# Consideration of electrodes and electrolytes for electrochemical gasification of coal by anodic oxidation

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Received 7 December 1979

It has been found that the performance of graphite anodes in coal electrolysis is closely comparable to that reported previously for anodes of Pt gauze; the sole exception discerned thus far is that the gases formed anodically at graphite are somewhat enriched in carbon monoxide and correspondingly depleted in carbon dioxide as compared to Pt anodes. No significant consumption or degradation of the graphite was observed during short experiments in which about half an equivalent of charge passed – the weight gain measured was about 0.05% of that computed from the assumption of formation of an equivalent quantity of surface oxides. Of several different electrolytes considered and tested, aqueous solutions of $H_2SO_4$ appear to cause the most rapid reaction rates.

## 1. Introduction

We have recently reported [1–3] a novel method of hydrogen production by the combined use of fossil fuel and electrical energy. In this new process, which we call electrochemical gasification of coal, solid particles of coal are oxidized within an aqueous electrolyte at the anode of an electrochemical cell. Other suitable names are 'coal assisted' or 'coal-depolarized' water electrolysis. Although coal contains a wide variety of organic molecules rich in carbon, we focus here only on the carbon of the coal; accordingly the reaction of the electrochemical coal-gasification process at the anode, in simplified stoichiometry, may be expressed as:

$$C_{(s)} + 2H_2O_{(l)} \rightarrow CO_2 + 4H^+ + 4e \qquad (1)$$
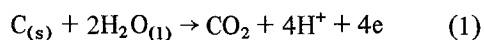
The corresponding reaction at the cathode may be represented as:

$$4H^+ + 4e \rightarrow 2H_{2(g)}. \qquad (2)$$
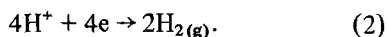
The sum of Reactions 1 and 2 is the overall reaction between carbon (coal) and water:

$$C_{(s)} + H_2O_{(l)} \rightarrow 2H_{2(g)} + CO_{2(g)}. \qquad (3)$$
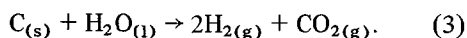
Supplying electrical energy by the application of an electrical potential permits this reaction to occur at moderate temperatures at which this reaction would otherwise be thermodynamically unfavourable by virtue of a large equilibrium free-energy barrier.

We have found that actual practical operating potentials must be about 0.85–1.0 V at room temperature where the thermodynamic reversible potential ($\Delta G^{\ominus}/nF$) for Equation (3) is only 0.21 V. Fig. 1 shows the effect of temperature on the reversible cell potential as computed from the thermodynamic $\Delta G$ and $\Delta H$ values of the Reaction (3). Compared to conventional coal gasification the advantage of this electrochemical process for gasification of coal is the production of gaseous oxides of carbon at the anode with few impurities and also the simultaneous production of essentially pure hydrogen separately at the cathode. Because each gaseous product is produced separately and essentially free of impurities such as sulphur, tar and ash, subsequent cleaning, separation and purification steps are minimized.

Previous reports [1, 2] have introduced the process of electrochemical gasification, provided initial data as to reactivities of various coals and distinguished the process from conventional water electrolysis. Moreover the energy requirement of conventional water electrolysis was compared in an elementary way with that of electrochemical gasification. Its relationship to conventional methods of coal gasification and the well known steam carbon reaction was also discussed. We
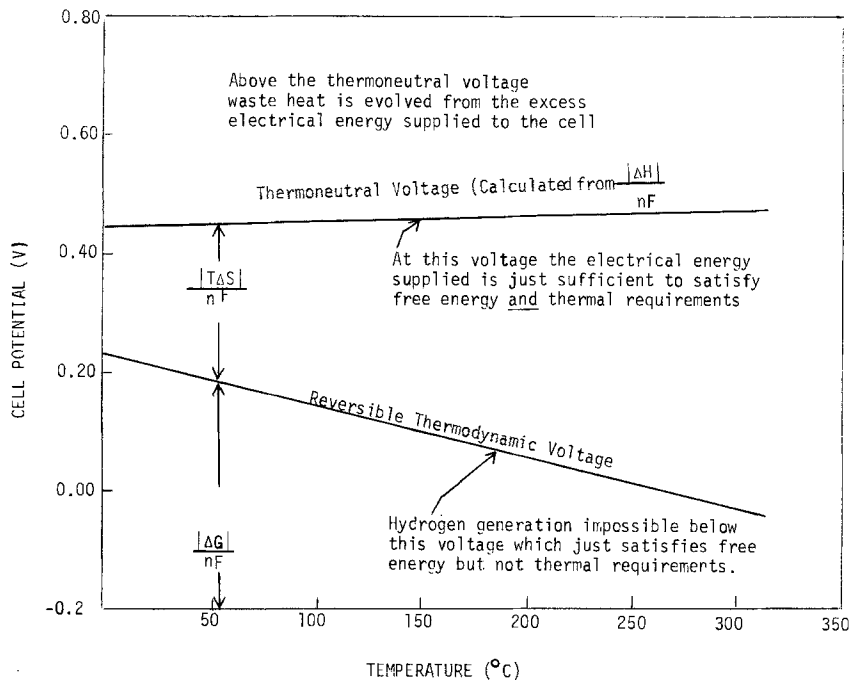
Fig. 1. Influence of temperature on thermodynamic reversible (open circuit) potential of reaction
$C(s) + H_2O(1) \rightarrow 2H_2(g) + CO_2(g)$.

reported earlier in [3] the effect of different operating conditions and variables. In all these studies platinum gauze was used as the electrode material. Less costly electrode material must be sought, however, to minimize capital investment costs with a view toward the lowest possible production cost of the gaseous products. With this in mind we investigated different forms of graphite electrodes as anode material and the findings are reported in this paper where we also compare the results with those obtained using a platinum electrode. Some considerations regarding the electrolyte are also discussed.

## 2. Experimental

Experiments were conducted in the simple cell shown in Fig. 2. The external emf was controlled by a potentiostat (both Models PAR 371 and PAR 179A were used) and the cathode was Pt gauze, gauge 52 (0.004 in. diameter wire) supplied by Mathey Bishop, Inc. The anodes were prepared from graphite rods and graphite felt supplied by Union Carbide Corporation and electrical connections were accomplished through a Pt wire cemented to the graphite material using conductive

epoxy resin (Epo-tek), supplied by Epoxy Technology Incorporated. Additional details of the experimental apparatus are given in [2]. Gas analysis was by chromatography employing a
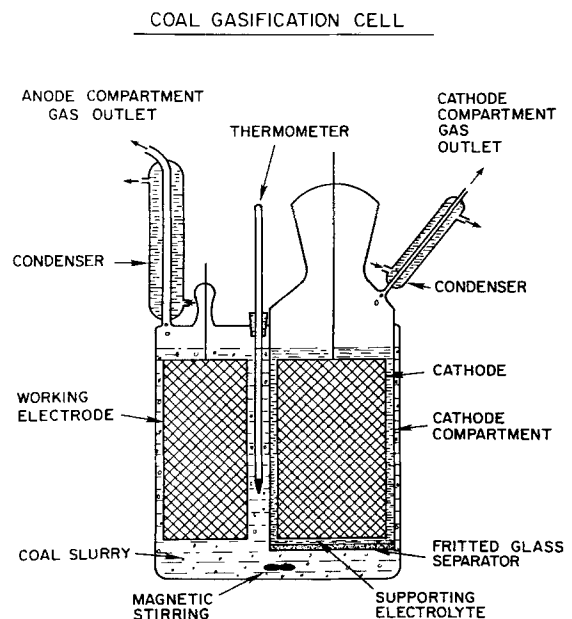


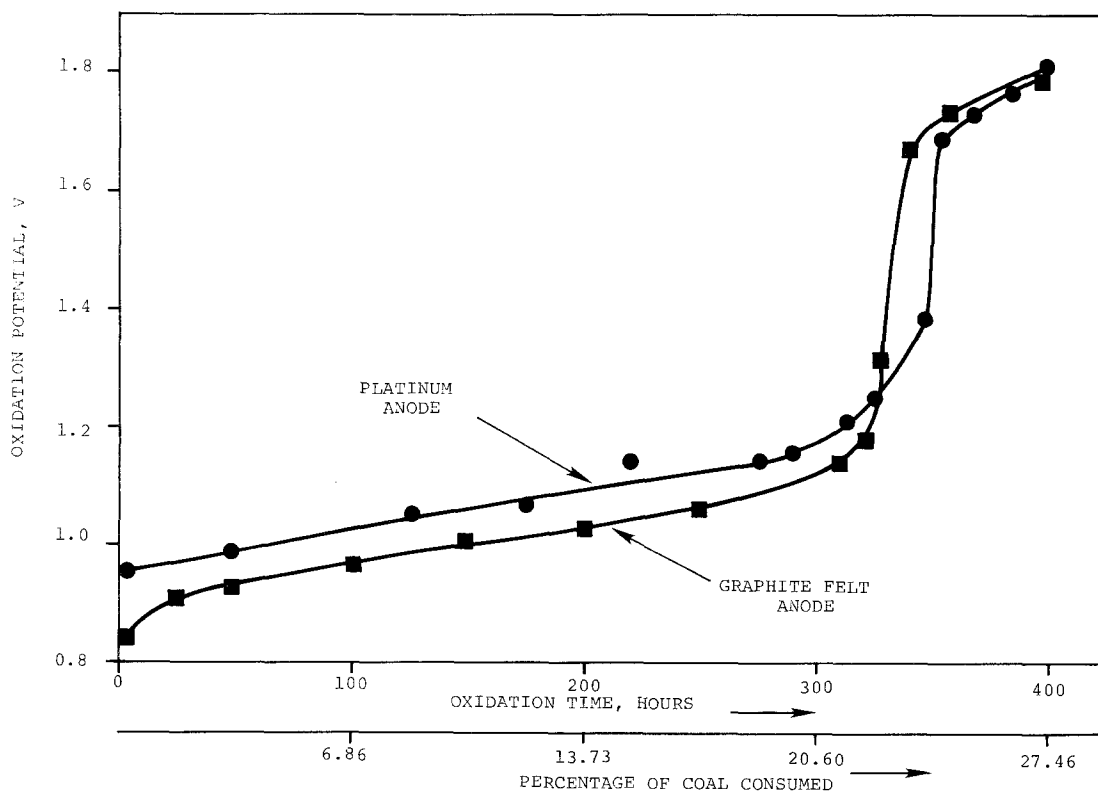Fig. 2. Simple electrolytic cell used for experiments.

Fig. 3. Change of cell potential during galvanostatic oxidation. NDL slurry conc.: 0.069 g cm$^{-3}$ (total volume: 477 cm$^3$); supporting electrolyte: 5.6 M H$_2$SO$_4$; particle size: 105–125 $\mu$m; temperature: 114° C; galvanostatic current: 101.5 mA; Pt anode area: 96.5 cm$^2$ (geometrical); graphite-felt area: 104 cm$^2$ (geometrical).

Spherocarb (100/120 mesh) separation column. Aqueous electrolytes were prepared with Baker analysed reagent grade sulphuric acid. Coal samples in pulverized form were obtained from the US Department of Energy Pittsburgh Energy Research Center; their ultimate analyses are given elsewhere [2].

### 3. Galvanostatic studies: effect on the cell potential

North Dakota lignite (NDL) was electrochemically gasified by anodic oxidation at a galvanostatic rate of 101.5 mA using both platinum and graphite felt anodes. For comparison, the corresponding cell potentials are plotted in Fig. 3 against the percentage of the total coal consumed* in the process. These results indicate clearly that the course of the reaction is similar with either Pt or graphite anodes. To maintain the desired galvanostatic rate

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* The expression 'coal consumption' used herein means the ratio of equivalents of electric charge passed divided by the number of equivalents of carbon originally present.

of reaction, the cell potential gradually rises regardless of whether a graphite or Pt anode is used because the coal particles become more and more unreactive as the reaction progresses (there may also be some effect due to decreasing coal concentration as well). This loss of reactivity of the coal particles is probably due to formation of oxygen-containing functional groups during the anodic oxidation of coal. It is also possible that during the course of the reaction these oxides increase in concentration on the surface of the coal particles toward a steady-state thereby rendering electron abstraction progressively more difficult, with subsequent rise of the oxidation potential. Preliminary results obtained by analysing the residual carbon values after partial electrochemical gasification of Pittsburgh coal also support this view. Based on the known decomposition temperatures reported [4] for such surface oxygen compounds on carbon it should be possible to reduce the loss of reactivity of the coal at 200–600° C. It seems reasonable to consider surface oxides as
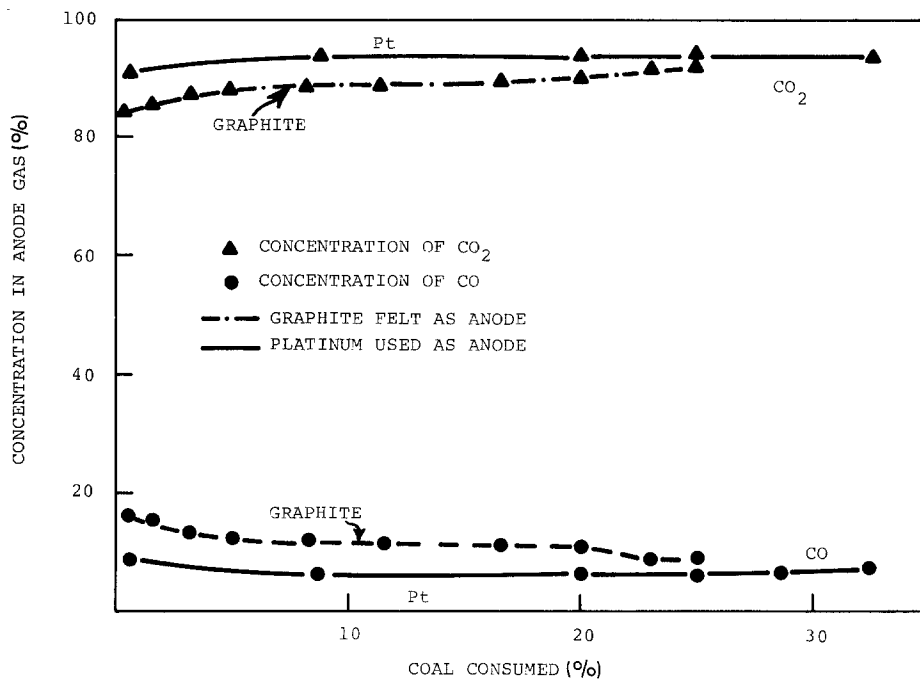
Fig. 4. Anode gas composition vs. percentage of coal consumed during galvanostatic oxidation. Fresh NDL; slurry conc.: 0.069 g cm$^{-3}$ (total volume: 477 cm$^3$), particle size: 105–125 $\mu$m; temperature: 114° C; galvanostatic current: 101.5 mA; Pt anode area: 96.5 cm$^2$ (geometrical), graphite-felt anode area: 104 cm$^2$ (geometrical); supporting electrolyte: 5.6 M H$_2$SO$_4$.

intermediates in the pathway from coal and water as reactants to gaseous oxides of carbon and hydrogen as reaction products. This loss of reactivity may also result in part from the accumulation on the coal particles of a tar-like coating that appears to form during electrochemical coal gasification. This tar-like substance is an additional reaction product probably formed by the scission of small aliphatic fragments from larger coal molecules during anodic oxidation. The potential required to maintain a constant current of 101.5 mA continues to rise in Fig. 3 to about 1.2 V (at this stage about 20% of the coal is consumed) whereupon the required potential rises quickly to about 1.7 V, thereby suggesting the onset of a different reaction mechanism. It should be noted that analysis of the anode gas revealed only CO$_2$ and CO and no trace of O$_2$ until the potential reached 1.98 V. It appears, therefore, that the discontinuity at 1.2 V may correspond to the onset of a different mechanism of electrochemical coal gasification; it cannot be explained as the onset of simple water electrolysis.

## 4. Gas analysis and ratio of anode-to-cathode gas volumes

During the oxidation of NDL at a graphite-felt anode the gas produced at the cathode was essentially pure H$_2$, and in each case the coulombic efficiency of H$_2$ production was close to 100% based on measured current integrated over time. The gas produced within the anode compartment was a mixture of CO$_2$ and CO. The composition of the anode gas, however, varies somewhat over the course of the gasification reaction; this behaviour may be associated with corresponding changes in population of surface oxides on the coal. Typical results are shown in Fig. 4 where the anode gas composition is plotted against percentage of coal reacted galvanostatically. (Note that the corresponding course of potential change is shown in Fig. 3.) It is evident that the gas generated at the anode is mostly CO$_2$, but with an initial CO composition of about 16% which then gradually decreases to about 11.5%. This latter value is obtained after about 7.5% of the
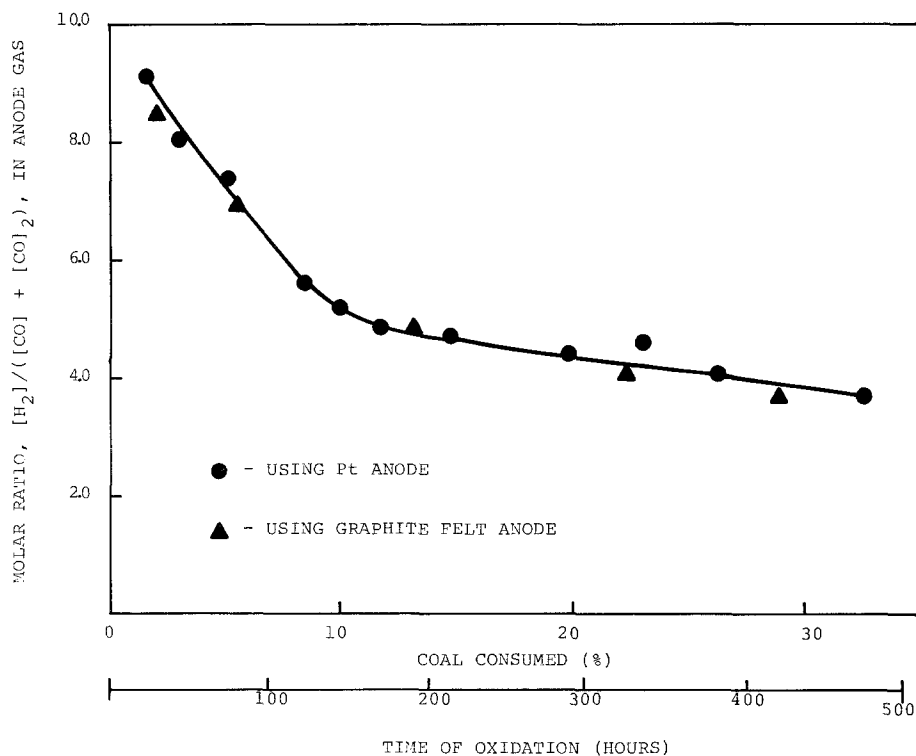
Fig. 5. Changes in composition of accumulated anode gas during reaction. Experimental conditions: slurry conc.: 0.069 g cm$^{-3}$ (total volume: 477 cm$^3$); particle size: 105–125 $\mu$m; temperature 114° C; NDL galvanostatic current: 101.5 mA; Pt anode area: 96.5 cm$^2$ (geometrical); graphite-felt anode area: 104 cm$^2$ (geometrical); supporting electrolyte: 5.6 M H$_2$SO$_4$.

coal is consumed and thereafter it decreases very slowly up to the point where the mechanism itself appears to change; beyond this point CO composition decreases further. It is also clear from Fig. 4 that the CO production rate is higher, if in place of platinum a graphite-felt anode is used. This behaviour may be attributed to the known better electrocatalytic behaviour of Pt as compared to graphite for CO oxidation and, therefore, a part of the CO is assumed to be further oxidized to CO$_2$ on the platinum electrode.

Another noticeable difference is the decrease in CO concentration when about 20% of the coal is consumed, whereas, using a platinum anode the CO concentration is observed to increase beyond the state of 20% consumed. The ratio of cumulative gas volume collected at the anode to that collected at the cathode during a galvanostatic experiment at a constant oxidation current of 101.5 mA, is plotted in Fig. 5 against the fraction of NDL consumed, for both platinum and graphite anodes. It is seen that the volume ratio of the gases collected at the cathode to those at the anode

ranged from about 9.1 to 3.7; the higher ratios were obtained at the beginning of the experiment but they then decreased. According to the stoichiometry of Reaction 3 this gas ratio should be about 2. Cathode-to-anode gas ratios greater than about 2.0 can be attributed mainly to accumulation of oxygen on the coal particles in the form of functional groups such as —COOH, —CHO, CH$_2$OH and the like. Moreover, higher relative amounts of H$_2$ may also be attributed in part as arising from the hydrogen content of the coal.

Preliminary approximate analysis of the residual carbon after NDL gasification reveals preferential consumption of volatile components which are expected to be rich in hydrogen. The production of anode gas is probably strongly related to the concentration of oxygen complexes in and on the surface of coal. Binder et al. reported [5] that during anodic oxidation of graphite a surface layer forms first and only thereafter does CO$_2$ evolution begin. As the oxidation process advances, surface oxides may build up to steady-state concentration on the coal, whereupon the
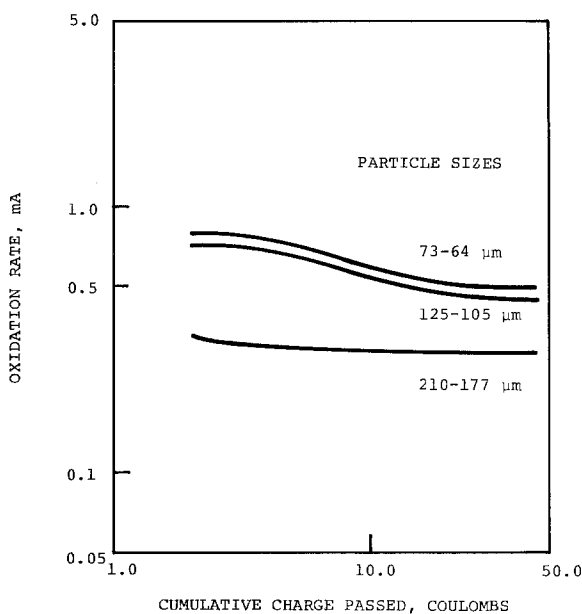
Fig. 6. Effect of particle size on the oxidation rate of Pittsburgh seam coal. Slurry conc. = 0.05 g cm⁻³; total electrolyte volume = 80 cm³; potential = 1.0 V; temp. + 40° C; supporting electrolyte: 4.1 M $H_2SO_4$; graphite rod area = 2.3 cm² (geometrical).

anode compartment gas generation rate increases. The initially higher concentration of CO as shown in Fig. 4, was also observed using Pt or graphite electrodes as well as with coals having different contents of volatile material, or with char and activated charcoal. Such higher CO concentration occurred in both potentiostatic and galvanostatic experiments. This behaviour probably has little connection with oxidation of the hydrocarbon portion of the coal. It is more plausible that the amounts and stabilities of oxygen complexes on the coal would most strongly affect the relative amounts of CO and $CO_2$ produced.

## 5. Effect of particle size

The particle size of coal affects the rate of electro-chemical oxidation of coal. Some hypotheses to explain this behaviour are:

(i) larger coal particles might encounter the anode more effectively. It has been shown [6], for example, that the depth of penetration of particles through the hydrodynamic boundary layer at an electrode depends on particle radius;

(ii) if the site of reaction is the particle surface, small particles with higher specific surface area should react more rapidly on a unit mass basis;

(iii) because of the mineral nature of coal and the inherent heterogeneity associated with its maceral structure, it is likely that sieving pul-

verized coal into several different fractions based on particle size might produce a corresponding classification of chemical reactivity according to particle size.

The contrary nature of effects (i) and (ii) should be noted as well as the difficulty of making a priori prediction of the direction of effect (iii). Using a graphite rod as anode North Dakota Lignite of three different particle-size ranges (177–210 $\mu$m; 105–125 $\mu$m; 64–73 $\mu$m) was anodically oxidized under the same conditions and the measured oxidation rates are plotted as current versus total charge passed in Fig. 6. These results show that smaller particles provide higher rates during electro-oxidation and this finding is in general accordance with that of conventional coal gasification and conventional notions of the chemical reactivity of solid particles, i.e. smaller particles are more reactive. When Pt gauze was used as anode, a more complex particle-size effect was observed [3] initially on the rate of electrochemical oxidation. The different nature of the surface of the different electrodes probably caused this.

## 6. Effect of coal concentration

Increasing coal-to-electrolyte loading increases the oxidation rate. Fig. 7 shows the interrelationship of oxidation current, temperature and coal-to-
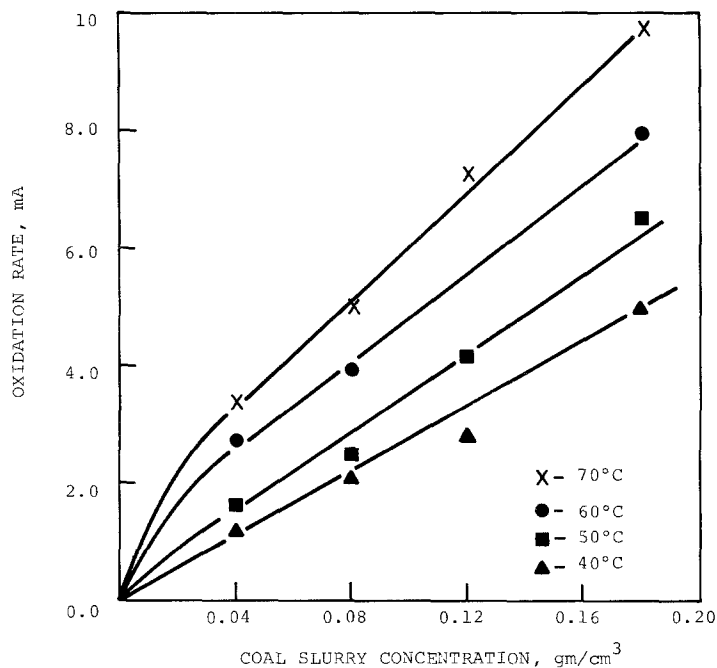
Fig. 7. Effect of coal concentration on the oxidation rate. Fresh NDL; total electrolyte volume: 80 cm³; supporting electrolyte: 4.1 M $H_2SO_4$; particle size $\leqslant 88\,\mu m$; potential: 1.0 V; graphite rod anode area: 2.3 cm² (geometrical). Control experiments with no coal gave currents less than 0.01 mA.

electrolyte concentration; increasing temperature causes current to increase as expected. The rate of change of oxidation current, $I$, due to change of coal concentration, $C$, in the coal concentration range 0.04 g cm⁻³ to 0.175 g cm⁻³ is constant at a particular temperature, and increases slightly as the temperature rises. Similar to our previous findings [3] with a platinum anode, when a graphite-rod anode is used, the reaction order

Table 1. Order of the potentiostatic oxidation reaction with respect to coal concentration

| Potential of oxidation | Temperature of oxidation (° C) | Order |
|---|---|---|
| 0.75 | 50 | 0.78 |
| 0.85 | | 0.80 |
| 1.00 | | 0.92 |
| 1.15 | | 0.93 |
| 1.0 | 40 | 0.96 |
| | 50 | 0.92 |
| | 60 | 0.78 |
| | 70 | 0.73 |

Electrolyte: 4.1 M $H_2SO_4$; total electrolyte volume = 80 cm³; coal sample: North Dakota Lignite of particle size: $\leqslant 88\,\mu m$; anode: graphite rod of external geometric area of about 2.3 cm²; coal slurry concentrations were in the range 0.04 to 0.18 g cm⁻³.

with respect to coal concentration, in the range 0.04 g cm⁻³ to 0.175 g cm⁻³, as shown in Table 1, was also found to decrease with increasing temperature (40 to 70° C) and to increase with increasing potential (0.85 to 1.15 V). Depending on the experimental temperature and potential, the order of reaction was in the range of 0.73 to 0.96.

## 7. Effect of potential on the rate of electro-oxidation

The higher the potential the greater the oxidation current. This behaviour was observed using a graphite rod anode and the experimental results are shown in Fig. 8 where the cell potential of electro-oxidation of coal is plotted against the rate of the reaction, at four different temperatures.

## 8. Activation energy

By application of Arrhenius' relationship to the measured rate data in the temperature range 25–110° C, the activation energy of electrochemical coal gasification at a graphite anode was estimated to lie between 9 and 11 kcal mol⁻¹, depending on the operating conditions. This activation energy is essentially independent of
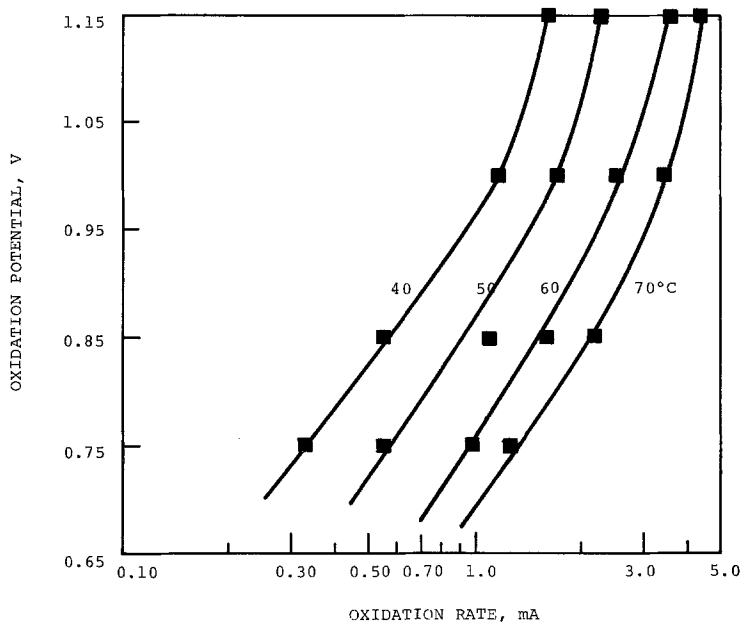
Fig. 8. Effect of cell potential on the oxidation rate at different temperatures. Fresh NDL; slurry conc. = 0.04 g cm$^{-3}$; particle size $\leqslant$ 88 $\mu$m; total electrolyte volume = 80 cm$^3$; supporting electrolyte: 4.1 M H$_2$SO$_4$; graphite rod anode area: 2.3 cm$^2$ (geometrical).

sulphuric acid concentration of the electrolyte; a small decrease of the activation energy was observed with increasing coal concentration.

## 9. Change of graphite electrodes during electro-oxidation of coals

In connection with the use of graphite as the anode electrode for electrochemical coal gasification it is known [5, 7] that graphite itself can undergo electrochemical oxidation, at the experimental conditions employed; such oxidation would be expected to make a corresponding contribution to the total measured reaction rate; simultaneous passivation of the electrode due to its oxidation, might also be expected. As Fig. 7 indicates, however, only very small such oxidation currents were observed due to anodic

reaction of the graphite electrode itself when no coal was present, but under conditions otherwise identical. In the absence of coal the oxidation currents were always less than 0.01 mA which is orders of magnitude less than the rate when coal was present in the anolyte. Moreover, Table 2 shows that the mass of the anode is essentially unchanged after oxidation of coal, even after the passage of significant charge. As Table 2 reveals, the weight change of the graphite-rod electrode can only account for a small portion of the total charge passed. This behaviour is not unexpected based on the well recognized relative chemical inertness of graphite compared to coal. The results of other experiments shown in Fig. 9 also demonstrate that graphite has lower electrochemical reactivity. In Fig. 9 reaction rates of Pittsburgh coal and of a graphite powder (obtained

*Table 2. Change of graphite anode during electrochemical oxidation*

| Wt anode before reaction $W_1$ (g) | Wt. anode after reaction $W_2$ (g) | Wt gain due to reaction $W_2 - W_1$ (g) | $Q_1$, charge equivalent to weight gain (C) | $Q_2$, charge actually passed during the exp. (C) | $(Q_1/Q_2)$ 100 (%) |
|---|---|---|---|---|---|
| 5.35 | 5.355 | 0.005 | 60.3* | 55 000 | 0.10 |

Graphite rod anode area: 2.3 cm$^3$ (geometrical); North Dakota Lignite slurry conc.: 0.04–0.18 g cm$^{-3}$; temperature: 40–70° C; potential: 0.75–1.0 V; electrolyte: 4.1 M H$_2$SO$_4$ (total volume: 80 cm$^3$).

*This calculation is based on the assumption that the weight increase is due to the formation of surface oxide on the graphite rod.
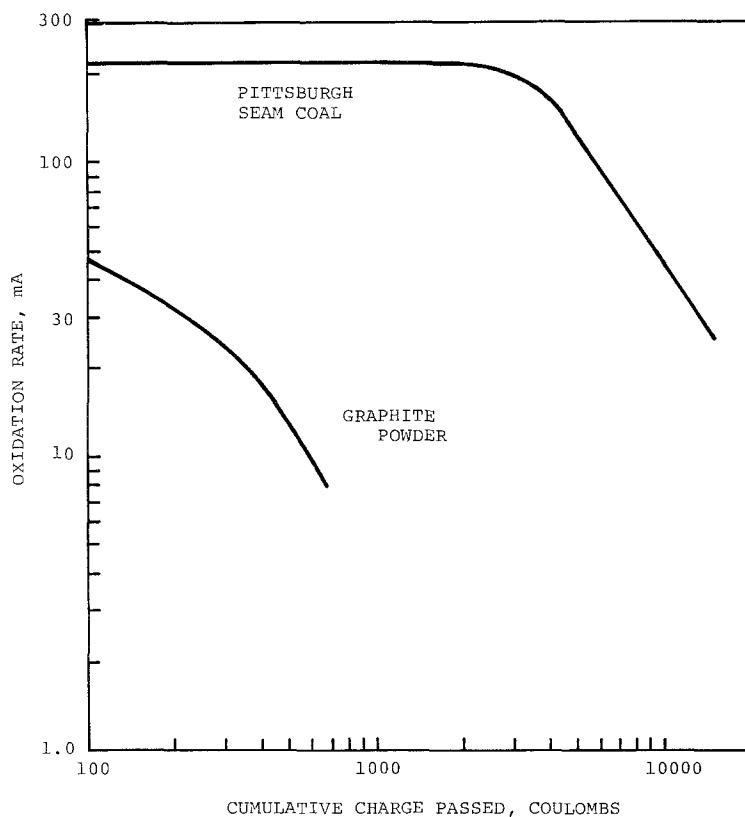
Fig. 9. Comparison of the oxidation rate of Pittsburgh coal and graphite powder. Slurry conc.: $0.069\,g\,cm^{-3}$; total volume $= 477\,cm^3$; supporting electrolyte: $5.6\,M\,H_2SO_4$; particle size $\leqslant 44\,\mu m$; temp. $114^\circ C$; potential $= 1.0\,V$; Pt anode area: $96.5\,cm^2$ (geometrical).

from Kropfmühle Graphitwerke, Germany) are plotted against cumulative coulombs passed. Furthermore no significant passivation of the graphite rod or the graphite felt material used as anodes owing to the formation of surface oxides, was observed by using the same electrode in consecutive experiments without any procedure to restore or pretreat the surface. After 400 h reaction time the graphite-felt anode of Fig. 3 was reused, without any surface pretreatment, to oxidize fresh samples of NDL coal with the result that the curve of Fig. 3 was essentially reproduced under the same conditions and with similar product formation.

A problem in the case of the felt anode, was the accurate measurement of changes in weight due to anodic oxidation because small coal particles lodge in the structure of the electrode and it is difficult to separate them. Scanning electron photomicrographs of the anode, both before and after reaction are shown in Fig. 10, at two different magnifications. The figures do not reveal thinning, deformation, rupture or any other observable

change in the fibres of the felt anode due to its use as anode for 400 h.

## 10. Electrolytes for coal gasification

In view of the expected dependence of coal electro-oxidation on water activity and the sensitivity of catalysts to poisoning, it may be expected that the rate could vary widely depending on the electrolyte used. This has motivated us to examine the performance of several electrolytes. Although we have found that electrochemical gasification of coal proceeds readily in alkaline as well as acidic electrolytes, the solubility of $CO_2$ in alkaline electrolytes could conceivably be problematic under some circumstances. As an alternative to a simple caustic electrolyte, buffer systems such as $CO_3^{2-}/HCO_3^-$ offer promise. This kind of medium is less corrosive than an oxidizing acid electrolyte such as $H_2SO_4$ and would presumably reduce costs of materials and, in principle, reject $CO_2$. Another alternative to the $H_2SO_4$ electrolyte in electrochemical coal
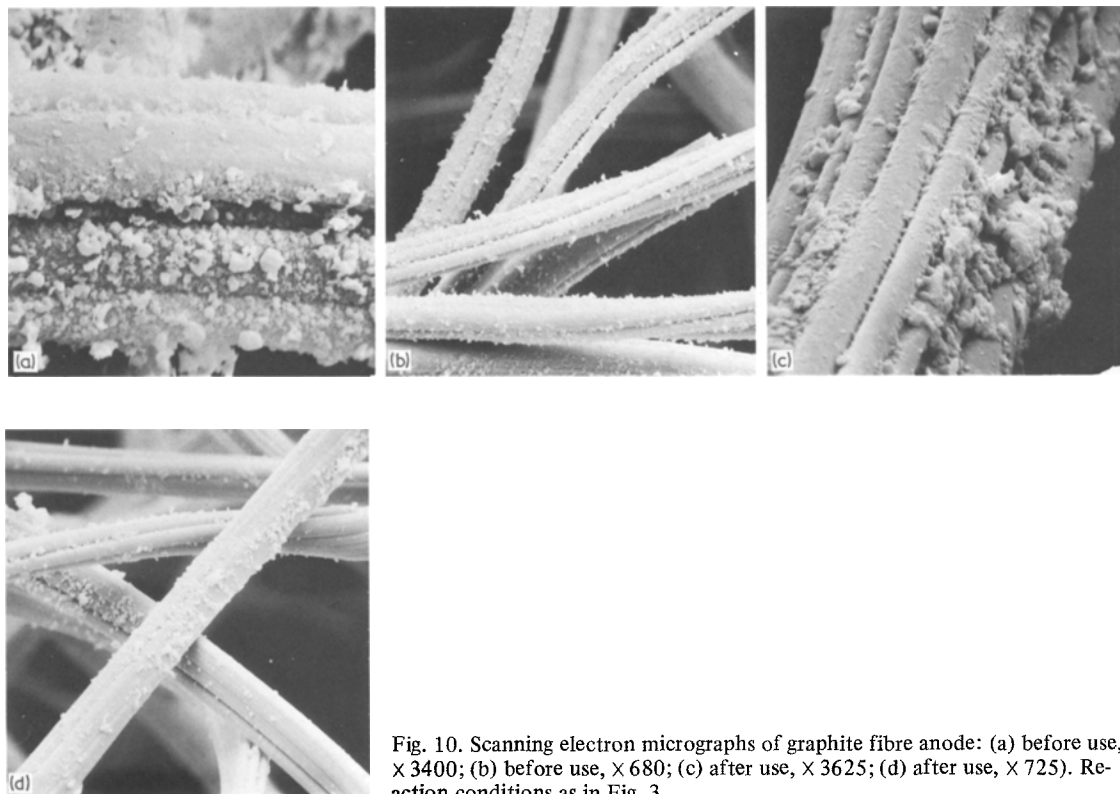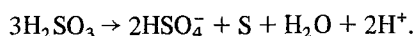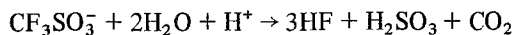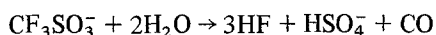
Fig. 10. Scanning electron micrographs of graphite fibre anode: (a) before use, ×3400; (b) before use, ×680; (c) after use, ×3625; (d) after use, ×725). Reaction conditions as in Fig. 3.

gasification is $H_3PO_4$ which can be used at much higher temperature than $H_2SO_4$. We have found, however, that the performance of this electrolyte was much poorer than that in $H_2SO_4$; similar results have been reported [8] for methanol oxidation. When phosphoric acid was used as an electrolyte currents for oxidation of Pittsburgh coal were about one tenth those observed with $H_2SO_4$ electrolytes under approximately comparable conditions. Trifluoromethane-sulphonic acid has been suggested as an attractive electrolyte for methanol electro-oxidation on smooth Pt [9] and this electrolyte has already proved quite successful for hydrogen electro-oxidation and oxygen electro-reduction [9]. In principle $CF_3SO_3H \cdot H_2O$ should be an ideal electrolyte – it has high ionic conductivity, has a weakly complexing anion and appears to be relatively stable and of acceptable corrosivity [10]. We examined aqueous solutions of $CF_3SO_3H$ in coal electro-oxidation and found reaction rates comparable to those observed with $H_2SO_4$ solution of similar concentration. Figs. 11 and 12 show that

the reaction rates in $H_2SO_4$ and TFMSA are roughly comparable under approximately the same conditions. Lower reaction rates were observed with TFMSA at higher temperatures ($> 80°$ C) which may be ascribed to the inherent instability of this reagent above $60°$ C. These findings are in accord with previous results of other workers [11] who evaluated $CF_3SO_3H \cdot H_2O$ and its aqueous solutions as electrolytes for methanol electro-oxidation. The aqueous $CF_3SO_3^-$ anion is probably decomposed in the electro-chemical cell as follows [12]:

$$CF_3SO_3^- + 2H_2O \rightarrow 3HF + HSO_4^- + CO$$

$$CF_3SO_3^- + 2H_2O + H^+ \rightarrow 3HF + H_2SO_3 + CO_2$$

$$3H_2SO_3 \rightarrow 2HSO_4^- + S + H_2O + 2H^+.$$

As an alternative to TFMSA, Brummer *et al.* [14] investigated a number of methane polysulphonic acids, i.e. $CX_2(SO_3H)_2$ and $CX(SO_3H)_3$ where X = H and Cl. The results showed that these acids were susceptible to both oxidation and reduction on a Pt electrode at higher temperature ($130°$ C).
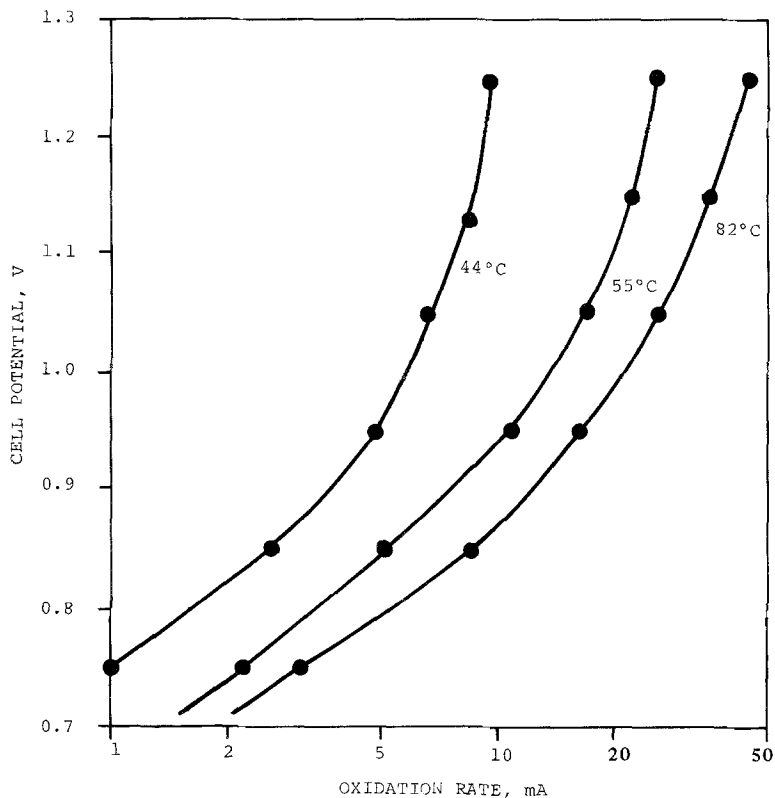
Fig. 11. Relationship between cell potential and oxidation current. Electrolyte: 10% $H_2SO_4$. N.D. lignite concentration 0.068 g cm$^{-3}$. Particle size: 63–74 $\mu$m. Electrode area: 5.3 cm$^2$ (geometric, superficial).
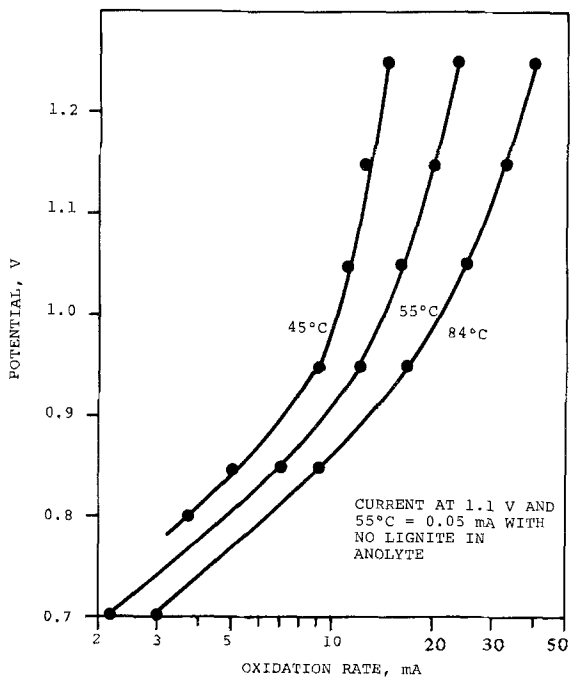


Fig. 12. Relationship between cell potential and oxidation current. Electrolyte: 10% TFMS. N.D. lignite concentration 0.069 g cm$^{-3}$. Particle size: 63–74 $\mu$m. Electrode area: 5.3 cm$^2$ (superficial, geometric).
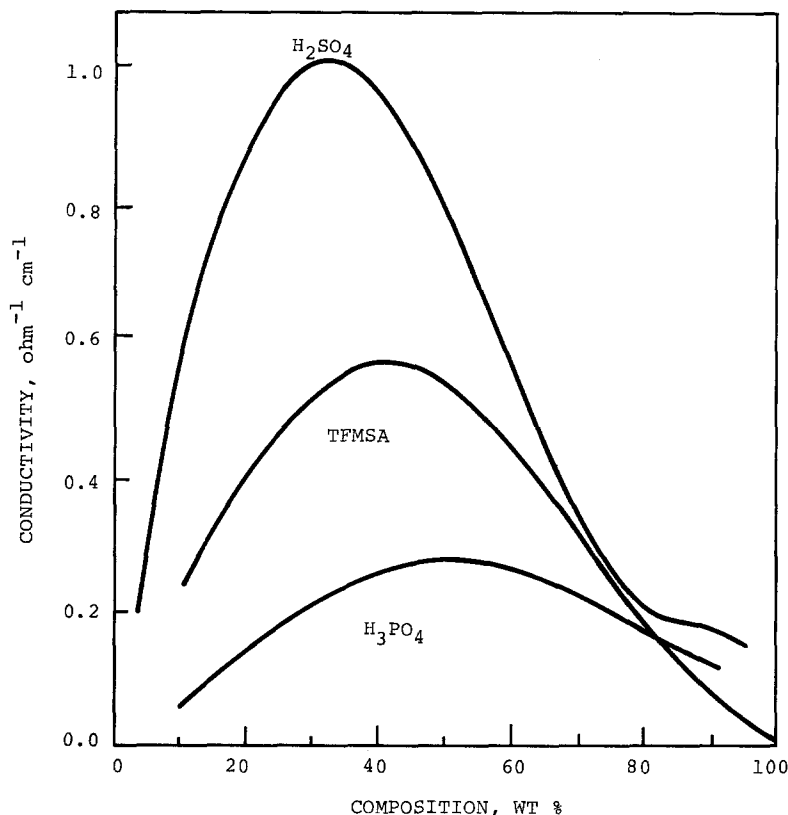
Fig. 13. Conductivities of
acid—water solutions at 40° C,
from [15].

The specific conductances of water solutions of
trifluoromethane sulphonic acid monohydrate,
$H_2SO_4$ and $H_3PO_4$ at 40° C are plotted in Fig. 13
which is taken from [13]. The conductive
superiority of $H_2SO_4$ solutions to $H_3PO_4$ or
TFMSA solutions is evident from this graph. In
view of this ranking it is not unexpected that
$H_2SO_4$ remains the most favourable electrolyte
for electro-oxidation of coal that we have yet
investigated.

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