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The Characterization of Charcoal and High-Density Carbon Pellets Produced from Douglas-Fir Bark

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ABSTRACT

High-density carbon pellets (HDCP) produced from Douglas-fir bark could provide an alternate source of carbon for industry. The production of HDCP in vertical retorts is discussed. Scanning electron microscopy (SEM), atomic absorption spectroscopy (AAS), carbon elemental analysis, and other analytical methods were used to characterize HDCP. HDCP produced from Douglas-fir bark in this work have 90% fixed carbon, an average density of 1.3 g/mL, and 1.18% average ash content with negligible metal impurities. These pellets should be particularly suited for use in the production of adsorbents, high-grade carbons, reductants, carbon black, carbon electrodes, and activated carbons.

Index Entries: Carbonization; charcoal; carbon pellets; pyrolysis; wood bark.

INTRODUCTION

Bark from Douglas-fir (*Pseudotsuga menziesii* [Mirb.] Franco) is a major byproduct of the lumber and plywood industries of the Pacific Northwest. The potential annual supply from Washington and Oregon sawmills is estimated at three million tons (*1*). Effective ways of using this enormous

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quantity of bark would not only solve the disposal problem but also would have economic advantages. The fuel value of bark is 40,000 kJ/kg, compared with 56,500 kJ/kg for coal and 88,000 kJ/kg for petroleum (2). The low energy content and low density of bark limit its general use for fuel. However, converting bark to charcoal by destructive distillation reduces its volume considerably. Compaction of charcoal to HDCP increases its density, making transportation easier and more economical. HDCP could be used to meet future demands for charcoal as reductants in metallurgical processes or as adsorbents in chemical processes. (The calorific value/unit weight of HDCP is greater than that of bituminous coal [3]).

The production of charcoal by destructive distillation of wood or wood wastes has been practiced for centuries. The simplest method uses a primitive underground or pit kiln, and the most advanced involves the use of a rotary kiln where charcoal is produced continuously. Recently, attempts have been made to use the pyroligneous liquor and tar oils generated from the pyrolysis of wood materials as a combustible oil. Although the oil has an energy content of about 23,000 kJ/kg, difficulties are experienced because of the highly oxygenated nature of the oil (*4*). Pyrolysis of bark has long been studied (*5,6*) as a possible method to convert bark into commercially useful organic chemicals, but a better understanding of the physical, mechanical, chemical, and thermal properties of bark components is needed. The principal objective of our work was to examine the nature and characteristics of charcoal produced from pyrolysis of bark and of HDCP produced from the subsequent densification of charcoal.

EXPERIMENTAL METHODS

Production of Charcoal

Douglas-fir bark was dried for 5 h until it reached a constant weight, and then weighed prior to pyrolysis. Twenty grams of oven-dry powdered bark with particle sizes ranging from 0.5 to 2 mm were loaded into an aluminum carbonizer (95 mm diameter × 245 mm length). Asbestos packing ensured an airtight seal between the carbonizer and the lid. The open end of the pipe connection from the lid was connected to a water-cooled copper condenser. The carbonizer, which was heated externally by an electric heater provided with a controller, was insulated with asbestos sheets and kept in a laboratory hood (for safety reasons). Processing was conducted at 500°C for 65 min on oven-dry bark, except when the effect of temperature, moisture, or time was being studied. Uncondensable gases were allowed to escape from the water-cooled copper condenser with no attempts made to analyze or to recirculate the gases, because data are already available about their chemical composition and uses (*7–9*). Cubes of oven-dry wood, approximately 2.5 cm/side, were also carbonized and cooled for 1 h before SEM studies.

Production of HDCP

Bark charcoal samples from several carbonization experiments were collected, oven dried for 2 h, and powdered in a mortar. Different particle sizes (75–300 μm) were collected and stored in airtight polyethylene bags. The pyroligneous liquor that was collected in sample bottles was kept undisturbed for 8 d. The dark brown viscous bottom layer (liquid tar) was separated and used as a binder to produce HDCP. Samples of bark charcoal powder, with particle sizes ranging from 75 to 300 μm, were mixed with liquid tar in varying proportions at room temperature and cold-pressed into pellets (12 mm diameter × 10 mm height). Pressure and hold-time were varied. Pellets were dried at 102°C for 2 h. The average densities of pellets were then determined.

Other Procedures

pH of Charcoal and HDCP

Ten grams of oven-dry charcoal were placed in a flask with 100 mL of boiled reagent water and refluxed for 15 min. The particles of carbon were filtered. The filtrate was cooled to 50°C, and its pH was measured. The procedure was repeated for oven-dry HDCP.

Equilibrium Moisture Content

The equilibrium moisture content was determined by subjecting samples of bark, bark charcoal, and HDCP to a range of humidity conditions at a constant temperature of 25°C.

Impregnation of Douglas-Fir Bark with Inorganic Chemicals

Oven-dry bark samples were impregnated with the inorganic chemicals NaCl, KCl, $ZnCl_2$, $CaCl_2$, Na_2CO_3, K_2CO_3, $CaCO_3$, NaOH, KOH, and $Ca(OH)_2$ (as aqueous solution) and then carbonized as described above to study the effect of secondary char formation.

Analytical Methods

Microstructure of Douglas-Fir Bark Charcoal

Series of Scanning Electron Micrographs (SEM) were taken of uncarbonized and carbonized bark. Specimens of oven-dried bark were stored in airtight polyethylene bags from 2 to 10 d. Some samples were stored for over 6 mo. Bark charcoal samples processed at 400 and 575°C were selected for SEM studies. Specimens carbonized at 575°C were sufficiently conductive and required no special coating or preparation before being placed in the scanning electron microscope (AMR, Bedford, MA, Model 1000A). Because neither bark nor charcoal specimens at 400°C were conductive, they were coated with metal (60% gold and 40% palladium) in a vacuum evaporator. The specimens were cut in an appropriate plane with a sharp blade and mounted on the specimen holder.

Analysis of Bark Charcoal and HDCP

Moisture, volatile matter, ash, fixed-carbon content, and density were determined by American Society of Testing Materials methods (*10–12*). If ASTM methods for charcoal were not available, appropriate methods for coal were adopted. Gas yields were determined by difference. Proximate analyses of HDCP were conducted by ASTM methods (*11*).

Analysis of Ash

Representative samples of oven-dry charcoal or HDCP were ground to <250 μm. Crucibles containing 1 g of each sample were placed in a cold muffle furnace and heated gradually so that the temperature reached 500°C in 1 h and 750°C in 2 h. Each sample was ignited at 750°C, cooled, and ground in a mortar to <74 μm. It was then reheated at 750°C for 1 h, ignited, and cooled in a desiccator. The procedure was repeated until a constant weight of ash was obtained. The resulting ash was analyzed by AAS following the proper ASTM method (*13*) to determine the Fe, Ca, Na, Mg, and K contents. The procedure was repeated for HDCP. The same procedure was followed for 2 g ground bark, particle size <425 μm.

RESULTS AND DISCUSSION

In this study, experimental data were collected to quantify the effects of pyrolysis temperature and time, and the initial moisture content of the bark, on the yield of charcoal. The data presented here are sufficient to determine the feasibility of utilizing bark to produce HDCP, but further work is required to characterize the bark charcoal for the production of other products, such as activated carbon.

Our results indicate that fixed-carbon content increases as pyrolysis temperature and time increase, but that yield of charcoal decreases drastically at higher temperatures. Pyrolysis time has very little effect on charcoal yield. Pyrolysis of oven-dry bark carried out for 65 min at 500°C yielded 35.1% (wt) charcoal, 34.5% (wt) pyroligneous liquor, 12.5% (wt) liquid tar, and 17.9% (wt) uncondensable gases (calculated by difference). Some properties of bark charcoal and HDCP are given in Table 1.

The most important point illustrated by Table 1 is that compaction into pellets considerably increased the density. The addition of tar as binder decreased the fixed-carbon content, because the tar contained a smaller percentage of carbon than did the charcoal. The volatile matter was higher for the HDCP, because the tar binder contained volatiles, whereas the charcoal did not. The difference in ash content should be negligible because the charcoal contained little ash, and the amount of binder used was small. The pH of the HDCP was lower because the tar binder undoubtedly contained some acids.

Table 1
Comparison of Properties of Charcoal and HDCP Produced from Douglas-Fir Bark Carbonized at 575°C

Property	Charcoal	HDCP
Density	1.13 g/mL	1.30 g/mL
Fixed carbon content	92.0% max	90.2% max
Volatile matter	6.2% min	8.0% min
pH	6.2 min	5.9 min

The average density of HDCP was 1.3 g/mL. This makes them easier than charcoal to transport at much lower costs, provides uniformity in composition, and increases energy content/unit volume.

Effect of Carbonization Variables

Temperature

Ovendry Douglas-fir bark, heated out of contact with air, decomposed into solids, liquids, and gases. Pyrolysis at 400°C yielded 41.2% (wt) bark charcoal, 33.5% (wt) pyroligneous liquor, 9.1% (wt) liquid tar, and 16.2% (wt) noncondensable gases. At 575°C, charcoal yield was only 33.1% (wt), whereas liquid tar yield increased to 13.0% (wt), pyroligneous liquor to 34.8% (wt) and noncondensable gases to 19.1% (wt) (Table 2). The carbonization time was kept constant at 65 min in all the experiments. The effect of temperature on shrinkage of Douglas-fir bark during carbonization was not studied in detail. However, we observed that at 400°C, the relative volume decreased to 48% of its original volume, and at 575°C, was down to 35% of its original volume.

Although Table 2 shows an apparent increase in metal content of the charcoal with increasing temperature, the amount of metal (and ash) present was essentially constant when weight loss of the substrate from pyrolysis was taken into consideration. This clearly indicates that inherent metals do not undergo changes during pyrolysis.

The proximate analyses of charcoals produced at different temperatures from 400 to 575°C showed that charcoal with a fixed-carbon content of 90% (wt) could be obtained at 575°C. Increase in pyrolysis temperature increased the ash content of the charcoal, but this increase, which resulted from burning off the organic matter, was marginal (Table 2).

Comparisons of SEM micrographs of bark before and after pyrolysis at different temperatures are difficult because of the complex anatomy of bark. Fig. 1A shows typical parenchyma cells in a cross-section of bark before pyrolysis. Figs. 1B and 1C are representative of bark after pyrolysis at 400 and 575°C. Bark structure was recognizable at 400°C, but was somewhat modified at 575°C. Figure 1D indicates the appearance of crystalline

Table 2
Effects of Carbonization Temperature on the Pyrolysis of Douglas-Fir Bark with a Carbonization Time of 65 min

	Carbonization temperature, °C							
	400	425	450	475	500	525	550	575
Product	Product yield, g/100 g bark							
Charcoal	41.2	39.6	38.3	36.7	35.1	34.3	33.6	33.1
Liquor	33.5	33.9	34.1	34.3	34.5	34.6	34.7	34.8
Tar	9.1	9.9	10.4	11.5	12.5	12.8	12.9	13.0
Gases	16.2	16.6	17.2	17.5	17.9	18.3	18.8	19.1
	Composition of charcoal, g/100 g charcoal							
Fixed carbon	83.6	84.6	85.1	86.3	87.7	88.2	88.5	90.0
Ash (excluding Ca, K, Na, Mg)	0.62	0.67	0.72	0.84	0.92	1.01	1.04	1.18
	Metal content of charcoal, ppm							
Ca	225	229	232	236	240	243	246	250
K	48	50	51	53	55	56	58	60
Na	35	38	40	42	43	46	47	50
Mg	20	21	24	26	28	30	33	35

material in a charred specimen at 575°C. Figure 1E shows typical sieve areas on a longitudinal surface of a sieve cell. The pores in the sieve areas appear a bit broken up or enlarged, but not too seriously. This fracture might have occurred as a result of increased pyrolysis temperature. Figure 1F indicates the formation of bubble-like structures in the parenchyma. Such bubble-like structures were not present in the specimens before pyrolysis. They were observed in specimens at 400°C, but were more predominant in charred specimens of 575°C, as seen in Fig. 1F. These bubble-like structures could be extractive depositions (on prenchyma walls) that have been affected by temperature.

Time

Short pyrolysis periods produced higher yields of charcoal (33.5% (wt) in 45 min) and lower yields of gas (19.3% (wt) in 45 min), whereas longer pyrolysis periods produced lower yields of charcoal (29.7% (wt) in 125 min) and higher yields of gas (22.7% (wt) in 125 min) (Table 3). Therefore, a short pyrolysis process is desired if the final product to be produced is charcoal. If gas is regarded as the main product, a long pyrolysis process should be adopted.

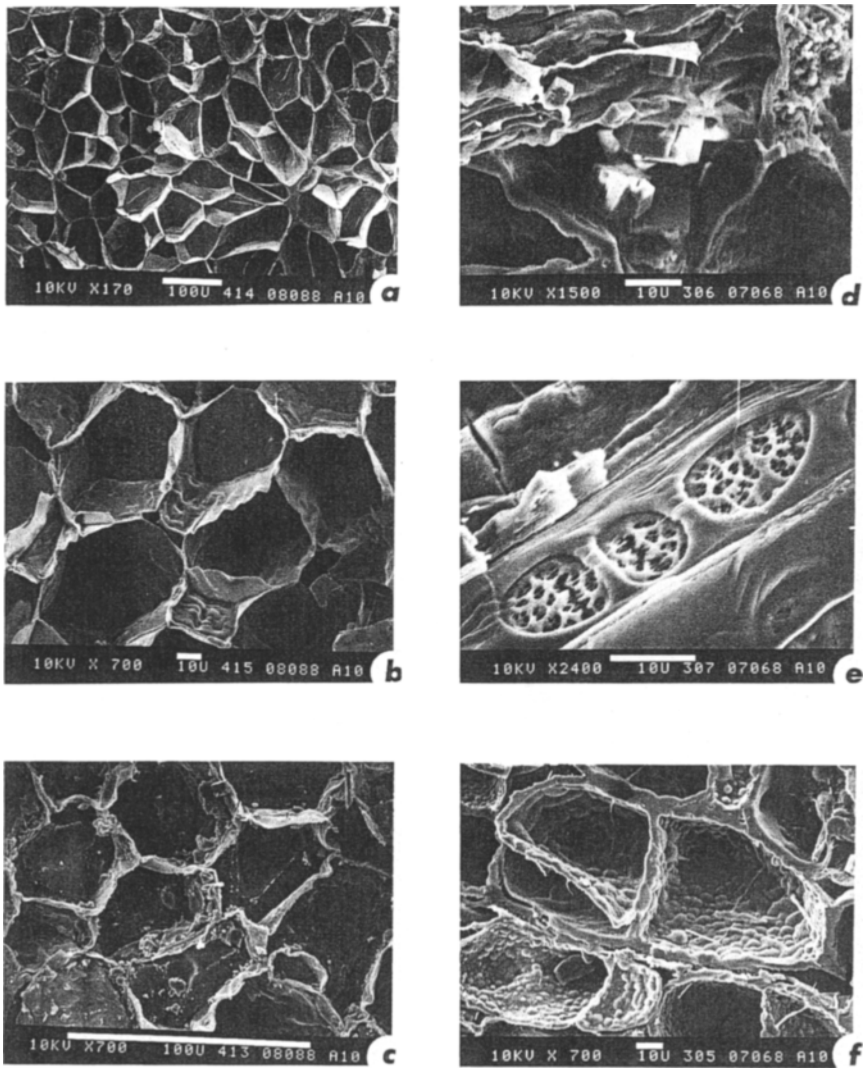

Fig. 1. SEM micrographs of Douglas-fir bark. A) Before pyrolysis. B) Carbonized at 400°C. C) Carbonized at 575°C. D) Presence of crystalline material in charred specimen at 575°C. E) Fractured sieve cells in charred specimen at 575°C. F) Presence of bubble-like structure in the parenchyma of a charred specimen.

Other Factors

Moisture Content

Pyrolysis on six bark samples with moisture contents from essentially zero (oven-dry) to 50% (wt) were carried out. Higher charcoal and lower gas yields are obtained with bark that is more moist (Table 4). The percentage of moisture present in the source material is an important factor

Table 3
Effect of Pyrolysis Time on Product Yield

Pyrolysis time, min	Product yield, g/100 g bark Charcoal	Liquor + tar	Gases
45	33.5	47.2	19.3
55	32.7	47.2	20.1
65	32.0	47.3	20.7
85	31.2	47.4	21.4
105	30.6	47.4	22.0
125	29.7	47.6	22.7

Table 4
Effect of Moisture Content of Bark on Product Yield at a Carbonization Temperature of 575°C

Moisture content, %	Product yield, g/100 g bark Charcoal	Liquor	Tar	Gases
50	34.4	35.6	12.4	17.6
40	34.2	35.5	12.6	17.7
30	34.1	35.5	12.6	17.8
20	33.6	35.2	12.8	18.4
10	33.3	35.0	12.9	18.8
Oven-dry	34.4	35.6	12.4	17.6

Table 5
Equilibrium Moisture Content at 25°C as a Function of Relative Humidity

Relative humidity, %	Equilibrium moisture content, % Bark	Bark charcoal	HDCP
30	9.6	6.9	5.1
50	11.7	7.3	6.2
65	13.1	8.2	7.0
90	27.3	10.1	8.3

in design considerations of equipment used for pyrolysis. A detailed economic analysis is required to determine the optimum extent of drying of the bark prior to carbonization.

The equilibrium moisture content of bark, HDCP, and bark charcoal is a function of the relative humidity of the ambient air. The variations are large, and hence significantly affect net mass (Table 5). Relative humidity is an important property that needs to be considered when purchasing bark and packaging HDCP.

Bark Treated with Inorganic Chemicals

Charcoal yields from carbonization at 500° for 65 min of various bark samples impregnated with inorganic chemicals were found to be higher than those from untreated samples. This appears to be caused by the enhancement of secondary charring reactions of heavy primary volatiles (*14*).

Cooling Rate

Bark charcoal can be cooled either by quenching or by slow cooling out of contact with air. Quenching the bark charcoal leads to surface cracking, resulting in uneven distribution of pore size, from micro- to macropores. Slow cooling results in only micropores.

PROPOSED PROCESS FOR THE UTILIZATION OF DOUGLAS-FIR BARK

Process Description

The steps in the manufacture of HDCP from Douglas-fir bark are illustrated in Fig. 2. The raw material can be obtained from sawmills and plywood mills, or directly from the forest. Procuring and processing bark from the forest usually costs more, but the choice rests on availability as well as economic considerations. The bark from sawmills and plywood mills can be stored until needed. The size of the bark from the sawmills and plywood mills is expected to vary from 1 to 50 mm diameter.

The first step in the proposed process for the utilization of Douglas-fir bark is the removal of unbound moisture in a hydraulic press. Next, much of the bound moisture is driven off in a drier that utilizes a hot gas stream. After being pulverized, the bark is made into briquets in an extruding machine, resulting in additional moisture removal. The dimensions of the extruded material are 100 mm diameter and 1200 mm length, and the moisture content is about 12%.

The bark briquets are then carbonized in vertical retorts. The hot gas can be mixed with air, combusted, and recycled to the bark drier. Liquid drained from the retorts contains two immiscible phases that can be separated into pyroligneous liquor and tar streams. The liquor can potentially be used as a liquid fuel or torched. Some of the heavy tar is used in the next step, but the rest must be burned or disposed of as a hazardous waste. The desired product of this step in the process is charcoal having a high fixed-carbon content.

Next, the charcoal is mixed with some of the carbonization tar, which acts as a binder. The charcoal/tar mixture is then sent to hydraulic multi-orifice extrusion presses operating at 250 psia, where 40 mm diameter × 40 mm long cylindrical pellets are produced. Some of the combustible gases from the vertical retorts can be used to dry the HDCP in tray driers, thus reducing air pollution.

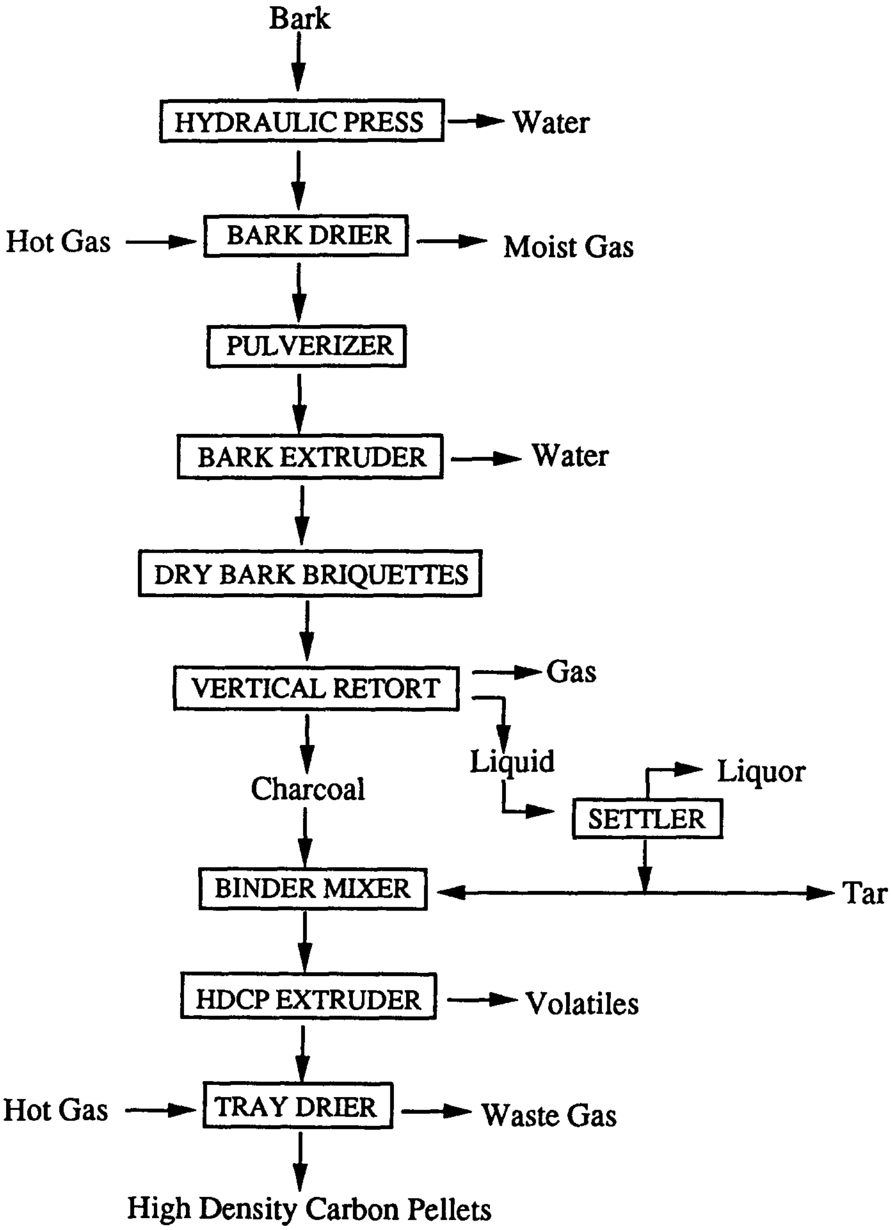

Fig. 2. Proposed bark-utilization process.

Finally, the desired product, dry high-density carbon pellets, exits the tray driers. Since the purpose of these tray driers is to heat-cure the pellets while driving off as much of the volatile matter (resulting from the tar used as binder) as possible, the waste gas stream from this equipment must be treated (e.g., torched) to minimize hydrocarbon pollution to the atmosphere.

Economic Considerations

The most important factor in the production of HDCP is the yield of charcoal. The yield is inversely related to the fixed-carbon content. In other words, the higher the fixed-carbon content, the lower will be the yield of charcoal. Lowering the fixed-carbon content would increase the yield, but would degrade the quality of the charcoal. For the best balance of the economics of the process, previous experience suggests that a charcoal yield of 35% be used. This should be attainable at a carbonization temperature of 500°C for 65 min (Table 2), which should give a fixed-carbon content of 85–90%. Even with the temperature variations that occur in vertical retorts, we should be able to obtain charcoal with a fixed-carbon content of at least 80%.

Environmental and energy considerations are also important. We anticipate that the combustible gases and pyroligneous liquor from the carbonization step (vertical retort in Fig. 2) will be more than sufficient to supply energy for the entire process. If the excess tar cannot be burned, but must be disposed of as a hazardous waste, then the added cost to the process could be important. For example, occasionally the condensation of pyroligneous vapors leads to organic acids that must be neutralized with lime.

Relative humidity data given in Table 5 indicate its strong influence on bark, charcoal, and HDCP. If the bark does not contain any unbound moisture, then exposure of the bark to humid air could lead to increased drying costs. Since charcoal from the retorts is essentially bone-dry, exposure to even slightly moist air will result in an increased tray-drier duty. Lastly, the HDCP should be packaged as they are produced, because exposure of the bone-dry pellets to air could adversely affect the product, depending on its application.

Other economic considerations include the effect of liquid tar on the fixed-carbon content of HDCP (this effect is generally small) and the strength and density of the product (i.e., the pressure in the HDCP extruder may have to be varied according to the desired properties of the product). Overall, the product composition and properties attained in our laboratory coupled with the low cost of the feedstock suggest that the proposed process may well be feasible and is worthy of further consideration.

CONCLUSIONS

The depletion of fossil fuels and the uncertainty in costs of petroleum products create a demand for substitutes from renewable resources. The manufacture of HDCP from bark has the advantage that it utilizes a waste material with high disposal costs. Process considerations suggest that HDCP production may be economically feasible. HDCP have very low ash content and should be a good substitute for coal and petroleum coke as a reducing agent in the metallurgical industries. HDCP can also be used for the manufacture of adsorbents, i.e., activated carbon. With proper design of carbonization processes, a charcoal of fixed-carbon content can be produced. The final fixed-carbon content depends on the pyrolysis temperature. Higher char formation for treated bark heated out of contact with air suggests secondary char formation from catalytic action of inorganic chemicals. This would help to increase the charcoal yield, which would ultimately help to achieve a better rate of return on investment.

Conversion of combustible gases from wood into energy is no longer in the theoretical stages of development, but has moved into the beginning phases of commercial application. If this developed technology is adopted in the present process, combustible gases, which amount to 18% (wt) of the raw material used, are likely to increase the profitability of the project to a great extent.

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