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Assessment of Heavy Metal and Oil-Contaminated Silty Sand Treatment by Electrical Resistance Heating Method

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Featured Application: The feasibility of the electrical resistance heating method developed in this study was evaluated for the remediation of multi-contaminated soils in terms of environmental and geotechnical aspects.

Abstract: The feasibility of the electrical resistance heating method developed in this study was evaluated for the remediation of multi-contaminated silty sand in terms of environmental and geotechnical aspects. The multi-contaminated silty sand sampled in this study was polluted with 21,081 mg/kg of heavy oils, as well as heavy metals. Silty sand, treated using the electrical resistance _heating method was environmentally, as well as geotechnically, compared with the multi-contaminated silty sand in terms of residual concentration, leaching, shear modulus and modified California bearing ratio (CBR). The remediation test was conducted with a target temperature of 700 °C. The removal efficiency of total petroleum hydrocarbon (TPH) was estimated as 99.99% after remediation in 48 h; most of the heavy metals, as some of the contaminants, were isolated as a crystal in treated silty sand without any harmful leakage, and heavy oil was fully extracted with a form of mist and dust. Moreover, it was also geotechnically found that the decontamination process, including the removal of heavy metals and oils, had an effect on the increase in the internal friction angle, shear modulus and modified CBR of treated silty sand. In conclusion, it is shown that the electrical resistance heating method developed in this study is an environmentally and geotechnically effective technology for the recovery of clean construction fill material from hazardous-waste-contaminated silty sand.

Keywords: electrical resistance heating; California bearing ratio; total petroleum hydrocarbon; multi-contaminated silty sand; vitrification

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1. Introduction

Soil contamination has become a global environmental issue, as it has seriously affected both the environment and human health. Despite this, its effect on the geotechnical properties of soil has been investigated, including whether the contaminated soil maintains the geotechnical performances needed for earthwork foundation or fill materials. Some experts have researched several contaminated soil samples with various oil types and concentrations. Shin et al. [1] studied the geotechnical properties (internal friction angle and permeability) of oil-contaminated Jumunjin standard sand of Korea with various relative densities of sand, oil types and concentrations. Some of these studies [2,3] found that the viscosity of oils promotes slippage between the soil particles due to the lubrication of soil particles. Al-Sanad et al. [2] carried out consolidation tests on clean and contaminated sand by heavy crude oil in the amounts of 0, 2 and 6% by weight. It was found that the compression index (C_c) increased with the increase in oil contents.

Shin and Das [3] studied the change in the shear strength and bearing capacity of unsaturated oil-contaminated sand using three types of oil: Oman crude oil, engine oil and lamp oil. They found that an increase in oil contents significantly reduced the bearing capacity and shear strength, with a decrease in the internal friction angle (φ).

Remediation technologies have been developed to treat hazardous contaminated sites because the chemical form of the contaminants can influence their solubility, mobility and toxicity on soil [4]. Remediation technologies are divided into physical, chemical, biological and thermal treatment methods. Each technology has been applied as an in situ or ex situ approach depending on the remediation cost, site conditions and contaminant characteristics.

There are several limitations of physicochemical and biological methods for the remediation of heavy oils, as well as heavy metals. In other words, these methods are less cost-effective and require a long-term period when individually used [5]. However, thermal treatment methods, which heat the contaminated soil regardless of contaminant types, can be applied to multi-contaminated sites [5]. Even though the typical thermal treatment method is technically effective for remediating the contaminated soil, its production and operating costs are much more expensive, and its maintenance requires much attention and labor.

Therefore, in order to overcome these limitations, electrical resistance heating technology, as one of the thermal treatment methods, was developed in this study for the remediation of heavy oils, as well as heavy metals, in multi-contaminated silty sand. An electrical resistance heating device simply consists of a power supply using the flow of AC electricity and a heating resistor. By allowing a high electric current to flow through a resistor installed in the soil, the soil volume is rapidly heated, resulting in an increase in temperature to remediate heavy oils, as well as heavy metals, in multi-contaminated soil.

This paper presents the results of an environmental and geotechnical laboratory experimental program before and after thermal treatment, using electrical resistance heating technology. Finally, it is estimated whether treated silty sand is suitable as a construction fill material based on the effect of decontamination on the geotechnical properties of multi-contaminated silty sand and the recovered geotechnical properties of treated silty sand.

2. Experimental Program

2.1. Experimental Materials

The soil used for this study was a mixture of sludge and typical soil at OO plant, Pohang, South Korea, which was exposed to oil and heavy metal contaminants. The basic properties of the soil used for this study are summarized in Table 1. Specific gravity is 2.56. Particle-size distribution curve is shown in Figure 1. The soil is classified as silty sand (SM) according to the Unified Soil Classification System (USCS). Disturbed soil samples were used for treatment and laboratory tests.

Table 1. Physical properties of multi-contaminated soil.

Item	Unit	Value	References
USCS classification	-	SM	ASTM D-2487 [6]
Optimum moisture contents	%	13.9	ASTM D-698 [7]
Max. dry unit weight	kN/m^3	17.16	ASTM D-698 [7]
Liquid limit †	-	* N.P.	ASTM D-4318 [8]
Plastic limit †	-	N.P.	ASTM D-4318 [8]
Plasticity index †	-	N.P.	ASTM D-4318 [8]
Specific gravity	g/cm^3	2.56	ASTM D-792 [9]
Coefficient of uniformity	-	10.7	ASTM D-6913 [10]
Coefficient of curvature	-	2.3	ASTM D-6913 [10]

* N.P.: non-plastic. † Atterberg limits: liquid limit, plastic limit and plasticity index.

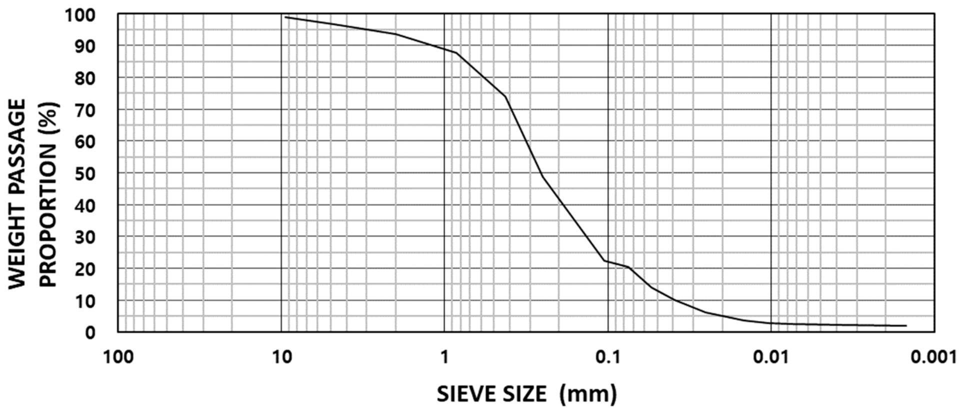

Figure 1. Particle size distribution curve of multi-contaminated soil sample (ASTM D-6913 [10]).

2.2. Analysis of Contaminants

Analysis of oil-contaminated specimens was conducted in accordance with the standard for soil contamination test (KS M ISO 16703 [10]). For pretreatment of the specimen, anhydrous Na_2SO_4 was placed in the oil-contaminated soil (10 g) to maintain the specimen in powder state, and 100 mL of dichloromethane (CH_2C_{l2}) was added. Extraction was repeated twice for 3 min using ultrasonic wave, dichloromethane obtained through the filter was concentrated to 2 mL, and 0.3 g of silica gel was added and mixed for 5 min to eliminate the impurities. Then, the supernatant was analyzed using gas chromatography [11].

Analysis of the pattern of total petroleum hydrocarbon (TPH) contained in the soil was carried out using gas chromatography (Agilent Technologies, 7890 A GC System). The column used was Ultra-2 (DB-1, 10 m × 0.10 mm × 0.10 micron). The initial temperature was set to 50 °C for 2 min and then increased to 320 °C at a rate of 8 °C/min; the specimen was kept at the final temperature for 10 min. The carrier gas was He (1.0 mL/min).

Heavy metals, such as zinc, nickel, cadmium, copper, arsenic, mercury, lead and hexavalent chromium, were analyzed according to the inductively coupled plasma-mass spectrometry method announced by Ministry of Korean Environment [5]. As shown in Table 2, mercury content was 21.63 mg/kg, exceeding the concern level (20 mg/kg), and TPH of oil contaminant was 21,081 mg, which was 10 times the allowed limit of 2000 mg/kg.

Table 2. Contaminants in multi-contaminated soil sample.

Type	Unit	Criteria	Results	Remarks
Copper (Cu)	mg/kg	2000	27.1	
Lead (Pb)	mg/kg	700	39.8	
Zinc (Zn)	mg/kg	2000	243.8	
Nickel (Ni)	mg/kg	500	15.7	
Hexavalent chrome (Cr^{6+})	mg/kg	40	0.8	
Cyanide (CN)	mg/kg	120	55.6	
Mercury (Hg)	mg/kg	20	21.63	Exceeded
Arsenic (As)	mg/kg	200	5.42	
Benzene	mg/kg	3	0.7	
TPH	mg/kg	2000	21,081	Exceeded
Benzo(a)Pyrene	mg/kg	7	0.901	

2.3. Experimental Design

As one of the thermal treatment methods, an electrical resistance heating technology developed for this study allowed for thermal desorption, as well as vitrification of multi-contaminated silty sand by heavy oils and heavy metals.

As seen in Figure 2, by allowing high electric current to flow through a resistor that was located in the soil, the soil volume was rapidly heated, resulting in an increase in temperature, which evaporated volatile (organic) contaminants and isolated (inorganic) heavy metal contaminants as crystal structures (solidification) in soil [5].

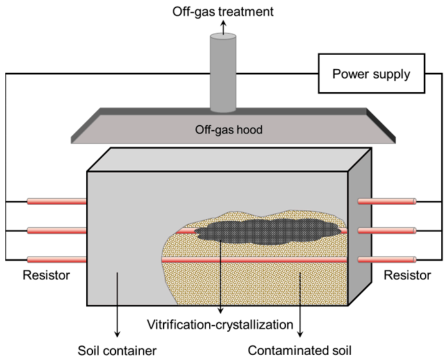

Figure 2. Schematic diagram of electrical resistance heating device.

The remediation experimental device, as shown in Figure 3a, simply consists of a power supply, with a capacity of 10 kW, and a heating resistor using the flow of AC electricity. The commercial voltage was designed to be 220 V/60 Hz. The secondary voltage was designed to ensure safety during the experiment by setting the maximum output voltage to 8 V. The experimental device could control the secondary voltage, from 0 to 8 V, in the primary voltage range of 0–220 V and adjust the input current. In addition, the current could be controlled within a range of up to 1250 A according to the load condition of the secondary current.

(**a**) (**b**) (**b**)

Figure 3. Diagram of (**a**) electrical resistance heating device; (**b**) side view of soil container; (**c**) top view of soil container.

The soil container was made of stainless steel with dimensions of 30 × 80 × 100 cm, and a polycarbonate panel was installed on the front side of soil container to observe internal changes. As shown in Figure 3b, there are holes in which resistors as a heat source can be installed at a horizontal distance of 10 cm and a vertical distance of 20 cm. It was designed to analyze heat transfer and thermal influence range according to the location of the heat source. Moreover, there is an outlet on the top of soil container as shown in Figure 3c, which was designed to discharge the gas generated by the volatilization of contaminants during the thermal treatment process.

An electrical resistance heating test was conducted for 24 and 48 h with a power consumption of 6 kW/h and target temperature of 700 °C. The temperature variations at each section in the soil container were measured using GRAPTHTEC's DATA LOGGER (GL840) and K-type thermocouple, as shown in Figure 4.

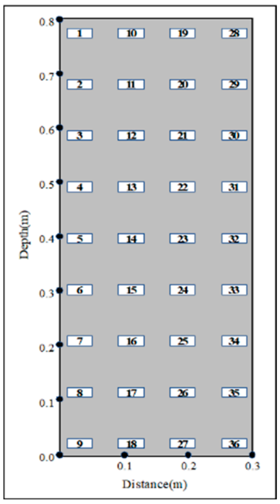

Figure 4. Location of temperature monitoring (side view).

3. Experimental Results

3.1. Soil Remediation Results

The heating element reached the target temperature of 700 °C in only 2 h, as seen in Figure 5. The temperature distribution throughout the remediation process was monitored, as presented in Figure 6. It was observed that the radius of 0.1 m from the heating element was immediately heated, and the heat transfer rapidly occurred in 24 h. Additionally, thermal behavior showed that vapor from the heated soil was released upward, and the heat was transferred in a typical convection direction.

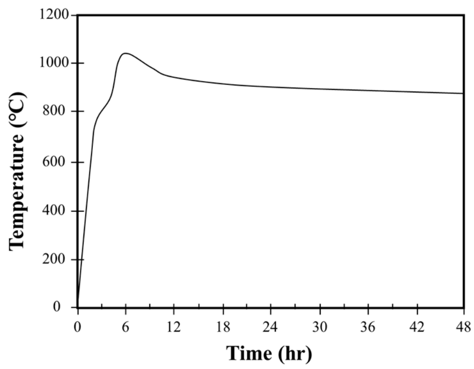

Figure 5. Temperature of heating element.

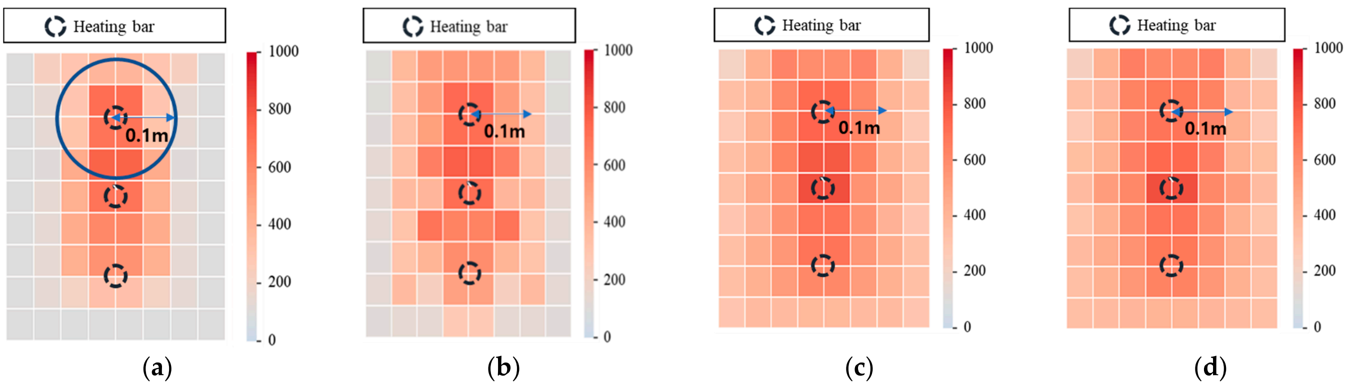

Figure 6. Heat map (side view) after: (**a**) 6 h; (**b**) 12 h; (**c**) 24 h; (**d**) 48 h.

While the temperature of silty sand was increased through thermal conduction, volatile contaminants contained in contaminated silty sand were purified, as seen in Figure 7. The TPH of multi-contaminated silty sand was entirely eliminated by 99% from 21,081 mg/kg, and the mercury was also eliminated below the concern level.

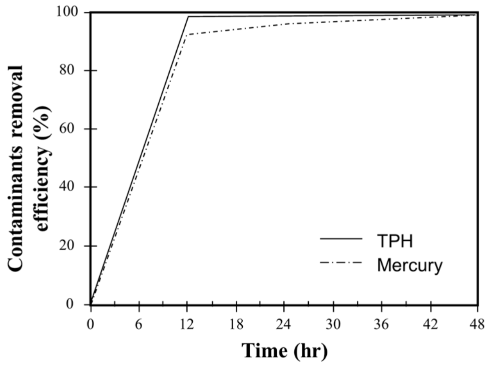

Figure 7. Removal efficiency of TPH and mercury.

Heavy metals, such as copper, lead, zinc, nickel, cyanide, arsenic and chromium, contained in contaminated silty sand were immobilized in the solid. In other words, all of the heavy metals contained in the contaminated silty sand were vitrified by high temperature from the electrical resistance heating technology, as shown in Figure 8.

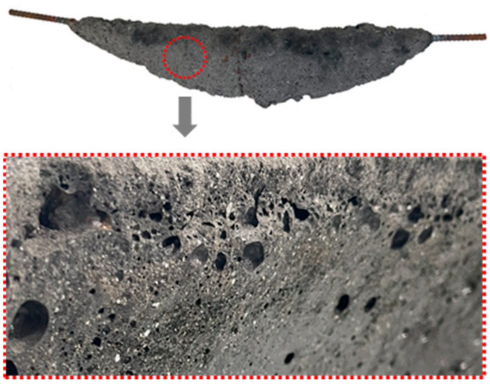

Figure 8. Vitrified heavy metals.

After thermal treatment in 48 h, the leaching of heavy metals, except for lead, was not detected, as presented in Table 3.

Table 3. Residual contaminants in treated soil with remediation time.

Type	Unit	Remediation Time (h)		
		0	24	48
Copper (Cu)	mg/kg	27.1	16.8	* N.D.
Lead (Pb)	mg/kg	39.8	27.3	0.01
Zinc (Zn)	mg/kg	243.8	204.4	N.D.
Nickel (Ni)	mg/kg	15.7	15.0	N.D.
Hexavalent chrome (Cr^{6+})	mg/kg	0.8	N.D.	N.D.
Cyanide (CN)	mg/kg	55.6	0.2	N.D.
Mercury (Hg)	mg/kg	21.63	0.78	N.D.
Arsenic (As)	mg/kg	5.42	5.2	N.D.
Benzene	mg/kg	0.7	N.D.	N.D.
TPH	mg/kg	21,081	N.D.	N.D.
Benzo(a)Pyrene	mg/kg	0.901	N.D.	N.D.

* N.D.: not detected.

3.2. Direct Shear Test

A direct shear test was intended to identify the effect of decontamination on the shear strength of treated soil. This direct shear test was carried out under the consolidated, drained condition in accordance with ASTM D3080 [12], for which the soil sample was compacted with 95% of the maximum dry unit weight (16.0, 15.9 and 16.0 kN/m^3 for soils contaminated and treated for 24 and 48 h, respectively). The void ratios of the tested soils (soils contaminated and treated for 24 and 48 h) were 0.56, 0.58 and 0.57, respectively.

Normal stress (σ) applied to a direct shear test should be designed according to the surface load considered at the construction site. As shown in Figure 9, the direct shear test was conducted with normal stresses of 49, 98 and 147 kPa, at which maximum shear stress was clearly determined as shear strength on a Mohr–Coulomb failure envelope.

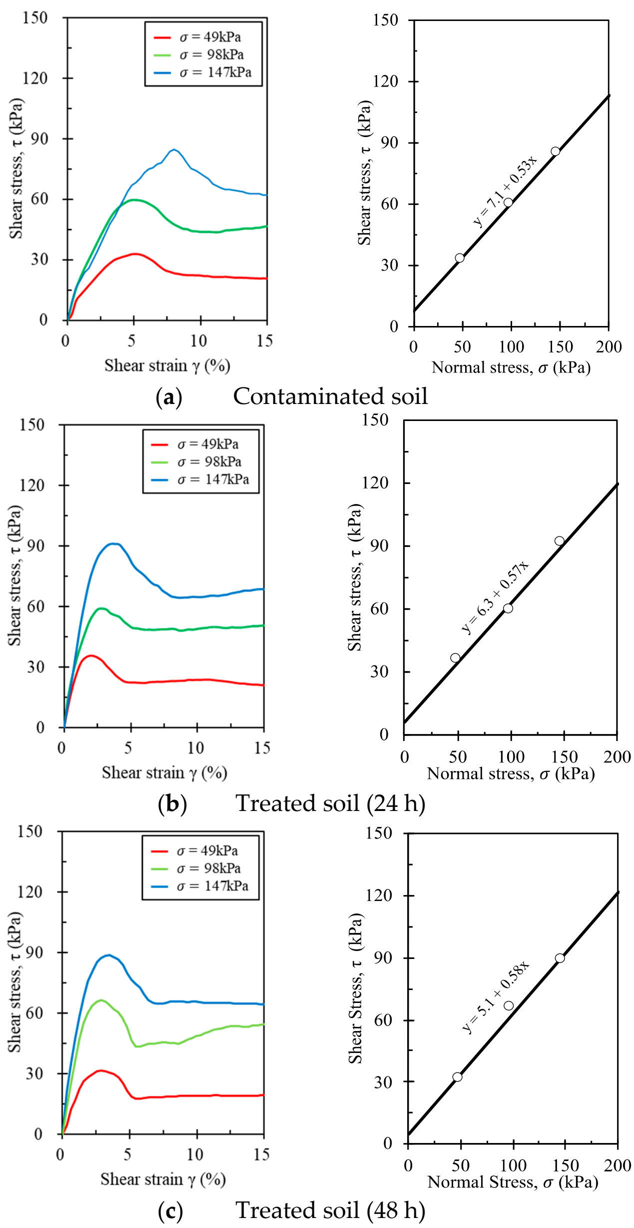

Figure 9. Shear strain–shear stress responses and Mohr–Coulomb failure envelope: (**a**) Contaminated soil; (**b**) treated soil (24 h); (**c**) treated soil (48 h).

As shown in Table 4, the cohesion of contaminated silty sand was 7.1 kPa, and the internal friction angle was 27.9°. The cohesion of remediated silty sand was 5.1 kPa, and the internal friction angle was 30.2° after treatment of 48 h, indicating that cohesion was reduced after remediation, while the internal friction angle was increased. In other words, it was found that while the viscosity according to oil concentration increased, the internal friction angle decreased. As shown in Figure 10, treated soils showed a more obvious dilative behavior under lower shear strain than in contaminated silty sand due to the particle climbing action (interlocking) during shearing.

Table 4. Shear strength of tested soils.

Sample	Cohesion (C)	Internal Friction Angle (∅)
Contaminated soil	7.1	27.9
Treated soil (24 h)	6.3	29.6
Treated soil (48 h)	5.1	30.2

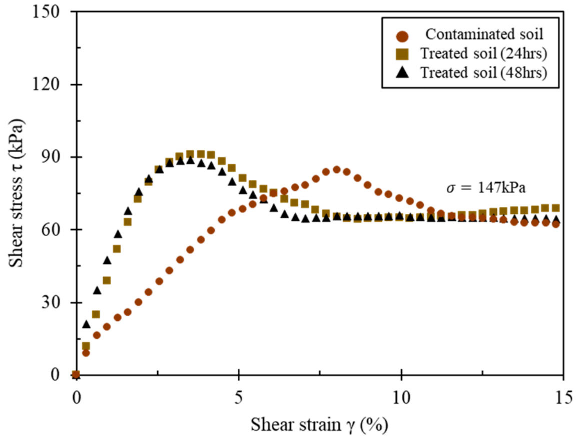

Figure 10. Comparison of shear strain–shear stress responses at σ = 147 kPa.

Moreover, it was found that decontamination caused an increase in the secant shear modulus (G_{sec}), which commonly represents soil stiffness, as shown in Figure 11:

$$G_{sec} = \frac{\tau}{\gamma} \quad (1)$$

Here, τ = shear stress and γ = shear strain.

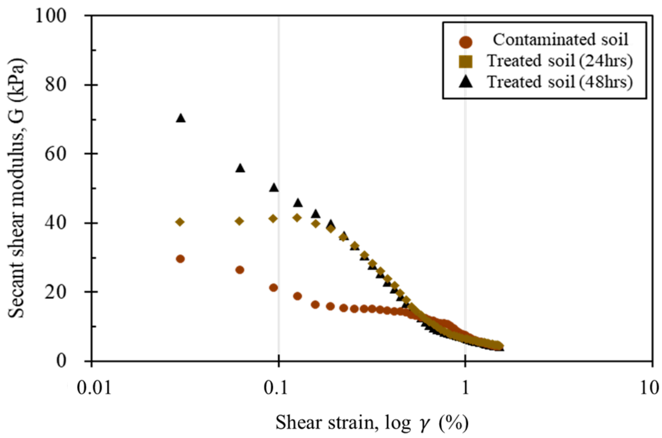

Figure 11. Comparison of secant shear modulus at σ = 147 kPa.

3.3. Modified California Bearing Ratio Test

The CBR test was conducted to determine whether treated soil was viable for use as a fill construction material. The specimen was prepared with the maximum dry unit weight and optimum moisture confirmed from the compaction test, according to ASTM D1883 [13] and KS F 2320 [14].

As shown in Figure 12, the modified CBR is defined as the CBR value in accordance with 95% of the maximum dry unit weight, which represents the field compaction conditions and is recognized as the field compaction maintenance criteria in Korea [14]. The modified CBR of the oil-contaminated soils was significantly decreased compared with the treated soils. Oil-contaminated soil particles tended to easily slide due to the lubrication effect, which may have caused a reduction in the modified CBR values [15]. Treatments carried out for 24 and 48 h increased the modified CBR value to 10.1 and 10.4%, while the value was 9.4% when the silty sand was multi-contaminated. The obtained result was attributed to the reduction in the lubrication effect of oil, which makes the soil a looser material [16]. When comparing these results with the criteria related to road construction fill materials, as shown in Table 5 [17], it was found that the soil treated for at least 12 h with the electrical resistance heating method was feasible for reuse in several applications, such as road subsoil, subgrade, subbase and anti-frost layer materials.

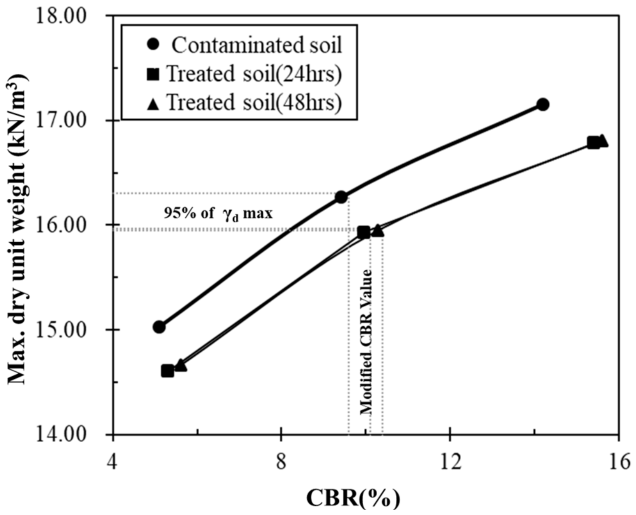

Figure 12. Modified CBR test results.

Table 5. Comparison of tested soils' modified CBR with criteria for road construction fill materials.

Type	Criteria (%)	Contaminated Soil	Treated Soil (24 h)	Treated Soil (48 h)
Subsoil	more than 2.5	satisfied	satisfied	satisfied
Subgrade	more than 10	not satisfied	satisfied	satisfied
Subbase	more than 10	not satisfied	satisfied	satisfied
Anti-frost layer	more than 10	not satisfied	satisfied	satisfied

3.4. Recyclability Assessment of Remediated Soil

It was determined whether the characteristics of the treated soil were suitable for fill materials according to the KS F codes presented in the recycled aggregate quality standards from the Ministry of Land, Infrastructure and Transport [17]. Based on the properties (particle size, Atterberg limits and modified CBR) of the remediated silty sand satisfying the criteria for construction fill material, as shown in Table 6, it was found that the treated silty sand in this study could be recycled and reused as construction fill material by simple electrical resistance heating [17].

Table 6. Assessment of recyclability of remediated soil.

Type	Category	Unit	Criteria	Test Result	Remarks
Fill material	Maximum aggregate size	mm	less than 100	4.75	satisfied
	5 mm Sieve pass rate	%	25~100	99.1	satisfied
	0.08 mm Sieve pass rate	%	0~25	21.7	satisfied
	Liquid limit †	%	less than 50	N.P.	satisfied
	Plastic limit †	%	less than 25	N.P.	satisfied
	Plasticity index †		less than 10	N.P.	satisfied
	Modified CBR		more than 10	10.4	satisfied

† Atterberg limits: liquid limit, plastic limit and plasticity index.

Compared with other thermal treatment approaches from the previous research results in 2016, as shown Table 7 [18], the energy cost of developed electrical resistance heating technology is similar with conventional ex situ thermal desorption methods in spite of remediating the full range of hydrocarbons with a temperature condition higher than at least 500 °C, and it is much cheaper than incineration methods under similar treatment (temperature) conditions.

Table 7. Comparison of remediation costs with thermal treatment methods (based on [18]).

Technology	Removal Mechanisms	Treatment Conditions	Contaminants Targeted	Cost (USD per Metric Ton)
Developed electrical resistance heating	Entrapment in molten glass, desorption, pyrolysis, oxidation	500–1300 °C	Full range of hydrocarbons	USD 35–USD 70 (2021)
Ex situ thermal desorption	Desorption (pyrolysis and oxidation often occur)	Low temp.: 100–300 °C High temp.: 300–550 °C	Volatile and semi-volatile hydrocarbons, creosote, and TPH	USD 46–USD 99 (2016)
Incineration	Oxidation	600–1600 °C	Full range of hydrocarbons	USD 150–USD 2900 (2016)

4. Conclusions

The feasibility of the electrical resistance heating method developed in this study was evaluated for the thermal desorption and vitrification of multi-contaminated silty sand by heavy oils and heavy metals in terms of environmental and geotechnical aspects. The target temperature of electrical resistance heating was designed at 700 °C in order to remediate contaminated soil with heavy metals and volatile organic compounds. The power consumption was set to 6 kW/h, and purification was performed for 24 and 48 h. The treated soil was analyzed through environmental and geotechnical tests in terms of TPH, leaching of heavy metals, shear strength, shear modulus and modified CBR:

(1) Based on the Korean Ministry of Environment standards [5], contaminants, such as heavy oil, organic matter and heavy metal, were not detected; the removal efficiency of TPH was estimated as 99.99% and heavy metals, as some of the contaminants, were isolated as a crystal in a treated soil without any harmful leakage.

(2) It was also geotechnically found that decontamination processes, such as the thermal treatment of heavy metals and heavy oils, had an effect on the increase in the internal friction angle, shear modulus and modified CBR of treated silty sand. The obtained results are attributed to the reduction in the lubrication effect of oil occupied between soil particles.

(3) The physical properties (particle size and Atterberg limits) and improved mechanical properties (internal friction angle, shear modulus and modified CBR) of remediated silty sand satisfied the criteria for construction fill materials.

(4) In conclusion, it was shown that the electrical resistance heating method developed in this study is an environmentally and geotechnically effective technology for the recovery of clean construction fill material from hazardous-waste-contaminated silty sand.

Author Contributions: C.K.: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing—original draft; T.K.: Conceptualization, Resources, Writing—review & editing, Funding acquisition, Supervision, Project administration; D.L.: Conceptualization, Resources, Writing—review & editing, Methodology, Visualization; D.P.: Conceptualization, Writing—review & editing, Supervision, Project administration. All authors have read and agreed to the published version of the manuscript.

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