction curves of natural, contaminated, and treated (CEM II and OPC) soils.](image)

The MDD of the treatment soil slightly increased when the soil was solidified with CEM II and OPC, reaching a peak at 1.69 g/cm$^3$ and 1.635 g/cm$^3$. A high increase in OMC could be observed compared to the OMC of natural soil, particularly in relation to the soil treated with CEM II. In comparison, the value of MDD of the natural soil was much higher than that for treated soils. By adding the cement to the polluted soil, the MDD of the stabilized contaminated soil increased due to the specific gravity of cement (commonly 3.15) compared with the contaminated soil (2.35). Meanwhile, the OMC increased since cement has a better absorption potential for water.
4.2. Direct Shear Test Results

The results from the direct shear tests are presented in Figure 5, which are illustrated in the form of shear stress against normal stress. It can be seen that all soils had almost the same trend: a noticeable increase in the treated soil was observed compared with other soils.

![Figure 5. Direct shear test results of natural, contaminated, and treated (CEM II and OPC) soils.](image)

The direct shear test results showed that the internal friction angle (Φ) decreased drastically from 28.5° to 12.7° for natural, treated (CEM II and OPC), and contaminated soils, respectively. Shin et al. [41] stated that oil contamination causes a noticeable decrease in the Φ value. Ghaly [42], Khamehchiyan et al. [10], and Nasehi et al. [17] reported that in the presence of a high crude oil content, the friction angle decreases. This inverse correlation might be explained by the coating of soil particles with crude oil, which acts as a lubricant that decreases the inter-granular contact force between the sand particles. However, Abousnina et al. [43] reported that, for samples containing 2% to 20% oil, no significant difference in the frictional angle of the sand was detected, which indicates that the sand particles were totally covered with crude oil at a level of more than 2%, and their frictional angle remained unchanged. For the stabilized soils, the treated soils indicated an increment in the angle of internal friction by stabilizing with CEM II and OPC to 30.7° and 25.0°, respectively, as shown in Figure 5. This increment could be related to the cement action that increases the agglomeration between grains and minimizes lubrication, increasing the contact force between particles. However, these results are different from the findings of Al-Rawas et al. [32].

The cohesion of the natural soil was 35 kPa. Crude oil contamination led to an increase in the cohesion value of this soil to 56 kPa. These findings match the results of Nasehi et al. [17], but are incompatible with the findings of Khamehchiyan et al. [9]. It is clear that crude oil’s ability to resist shear force is greater than water, since its viscosity is more. Therefore, during the application of shear force to the contaminated specimen, crude oil resists a portion of that shear force besides the soil particles and, in turn, increases the soil’s apparent cohesion.

In soil stabilized with CEM II, the rise in cohesion was dramatic, changing to 81 kPa, while the cohesion in soil treatment with OPC reached a low value of 27 kPa. This is due to the increase in the material’s cohesiveness as a result of the cementing action caused by the hydration process. This is in line with the finding that CEM II hydrated more quickly
and provided a higher strength than OPC. Since the $C_2S$ is responsible for the subsequent rise after the first week in the strength of the cement’s hydraulic components, its value is 25.72% in CEM II, compared to 9.79% in OPC.

### 4.3. Permeability Tests

Table 5 shows the permeability test results for the natural, contaminated and treated soils. As expected, the permeability has a reverse correlation with oil content. The coefficient of permeability ($k$) for contaminated soil ($7.34 \times 10^{-6}$ cm/s) was lower than for natural soil ($3.47 \times 10^{-5}$ cm/s). However, even at 14% crude oil, the decrease in the value of $k$ is not as high as expected. It is clear that oil-contaminated soil decreases the $k$ due to the occupation of the crude oil for the pore spaces, which causes a reduction in the flow rate through the soil by minimizing the volume of the pores responsible for facilitating the movement of fluids within the soil. Similar results are presented by Kamehchiyan et al. [9] and Abousnina et al. [43].

| Soil Properties          | Natural Soil | Contaminated Soil | Treatment Soil with CEM II | Treatment Soil with OPC |
|--------------------------|--------------|-------------------|----------------------------|-------------------------|
| Coefficient of permeability $k$ (cm/s) | $3.47 \times 10^{-5}$ | $7.34 \times 10^{-6}$ | $4.55 \times 10^{-8}$ | $4.87 \times 10^{-6}$ |

The results for the treated soils indicated a decrease in the value of $k$, compared to the natural and contaminated ones. By adding 8.7% cement, the permeability of CEM II and OPC decreased to $4.55 \times 10^{-8}$ cm/s and $4.87 \times 10^{-6}$ cm/s, respectively. With the addition of cement to the content, a cement product, such as a bonding gel, was produced, which reduced the porosity that binds the soil particles together and hindered the passage of water into the soil. Consequently, the permeability coefficient was reduced. Similar results were found by Al-Rawas et al. [32].

### 4.4. CBR Tests

The CBR is a test usually performed to assess the strength of subgrade soils and base course materials in pavement work. As summarized in Table 6 and Figure 6, the CBR values of the crude oil contaminated soils under un-soaked conditions significantly decreased compared with the natural ones. This reduction is probably due to the combination of excessive oil presence and the low maximum density of the contaminated soil. These results are consistent with those of Al-Sanad et al. [40] and Nasr [31]. In contrast, the values of CBR for natural and contaminated soils under soaked conditions were similar.

| Soil Identification      | Maximum Dry Unit Weight (kN/m$^3$) | Optimum Moisture Content % | CBR % Unsoaked | CBR % Soaked |
|--------------------------|------------------------------------|----------------------------|----------------|--------------|
| Natural Soil             | 17.60                              | 12.6                       | 41.88          | 25.25        |
| Contaminated Soil        | 15.89                              | 12.2                       | 26.24          | 23.72        |
| Treatment Soil with CEM II | 16.55                            | 18.7                       | 75.16          | 38.26        |
| Treatment Soil with OPC  | 16.01                              | 14.5                       | 60.35          | 34.68        |

Figure 6 also shows that, for both soaked and un-soaked treated (CEM II and OPC) soils, the values of CBR were significantly improved in comparison to those for the natural soil. Based on the review of Wang [44], cement contains hydration products that increase therapy strength and performance. The enhancement in un-soaked and soaked CBR values is due to the production of cementitious components, such as calcium silicate hydrates and calcium aluminate hydrates in the contaminated soil stabilized/solidified by cement [45].
4.4. CBR Tests

The CBR is a test usually performed to assess the strength of subgrade soils and base courses. The results of CBR tests are presented in Table 6. The CBR values for contaminated soil stabilized with OPC and CEM II are higher than those for natural soil. The CBR values for treated soils are significantly improved compared to those for natural soil.

Figure 6. Stress versus vertical displacement for (a) un-soaked and (b) soaked samples.

4.5. SEM Analysis

SEM is a technique that provides many magnified images and explains differences that soil enhancement produces in physical, chemical, and mechanical behavior, including shape, size, composition, and crystallography properties [46]. SEM was used in this study to investigate the microstructure particles for the natural, contaminated, and treated soils, in order to detect the structure of the bonding between sand particles in the previous cases.

Figures 7a–d and 8a–d illustrate the geometric arrangement for the natural, contaminated, and treated soils, respectively. In natural soil fabric, the diameter in the singular grains can be observed. However, it is not possible to distinguish individual floccules in these micrographs, as indicated in Figure 7a. Subsequently, the morphological shape of the natural soil, as shown in Figure 8a, indicated the appearance of burrs in the soil grains, confirming its non-coated properties.

Crude oil firmly coated the singular soil particles via hydrogen bonding and van der Waals forces. As a result, it was shown in the form of a dense-packed structure with almost no visible voids, as shown in Figure 7b, since the lining oils created a water-resistant layer that blocked the voids, causing a reduction in permeability. Moreover, in the photomicrograph of Figure 8b, the surface of the contaminated soil appeared as one flock with no distinct pore spaces, indicating that it was filled with oil.

Figure 7c illustrates a significant improvement in the soil treatment with CEM II. The similarity of the microscopic surface of the treated soil to the natural soil was obvious, in addition to agglomerated morphology of the soil sample. The change in color from dark to light in the samples signifies that the crude oil was removed from the soil in a satisfactory proportion. These results are in agreement with the chemical results shown in Table 7, which indicate a decline in total organic carbon from 11.7% to 0.8% and in oil and grease from 14% to 0.96%, simultaneously, which is a significant performance. However, the structural features for the soil sample in Figure 8c showed a small proportion of oil covering some of the grains with the presence of apparent voids in the surface element.

In comparison, no considerable improvement was noticed in the soil that was stabilized with OPC, as shown in Figure 7d, compared to CEM II. The microscopic surface of the treated soil was more similar to the contaminated soil than it was to the natural soil. The crude oil still coats the soil particles. If we combine the impact of the oil and cement, the influence of oil is still dominant. This result is consistent with the chemical results; soil treated with OPC had 8.2% total organic carbon and 9.8% oil and grease. Although the soil grains became aggregated, they were formed in the shape of flocks coated with oil, as shown in the microscopic image in Figure 8c.
Figure 7. SEM photograph of 200 nm: (a) natural soil; (b) contaminated soil; (c) soil stabilized with CEM II; and (d) soil stabilized with OPC.

Figure 8. Cont.
pollutants, such as alkalinity, bicarbonate, sulfate, chloride, total organic matter, and oil and grease in the polluted samples were higher than for the clean sample. It is clear that oil disposal caused the contamination of the soil at the Khurmala oil field. The research conducted by Ergozhin et al. [47], Kuany et al. [48], Wang et al. [49], Trejos-Delgado et al. [50], and Jabbarov et al. [51] confirms the obtained results. All pH values were in the normal range, except the pH value for the soil sample treated with OPC, which is classified in the high alkaline range (pH = 12.74). Limited variation in the alkalinity, carbonate, and bicarbonate values for normal, polluted, and treated soil samples was reported.

Treatment using CEM II led to a decrease in the pH, chloride, total organic matter, oil and grease in the treated soil sample, while treatment via OPC resulted in a decrease in pollutants, such as alkalinity, bicarbonate, sulfate, chloride, total organic matter, and oil and grease in the processed soil sample. Generally, treatment with CEM II and OPC caused a decrease in contaminants, especially chloride, total organic matter, and oil and grease. A fluctuation in pH, electrical conductivity, alkalinity, and sulfate values was observed; this may be due to the chemical reactions between pollutants and the components of the treatment materials. Sulfate and chloride figures after treatment became lower than those of the normal soil sample. Results revealed that the application of CEM II for the treatment of the polluted soil samples was often superior to that of the OPC.

### 4.7. XRD Tests

XRD is a powerful nondestructive method for symbolizing crystalline materials. It offers information on the structures, stages, preferred crystal locations (texture), and other structural factors. XRD peaks are formed by the productive interference of a monochromatic beam of X-rays distributed at definite angles from each set of lattice planes in an
illustration. The highest strengths are found using the atomic positions within the lattice planes. Accordingly, the XRD pattern is the print of periodic atomic arrangements in a specified material [52].

XRD test results for the soil samples are illustrated in Table 8 and Figure 9. The pollutants changed the shape of Figure 9b, when compared with Figure 9a; silicon oxide and calcium carbonate values were increased in the polluted soil sample, when compared with the normal soil sample (Table 8). Furthermore, calcium aluminum silicate also increased, while silicon oxide and calcium carbonate decreased after both treatment methods. Additionally, treatment using CEM II is shown in Figure 9c. Using OPC for the treatment of the polluted soil sample affected the soil components, as shown in Figure 9d. Values of aluminum calcium silicon, magnesium aluminum silicate, iron silicate hydroxide, magnesium dialuminium disilicide–U1, and sodium aluminum silicate hydrate increased in the treated samples (Table 8). The obtained results shown in Table 7 are in coincidence with the illustration of XRD results. The variation of values in Figure 9a–d agrees with the obtained results in Table 8. The present results agree with the published work of Aziz [53].

Table 8. XRD test results of the natural, contaminated, and tread soil samples.

| Soil Type     | Silicon Oxide | Calcium Carbonate | Albite Low | Calcium Aluminum Silicate | Aluminum Calcium Silicate | Magnesium Aluminum Silicate | Iron Silicate Hydroxide | Magnesium Dialuminium Disilicide–U1 | Sodium Aluminum Silicate Hydrate |
|---------------|---------------|-------------------|-----------|---------------------------|---------------------------|----------------------------|---------------------------|---------------------------------|---------------------------------|
| Natural       | 76            | 73                | 66        | 61                         | 54                        | 69                         | 66                        |                                 |                                 |
| Contaminated  | 82            | 74                |           |                           | 53                        | 41                         | 54                        | 69                              | 66                              |
| OPC           | 66            | -                 |           | 61                         |                           |                            |                           |                                 |                                 |
| Stabilized OPC|               |                   |           |                           |                           |                            |                           |                                 |                                 |
| Stabilized CEM II | 71     | 69                |           |                           |                           |                            |                           |                                 |                                 |

Figure 9. XRD test for the soil sample: (a) natural soil; (b) contaminated soil; (c) soil stabilized with CEM II; and (d) soil stabilized with OPC.
5. Conclusions

According to the study’s results presented above, the following conclusions can be drawn:

- The disposal of crude oil resulted in soil contamination at the Khurmala oil field.
- Compaction characteristics and CBR values deteriorated with the presence of crude oil content. At the same time, when the contaminated soil was treated with a stabilization agent containing both types of cement (i.e., CEM II and OPC), an increase in the MDD and OMC and CBR values was observed, but the best result was achieved with CEM II.
- The greatest improvement in the shear strength parameters ($c'$ and $\Phi'$) was achieved when the contaminated soil was treated using CEM II.
- Generally, the contamination of sandy soil with crude oil induced a permeability reduction, and a further decrease in permeability was detected as the soil solidified with cement.
- A substantial reduction in the oil and grease of the treated soil was achieved using CEM II, compared to soils treated with OPC. The SEM results confirm this.
- The solidification/stabilization (S/S) method provides an effective remediation method for processing waste to produce a safe, dry material acceptable for on-site burial. The application of the S/S process via utilizing cement has an influential role in strengthening the geotechnical characteristics for the contamination of soils with crude oil.
- The remediation of contaminated soil with crude oil utilizing CEM II resulted in a larger improvement compared to when using OPC.

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References

1. Wang, M.; Zhang, B.; Li, G.; Wu, T.; Sun, D. Efficient Remediation of Crude Oil-Contaminated Soil Using a Solvent/Surfactant System. *RSC Adv.* 2019, 9, 2402–2411. [CrossRef]
2. Urum, K.; Pekdemir, T.; Çopur, M. Screening of Biosurfactants for Crude Oil Contaminated Soil Washing. *J. Environ. Eng. Sci.* 2005, 4, 487–496. [CrossRef]
3. Shah, S.J.; Shroff, A.V.; Patel, J.V.; Tiwari, K.C.; Ramakrishnan, D. Stabilization of Fuel Oil Contaminated Soil—A Case Study. *Geotech. Geol. Eng.* 2003, 21, 415–427. [CrossRef]
4. Cheng, L.; Shahin, M.A. Stabilisation of Oil-Contaminated Soils Using Microbially Induced Calcite Crystals by Bacterial Flocs. *Geotech. Lett.* 2017, 7, 146–151. [CrossRef]
5. Rao, S.M.; Rao, S.S. Ground Heave from Caustic Soda Solution Spillage—A Case Studt. *Soils Found.* 1994, 34, 13–18. [CrossRef]
6. Rahman, Z.A.; Hamzah, U.; Taha, R.; Ihnain, N.S.; Ahmad, N. Influence of Oil Contamination on Geotechnical Properties of Basaltic Residual Soil. *Am. J. Appl. Sci.* 2010, 7, 954–961. [CrossRef]
7. Meegoda, J.N.; Chen, B.; Gunasekera, S.D.; Pederson, P. Compaction Characteristics of Contaminated Soils-Reuse as a Road Base Material. *Geotech. Spec. Publ.* 1998, 79, 195–209. [CrossRef]
8. Ratnaweera, P.; Meegoda, J.N. Shear Strength and Stress-Strain Behavior of Contaminated Soils. *Geotech. Test. J.* 2006, 29, 133–140. [CrossRef]
9. Kamehchiyan, M.; Hossein Charkhabi, A.; Tajik, M. Effects of Crude Oil Contamination on Geotechnical Properties of Clayey and Sandy Soils. *Eng. Geol.* 2007, 89, 220–229. [CrossRef]
10. Estabragh, A.R.; Beytolahpour, I.; Moradi, M.; Javadi, A.A. Consolidation Behavior of Two Fine-Grained Soils Contaminated by Glycerol and Ethanol. *Eng. Geol.* 2014, 178, 102–108. [CrossRef]
11. Akinvumi, I.I.; Diwa, D.; Obianigwe, N. Effects of Crude Oil Contamination on the Index Properties, Strength and Permeability of Lateritic Clay. *Int. J. Appl. Sci. Eng. Res.* **2014**, *3*, 816–824. [CrossRef]

12. Wang, Y.; Feng, J.; Lin, Q.; Lu, X.; Wang, X.; Wang, G. Effects of Crude Oil Contamination on Soil Physical and Chemical Properties in Momoge Wetland of China. *Chin. Geogr. Sci.* **2013**, *23*, 708–715. [CrossRef]

13. Oluwemi, J.; Osouloale, O. Oil Contaminated Soil as Potential Applicable Material in Civil Engineering Construction. *J. Environ. Earth Sci. Int.* **2014**, *4*, 87–100.

14. Kermani, M.; Ebadi, T. The Effect of Oil Contamination on the Geotechnical Properties of Fine-Grained Soils. *Soil Sediment Contam.* **2012**, *21*, 655–671. [CrossRef]

15. Evgin, E.; Das, B.M. Mechanical Behavior of an Oil Contaminated Sand. In Proceedings of the Mediterranean Conference on Environmental Geotechnology, Izmir, Turkey, 25–27 May 1992; pp. 101–108.

16. Alifach, M.T.; Wilkinson, S. Effect of Crude-Oil-Contaminated Soil on the Geotechnical Behaviour of Piles Foundation. *Geotech. Res.* **2020**, *7*, 76–89. [CrossRef]

17. Nasehi, S.A.; Uromeihy, A.; Nikudel, M.R.; Morsali, A. Influence of Gas Oil Contamination on Geotechnical Properties of Fine and Coarse-Grained Soils. *Geotech. Geol. Eng.* **2016**, *34*, 333–345. [CrossRef]

18. Khosravi, E.; Ghaseinzadeh, H.; Sabour, M.R.; Yazdani, H. Geotechnical Properties of Gas Oil-Contaminated Kaolinite. *Eng. Geol.* **2013**, *166*, 11–16. [CrossRef]

19. Yan, G.; Cai, B.; Chen, C.; Yue, Y.; Wang, Q.; Deng, H.; Liu, S.; Guo, S. Bioremediation of Crude Oil Contaminated Soil. *Pet. Sci. Technol.* **2015**, *33*, 717–723. [CrossRef]

20. Gao, Y.C.; Guo, S.H.; Wang, J.N.; Li, D.; Wang, H.; Zeng, D.H. Effects of Different Remediation Treatments on Crude Oil Contaminated Saline Soil. *Chemosphere* **2014**, *117*, 486–493. [CrossRef]

21. Kang, C.U.; Kim, D.H.; Khan, M.A.; Kumar, R.; Ji, S.E.; Choi, K.W.; Paeng, K.; Park, S.; Jeon, B.H. Pyrolytic Remediation of Crude Oil-Contaminated Soil. *Sci. Total Environ.* **2020**, *713*, 136498. [CrossRef]

22. Russell, D.L. *Remediation Manual for Petroleum Contaminated Sites*; CRC Press, Taylor & Francis Group: Lancaster, UK, 1992; ISBN 9780877622876.

23. Balba, M.T.; Al-Awadhi, N.; Al-Daher, R. Bioremediation of Oil-Contaminated Soil: Microbiological Methods for Feasibility Assessment and Field Evaluation. *J. Microbiol. Methods* **1998**, *32*, 155–164. [CrossRef]

24. Moon, D.H.; Grubb, D.G.; Reilly, T.L. Stabilization/Solidification of Selenium-Impacted Soils Using Portland Cement and Cement Kiln Dust. *J. Hazard. Mater.* **2009**, *168*, 944–951. [CrossRef] [PubMed]

25. Estabragh, A.R.; Khatibi, M.; Javadi, A.A. Effect of Cement on Treatment of a Clay Soil Contaminated with Glycerol. *J. Mater. Civ. Eng.* **2016**, *28*, 04015157. [CrossRef]

26. Alpaslan, B.; Yukselen, M.A. Remediation of Lead Contaminated Soils by Stabilization/Solidification. *Water Air Soil Pollut.* **2002**, *133*, 253–263. [CrossRef]

27. Dermatas, D.; Meng, X. Utilization of Fly Ash for Stabilization/Solidification of Heavy Metal Contaminated Soils. *Eng. Geol.* **2003**, *70*, 377–394. [CrossRef]

28. Akinwumi, I.I.; Booth, C.A.; Diwa, D.; Mills, P. Cement Stabilisation of Crude-Oil-Contaminated Soil. *Proc. Inst. Civ. Eng. Geotech. Eng.* **2016**, *169*, 336–345. [CrossRef]

29. Yu, C.; Liao, R.; Zhu, C.; Cai, X.; Ma, J. Test on the Stabilization of Oil-Contaminated Wenzhou Clay by Cement. *Adv. Civ. Eng.* **2018**, *2018*, 9675479. [CrossRef]

30. Wang, J.; Guo, Y.; Shang, J.Q.A. Portland Cement Stabilization of Canadian Mature Fine Oil Sands Tailings. *Environ. Geotech.* **2021**, *8*, 1–43. [CrossRef]

31. Taylor, P.; Nasr, A.M.A. Utilisation of Oil-Contaminated Sand Stabilised with Cement Kiln Dust in the Construction of Rural Roads. *Int. J. Pavement Eng.* **2014**, *15*, 889–905. [CrossRef]

32. Graettinger, A.J.; Johnson, P.W.; Sunkari, P.; Duke, M.C.; Al-Rawas, A.; Hassan, H.F.; Taha, R. Stabilization of Oil-Contaminated Soils Using Cement and Cement by-Pass Dust. *Manag. Environ. Qual. Int. J.* **2009**, *16*, 670–680. [CrossRef]

33. ASTM. *Annual Book of ASTM Soil and Rock*; ASTM: West Consh, PA, USA, 2017.

34. ASTM D4007-11e1. *Standard Test Method for Water and Sediment in Crude Oil by the Centrifuge Method (Laboratory Procedure)*; ASTM Int.: West Consh, PA, USA, 2016.

35. ASTM D4294-16e1. *Standard Test Method for Sulfur in Petroleum and Petroleum Products by Energy Dispersive X-Ray Fluorescence Spectrometry*; ASTM Int.: West Consh, PA, USA, 2016.

36. ASTM D3230-19. *Standard Test Method for Salts in Crude Oil (Electrometric Method)*; ASTM Int.: West Consh, PA, USA, 2019.

37. ASTM D1298-12b. *Standard Test Method for Density, Relative Density, or API Gravity of Crude Petroleum and Liquid Petroleum Products by Hydrometer Method*; ASTM Int.: West Consh, PA, USA, 2017.

38. ASTM D7042-21. *Standard Test Method for Dynamic Viscosity and Density of Liquids by Stabinger Viscometer (and the Calculation of Kinematic Viscosity)*; ASTM Int.: West Consh, PA, USA, 2021.

39. Head, K.H.; Epps, R.J. *Manual of Soil Laboratory Testing*, 3rd ed.; Whittles Publishing, CRC Press, Taylor & Francis Group: Boca Raton, FL, USA, 2011.

40. Al-Sanad, H.A.; Eid, W.K.; Ismael, N.F. Closure to “Geotechnical Properties of Oil-Contaminated Kuwaiti Sand” by Hasan A. Al-Sanad, Walid K. Eid, and Nabil F. Ismael. *J. Geotech. Eng.* **1996**, *122*, 787–788. [CrossRef]
41. Shin, E.C.; Omar, M.T.; Tahmaz, A.A.; Das, B.M. Shear Strength and Hydraulic Conductivity of Oil-Contaminated Sand. In the Fourth International Congress on Environmental Geotechnics; de Mello, L.G., Almeida, M., Eds.; Balkema Publishers: Rio de Janeiro, Brazil, 2002; pp. 9–13.
42. Ghaly, A.M. Strength Remediation of Oil Contaminated Sands. In The 17th International Conference on Solid Waste Technology and Management; Zandi, I., Mersky, R.L., Shieh, W.K., Eds.; Widener University: Philadelphia, PA, USA, 2001; pp. 289–298.
43. Abousnina, R.M.; Manalo, A.; Shiau, J.; Lokuge, W. Effects of Light Crude Oil Contamination on the Physical and Mechanical Properties of Fine Sand. Soil Sediment Contam. 2015, 24, 833–845. [CrossRef]
44. Wang, L. Cementitious Stabilization of Soils in the Presence of Sulfate. Ph.D. Thesis, Louisiana State University, Baton Rouge, LA, USA, 2002.
45. Prusinski, J.R.; Bhattacharja, S. Effectiveness of Portland Cement and Lime in Stabilizing Clay Soils. Transp. Res. Rec. 1998, 1652, 215–227. [CrossRef]
46. Ural, N. The Significance of Scanning Electron Microscopy (SEM) Analysis on the Microstructure of Improved Clay: An Overview. Open Geosci. 2021, 13, 197–218. [CrossRef]
47. Ergozhin, Y.; Dzhusipbekov, U.; Teltayev, B.; Nurgalieva, G.; Shakirova, A.; Khudaibergenova, K.; Izmailova, G.; Yelshibayev, N. Crude Oil Contaminated Soil: Its Neutralization and Use. Sustainability 2020, 12, 3087. [CrossRef]
48. Bol, P.; Kuany, G.; Zhou, X.; Abdelhafez, I.A.; Abdelhafez, A.A. Oil Contaminated Soil, Global Environmental Impact (Overview). Int. J. Curr. Sci. Eng. 2019, 1, 124–129.
49. Wang, S.; Xu, Y.; Lin, Z.; Zhang, J.; Norbu, N.; Liu, W. The Harm of Petroleum-Polluted Soil and Its Remediation Research. AIP Conf. Proc. 2017, 1864, 020222. [CrossRef]
50. Trejos-Delgado, C.; Cadavid-Restrepo, G.E.; Hormaza-Anaguano, A.; Agudelo, E.A.; Barrios-Ziolo, L.; Loaiza-Usuga, J.C.; Cardona-Gallo, S.A. Oil Bioremediation in a Tropical Contaminated Soil Using a Reactor. An. Acad. Bras. Cienc. 2020, 92, 1–18. [CrossRef]
51. Jabbarov, Z.; Abdrahmanov, T.; Putilov, A.; Kováčik, P.; Pirmatov, K. Change in the Parameters of Soils Contaminated by Oil and Oil Products. Agriculture 2019, 65, 88–98. [CrossRef]
52. Kohli, R.; Mittal, K.L. Developments in Surface Contamination and Cleaning; Elsevier: Amsterdam, The Netherlands, 2019; Chapter three; pp. 23–105, ISBN 9780128160817.
53. Aziz, S.Q. Landfill Leachate Treatment Using Powdered Activated Carbon Augmented Sequencing Batch Reactor (SBR) Process. Ph.D. Thesis, Universiti Sains Malaysia, Gelugor, Malaysia, 2011.