

Thermal Remediation of Tar-Contaminated Soil and Oil-Contaminated Gravel

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Abstract

Pilot plant studies were carried out to evaluate the feasibility of remediation of coal tar-contaminated soil and oil-contaminated gravel by incineration with environmentally acceptable performance. In addition to adequate decontamination and emission control, effort was devoted to treating the special feedstocks with difficult handling properties by use of conventional combustion and feeding systems. Analysis of the results showed that satisfactory decontamination of the soil and gravel could be achieved and emission of pollutants in the flue gas could be effectively controlled. It would also be possible to recover the heat energy from the incinerated material to make the remediation process more cost-effective.

1. Introduction

Pollutants such as oil and other chemical substances may contaminate large quantities of soil *via* spills, leakage, or other incidents. A relatively recent example was the sinking of the oil tanker Erika off the west coast of France, spilling 14,000 tons of heavy oil. As a result, 250,000 tons of sand contaminated by the heavy oil was collected and transferred for decontamination treatment [1].

For decontamination of contaminated soil, high-temperature treatment is commonly considered, since it has the advantages of reliability, high capacity, and effective destruction of the hazardous material with subsequently reduced long-term liability [2-5]. Any such treatment should ensure thorough destruction of the hazardous substances while avoiding pollution of air, preferably using existing facilities. In the present work we have investigated remediation of soil contaminated by coal tar and gravel contaminated by oil, using a pilot-scale combustor adapted to provide the required treatments.

1.1. Incineration of coal tar-contaminated soil in a moving-bed furnace

Coal tar-contaminated soil has accumulated over several decades in the vicinity of a calcination plant owned by a coal corporation in western Canada. The simplest route for decontamination would be incineration of the soil in the calciner.

The tar-contaminated soil is highly sticky, and cannot be handled directly with conventional feeding and combustion systems. To resolve this difficulty, the soil was mixed with a local lignite coal for the present work. The lignite coal served both to reduce the stickiness and as an auxiliary fuel. Lignite was easily crushed into a suitable size range to ensure good mixing with the tar/soil, and hence reduce the stickiness of the feedstock both in the feeding system and the incinerator. It is believed that in a high-temperature solid bed, the burning of lignite should start first because of its lower ignition temperature (estimated to be below 300°C), thus supplying heat for burning the tar and meanwhile reducing the possibility of clinkering in the incinerator. Additionally, it is expected that the tar is effectively dispersed by the lignite and hence its burning could be facilitated in the co-combustion process. Tests in a modified pilot-scale furnace were carried out to evaluate the effectiveness of this approach, and the potential of decontamination of the soil in a rotary calciner.

1.2. Incineration of oil-contaminated gravel in a fluidized bed combustor (FBC)

Many sites in North America are contaminated by frequent oil spills [6,7]. It is reported that there are about 12 spills of 4,000 liters or more each day in Canada, typically one of which occurs in navigable waters [8]. The government agency under Environment Canada has sponsored an investigation of the use of incineration technology for remediation of oil-contaminated gravel and debris.

Fluidized bed combustion technology can be used for combustion or incineration of a wide range of materials, including heavy residues such as bitumen, tars, pitches, and other oily wastes [9-13]. This technology has been tested for treatment of sludge from the Sydney Tar Ponds [14], whose remediation is an immediate priority for the government of Canada [15]. The technology has also been evaluated, with the support of the government of France, for treating the oil-contaminated sand produced by the previously mentioned shipwreck of the Erika [16].

In the present work incineration of oil-contaminated gravel has been carried out with a pilot-scale FBC, as part of an Environment Canada program. To accommodate oiled shoreline gravel and weathered oiled gravel, the effects of salt water addition and weathering on the incineration have also been studied.

2. Experimental

2.1. Test facilities and methods

A pilot-scale bubbling FBC for general combustion research was used for the incineration of the oil-contaminated gravel (Fig. 1). The unit comprises a combustor, fuel and limestone feed systems, preheat burner, multicyclone dust collector, flue-gas-to-air heat exchanger, baghouse and stack. The feedstocks are charged into a storage bin. The bin is placed on a weigh feeder, which meters and controls the rate of the feed material. The combustor is 5 m high, with an internal cross-section of 0.380 x 0.405 m (0.154 m²). The burner preheats the combustor, using propane, to the ignition temperature of the test fuel. For the oil-contaminated gravel, as the heating value was very low, the propane burner was continuously fired to maintain the combustor temperature. The amount of air supplied to the propane burner was kept to a minimum while allowing the burner to continue to operate. The rest of the air was supplied as fluidizing air through the air distributor at the bottom of the combustor to maintain good fluidization. This resulted in excess oxygen content of about 10% in the flue gas.

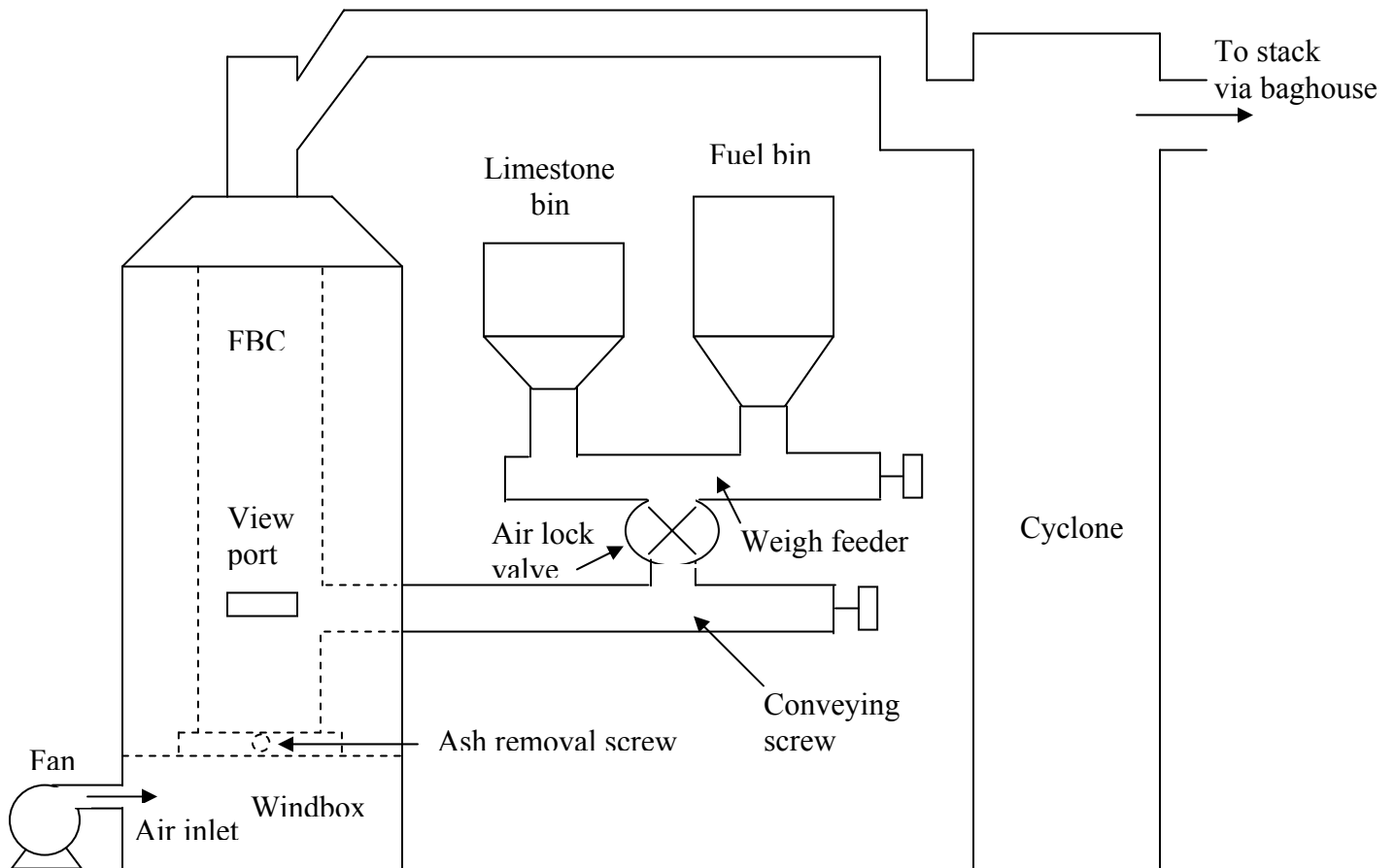


Fig. 1. Schematic of the pilot plant fluidized combustor used for remediation of oil-contaminated gravel.

Temperatures and pressures are measured at strategic locations throughout the unit. On-line continuous monitoring of flue gas concentrations of CO, CO₂, NO_x, SO₂ and O₂ is carried out using a flue gas conditioning system and a bank of gas analyzers. Concentration of total hydrocarbons is monitored by a hydrocarbon analyzer. The sampling port is near the exit of the cyclone. The measurement errors are estimated to be within $\pm 5\%$.

For incineration of the coal tar contaminated soil, the same FBC unit was modified to evaluate the potential of decontaminating this material using a rotary calciner. The modified unit is shown in Fig. 2. The bottom windbox and air distributor section of the FBC were replaced with a sloping grate section. In the incineration tests a solid bed was maintained. The tar-soil was fed above the bed and a mixture of ash and char resulting from the incineration was discharged from the bottom. The burner for preheating the unit and supporting combustion was located above the bed, along with the inlet of combustion air. The burner was shut off when the feed could sustain combustion at the desired temperature. When combustion support was required, fuel oil or propane was fired through the burner.

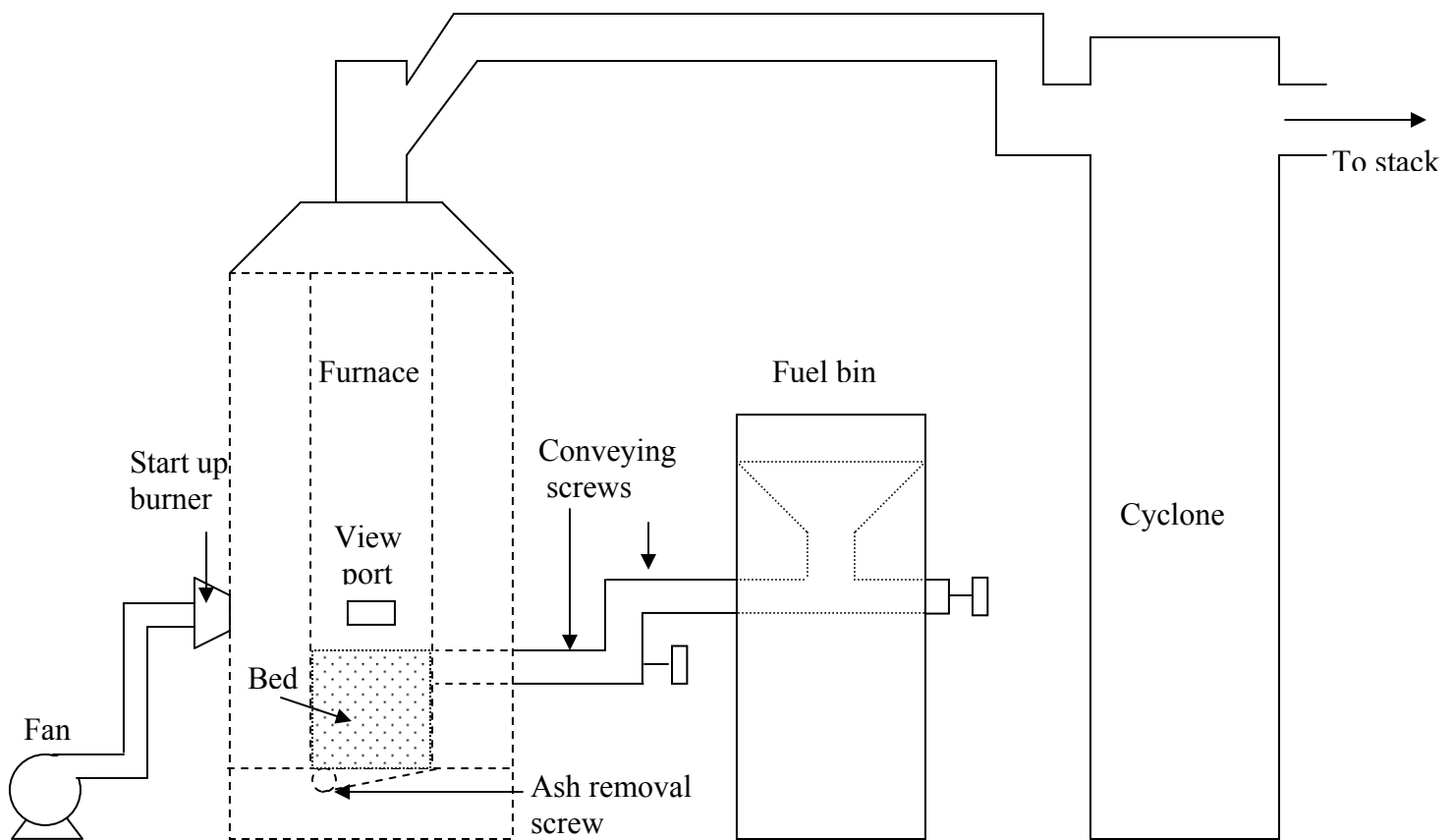


Fig. 2. Modified pilot-scale furnace for incineration of coal tar-contaminated soil.

2.2. Feedstocks of coal tar-contaminated soil

Three batches of coal tar-contaminated soil were provided and the analytical data for the samples are shown in Table 1. The first two batches were rich in coal tar, whereas the third batch contained less tar.

The tar-soil was mixed with lignite in varied proportions as the feed to the furnace. The properties of the lignite are shown in Table 2. Prior to mixing the lignite was dried and crushed. Through testing it was determined that lignite crushed to below 6 mm ensured good mixing.

Table 1. Analytical data for coal tar-contaminated soil samples

	Sample 1	Sample 2	Sample 3
Proximate analysis (wt %)			
Moisture	19.40	13.80	30.20
Ash	22.57	23.96	47.32
Ultimate analysis (wt %)			
Carbon	46.51	51.72	17.20
Hydrogen	3.83	4.47	1.43
Nitrogen	0.58	0.67	0.38
Sulphur	0.56	0.72	0.49
Calorific analysis (MJ·kg⁻¹)	20.13	22.50	6.50

Table 2. Analytical data for the lignite coal (dry basis) which was mixed with the coal tar-contaminated soil

Proximate analysis (wt%)	
Ash	16.72
Volatiles	37.27
Ultimate analysis (wt%)	
Carbon	63.32
Hydrogen	3.86
Nitrogen	1.17
Sulphur	0.63
Oxygen (by difference)	14.30
Calorific analysis (MJ·kg⁻¹)	23.93

2.3. Gravel feedstocks

Three batches of gravel (size range 9.5 to 16 mm) were provided by Environment Canada. In the first batch 2 wt% of fresh Saudi crude oil was added to simulate fresh oil-contaminated gravel. In the second batch 2 wt% of an emulsion of 25% Saudi oil and 75% salt water was added to simulate shoreline gravel contaminated by oil. The third batch was prepared by adding 2 wt% of Saudi oil to the gravel and allowing the material to weather for two weeks before storing in sealed barrels. Each batch was about 800 kg. The analysis of the Saudi oil is given in Table 3. The calorific values of the samples are 0.893, 0.223 and 0.878 MJ/kg for the 2% oil batch, 2% emulsion batch and the weathered batch, respectively.

3. Results and discussion

3.1. Incineration of the coal tar-contaminated soil in the moving-bed furnace

As noted earlier, a major concern was the stickiness of the feed. Effective mixing and handling of the feed were explored as well as the combustion in six test runs. The conditions of the tests and the results of emission measurements and opacity observations for the flue gas are summarized in Table 4. Analytical data for the ashes taken after the incineration tests are shown in Table 5. For the two test runs (Nos. 2 and 4) with lower lignite fraction (50 wt%), the feed was sticky, causing “hanging up” a few times in the feed bin. In particular, in test No. 4 stable conditions in the furnace were not achieved. In this run clinkering occurred and removal of ash and char residue from the furnace bottom was difficult and eventually stopped. All these occurrences were clearly related to the sticky nature of the feed material, and highlighted the importance of reducing the stickiness both in the feeding system and in the incinerator. With higher lignite fractions (75 and 80%), this problem did not occur.

Table 3. Analytical data for Saudi crude oil

	Fresh Saudi oil	Weathered Saudi oil
Ultimate analysis (wt%)		
Carbon	84.8	85.3
Hydrogen	13.0	12.5
Nitrogen	0.1	0.1
Sulphur	1.94	2.29
Oxygen	< 0.5	< 0.5
Calorific value (MJ·kg⁻¹)	44.64	43.92

Table 4. Test conditions and results of flue gas emissions and opacity observation for treating the coal tar-contaminated soil

	Test 1	Test 2	Test 3	Test 4 ^a	Test 5 ^b	Test 6
Feed Rate (kg·h ⁻¹)	23.2	40.0	20.0	32.7	34.5	20.4
Lignite-soil ratio	75/25	50/50	75/25	50/50	75/25	80/20
Gas temperature (°C)	793	966	872	1078	929	872
Bed temperature (°C)	318	204	222	368	121	231
Gas residence time (s)	4.9	3.2	5.1	-	3.7	5.0
Bed depth (mm)	100	200	200	-	130	150
O ₂ (%)	5.0	4.4	4.1	2.5	4.6	5.7
CO ₂ (%)	10.7	12.0	11.7	11.0	11.3	8.3
CO (ppm)	112.0	124.0	186.0	206.0	78.0	130.0
SO ₂ (ppm)	354.0	345.0	388.0	197.0	109.0	214.0
NO _x (ppm)	115.0	166.0	169.0	58.0	151.0	108.0
Hydrocarbons (ppm)	0.0	0.0	4.6	3.5	19.0	10.0
Opacity	very slight to nil	invisible	slight	-	slight	slight

^a During this run the conditions were unstable and clinkering in the furnace occurred. The removal of ash and char residue from the furnace bottom was difficult and eventually stopped; see text.

^b With the batch that was lean in tar content.

Table 5. Analysis of ash samples after incineration of coal tar-contaminated soil

Proximate analysis (wt%)	Test 1	Test 2	Test 3	Test 4	Test 5	Test 6
Moisture	2.23	3.53	0.91	1.55	6.91	4.59
Ash	55.60	39.93	42.90	36.16	41.02	33.02
Volatiles	10.57	20.12	10.49	19.53	24.56	26.35
Fixed carbon	31.60	36.42	45.70	42.76	27.51	36.02

The bed temperature could only be measured locally by thermocouples at the wall of the furnace. Thus, unlike the homogeneous gas temperature, the bed temperature merely reflected the temperature at the thermocouple tip. The apparently low bed temperature may largely be attributed to the thermocouples being shielded or covered by inactive bed material near the wall. In a rotary kiln the temperature of the bed near the wall would be much higher owing to the refractory roof that provides a substantial radiant heat source, and could approach the gas temperature.

With the solid bed configuration of the modified furnace, the major operational conditions that determine the completeness of the burning of the tar would be temperature, feed rate and bed height. As the ash was continuously withdrawn from the bottom of the furnace, the feed moved from the top of the bed to the bottom with the

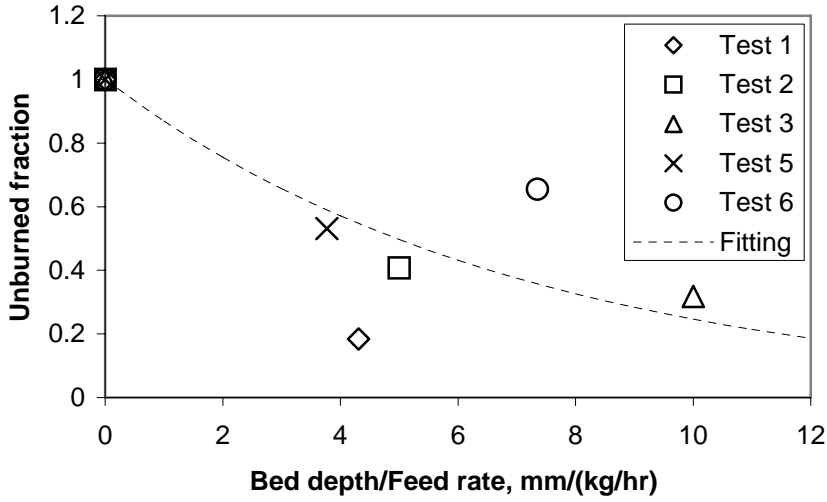


Fig. 3. Unburned fraction of combustibles in the ash withdrawn from the bottom of the furnace as a function of the ratio of bed depth to feed rate. The dashed curve represents the description by Eq. (2).

burning proceeding. As a first approximation, the fraction of unburned combustibles in the bed may be expressed as

$$\frac{dY}{dz} = -\frac{A}{Q} k_s Y \quad (1)$$

where Y is the fraction of the unburned combustibles, z is the bed height, A is the cross-sectional area, and Q is the volumetric flow rate of the solids. k_s is an apparent rate coefficient for the solid phase which is a function of oxygen concentration and temperature, and includes other effects such as porosity or specific area of the aggregate and diffusion. From this relation we obtain

$$Y = Y_0 \exp\left(-\frac{A}{Q} \bar{k}_s z\right) \quad (2)$$

where \bar{k}_s takes the mean value of k_s over the total bed height:

$$\bar{k}_s = \frac{\int_0^Z k_s dz}{Z} \quad (3)$$

Thus, for fixed bed height and feed rate of tar-soil, the unburned fraction of the combustibles depends on \bar{k}_s . When both bed height and feed rate are varied, as is the case in the present study, the unburned combustible fraction can be given as a function of the ratio z/Q , according to Eq. (2). Figure 3 shows the dependence of the unburned fraction, which is calculated from a material balance using the data of the feed and ash,

on z/\dot{M} , where \dot{M} is the mass flow rate of the feed and hence z/\dot{M} is proportional to z/Q . The curve represents the result of fitting Eq. (2) to the data, where \bar{k}_g is taken as an averaged value. With the curve one can estimate the completeness of the burning as a function of residence time of the solids. The estimated range of residence time for the present tests was 20-50 min. In the targeted rotary calciner the residence time of the solids will be 2-3 h, and the temperature of the solids will be much higher owing to the refractory roof, as has been mentioned earlier. Accordingly, the burning of the combustibles is expected to be complete.

For the flue gas, the constituents that are related to incomplete combustion and will not be affected by flue gas cleaning are CO and hydrocarbons. The level of CO is seen to be quite low compared with the regulated upper limit (250 ppm), and hence the major concern is that of hydrocarbons, in the event that it correlates with emissions of toxic polycyclic aromatic hydrocarbons (PAH). From Table 4 it can be seen that, for the present system, there is no apparent relation between CO and hydrocarbon concentrations. An examination of the data suggests that the hydrocarbon concentration is not strongly dependent on gas temperature in the present system. The concentration should be a function of gas residence time, and also appears to be dependent on the unburned fraction of the fuel, which is indicative of inefficient combustion. To project the hydrocarbon level in the rotary calciner, we assume plug flow for the gas and express the concentration of the hydrocarbons as a function of the gas residence time

$$\frac{dC}{dt} = -k_g C \quad (4)$$

where C is the hydrocarbon concentration, t is the residence time, and k_g is a coefficient for the gas phase reaction, which includes the effect of oxygen concentration. It should be noted that when the flow deviates from plug flow, the present treatment leads to a lower value of the rate coefficient estimated from the data. This will result in a more conservative projection of the decontamination rate, as can be seen later. From Eq. (4) we obtain

$$C = C_0 \exp(-\bar{k}_g t) \quad (5)$$

where \bar{k}_g is the mean value of k_g defined in a similar way to Eq. (3). C_0 is the initial concentration, which is assumed to be proportional to the unburned combustible fraction of the solids, following the earlier discussion. Thus the hydrocarbon concentration can be given as

$$C = \gamma F \exp(-\bar{k}_g t) \quad (6)$$

where γ is the proportionality coefficient and F is the unburned fraction, which is shown in Fig. 3. According to this relation, there should be a linear relationship between $\ln(C/F)$

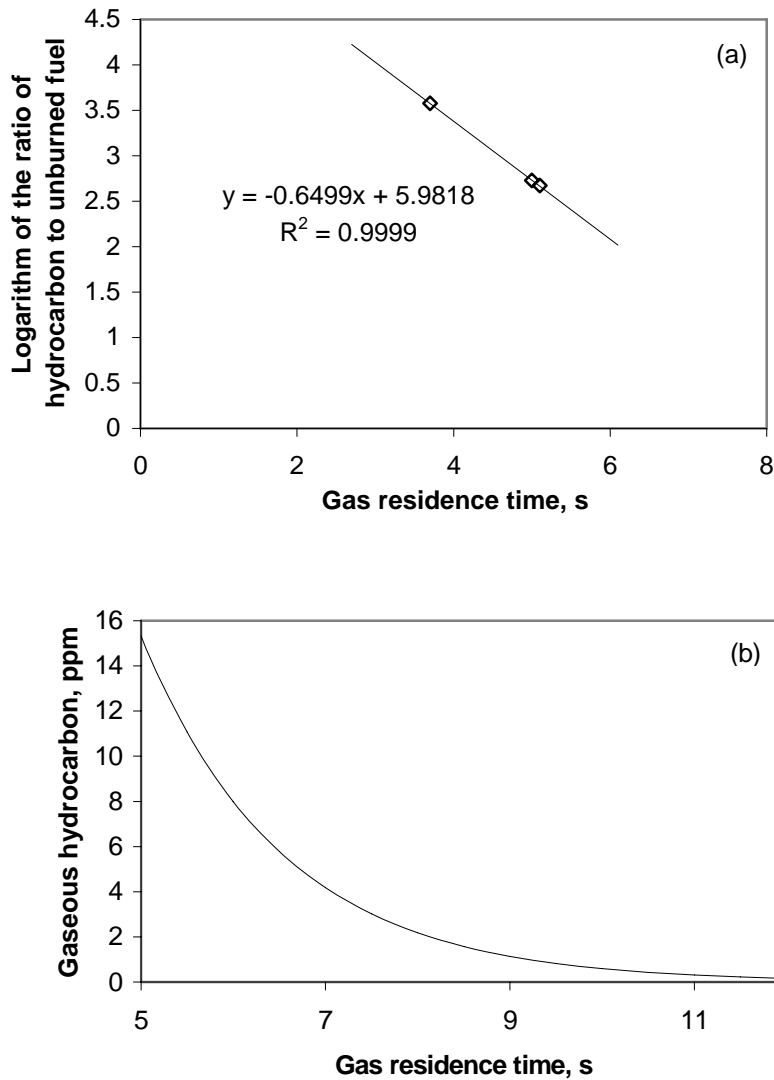


Fig. 4. (a) Logarithm of the ratio of gaseous hydrocarbon to unburned combustibles in the solids as a function of gas residence time. The equation is for the trend line and R^2 is the square of the correlation coefficient. (b) Projected gaseous hydrocarbon concentration, based on the information from (a), as a function of the gas residence time.

and the residence time t when \bar{k}_g is treated as a constant, which takes a mean value over the measured temperature range. A plot of $\ln(C/F)$ against t for the available data of unburned gaseous hydrocarbons is given in Fig. 4a, showing good linearity. From the slope and the intercept of the line the values of \bar{k}_g and γ are evaluated in terms of Eq. (6). Fig. 4b shows the projected hydrocarbon concentration in terms of Eq. (6) for the upper-limit case $F = 1$. It can be seen that for $t > 10$ s, which is expected in the targeted calciner, the projected hydrocarbon concentration is below 0.5 ppm. In the calciner the gas temperature can also be higher, giving rise to faster burning rate and lower

Table 6. Test conditions and results of emissions measurements and oil residue analysis for incineration of the oil-contaminated gravel

	Test 1	Test 2	Test 3
Feed	gravel/fresh oil	gravel/oil emulsion	weathered gravel/oil
Average bed temperature (°C)	877	840	824
Feed rate (kg·h ⁻¹)	50	50	50
Superficial gas velocity (m·s ⁻¹)	2.2	2.1	2.0
O ₂ (%)	10.4	12.0	11.8
CO ₂ ^a (%)	14.7	16.0	15.4
CO ^a (ppm)	50	56	71
SO ₂ ^a (ppm)	100	119	119
NO _x ^a (ppm)	56	59	66
N ₂ O ^a (ppm)	0.52	0.59	0.57
PAH ^b (ng·m ⁻³)	636	1022	755
PCDDs/PCDFs ^b (TEQ, pg·m ⁻³)	3.28	4.85	4.09
Oil remaining on gravel (by Xylene extraction)	Nil	Nil	Nil

^a Values normalized to 3.5% oxygen

^b Sampled and measured by Environment Canada

hydrocarbon concentration. With more complete combustion, the opacity of the flue gas, which was not significant in the present study, would be further reduced.

Thus, based on the above results, decontaminating the soil in the targeted calciner with environmentally acceptable performance should be achievable. It should be noted that, although recovery of the energy from the co-combustion of the tar and lignite has not been evaluated in the present study, it is in principle possible and this could make the remediation process more cost-effective.

3.2. Incineration of the oil-contaminated gravel in the FBC

Three tests, one for each batch of the oiled gravel, were carried out under essentially the same conditions. No major difficulty was encountered in conveying the material through the feeding system, although the feedstocks jammed several times in the air lock valve of the feeder. In addition, defluidization occurred to a bed height of 200 mm by large gravel particles that accumulated in the lower section of the bed. In a full-scale unit the air lock valve would be much larger and such blockage would be avoided (new designs are available for feeding and removing noncombustibles up to 10 cm in size [17], which will also solve the defluidization problem). Alternatively, the air lock valve may not be needed if overbed feeding is used.

The test data are given in Table 6. A xylene extraction method was used to determine whether any oil remained on the gravel. The results showed that the gravel was

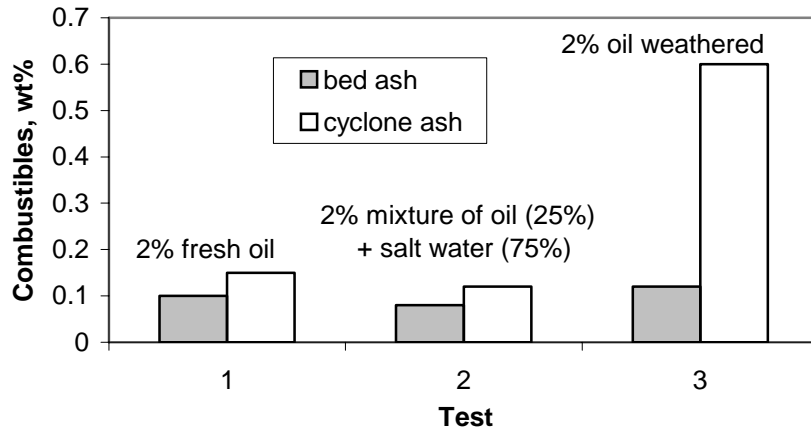


Fig. 5. Content of combustibles in the incineration ashes of oil-contaminated gravel.

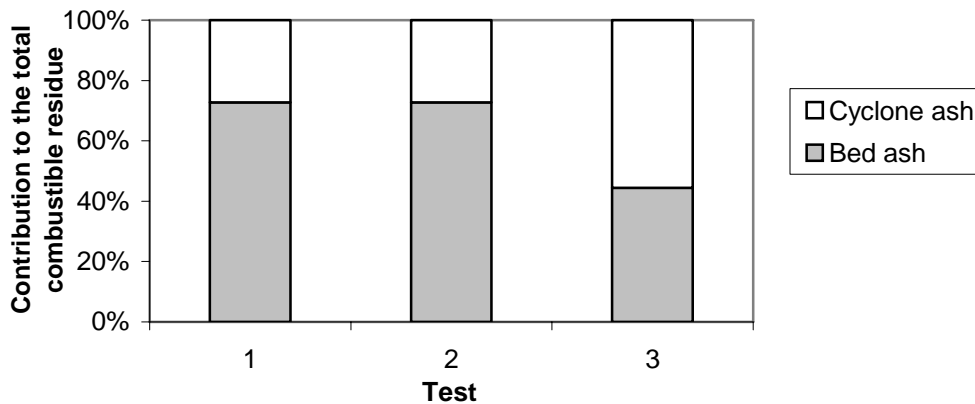


Fig. 6. Contribution from bed ash and cyclone ash to the total combustible residue in incineration of oil-contaminated gravel. (Test 1 = 2% fresh oil; test 2 = 2% mixture of oil (25%) and salt water (75%); test 3 = 2% weathered oil)

effectively decontaminated and ready for reuse or return to the natural environment. The samples of the solids removed from the combustor and the cyclone ash were also analyzed (ASTM D3174-73 method) for combustibles. The results indicated 0.1, 0.08 and 0.12% combustibles in the bed ash and 0.15, 0.12 and 0.6% combustibles in the cyclone ash for the three tests, respectively, as shown in Fig. 5. This level of combustibles is not considered to be problematic from the point of view of disposal of the gravel, but analysis of the results can provide valuable information on the effect of the contaminant characteristics. The combustible content in the cyclone ash is notably higher than in the bed ash, and can be attributed to the fact that cyclone ash is generally finer and has shorter residence time in the combustor. The combustible content in the fly ash of the weathered sample is particularly high, indicating a strong effect of the weathering. The contributions from the bed ash and cyclone ash to the total combustible residue are shown in Fig. 6. Whereas in the fresh oil batch and oil-emulsion batch the contribution from the cyclone ash was lower, in the weathered batch the contribution from the cyclone

ash is higher by 10% than that from the bed ash, suggesting the importance of fly-ash capture in the incineration of weathered oiled gravel.

It is noteworthy that the decontamination rate of the sample containing salt water appears to be substantially lower. The rate is evaluated by taking the fluidized bed as a backmix reactor, for which the rate can be given as

$$R = \dot{M}(X_0 - X_{ash})/V \quad (7)$$

where \dot{M} is the feed rate, X_0 and X_{ash} are the weight fractions of combustibles in the feed and the bed ash, respectively, and V is the bulk volume of the bed. Using the data of Fig. 5, relative decontamination rate in the bed based on Eq. (7) is calculated and shown in Fig. 7. It can be seen that the rate for the salt water-containing batch was only 22% of the rate for the fresh-oil batch. Moreover, PAH emissions from this batch are significantly higher (Table 6).

The mechanism of the effect of salt water on the decontamination rate is not clear. Chlorides have been found to interfere with CO oxidation [13,18-20], but this effect does not appear to be operative for the present system, as is seen from the data in Table 6. The weight fraction of the salt water was 1.5%. As the specific weight of gravel is high (about twice of that of lignite), the volume fraction of the water is higher and this might play a role in lowering the oil burning rate. Another contributing factor could be that oil penetration into the micropores of the gravel was facilitated by water, particularly when dissolution of the oil was enhanced by heating. Although elucidation of the mechanism is not in the scope of the present work, the observed effect of the salt water on the decontamination rate is significant and should be considered when setting operating conditions for remediation of shoreline gravel contaminated by oil.

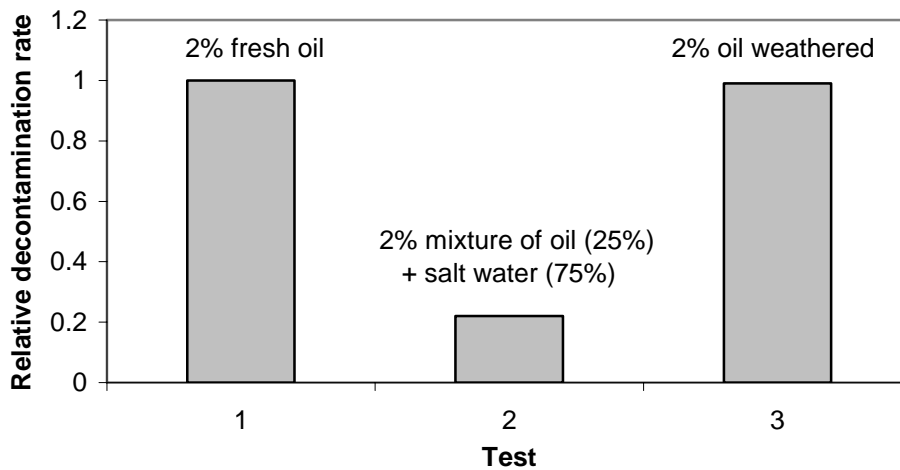


Fig. 7. Relative decontamination rate in the bed in incineration of oil-contaminated gravel. The rate of test 1 (2% fresh oil) is taken as the reference value.

The emissions of CO, NO_x, SO₂ and N₂O shown in Table 6 were all well below Canadian environmental guidelines. The emissions of PAH and polychlorinated dibenzo-dioxins and polychlorinated dibenzo-furans (PCDDs/PCDFs) in terms of their toxic equivalency quantities (TEQ), which were measured by Environment Canada, were also well below the values of government guidelines (5 µg/m³ for PAH and 80 pg/m³ for PCDDs/PCDFs for hazardous waste incineration facilities), indicating satisfactory emissions performance.

There are commercial incineration processes for remediation of polluted soils, particularly for PCB (polychlorinated biphenyls) contaminated soils [23, 24]. Our results should be informative to the application of the technologies to treating materials similar to those studied in the present work

4. Conclusions

The remediation tests targeted treatment of two types of contaminated materials using two representative incineration technologies—rotary kiln and fluidized bed [21, 22]. In both cases promising results were obtained.

Lignite can be used to reduce the stickiness of coal tar-contaminated soil, so that the incineration can be carried out with conventional combustion and feeding systems. Based on the results of the incineration tests with the present facilities, complete burning of the combustibles in both solid and gas phases using a rotary calciner is expected.

Incineration results of the oil-contaminated gravel show that the gravel used in the present study was effectively decontaminated in the FBC. However, it is noteworthy that weathering of the gravel could lower the decontamination rate, particularly in the fly ash. Furthermore, addition of salt water resulted in significantly lowered decontamination rate. This effect would be important in remediation of oil-contaminated shoreline gravel.

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