

# Coal agloflotation: a remediation technique for cleaning of town gas soils

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## Abstract

The use of a “clean” fuel and the treatment of contaminated soils are two factors that can greatly improve the environment. Coal, which occurs in great abundance in the USA, is widely used both in the small-scale domestic and in the large-scale industrial market. Apart from high sulfur content, a high ash content of coal is undesirable. Soils contaminated with polycyclic aromatic hydrocarbons (PAHs) are a potential threat to health and the environment and need to be treated. The present study investigates the use of a hydrophobic coal surface for the adsorption and removal of oily/tarry contaminants such as PAHs from town gas soils. The agloflotation process uses a coal–soil–water mixture to achieve simultaneous removal of mineral matter from coal. The cobeneficiation achieves 90% and higher decontamination for soil and 40% and higher ash removal from coal.

*Keywords:* Agloflotation; Clean soil process; Coal beneficiation; Polycyclic aromatic hydrocarbon

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## 1. Introduction

Soils contaminated with toxic organic compounds are a potential environmental hazard to the global community. For decades, organic pollutants trapped in the soil matrix have leached out and migrated, ultimately making their way to ground water aquifers. The ground waters then carry these toxins through the ecological system, bringing them into the food chain, which is a matter of grave concern because many of these compounds are suspected to be carcinogenic and mutagenic in nature and have a high bioaccumulation potential [1].

One major source of such organic pollutants has been the coal gasification facilities

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which have existed since the 1850s. A typical process for manufactured gas, also known as town gas, used the reaction of coal or coke with air and steam in large, oven-like chambers. The gas plants, commonly called town gas works, produced large quantities of byproducts including complex mixtures of coal tars, sludges, oils, and other chemicals from early gas-scrubbing systems. Residues of coal tars and other undesired effluents were then disposed of in unlined pits on site, in ponds, or as landfills [2,3].

The composition of coal tar, although vastly variable, usually contained polycyclic aromatic hydrocarbons (PAHs) such as benzo[*a*]pyrene, naphthalene, phenanthrene, and benz[*a*]anthracene, phenolic compounds, light aromatics such as benzene, toluene, xylenes, miscellaneous organics such as dibenzofuran, and small quantities of such inorganic chemicals as iron, lead, copper, zinc, various sulfides, cyanides and nitrates [4,5]. Several studies have indicated that many of these organic pollutants, especially PAHs, pose a serious threat to the environment and human health and thus make the immediate cleanup of such contaminated sites imperative. Several technologies have been studied for the remediation of hydrocarbon contaminated soils, including physico-chemical processes such as washing and leaching, incineration, biological treatment and other miscellaneous techniques. However, the application and success of most of these methods are highly site-specific [6,7].

Another issue of great concern is the use of cleaner and environmentally acceptable solid fuel; this has focused much attention on methods of coal cleaning, as coal is widely used for many household and industrial applications. Coal is one of the vital natural resources that could replace imported energy supplies and is the key to a smooth transition from our present petroleum based energy economy to renewable, non-polluting energy sources in the 21st century. Coal beneficiation involves removing such contaminants as ash-forming minerals, sulfur, and other undesirable admixtures from coal without substantially changing coal properties [8].

Coal agglomeration, which utilizes the principle of coal–oil agglomeration, takes advantage of the hydrophobic nature of the coal surface and the essentially hydrophilic nature of the mineral matter particles. Because of its characteristic hydrophobicity, the surface of coal particles can be easily wetted by a non-polar liquid such as oil. When oil is added to an aqueous suspension of finely ground coal, followed by vigorous agitation of the coal–water–oil mixture, the coal particles become coated with thin layers of oil and are bonded together to form agglomerates. These agglomerates can easily be separated from the mineral impurities remaining in the water phase. Coal agglomeration is an efficient method of soil remediation which can be utilized successfully to decontaminate soils contaminated by polycyclic aromatic hydrocarbons (PAHs) [9].

The agglomeration process was initially developed by the Alberta Research Council and later modified for soil cleaning application. This technology known as the “Clean Soil Process” has been licensed to Thermo Design Engineering of Canada by EPRI. Thermo Design Engineering has built a 200 ton per day mobile commercial unit which is now ready for demonstration [10].

### *1.1. Process mechanism*

The process is based primarily on abrasion mass transfer and can be divided into four stages, as shown in Fig. 1 [11,12]. The first stage is the layering. Here the coal and soil

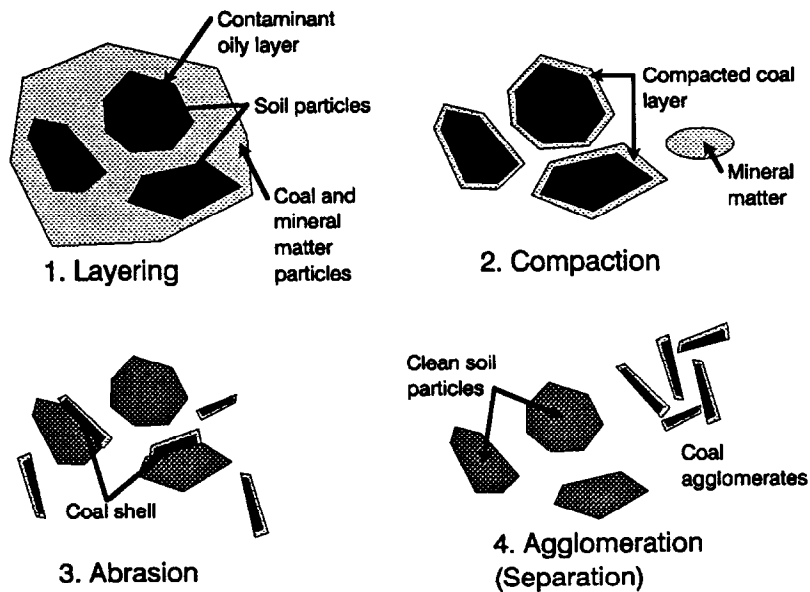


Fig. 1. Process mechanism.

particles are gently tumbled in a tumbler where the coal particles stick around the contaminated soil particles. This stage is completed when all oil-containing surfaces are covered with coal particles. The next stage is compaction, and here the slurry mixture undergoes compaction or reduction in void volume, changing the oil saturation in the coal layer. At this stage the strength of the coal particles adhering to each other increases so that a strong and hard coal layer is formed around the soil particles. It is during these two stages that the contaminants get adsorbed on to the coal particles and the soil is cleaned.

The next stage is the abrasion stage, where the coal and the soil particles are physically separated by high shear stripping. The slurry mixture is transferred from the tumbler to the flotation cell. The mixture is then subjected to high shear mixing, where the bonded coal particles (which are now oil rich) are liberated. Finally comes the agglomeration stage. This is an important stage which helps to separate the coal particles easily. This is done by high shear mixing and by the addition of oil to increase the agglomeration of the coal particles. This improves the ease of handling of the final product [11,12].

## 2. Materials and methods

### 2.1. Soil samples

Experiments were conducted using contaminated soil samples obtained from industrial sites. The soil was drained to remove excess water, air-dried, and sieved to remove

twigs, rocks, and other miscellaneous objects. The soil was then hand-mixed thoroughly to attain homogenization. To determine the contaminants and their concentrations in the soil, several sample replicates were taken from the homogenized soil and subjected to Soxhlet extraction using methylene chloride. The soil extract was then concentrated using a rotavapor assembly. The dried extract was resuspended in a 50:50 methylene chloride:methanol mixture or in toluene for analysis. The contaminants in the soil extract were characterized using high performance liquid chromatography.

## 2.2. Coal samples

Midwestern bituminous coals, Indiana No.5 (Petersburg) and Indiana No.5 (Hawthorne), were used. These contained approximately 4.5–7% sulfur. The coal samples were dried and sieved into sizes ranging from –35 to 150 Tyler mesh.

## 2.3. Flotation cell

The standard Wemco flotation cell (Model 71260-01) used was a 5.5 l polymeric mechanical cell with a side projection for easy froth removal. A rotor and stator dipping into the center of the cell from an overhead support framework imparted high shear to the slurry under operating conditions and also had an air trap attached to it to aid froth formation. This avoided the need for a separate air sparger and the associated power costs. The cell was operated in batch mode.

## 2.4. Experimental

A typical run was initiated by mixing 500 g of air-dried contaminated soil with 200 g of pulverized Indiana No.5 coal in the 5.5 l tumbler. Next, 1 l of hot water (80°C) was added to the coal–soil mixture and the resulting suspension was subjected to tumbling at 60 rpm for 5 min. Witcodet 100 frother was added to the slurry mixture [12,13] and the

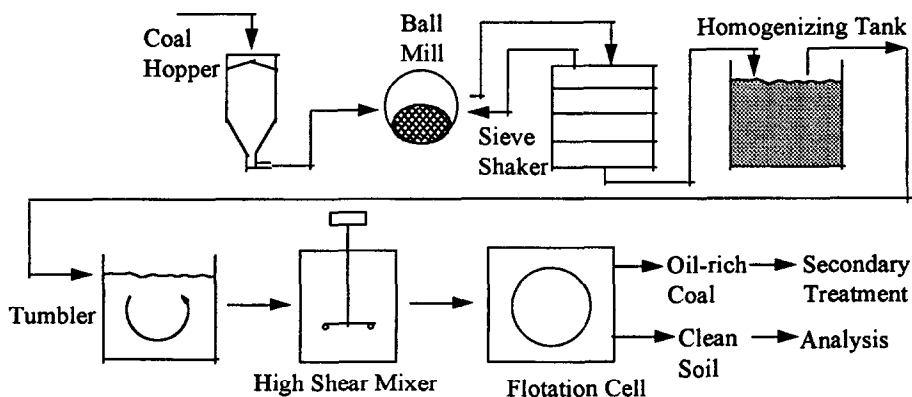


Fig. 2. Process flowsheet for coal agglomeration.

slurry was charged to 1500 rpm in the flotation cell and then held at 1000 rpm for 10–20 min. Air was sparged into the mixture at the end of treatment at a rate of  $\approx 3 \text{ l min}^{-1}$  to produce froth, which was skimmed off. The lighter coal particles adhered to the froth and were thus skimmed off. A process scheme is shown in Fig. 2 [12].

### 2.5. Proximate analysis

A Fisher Coal Analyzer (Model 490) was used to determine the ash, volatiles, and fixed carbon. Raw coal and treated coal were analyzed to determine the performance of the process in terms of the recovery of contaminants and of ash reduction.

### 2.6. HPLC analysis

Methylene chloride extracts of soil samples obtained after Soxhlet extraction were concentrated in a rotavapor and these were analyzed qualitatively and quantitatively using HPLC. A Hewlett-Packard HPLC 1050 instrument with a UV–VIS detector was fitted with a capillary column (VYDAC PAH column, No. 79918PAH-584EA). The samples were analyzed using a gradient method with acetonitrile and water. A standard Supelco PAH test mix was used for calibration.

## 3. Results and discussion

### 3.1. Soil contaminant characterization

The soil analysis using GC/MS and HPLC indicated that the contamination level varied from 3% to 16% (w/w). The soil contained mainly polyaromatic compounds such as benz[*a*]anthracene, chrysene, phenanthrene, and benzo[*a*]pyrene, among other hydrocarbons. Table 1 lists the contamination levels in the three soils used for this study. These soils were received in five-gallon containers and were labeled Site 1-1, Site 1, and Site 2 respectively. A preliminary GC/MS analysis of the soils showed that the soil typically contained substituted alkanes ( $C_{10}$  and higher), substituted aromatic hydrocarbons and polycyclic aromatic hydrocarbons (PAH). Further contaminant characterization was carried out by high performance liquid chromatography, and the PAH standard test mix from Supelco was used to determine the concentrations of characterized contaminants. Table 2 lists typical concentrations of various HPLC-characterized contaminants

Table 1  
Total contamination in soil samples

Soil source	Concentration (wt%)
Site 1-1	16.05
Site 1	3.37
Site 2	14.78

Table 2  
HPLC-characterized contaminants

Compounds	Concentration ( $\mu\text{g g}^{-1}$ in soil)
2-Fluorophenol	3.862
Acenaphthylene	7.601
Acenaphthene	15.493
Fluorene	8.947
Phenanthrene	2.689
Anthracene	0.364
Fluoranthene	2.676
Pyrene	1.939
4-Terphenyl- $\text{d}_{14}$	1.558
Chrysene- $\text{d}_{12}$	1.432
Benz[ <i>a</i> ]anthracene	1.450
Chrysene	1.947
Benz[ <i>b</i> ]fluoranthene	3.332
Benz[ <i>k</i> ]fluoranthene	1.632
Benz[ <i>a</i> ]pyrene	1.494
Dibenz[ <i>ah</i> ]anthracene	1.418
Benz[ <i>ghi</i> ]perylene	1.059
Indeno[123- <i>cd</i> ]pyrene	0.412

Note: Site 1-1 was used.

commonly found in the soils under study, although the concentrations varied for different soils.

### 3.2. Effect of agglomeration time

The soil decontamination and ash removal from coal were found to vary significantly depending on the mixing time used during agglomeration. Table 3 lists the various levels of decontamination and ash removals obtained using different mixing times. It is seen that both the decontamination efficiency and the ash removal increased rapidly during 20 min. Beyond 20 min, the increase in decontamination efficiency with time was very gradual and varied for different soils. Increased agglomeration time also enhanced the separation of contaminant-rich coal from soil and recovery of the coal in the liquid phase. A 90% or higher contaminant removal was achieved for all the soils at 20 min residence time, with a corresponding ash removal of 40% for coal.

Table 3  
Process performance with time

Time (min)	Coal recovery (%)	Ash removal (%)	Decontamination efficiency (%)
2	25	15	–
5	60	25	70
10	64	36	78
20	74	40	92

### 3.3. Effect of coal to soil ratio

The contaminant removal depends on the coal surface area available for contaminant adsorption. A high coal to soil ratio provides more coal surface for PAH enrichment, but it also increases the power requirement for slurry mixing and coal–soil separation. It was of interest to determine the coal to soil ratio that would maximize the contaminant removal. To this effect, three coal to soil ratios were used, namely, 10, 25 and 40% on a weight basis. The contaminant removal attained for these ratios is shown in Fig. 3.

It is seen from the results that an optimum decontamination of 80% or higher was obtained for all the PAHs in the soil. The optimum removal occurred at a coal to soil ratio of 25%. Addition of more coal actually decreased the contaminant removal. This may occur because the excess coal quantities result in coal particle clumps, which effectively reduce the surface area available for contaminant adsorption.

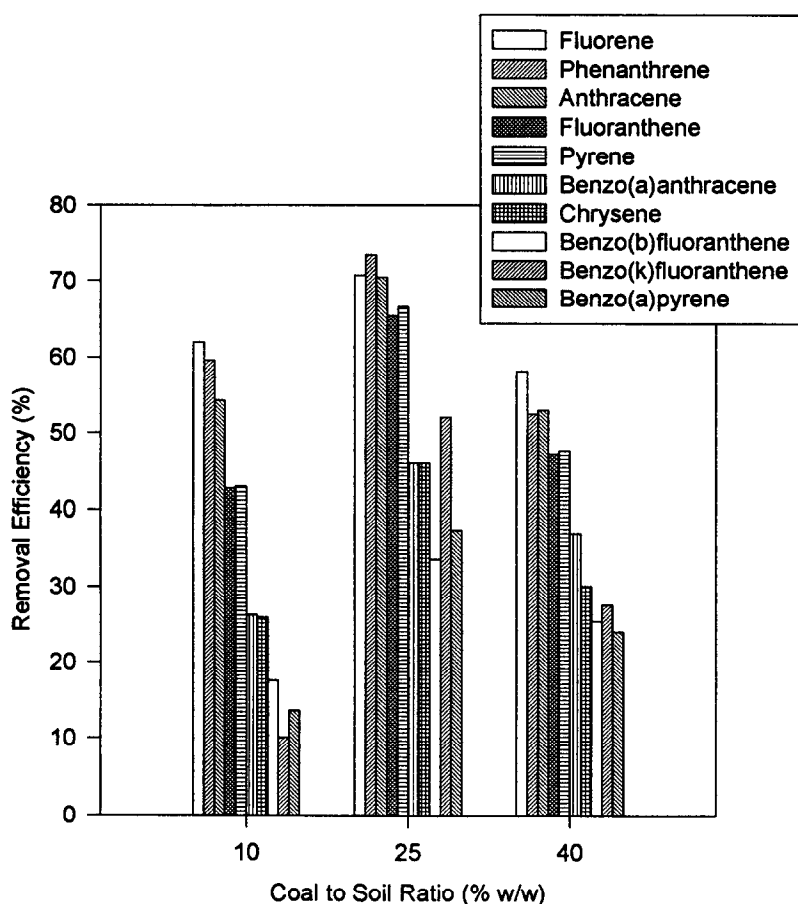


Fig. 3. Effect of coal to soil ratio on contaminant removal.

Table 4  
Effect of impeller speed on decontamination efficiency

Impeller speed (rpm)	Removal (%)
600	72
1000	95

Table 5  
Agglomeration process efficiency on various soil samples

Soil source	Contaminant concentration (wt%)		Removal (%)
	Original soil	Treated soil	
Site 1-1	16.05	0.558	96.5
Site 1	3.37	0.394	88.3
Site 2	14.78	0.780	94.7

### 3.4. Effect of impeller speed

As the separation of agglomerated coal contaminants from soil is achieved by shear forces, impeller speed was considered as a parameter of interest. The experiments were conducted at two impeller speeds, 600 and 1000 rpm, and the results are shown in Table 4. It is seen that higher impeller speed does give a higher decontamination efficiency. This further indicates that induction of high shear, either via increased impeller speed or by improved mixer design, is a key factor in enhancing the performance.

### 3.5. Process performance

The contaminant removal from soil and ash removal from coal for various soils are shown in Table 5. It is seen that a 95% contaminant removal was obtained for both Site 1-1 and Site 2 soils. The slightly lower removal efficiency for Site 1 is due to high chars and clay content in the soil.

Table 6 shows the proximate analysis results obtained using the two midwestern

Table 6  
Beneficiation of Midwestern coals

	Indiana No.5 Petersburg			Indiana No.5 Hawthorne		
	Raw	Treated	Removal (%)	Raw	Treated	Removal (%)
Ash	41.92	25.16	18	14.12	8.62	39
Total S /%	7.970	5.240	34	4.450	3.600	19
Pyritic /%	4.415	2.099	52	2.709	1.700	37
Sulfate S /%	2.277	1.696	26	1.400	0.669	53
Organic /%	1.278	1.445	–	0.331	1.321	–



coals, before and after treatment. It is seen that ash removals of 20% and 40% were obtained for Indiana No.5 Petersburg and Hawthorne, respectively. A 20–50% removal of pyritic and sulfate sulfur was achieved, but the organic sulfur content of the coal showed an increase after treatment. This is not clearly understood but one possible reason could be that the free sulfur might react with the tar contaminants present in the soil and may be induced into the coal matrix. This needs further investigation and is not the focus of this work. Overall, the coal agglomeration process did achieve coal beneficiation mainly via ash removal and also via removal of pyritic and sulfate sulfur.

#### 4. Conclusions

It is seen that the coal agglomeration process can effectively remove contaminants from soils and also beneficiate coal by ash removal. A 95% or higher contaminant removal was achieved for town gas soils contaminated with PAHs and other hydrocarbons. The decontamination efficiency was decreased for soils containing higher amounts of clay and chars. A 40% ash removal was achieved using Indiana No.5 Hawthorne coal. To obtain shorter residence times, an improved design of mixer and flotation cell to enhance coal–soil separation is desired.

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