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Low severity coal conversion by an electroreduction route

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The feasibility of electrochemical conversion of coal to low molecular weight hydrocarbons through an electroreduction route was demonstrated for the first time by Energy Research Corporation. This electroreduction process involves reaction of hydrogen ions with the coal surface leading to hydrogenation of coal molecules at low severity operating conditions. This process produced lower molecular weight hydrocarbons, similar to those obtained in conventional liquefaction by chemical reaction. In this proof-of-concept study, at low severity conditions (101 kPa and 250° C), the electroreduction behaviour of five coals as well as a charcoal and a devolatilized coal was investigated. The liquid product compositions (at room temperature) were dependent upon the parent coal and comprised a variety of aliphatic and aromatic compounds with phenolic aromatic compounds predominating. These compounds were found to fall in a low (100–400) molecular weight range which corresponds to oils (< 400). The effects of the process variables were also investigated. Coal type was found to be the most important parameter affecting the product spectrum. The volatile components in the coal appeared to play an important role in controlling electroreduction products. No clear-cut relationship was established between temperature, volatile contents, structural origin (maceral composition), or applied potential and product quantity or composition.

1. Introduction

Coal provides an abundant source of energy. Clean utilization of coal is very much desired in order to minimize harmful emissions. Conversion of coal to more useful forms of fuel such as H_2 and CO, synthetic natural gas, and liquid fuels (such as gasoline, kerosene and diesel) appears to be the most efficient approach for providing clean fuels from coal and maximizing its utilization.

Coal may be simply modelled as CH_n, where n is ~ 0.8 for bituminous coals. The manufacture of more easily usable fuel from coal involves increasing the numer of hydrogen atoms per atom of carbon and can be regarded as a process of *hydrogenation*. The conventional routes for conversion of coal to more easily usable fuel forms are: (1) *gasification*, generally involving reactions with steam, (2) *direct liquefaction* (direct addition of hydrogen), and (3) *pyrolysis* (thermal decomposition). Coal gasifiers are commonly operated at elevated pressure and in the temperature range of 600 to 1300° C. Active thermal decomposition of coal begins above 400° C. Direct hydrogenation processes under development are carried out at temperatures from 450 to 475° C and at pressures from 10 to 20 MPa and up to 30 MPa. Pressure favours hydrogenation through (a) inhibiting gas-forming dehydrogenation reactions and (b) increased partial pressures.

The coal conversion schemes mentioned above are complex and the economic penalties are high because of the severity of the operating conditions. An electrochemical approach to coal conversion has the potential to provide gasification and/or hydrogenation of coal at less severe conditions. In this approach, the energy of activation for the reaction is supplied in the form of electricity at a desired temperature. The controllability of temperature and input energy may allow a broader flexibility over the product selectivity.

Production of coal-derived compounds by both electrooxidation and electroreduction routes through cleavage of bonds and hydrogenation is possible. These two electrochemical coal conversion concepts are illustrated in Fig. 1. In the electrooxidation approach, the coal molecules are oxidized with H_2O molecules, leading to conversion of coal to smaller hydrocarbon molecules.

A parallel electrooxidation study was also conducted recently and reported separately [1]. The electroreduction process, which involves reaction of hydrogen ions and coal molecules similar to conventional coal liquefaction, may lead to saturation of aliphatic and aromatic bonds and reduction of other functional groups. This is expected to result in the lowering of coal molecular weight through cleavage of oxygen, sulphur and other bonds. An electrochemical reaction in aqueous electrolyte involving coal and H^+ may take place through the following reactions:

$$\text{Anode:}\quad H_2 \longrightarrow 2H^+ + 2e^- \tag{1}$$

$$\text{Cathode:}\quad \text{Coal} + 2H^+ + 2e^- \longrightarrow \text{Coal} - H_2 \tag{2}$$

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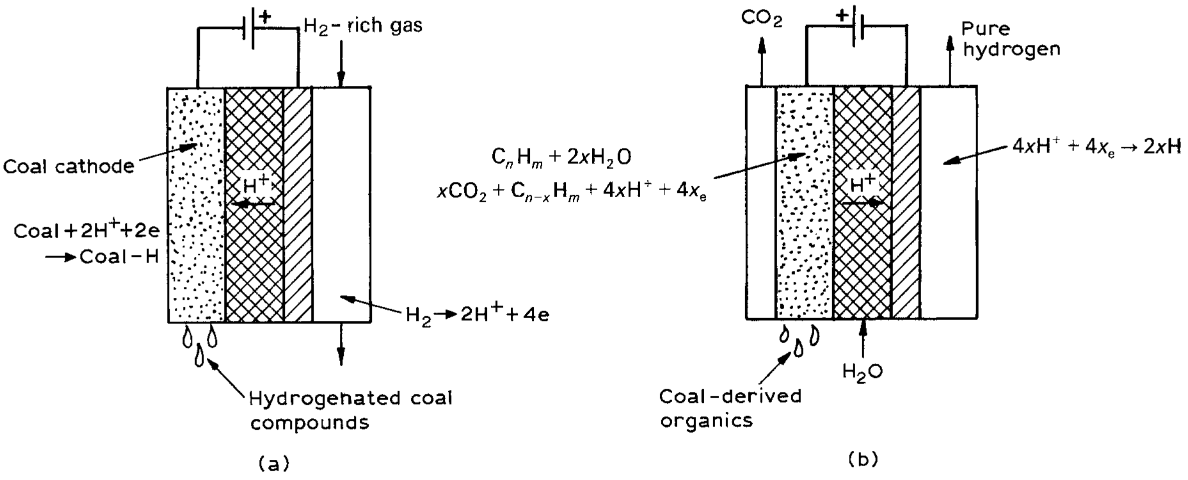

Fig. 1. Schematic of electroreduction of coal: (a) electroreduction, (b) electrooxidation.

In the electroreduction of coal the following electrochemical reactions of H_2 with carbon and unsaturated carbon–hydrogen bonds may occur, and others are possible:

Coal Electrode (Cathode):

$$\text{-C}{\equiv}\text{C-} + 2H^+ + 2e^- \longrightarrow \text{-CH}{=}\text{CH-} \qquad (3)$$

$$\text{-CH}{=}\text{CH-} + 2H^+ + 2e^- \longrightarrow \text{-CH}_2\text{-CH}_2\text{-} \qquad (4)$$

$$\text{-CH}_2\text{-CH}_2 + 2H^+ + 2e^- \longrightarrow 2[\text{-CH}_3] \qquad (5)$$

For all cases the anodic reaction is the oxidation of hydrogen to hydrogen ions and electrons.

The electroreduction cell potential is expected to be very small and dependent only on the cathode potential because the anode overpotential is negligible (less than 5 mV per 100 mA cm^{-2}). The selectivity and extent of the hydrogenation of coal may depend on the cathode potential. Hydrogen evolution overvoltage on carbon is < -350 mV with reference to RHE at 5 mA cm^{-2}, which may prevent hydrogen evolution from competing with the desired hydrogenation reaction at cathodic potentials. It is likely that most of these saturation and cleavage reduction reactions will require the supply of a small amount of external power.

Some limited attempts have been made to investigate coal electroreduction in organic electrolytes [2–8]. Early coal and coal-extract related reduction experiments were performed in nonaqueous solvents, mainly using polarographic methods [3–6]. Electrochemical reduction of a vitrain from low volatile bituminous coal in ethylene diamine saturated with lithium chloride resulted in hydrogen addition to the vitrain sample [8], providing extracts having molecular weights of 800 to 900. Recent work by Park [7] focused on the electroreduction of coal in a rigorously dried nonaqueous medium. This process has not been successful in the production of coal derived products. Limitations on the choice of electrolyte media, cell temperature, and the apparatus restricted the observations of these earlier studies.

Previous attempts to electrochemically hydrogenate olefinic compounds were limited to the use of water electrolysis [9] as the source for H^+:

$$H_2O \longrightarrow 2H^+ + 2e^- + \tfrac{1}{2}O_2 \qquad (6)$$

Use of this type of anode reaction leads to a significant consumption of power (adding ~2 V to the cell potential) and also causes product loss through oxidation of organics at the high anodic potential.

Electroreduction of coal in aqueous media had not been investigated prior to the present work. Furthermore, production of coal-derived liquid products by the electroreduction route has not been demonstrated previously.

This paper presents the findings of a study intended for proof-of-concept demonstrations of coal hydrogenation by electroreduction leading to formation of useful liquid products. An interim status report for both the electroreduction and the previously mentioned electrooxidation investigations was presented at a Contractors Review Meeting [10], and a final report has been issued [11].

2. Experimental details

The coal electroreduction experiments were conducted at 1 atm, 150 to 250° C, and at cathodic overpotentials of 0 to 900 mV/RHE using phosphoric acid (H_3PO_4) electrolyte. Hydrogen from a pure hydrogen stream was oxidized to H^+ using a fuel cell type gas diffusion anode, transported through the electrolyte immobilized in a porous matrix structure, and reduced on a porous coal electrode. The products of the coal electroreduction reactions were identified and the effects of process variables on electroreduction kinetics and product spectra were investigated.

2.1. *Test setup*

The experimental apparatus is illustrated in Fig. 2. The chemical compounds which were produced and evolved at the cell operating conditions were passed through a cold trap in which condensable products of the electroreduction reaction were collected

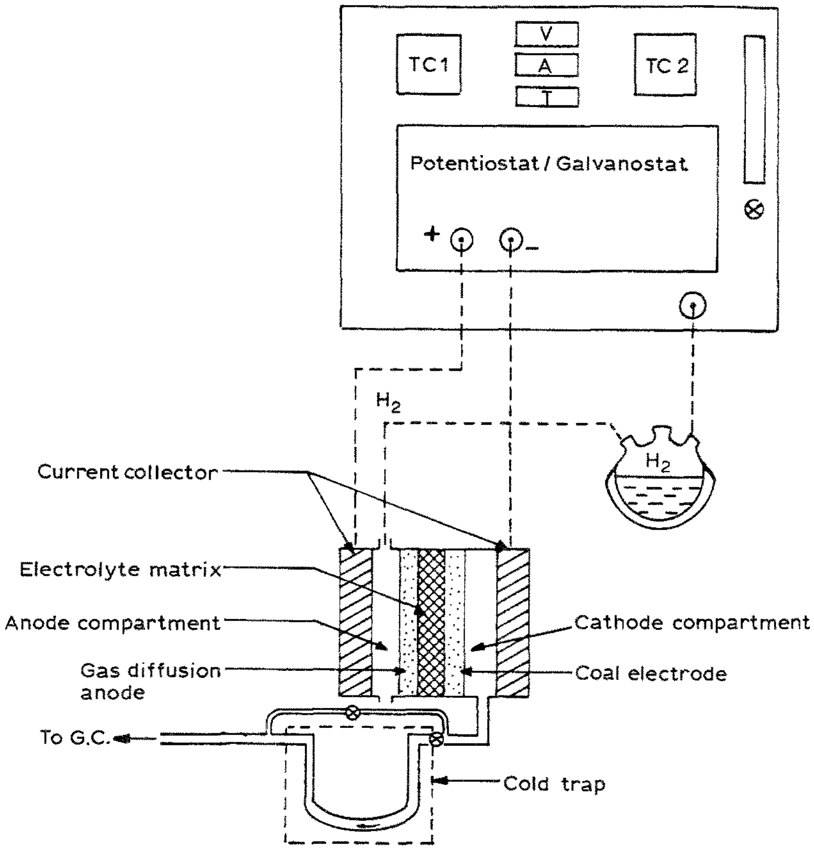

Fig. 2. Apparatus for electroreduction of coal.

for appropriate analyses. The gas production was measured and gases were analysed by gas chromatography. A Princeton Applied Research (PAR Model 173) potentiostat/galvanostat was used to supply power for the potentiostatic electroreduction experiments.

The cell design is shown in Fig. 3. This type of cell hardware has been utilized in phosphoric acid fuel cells (PAFCs) for many years [12]. The two end-plates, made from heat-treated graphite–glassy carbon composite material, provide current collection and flow fields for hydrogen (reactant) gas on the anode side and for product on the cathode side. The cell geometry is 9.5 cm on each side; the active cell area being 25 cm^2. The hydrogen flow to the anode was 10 to 15 cm^3 min^{-1}. No purge gases were employed.

The anode is a state-of-the-art gas diffusion electrode, comprised of a porous thin layer of supported platinum catalyst laminated on a porous, conductive graphite support layer. The matrix consists of a thin porous layer of silicon carbide (SiC) soaked with H_3PO_4 electrolyte. In the coal electrode (cathode), a porous thin layer made from pulverized coal using a rolling press is laminated on a porous wet-proofed graphite support layer. The coal loading in the test electrodes is ~ 0.03 mg cm^{-2} of geometric electrode area. The Teflon binder content was 20%; attempts to prepare coal electrodes with less or no Teflon resulted in non-uniform coal layers which did not adhere to the porous carbon paper and were considered inadequate for use in the electroreduction experiments.

Only potentiostatic electroreduction reactions were conducted in this study. In all experiments, the anode of the cell was used to control the electroreduction cathode potential. The hydrogen anode overpotential is known to be $\simeq 5$ mV per 100 mA cm^{-2} for the supported platinum catalyst. Cell resistance was measured using a milliohmmeter. From these values,

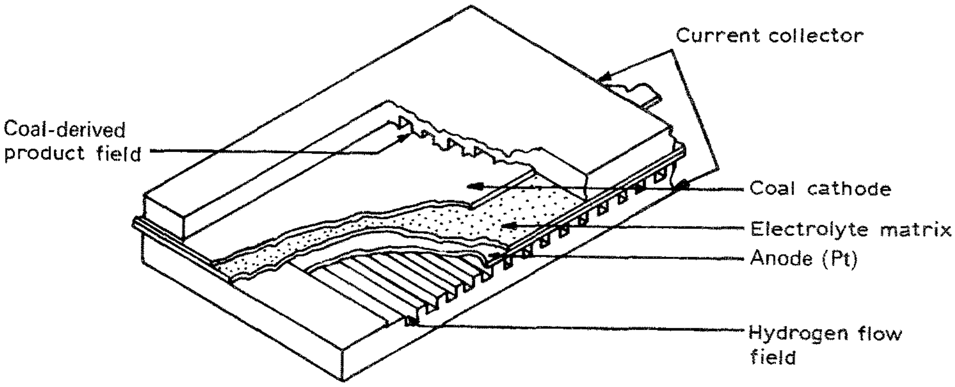

Fig. 3. Coal electroreduction cell design, cutaway view.

Table 1. Average analysis of coals tested

*Property**	*Illinois #6*	*Illinois #2*	*Pittsburgh*	*Charcoal*	*Montana Rosebud*	*North Dakota Velva Lignite*
PSOC #	1493	N/A	320	N/A	1485	N/A
Moisture	9.43	13.62	2.03	5	25.37	28.7
Volatile Matter	37.91	43.34	21.72	18–23	36.75	40.80
Fixed Carbon	46.92	49.92	62.50	–	51.81	46.4
Ash	15.17	6.66	15.78	5	11.44	12.8
Carbon	66.23	73.31	74.31	75	67.11	60.21
Hydrogen	4.17	5.21	4.26	–	4.69	3.98
Nitrogen	1.27	1.47	1.86	–	1.05	0.91
Oxygen	8.18	10.09	2.63	–	14.83	21.90
Sulphatic Sulphur	0.04	0.10	0.07	0	0.00	N/A
Pyritic Sulphur	2.61	2.34	0.45	0	0.02	N/A
Organic Sulphur	2.33	0.92	0.64	0	0.86	N/A
Total Sulphur	4.98	3.23	1.16	0	0.88	0.20

* Values are reported on a moisture-free basis except for the moisture.

the true potential of the cathode was easily ascertained without requiring a separate reference electrode.

2.2. *Test variables*

The effects of process variables such as temperature (200 to 250° C), reduction potential, and coal type on product selectivity were investigated. The tests were conducted for 65 to 500 h. The analysis of the coals used in this study is provided in Table 1. Two hydrogen donors, 9,10-dihydroanthracene and 1,2,3,4-tetrahydroquinoline, were considered for the investigation of the effect of hydrogen donor on the electroreduction of coal, based on their outstanding hydrogen donating ability in the liquefaction of bituminous and brown coals [13]. These compounds were, however, determined to be incompatible with the electroreduction test temperature in out-of-cell tests and were eliminated from further considerations.

3. Results

The electroreduction behaviour of the following materials was investigated: two high-volatile bituminous coals (Illinois #6 and Illinois #2), one medium volatile bituminous coal (Pittsburgh), one sub-bituminous coal (Montana Rosebud), and one lignite (North Dakota Velva), as well as one charcoal and one devolatilized coal. As mentioned previously, their average analyses are presented in Table 1. Current densities for the electroreduction reactions ranged from 3 to 40 mA cm^{-2} of geometric electrode area.

Certain similarities were noted in the electroreduction products from each of the materials studied. Liquid products as well as a solid waxy substance were obtained with each; however, no gaseous products were formed. In addition, pyritic sulphur was found to be removed during the electroreduction treatment.

The compositions of the liquid products are dependent upon the parent coal, and vary greatly. Short and long chain aliphatic, cyclic, aromatic, and fused ring aromatic hydrocarbons were detected.

To ascertain whether the liquid products are a consequence of chemical rather than electrochemical reaction, a blank test was conducted with Illinois #6 coal, in which no voltage was imposed on the coal electrode. Hydrogen was passed over the coal electrode to provide a substitute for the evolved H_2 which is present during the electroreduction reactions. Conditions were otherwise similar to those under which liquid product had been obtained with Illinois #6 coal. In the blank test, no liquid product and negligible solid waxy material were formed. It was concluded that the liquid produced is indeed the result of electrochemical rather than chemical reaction.

Some water was always collected along with the liquid products from coals; the products were separated with chloroform from the water associated with them. The chloroform extracts were analysed by gas chromatography/mass spectroscopy (GC/MS). The mass spectrometer was not activated until the chloroform had been eluted. A blank chloroform sample was also analysed by this technique to establish the background, which was found to be negligible.

Descriptions of the products derived from the various coals tested are given in the following paragraphs.

Illinois #6 provided the most varied as well as the greatest quantity of product. Eighty-one compounds were separated; of those present in quantity sufficient for identification (listed in Table 2), aliphatics and aromatics occur in a 1 : 1 volume ratio. A typical chromatographic analysis of the liquid products from Illinois #6 is given in Fig. 4. There are high concentrations of phenol derivatives and long chain hydrocarbons and mono-alcohols. One cyclic aliphatic compound was also detected. The estimated/calculated molecular weights (108 to 354) of coal liquefaction products correspond to oils ($\leqslant 400$) and asphaltenes (300 to 1000). Liquid products appeared at 250° C, at a relatively low electrode potential (– 335 mV/RHE). A 25% current efficiency for product reaction was estimated, based on hydrogen balance. A portion of the current consumed is accounted for by the cathodic

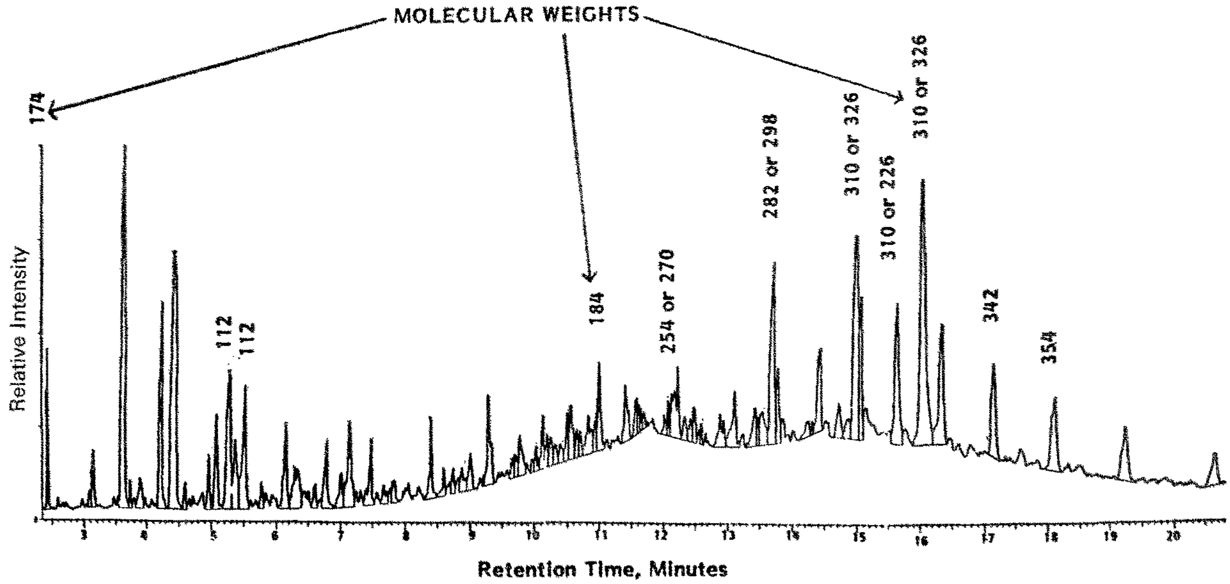

Fig. 4. Gas chromatogram of liquid products obtained during electroreduction of Illinois #6 coal.

production of hydrogen gas, which is collected and measured. The remainder of the current is assumed to be consumed in the formation of the liquid products from the electroreduction of coal.

$$\begin{aligned}\text{Total current} &= \text{H}_2 \text{ transfer current}\\ &\quad + \text{electroreduction reaction current}\end{aligned}$$

Current efficiency

$$= \frac{\text{Electroreduction reaction current}}{\text{Total current}}$$

This was not verified by an alternative route such as product balance.

With *Illinois #2*, a moderate quantity of liquid products was obtained, at 250° C and an electrode potential of $-500\,\text{mV/RHE}$. The products are comprised almost entirely of aromatics (Table 3), and are predominantly phenols with small quantities of acid and ketones.

Table 2. Illinois #6 coal derived liquid product analysis

Major Components	*Concentration* (%)	*Molecular Weight* (*Estimated/Calculated*)
Aromatic		
4-hydroxybenzene-sulphonic acid	6.8	174
2-methylphenol	3.0	108
methylphenol derivative	7.2	–
3-ethylphenol	3.0	112
2,4-dimethylphenol	2.3	112
isobenzofuranone	2.0	–
Aliphatic		
4,5-dimethylundecane	1.3	184
C_{18} hydrocarbon or alcohol	1.9	254 or 270
C_{20} hydrocarbon or alcohol*	3.8	282 or 298
C_{22} hydrocarbon or alcohol*	5.4	310 or 326
C_{22} hydrocarbon or alcohol	3.4	310 or 326
C_{22} hydrocarbon or alcohol	3.1	310 or 326
C_{22} hydrocarbon diol	2.3	342
C_{24} hydrocarbon alcohol	2.1	354
	47.6	
impurities	8.8	
other unidentified compounds in small quantities	43.6	

* Cyclic compounds. (Remaining aliphatic compounds are straight chain.)

Table 3. Illinois #2 coal derived liquid product analysis

Major Components	*Concentration* (%)	*Molecular Weight*
Aromatic		
butyl-2-methylpropyl-1,2-benzene-dicarboxylic acid	2.7	280
2,4-bis[(trimethylsilyl)oxy]-tri-benzoic acid	1.2	–
p-2(2-methylallyl)-phenol	0.6	–
1-(2,4-dimethylphenyl)ethanone	0.8	–
isobenzofuranone	4.5	–
2,5-dimethylbenzaldehyde	2.8	134
2,3-dihydro-1H-inden-5ol	3.7	–
3-propylphenol	2.6	136
7-chloro-1,3-dihydro-2H-1,4-benzodiazepin-2-one	3.0	–
1-methylene-1H-indene	3.4	–
2,3-dimethylphenol	2.2	122
2-ethylphenol	9.3	122
3-methylphenol	20.5	108
3,3-dimethylpentylcyclohexane	0.7	183
2-methylphenol	8.1	108
phenol	17.7	94
Aliphatic		
1,1′-oxybis[2-methoxy]ethane	1.1	
	84.9	
Impurities	8.7	
Other unidentified compounds in small quantities	6.4	

In the case of the medium volatile *Pittsburgh* coal, only a small quantity of product was collected. Ten compounds were separated; the aromatics, which account for 80% of the product volume, include phenol derivatives, acids and esters. An electrode potential of −600 mV/RHE and a temperature of 250° C were required for product formation by the electroreduction reaction.

Montana Rosebud, a sub-bituminous coal, likewise yielded a small quantity of liquid product, at 250° C. A high potential (−800 mV/RHE) was required. Sixty-seven compounds were separated; all of the identified compounds are phenol derivatives, within a fairly narrow molecular weight range (94 to 136).

North Dakota Velva lignite was the only coal tested from which liquid products were obtained at 200° C. The electrode potential required was also relatively low (−300 to 500 mV). A moderate quantity of liquid was collected, from which 26 compounds were separated. The identified products (Table 4) are also predominantly aromatic (90%). This was the only coal product in which a fused aromatic compound, naphthalene, was detected. The remainder of the aromatic compounds are phenol derivatives. A cyclic aliphatic compound is present, as well as straight chain diols which are approximately half the length of the long chain aliphatic mono-alcohols found in the Illinois #6 product.

The electroreduction behaviour of a *charcoal* sample was also investigated. The liquid products were formed only at 250° C and at high electrode potentials, −700 to −900 mV. Only a small quantity of product was collected; it is quite varied, however, with 79 compounds separated. Fused aromatics (substituted naphthalenes) are present in a fairly high concentration (~8%). The other aromatic constituents contain phenol, aldehyde, acid and ester groups.

The role of the structural origin (as reflected in the maceral composition, particularly active components) of coal in the electroreduction of coal was examined. No correlation was observed between the maceral compositions and the reactivities of the coals tested.

Table 4. Velva lignite coal derived liquid product analysis

Major Components	*Concentration* (%)	*Molecular Weight*
Aromatic		
phenol	26.1	94
2-methylphenol	7.8	108
3-methylphenol	21.5	108
2-ethylphenol	1.0	122
2,3-dimethylphenol	2.5	122
3-ethylphenol	6.7	122
naphthalene	3.7	128
3-propylphenol	1.1	136
Aliphatic		
1-methyl-3-propylcyclohexane	4.1	140
4-methylheptane	4.1	114
1,14-tetradecanediol	1.8	230
1,12-dodecanediol	2.0	202
Total identified	82.4	
Other unidentified compounds in small quantities	17.6	

To provide clarification of the contribution of the volatile materials in the formation of the liquid products obtained in the electroreduction of coal, a portion of Illinois #6 coal was devolatized by heat-treatment. Proximate and ultimate analyses performed on the devolatilized Illinois #6 coal indicated that the volatile content was almost completely removed by the heat-treatment.

A TGA analysis performed on Illinois #6 coal determined that the temperature range in which the volatiles are expelled is 400 to 900° C. The volatiles were analysed by GC/MS to identify the constituents, which are listed in Table 5. The majority have much lower molecular weights than the compounds produced in the electroreduction of Illinois #6 coal; therefore, the liquid product collected from Illinois #6 coal is not the result of simple devolatilization during the test.

During electroreduction of the devolatilized Illinois #6 coal (at 250° C), a much smaller quantity of liquid product was obtained than with the parent coal. There were also much fewer compounds isolated: 20 compared to 81 with the parent coal. A description of the products identified in the product from the devolatilized Illinois #6 coal is given in Table 6. There are significant differences in the compositions of the products with respect to both aromatic and aliphatic compounds. In the devolatilized product the longest chain aliphatic hydrocarbon is hexane, whereas C_{22} and C_{24} chains were found in the parent Illinois #6 coal product. The percentage of aromatic compounds increased from 50 with Illinois #6 coal to 65 with the devolatilized coal. In addition, compounds with much lower molecular weights were reported for the devolatilized product, in which the molecular weights of all components are less than 150; in the Illinois #6 product, the molecular weights of the aliphatics range from 184 to 354. Considering that there is only one compound in common (2-methylphenol) between the devolatilized and Illinois #6 coal products together with the great differences between them, the volatiles appear to play a large role in the electroreduction of Illinois #6 coal.

Table 5. Volatile compounds eluted from Illinois #6 coal (900° C) during heat-treatment

Hydrogen sulphide
Long chain unsaturated hydrocarbon
Long chain organic alcohols or acids
Hexadecanoic acid
Xylene
Toluene
Acetic acid
Sulphur dioxide
Propionic acid
Formic acid
Pentyl alcohols
Low mol. wt sulphur compound (possibly S_2)

Table 6. Devolatilized Illinois #6 coal derived liquid product analysis

Major Components	*Concentration* (%)	*Molecular Weight*
Aromatic		
2-methylphenol*	11.5	108
2,6-dimethylphenol	20.2	122
chlorocresol	6.9	140
Aliphatic		
3,3,4,4-tetramethylhexane	17.8	142
3-ethyl-5-methylheptane	3.6	142
2,3,4-trimethylpentane	0.6	114
Total identified	60.6	
Other unidentified compounds in small quantities	39.4	

* Only this compound was present in the liquid products from both as-received and devolatilized coals.

The white solid waxy deposits described previously were formed in the water in which the coal electrode exit gases were collected and were found to be partially soluble in acetone and chloroform, and insoluble in sulphuric, nitric, or hydrofluoric acids and sodium hydroxide. A deuterated chloroform extraction of a sample of the deposits was utilized in analyses by nuclear magnetic resonance (NMR) and infrared spectrophotometry (IR). The NMR analysis indicated that the aliphatic to aromatic C–H bond ratio is approximately 5:1. The extract and the residue were both analysed by IR. The spectrum of the organic portion of the deposit, which was extracted in the deuterated solvent, is typical of long chain esters or ester alcohols. The characteristic aromatic bands are absent, which is consistent with the NMR data. The residue from the extraction is typical of a silicate or related compound. The waxy colloidal deposits consist predominantly of the silicates, although the relative concentrations were not determined. These results help to explain the solubility behaviour of the waxy deposits. The colloidal particles appear to be comprised of silicate nuclei, coated with the organic material which forms a protective layer with respect to the acids and bases.

To determine how the sulphur contained in the coal is affected by the electroreduction reaction, sulphur analyses were performed on Illinois #6 coal electrodes in untested, blank tested (in the blank test described earlier) and electroreduction tested conditions. Sulphur analyses performed with a Parr bomb indicated that the pyritic sulphur was essentially completely removed during the electroreduction test and partially removed during the blank test. The applied voltage in the electroreduction test resulted in somewhat greater sulphur removal. (These results are supported by energy dispersive X-ray spectroscopy. Sulphur contents are significantly reduced in both blank and electroreduction tested electrodes compared to the untested electrode.) If the sulphur content in the untested electrode is assumed to represent total sulphur, the sulphur *lost* in the electroreduction tested electrode corresponds closely to the pyritic portion of the total sulphur.

4. Discussion

Production of liquid hydrocarbon products by electrochemical reduction of coal at low severity conditions has been successfully demonstrated. Liquid hydrocarbon products in the 100 to 300 molecular weight range were obtained from each of the coals tested in this study. An overall summary of observations is presented in Table 7. Phenolic aromatic compounds apear to be the predominant product. The products vary greatly and depend on the coal type. Methylated phenol is the only compound which occurred in the product from each coal. The current efficiency of the electroreduction reaction, estimated by hydrogen balance, was less than 25% for all cases.

One high-volatile bituminous coal, Illinois #6, provided the greatest quantity of product, which is also the product having the greatest variation along with the largest percentage of aliphatics (mostly long chain). The products (predominantly phenols) obtained with the other high-volatile bituminous coal, Illinois #2, are quite different from the Illinois #6 products. With the other coals tested, there was no obvious qualitative correlation between the volatile content and the products. This is in accordance with coal liquefaction experience, where there is also no clear relationship between conversion and volatile matter content of the coal [14].

In the electroreduction of devolatilized Illinois #6 coal, only a very small quantity of product (compared with the other coals) was obtained, and a higher potential was required for liquid production than with the as-received Illinois #6 coal. There are significant differences, as well, between the compounds removed from Illinois #6 coal by devolatilization and those produced in the electroreduction of as-received Illinois #6 coal; in particular, the products resulting from the devolatized coal are of much lower molecular weights. This demonstrates that the liquid products obtained in the electroreduction reaction at $< 250°$ C are not the result of simple devolatization of the coal. Further important differences between the liquid products obtained with the electroreduction of the devolatized and as-received Illinois #6 coals indicate that the volatiles do, however, play an important role in the electroreduction process.

There was some influence of coal rank on the product composition. No direct influence of structural origin (maceral composition) was deduced in the coal electroreduction experiments conducted in this programme. Likewise, no direct correlation was observed between the applied potential required for the electroreduction reaction and the composition of the products.

This investigation focused on a proof-of-concept demonstration of coal conversion by electrochemical reduction. The cell design used in this study is not

Table 7. Coal electroreduction observations: products depended on coal type; no other correlations were deduced

Coal Type	*'Reactivity'**	*Product Description*
Illinois #6 (HVB)	Highest; product collected at −335 mV (RHE)	81 compounds; High concentration of phenols and long chain hydrocarbons and alcohols
Illinois #2 (HVB)	Product collected at −500 mV (RHE)	32 compounds; Predominantly phenols with small quantities of acid and ketones
Pittsburgh (MVB)	Product collected at −600 mV (RHE)	10 compounds; 20% aliphatic/80% aromatic
Montana Rosebud (Sub-bituminous)	Product collected at −800 mV (RHE)	67 compounds; All phenols
North Dakota Velva Lignite*	Product collected at −300 to −500 mV (RHE)	26 compounds; Predominantly aromatic (90%) Fused aromatics present
Charcoal	Product collected at −700 to −900 mV (RHE)	79 compounds; Predominantly aromatic (80%) Fused aromatics ≃ 8%
Devolatilized Illinois #6	Product collected at −355 to −400 mV (RHE)	20 compounds; Phenols and short chain hydrocarbons

* North Dakota Velva lignite was the only coal from which liquid product was produced at 200° C; others at 250° C.

suitable for detailed mass balance, permitting estimation of coal conversions and product quantification. An economic analysis has not been pursued in this study because of the absence of sufficient rate and conversion information. Mass balance and economic evaluation should be addressed in a future study to evaluate the attractiveness of this novel process of coal conversion.

5. Conclusions

This paper reports the successful production of liquid hydrocarbon products from different coal types at low severity conditions (1 atm pressure and less than 250° C) by the electroreduction route of coal conversion. The coal electroreduction products indicated a low (100 to 400) molecular weight range which corresponds to oils (< 400). A variety of aliphatic and aromatic compounds have been identified in the products. Coal type has been observed to be the most important parameter affecting the product spectrum.

No attempt has been made to pursue quantitative characterization of the process and evaluate its commercial feasibility. The cell design used in this study is not conducive to a detailed mass balance analysis permitting estimation of coal conversion. Therefore, future studies should focus on detailed mass balance and the commercializable cell design development aspects.

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