OPTIMIZATION OF PT-DOPED KOCITE® ELECTRODES

IN H₃PO₄ FUEL CELLS

by

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Optimization of Pt-Doped KOCITE Electrodes in H₃PO₄ Fuel Cells.

Electrochemistry, fuel cells, electrocatalysts, Kocite® materials, pyropolymer, phosphoric acid electrolyte.

The use of UOP Inc. Kocite® electrocatalysts as low-cost air and/or fuel electrocatalysts in phosphoric acid electrolyte fuel cells is being optimized with respect to some of the electrocatalyst and electrode structure parameters. Kocite electrocatalysts are made from Kocite® materials, which are composite structures consisting of pyropolymer chemically bonded to refractory substrates. Fuel cell electrodes are fabricated from these electrocatalysts and normally tested as anodes or cathodes in model fuel cells with standard platinum-black.
Different types of Kocite electrocatalysts were produced by first leaching of the alumina substrate from a Kocite material and then impregnating the resulting pyropolymer structure with platinum using one of several different techniques. Kocite electrocatalysts fabricated by these techniques normally have a 650 m²/g surface area for a 15 wt-% platinum loading.

Teflon-bonded gas diffusion electrodes, with a porous graphite-paper backing were fabricated from these Kocite electrocatalysts by both machine-calendering and sheet molding techniques. Typical Kocite electrode platinum loadings were between 0.5 and 0.6 mg/cm². Model fuel cell test results were normally obtained at 180°C with air cathodes and H₂ anodes operated at atmospheric pressure. The best performance levels obtained using a Kocite cathode were 0.62 volts at 200 mA/cm² with a 65 mV oxygen gain at that current. With a cell having both a Kocite anode and cathode, 0.60 volts at 200 mA/cm² was achieved. Limited long term testing showed that Kocite electrodes can perform with good stability for at least 4500 hrs. at 180°C.

*"Kocite" is a registered trademark of UOP Inc.
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</tr>
</tbody>
</table>
I. INTRODUCTION

1.1 Objectives

The objective of this program is to optimize the performance of low-cost air and fuel electrodes fabricated from platinum-impregnated UOP Kocite materials for use in phosphoric acid electrolyte fuel cells operating at temperatures near 180°C. To accomplish this objective, various Kocite electrocatalysts are produced and fabricated into state-of-the-art fuel cell electrodes. These electrodes are tested as cathodes or anodes in model fuel cells using conventional platinum black electrodes as counter electrodes. The cell testing determines both the performance level and long term endurance attainable with Kocite electrodes.

This report summarizes the results obtained during the last nine months. Both the Kocite electrocatalyst formulations prepared during this period and the performance levels attained with Kocite electrodes made from these electrocatalysts will be discussed, along with recommendations for future research.

1.2 Background

The current research effort continues the optimization of Kocite electrocatalysts for use in phosphoric acid electrolyte fuel cell electrodes. The program organization established earlier is being continued. This involves a joint program with UOP Inc. as prime contractor and Energy Research Corporation (ERC, a subsidiary of St. Joe Minerals Corporation) as subcontractor. A broad division of effort is made on the basis of the UOP capability to produce Pt-impregnated Kocite electrocatalysts and the ERC capability to fabricate and optimize electrode structures. Testing and diagnostic evaluations of these electrodes are carried out jointly by UOP and ERC.

1.3 Review of Previous Results

In this section the status of this program at the beginning of the present contract period will be briefly reviewed to provide a background for a discussion of the effort made during the last nine months.

The nature of Kocite materials and many of the process conditions for preparing these materials have been described in earlier reports.1,2 During the previous contract periods a number of different Kocite ma-


Materials were examined as electrocatalyst supports. This evaluation led to the choice of a Kocite material using Alcoa's Hydral 705 as the starting substrate material. Relevant properties of this type of Kocite material are given in Table I. All Kocite materials used during the present contract period are of this type.

### Table I

**Kocite Material Properties**

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Substrate</td>
<td>Hydral 705</td>
</tr>
<tr>
<td>Pyropolymer Precursor</td>
<td>Benzene</td>
</tr>
<tr>
<td>Carbon Content</td>
<td>20 to 26 wt-%</td>
</tr>
<tr>
<td>Resistivity at 25°C</td>
<td>0.08 to 0.20 Ω·cm</td>
</tr>
<tr>
<td>Surface Area</td>
<td>80 to 120 m²/g</td>
</tr>
<tr>
<td>Average Pore Diameter</td>
<td>5.5 to 6.0 nm</td>
</tr>
<tr>
<td>Average Particle Diameter</td>
<td>0.5 μm</td>
</tr>
</tbody>
</table>

All Kocite electrocatalysts prepared during the previous contract period were made by the platinum impregnation of Kocite materials. Toward the end of that period it was ascertained that the alumina substrate was leached by phosphoric acid from small Kocite particles during fuel cell operation. As a result, experiments were initiated to determine the properties of leached Kocite materials. The initial preparations of these leached materials indicated a high surface area (~700 m²/g) was attainable, but no Kocite electrocatalysts were prepared from these materials.

A substantial Kocite electrode optimization program was undertaken during the previous contract period. Variations in Teflon content, catalyst layer density, catalyst layer thickness, and electrode prewetting (with H₃PO₄) were studied and numerous cells tested at UOP and ERC. Generally ERC's Kynol matrices were used but one cell was assembled using a SiC matrix prepared by ERC. The best cell performance results obtained up to the inception of the present contract period are listed in Table II. For both cells the Kocite electrode was used as a cathode at 180°C and had a Pt-loading of ~0.5 mg/cm².

### Table II

**Best 1976 Cell Performance Results**

<table>
<thead>
<tr>
<th>Kocite Electrode</th>
<th>Matrix Material</th>
<th>Terminal Voltage on air 100 ma/cm²</th>
<th>Terminal Voltage on air 200 ma/cm²</th>
<th>O₂ Gain at 100 ma/cm²</th>
<th>O₂ Gain at 200 ma/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>3380-156G</td>
<td>Kynol</td>
<td>0.65 V</td>
<td>0.565 V</td>
<td>60 mV</td>
<td>70 mV</td>
</tr>
<tr>
<td>3380-156G</td>
<td>SiC</td>
<td>0.66 V</td>
<td>0.57 V</td>
<td>70 mV</td>
<td>100 mV</td>
</tr>
</tbody>
</table>
1.4 Program for the Past Nine Month Period

The primary effort during this period was the preparation and evaluation of Kocite electrocatalysts made from leached Kocite materials and catalyzed by various Pt-impregnation techniques. This required an optimization of the leaching process by which the alumina substrate is leached from the Kocite material by H$_3$PO$_4$ as well as substantial characterization of the leached materials by various techniques. The main efforts of the last nine months have been in the following areas:

(A) Preparation and Leaching of Kocite Materials

A large number of Kocite materials were prepared and leached in H$_3$PO$_4$. By varying the leaching conditions and examining the product, desirable leaching conditions were established by which a high-surface-area leached Kocite material can be obtained.

(B) Kocite Electrocataylst Preparation

A large number of Kocite electrocatalysts were prepared both by first leaching the Kocite material and then impregnating with Pt or Pt-impregnation followed by leaching. Kocite electrocatalysts were prepared by different techniques involving the use of

(1) Chloroplatinic Acid,

(2) Platinum Diaminedinitrite,

(3) Platinum Acetylacetonate.

(C) Kocite Electrode Fabrication

Several Kocite electrocatalysts were delivered to ERC for incorporation into fuel cell electrodes. Several machine calendared electrode batches were fabricated for testing and a number of experimental electrode batches fabricated by a sheet molding process were also prepared. During this last nine month period less effort was placed on Kocite electrode structure optimization than during 1976.

(D) Kocite Electrode Testing

Kocite electrodes were tested almost exclusively as cathodes using 2 mg/cm$^2$ Pt black counter electrodes. The cell performance goal of 0.600 V at 200 ma/cm$^2$ with a Kocite cathode of ~0.5 mg/cm$^2$ Pt-loading was met with cells operating on air and H$_2$ at 180°C. Endurance data were accumulated on several cells.

A detailed discussion of the results obtained in each of these areas is presented in the following sections.
II. PREPARATION AND LEACHING OF KOCITE MATERIALS

2.1 Kocite Materials

The preparation of the Kocite materials used during the present contract period is reported in this section. In all cases Alcoa's Hydral 705 alumina substrate was used with a benzene pyropolymer precursor. Some of the preparation conditions as well as some of the chemical and physical properties of these Kocite materials are listed in Table III. Equipment replacement and recalibration led to some variation in the properties of Kocite materials made during the early part of this contract period, however, with the exception of Kocite batch 3289-195, Kocite materials used for electrocatalyst preparation had starting surface areas between 82 and 124 m²/g.

A pore volume distribution was obtained on two Kocite materials by nitrogen adsorption for later comparison with the pore volume distribution of the resulting Kocite electrocatalyst. The pore volume distribution of Kocite batch 3289-120 is shown in Figure 1. The most important characteristic of this distribution is the relatively narrow range of pore volumes. The distribution maximum occurs at 4.0 nm diameter pores and two shoulders in the distribution are found at pore diameters of 8.0 and 12.5 nm. For this Kocite material the dominant surface area contributions come from pores with diameters less than ~5.0 nm diameter.

An attempt was also made to characterize the structure of Kocite particles by Scanning Electron Microscopy (SEM). An example of the results obtained with Kocite batch 3576-60 is shown in Figure 2. In this figure, Kocite powders are shown at a magnification of 100,000X. Based on this micrograph and others, Kocite powders made with the Hydral 705 substrate appear to have a platelet structure with many particles having their largest dimension smaller than 200 nm. Micrographs of this type not only indicate that further study of Kocite materials and electrocatalysts at these magnifications are warranted, but that the platelet structure and proper sample preparation may allow useful Transmission Electron Micrographs (TEM) to be obtained. Higher magnification studies with TEM may allow the pore structure not visible in Figure 2, but indicated by Figure 1, to be resolved.

2.2 Leaching of Kocite Materials

To determine the most desirable conditions for leaching the alumina substrate from Kocite material, a variety of small batch experiments were run in which the ratio of H₃PO₄ to Kocite powder, leaching temperature, and leaching time were varied. The range of parameter variations

---

3 Pictures taken at Scanning Electron Microscope Users' Laboratory at the University of Chicago. The lab is operated on a resource grant from the National Institutes of Health.
<table>
<thead>
<tr>
<th>Batch Number</th>
<th>3289-120</th>
<th>3289-140</th>
<th>3289-195</th>
<th>3576-10</th>
<th>3576-20</th>
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<tr>
<td>Preparation</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Substrate</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
</tr>
<tr>
<td>Pyropolymer Precursor</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
</tr>
<tr>
<td>Pyrolysis Temperature (°C)*</td>
<td>840</td>
<td>870</td>
<td>830</td>
<td>810</td>
<td>840</td>
</tr>
<tr>
<td>Characteristics</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon Content (Wt-%)</td>
<td>21.52</td>
<td>26.22</td>
<td>27.35</td>
<td>23.8</td>
<td>26.62</td>
</tr>
<tr>
<td>Resistivity, ρ, @ 25°C (Ω-cm)</td>
<td>0.17</td>
<td>0.22</td>
<td>0.086</td>
<td>0.183</td>
<td>0.080</td>
</tr>
<tr>
<td>Apparent Bulk Density (g/ml)</td>
<td>0.30</td>
<td>0.30</td>
<td>0.267</td>
<td>0.288</td>
<td>0.265</td>
</tr>
<tr>
<td>Surface Area (m²/g)</td>
<td>107</td>
<td>122</td>
<td>38</td>
<td>124</td>
<td>77</td>
</tr>
<tr>
<td>Pore Volume (ml/g)</td>
<td>0.15</td>
<td>0.17</td>
<td>0.06</td>
<td>0.15</td>
<td>0.11</td>
</tr>
<tr>
<td>Average Pore Diameter (nm)</td>
<td>5.6</td>
<td>5.6</td>
<td>6.3</td>
<td>4.8</td>
<td>5.7</td>
</tr>
<tr>
<td>Pore Volume Distribution Measured</td>
<td>Yes</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>

*Temperature changes caused in part by thermocouple repair and repositioning at mid-bed.
<table>
<thead>
<tr>
<th>Batch Number</th>
<th>3576-40</th>
<th>3576-60</th>
<th>3576-65</th>
<th>3576-70</th>
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<td>Preparation</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Substrate</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
<td>Hydral 705</td>
</tr>
<tr>
<td>Pyropolymer Precursor</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
<td>C₆H₆</td>
</tr>
<tr>
<td>Pyrolysis Temperature (°C)*</td>
<td>840</td>
<td>865</td>
<td>870</td>
<td>865</td>
</tr>
<tr>
<td>Characteristics</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Carbon Content (Wt-%)</td>
<td>24.17</td>
<td>25.33</td>
<td>24.77</td>
<td>22.92</td>
</tr>
<tr>
<td>Resistivity, ρ, @ 25°C (Ω-cm)</td>
<td>0.12</td>
<td>0.14</td>
<td>0.14</td>
<td>0.13</td>
</tr>
<tr>
<td>Apparent Bulk Density (g/ml)</td>
<td>0.290</td>
<td>0.278</td>
<td>0.278</td>
<td>0.292</td>
</tr>
<tr>
<td>Surface Area (m²/g)</td>
<td>94</td>
<td>123</td>
<td>82</td>
<td>88</td>
</tr>
<tr>
<td>Pore Volume (ml/g)</td>
<td>0.14</td>
<td>0.17</td>
<td>0.11</td>
<td>0.12</td>
</tr>
<tr>
<td>Average Pore Diameter (nm)</td>
<td>6.0</td>
<td>5.5</td>
<td>5.4</td>
<td>5.5</td>
</tr>
<tr>
<td>Pore Volume Distribution Measured</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
</tbody>
</table>

*Temperature changes caused in part by thermocouple repair and repositioning at mid-bed.
Figure 1
Differential Pore Volume Distribution of Kocite Material 3289-120

[Graph depicting differential pore volume distribution with pore diameter on the x-axis and differential pore volume on the y-axis.]
and the leaching conditions considered optimal are indicated in Table IV. These experiments indicate that desired product of Kocite material starting with a surface area of \( \approx 100 \text{ m}^2/\text{g} \) and a 25 wt-% carbon content is a leached Kocite material with a surface area of \( \approx 800 \text{ m}^2/\text{g} \) and a 0-5 wt-% ash content relative to carbon. Such a material is readily obtained with the process conditions given in Table IV.

For the preparation of electrocatalyst supports, the equipment used for the leaching process was scaled up to handle 250 g Kocite batches (50-70 g of product). The Kocite powders are leached in a 5 L heated flask and stirred during the leaching process. The resultant product is washed and dried several times to remove all traces of \( \text{H}_3\text{PO}_4 \) prior to Pt impregnation.

The characteristics of the leached Kocite materials used in electrocatalyst preparation are listed in Table V. In general, leached Kocite materials are denoted by the addition of an "L" following the Kocite batch identification number. In two cases Kocite electrocatalysts were prepared by leaching subsequent to Pt-impregnation. In these cases the electrocatalyst identification number ends in an "L".

For selected materials, ash content, resistivity and a surface area analysis were performed. Ash contents less than 5 wt-% were normally obtained as indicated in Table V. The resistivity of a leached Kocite material showed no significant change from that of the starting Kocite material. Surface area measurements indicated that leached Kocite material surface areas are in the expected range.
Table IV  
Production of Leached Kocite Materials

| Production Conditions |  |
|-----------------------|-----------------
| 10 ml of 85% H₃PO₄ per gram Kocite material |  |
| 140°C for 24 hrs. |  |

<table>
<thead>
<tr>
<th>Characterization of Material</th>
<th></th>
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<tbody>
<tr>
<td>Surface Area</td>
<td>750–1000 m²/g</td>
</tr>
<tr>
<td>Inorganic Residue</td>
<td>0–5 wt-%</td>
</tr>
<tr>
<td>ABD</td>
<td>0.05 g/ml</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Leaching Variations Investigated</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Temperature (water content)</td>
<td>100°C (15%) to 180°C (0%)</td>
</tr>
<tr>
<td>H₃PO₄ to Kocite material Ratio</td>
<td>25 ml/g to 5 ml/g</td>
</tr>
<tr>
<td>Leaching Time</td>
<td>8 hr. to 4 days</td>
</tr>
<tr>
<td>Batch Number</td>
<td>3289-120L</td>
</tr>
<tr>
<td>--------------</td>
<td>-----------</td>
</tr>
<tr>
<td>Preparation</td>
<td></td>
</tr>
</tbody>
</table>
| Kocite Material or Electro-
  catalyst Batch Used* | 3289-120  | 3289-140  | 3289-195  | 3576-10  | 3576-20A  |
| Leaching Temperature (°C) | 140        | 140        | 141        | 142      | 143       |
| Leaching Time (Hrs)        | 24         | 22         | 24         | 24       | 24        |
| Characterization           |           |           |           |          |           |
| Ash Content (Wt-%)         | 4.9        | 0.0        | 2.0        | 0.0      |           |
| Resistivity, ρ, @ 25°C (Ω·cm) | 0.107     | 0.107     |           |          |           |
| Surface Area (m²/g)        | 989        |           |           |          |           |
| Pore Volume (ml/g)         | 1.76       |           |           |          |           |
| Average Pore Diameter (nm) | 7.1        |           |           |          |           |
| Pore Volume Distribution Measured | No       | No       | No        | No       | No        |

*Batch 3576-20AL was Pt impregnated prior to leaching. See Tables VI and VII.
Table V (Continued)

Leaching Parameters of Kocite materials and Electro catalysts

<table>
<thead>
<tr>
<th>Batch Number</th>
<th>3576-44AL</th>
<th>3576-60L</th>
<th>3576-65L</th>
<th>3576-70L</th>
</tr>
</thead>
<tbody>
<tr>
<td>Preparation</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kocite Material or Electro- Catalyst Batch Used*</td>
<td>3576-44A</td>
<td>3576-60</td>
<td>3576-65</td>
<td>3576-70</td>
</tr>
<tr>
<td>Leaching Temperature (°C)</td>
<td>141</td>
<td>142</td>
<td>145</td>
<td>143</td>
</tr>
<tr>
<td>Leaching Time (Hrs)</td>
<td>24</td>
<td>24</td>
<td>24</td>
<td>24</td>
</tr>
</tbody>
</table>

Characterization

| Ash Content (Wt-%) | 4.8 |
| Resitivity, ρ, @ 25°C (Ω-cm) | 0.166 |
| Surface Area (m²/g) | 881 |
| Pore Volume (ml/g) | 1.57 |
| Average Pore Diameter (nm) | 7.1 |
| Pore Volume Distribution Measured | No | No | No | No |

*Batch 3576-44AL was Pt impregnated prior to leaching. See Tables VI and VII.
III. KOCITE ELECTROCATALYSTS

3.1 Kocite Electro cataly st Preparation

During the last nine month contract period about 40 Kocite electrocatalyst batches were prepared from the leached and unleached Kocite materials listed in Tables IV and V, including a number of small (<10 g) batches prepared to evaluate the various impregnation techniques. Several larger Kocite electrocatalyst batches have been prepared and delivered to ERC for incorporation into Kocite electrodes. Some of the preparation details and physical properties of electrocatalysts sent to ERC are listed in Tables VI and VII, along with those of other batches from which significant characterization results were obtained.

Details relating to the Pt-impregnation of Kocite materials (both leached and unleached) are given in Table VI. Also listed are surface area analysis results on selected electrocatalysts. Pore volume distributions were obtained for two electrocatalysts and these results will be discussed in the next section.

As indicated in Table VI, most impregnations were made using chloroplatinic acid (CPA) with either ethanol or water solvents. However, a number of Kocite electrocatalysts were also prepared using either Pt diaminedinitrite (PDD) or Pt acetylacetonate (PAA). In general, electrocatalysts were impregnated at a level of ~15 wt-% relative to the carbon content.

To discuss the physical properties of the Kocite electrocatalysts listed in Table VI and the relative merits of the various impregnation techniques, the characteristics of these Kocite electrocatalysts are listed in Table VII by impregnation technique. Also indicated is whether the electrocatalyst was prepared by first leaching the Kocite material and then impregnating with Pt or the reverse order. In Table VI results are given as well for electrocatalyst Pt crystallite size as determined by X-ray diffraction line width broadening. For the larger (60 g) batches various steps of the impregnation process were performed in different equipment than for the smaller batches. It is believed that this may have led to larger Pt crystallites being obtained with larger batches due to inadequate mixing; a problem that is currently being solved.

Several different types of catalyization experiments were made to determine the best method for producing a Kocite electrocatalyst with small Pt crystallite size. The most important results of these experiments are apparent in Table VII and are summarized as follows:

(A) Using the CPA/C₂H₅OH impregnation technique several electrocatalyst batches were prepared by either leaching then impregnating or vice versa. Pt crystallite size results indicate that more highly dispersed Pt is obtained by leaching Kocite material prior to impregnation.
<table>
<thead>
<tr>
<th>Batch Number</th>
<th>3289-127A</th>
<th>3289-144A</th>
<th>3289-144C</th>
<th>3289-197A</th>
<th>3576-12A</th>
<th>3576-12C</th>
<th>3576-12E</th>
<th>3576-12F</th>
</tr>
</thead>
<tbody>
<tr>
<td>Preparation</td>
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<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Kocite Material Used</td>
<td>3289-120L</td>
<td>3289-140L</td>
<td>3289-140L</td>
<td>3289-195L</td>
<td>3576-10L</td>
<td>3576-10L</td>
<td>3576-10L</td>
<td>3576-10L</td>
</tr>
<tr>
<td>Platinum Source*</td>
<td>CPA</td>
<td>CPA</td>
<td>PAA</td>
<td>CPA</td>
<td>PDD</td>
<td>PDD</td>
<td>CPA</td>
<td>CPA</td>
</tr>
<tr>
<td>Solvent</td>
<td>C$_2$H$_5$OH</td>
<td>C$_2$H$_5$OH</td>
<td>C$_6$H$_6$</td>
<td>C$_2$H$_5$OH</td>
<td>30% HNO$_3$</td>
<td>30% HNO$_3$</td>
<td>H$_2$O</td>
<td>H$_2$O</td>
</tr>
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<td>Reduction Atmosphere</td>
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<td>H$_2$</td>
<td>N$_2$</td>
<td>H$_2$</td>
<td>N$_2$</td>
<td>N$_2$</td>
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<td>260</td>
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<td>260</td>
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<td>260</td>
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<td>2.0</td>
<td>2.0</td>
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<td></td>
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<tr>
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<td>15</td>
<td>15</td>
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<td>15</td>
<td>14.8</td>
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<td>Surface Area (m$^2$/g)</td>
<td>661</td>
<td>610</td>
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<td></td>
<td></td>
<td>642</td>
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<td>Pore Volume (ml/g)</td>
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<td>Pore Volume Distribution Measured</td>
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<td>Yes</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Resistivity, ρ, @ 25°C (Ω·cm)</td>
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</tr>
</tbody>
</table>

*CPA = chloroplatinic acid; PDD = platinum diaminedinitrite, PAA = platinum acetylacetonate.
Table VI (Continued)

Platinum Impregnation of Kocite Materials

<table>
<thead>
<tr>
<th>Batch Number</th>
<th>3576-20AL</th>
<th>3576-44AL</th>
<th>3576-61A</th>
<th>3576-61B</th>
<th>3576-61C</th>
<th>3576-61D</th>
<th>3576-61E</th>
<th>3576-61F</th>
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<td>Kocite Material Used</td>
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<td>3576-60L</td>
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<td>PDD</td>
<td>CPA</td>
<td>CPA</td>
<td>CPA</td>
<td>CPA</td>
<td>PDD</td>
<td>CPA</td>
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<td>Solvent</td>
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<td>C₂H₅OH</td>
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<td>H₂O</td>
<td>H₂O</td>
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<td>H₂O</td>
<td>H₂O</td>
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<td>Reduction Atmosphere</td>
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<td>H₂</td>
<td>H₂</td>
<td>H₂</td>
<td>H₂</td>
<td>H₂</td>
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</tr>
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<td>10</td>
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<td>30</td>
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<td>Platinum Concentration, Pt/(Pt+C) (Wt-%)</td>
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<td>15</td>
<td>11.4</td>
<td>16.3</td>
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<td>15</td>
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<td></td>
<td>620</td>
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<td>Pore Volume (ml/g)</td>
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<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
<td>No</td>
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<td>No</td>
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<tr>
<td>Resistivity, ρ, @ 25°C (Ω-cm)</td>
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<td>0.15</td>
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*CPA = chloroplatinic acid; PDD = platinum diaminedinitrite, PAA = platinum acetylacetonate.
### Table VII

**Characteristics of Kocite<sup>R</sup> Electrocatalysts**

<table>
<thead>
<tr>
<th>Impregnation Technique</th>
<th>Electro catalyst Preparation Method</th>
<th>Batch Number</th>
<th>Batch Size (g)</th>
<th>Platinum Concentration (Wt-%)</th>
<th>Platinum X-Ray Analysis</th>
<th>Electodes Fabricated</th>
</tr>
</thead>
<tbody>
<tr>
<td>CPA/C&lt;sub&gt;2&lt;/sub&gt;H&lt;sub&gt;5&lt;/sub&gt;OH</td>
<td>Leach-Impreg</td>
<td>3289-127A</td>
<td>60</td>
<td>17.3</td>
<td>37</td>
<td>6.7</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3289-144A</td>
<td>60</td>
<td>15</td>
<td>43</td>
<td>11.7</td>
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<td></td>
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<td>3289-197A</td>
<td>60</td>
<td>15</td>
<td>40</td>
<td>10.6</td>
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<td></td>
<td>Impreg-Leach</td>
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<td>14.9</td>
<td>57</td>
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<td>3576-44AL</td>
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<td>15</td>
<td>54</td>
<td>7.4</td>
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<td>CPA/H&lt;sub&gt;2&lt;/sub&gt;O</td>
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<td>3576-61B</td>
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<td>16.3</td>
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<td></td>
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<td>12</td>
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<td>49</td>
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<td>Pt Diamine-dinitrite</td>
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<td>15</td>
<td>69</td>
<td>8.4</td>
</tr>
</tbody>
</table>

*These results are standardized as discussed in Reference 1 except that Kocite electrocatalyst 3576-66A (not listed) is used as the standard.

b Mean size of crystallites over 2.5 nm diameter.
(B) Both C₂H₅OH and H₂O solvents were tried with CPA. In general, slightly better Pt dispersions are obtained with H₂O solvents.

(C) Pt crystallite sizes on Kocite electrocatalysts prepared using PDD are much smaller than those obtained by any other impregnation technique.

(D) Attempts to prepare Kocite electrocatalysts using PAA have met with only limited success to date.

With the results obtained from these electrocatalyst preparations, the data of Table VII suggest the electrocatalyst with the highest Pt dispersion should be obtained by first leaching Kocite material and then impregnating with PDD.

3.2 Kocite Electrocatalyst Structure

A preliminary determination of the structural properties of several Kocite electrocatalysts was made in addition to the structural property determinations of Kocite materials discussed in section 2.1. As an example, the pore volume distribution of Kocite electrocatalyst 3289-127A is shown in Figure 3. Comparison of this distribution with that shown in Figure 1 for Kocite material 3289-120 indicates the changes occurring in the pore sizes both upon leaching and Pt impregnation. The peak in the PVD shifts from 4.0 to 7.0 nm and the pore volume at all pore sizes increases substantially for the much higher surface area Kocite electrocatalyst (on a weight basis). In Figure 3, shoulders in the distribution are apparent at 3.0 nm and 12.5 nm. Similar results were obtained with Kocite electrocatalyst 3289-144A.

The changes in pore volume distribution between Kocite electrocatalyst 3289-127A and Kocite material 3289-120 are most clearly shown in Figure 4, where the ratio of the pore volume distributions is plotted. This figure shows that there has been more than a 15-fold increase in the number of pores having diameters between 7.5 and 10.0 nm, while the number of pores at other diameters shows a substantially smaller increase. It is believed that these changes in the PVD from Kocite material to electrocatalyst are determined by the structural configuration of the alumina substrate during pyropolymer deposition.

An SEM micrograph of Kocite electrocatalyst 3576-61B on an electrode surface is shown in Figure 5. This micrograph was obtained at the same magnification, 100,000X, as the micrograph of Kocite material 3576-60 shown in Figure 2. Comparison of these two micrographs suggests that the gross catalyst particle structure is similar to the initial Kocite particle structure. Further SEM and TEM studies will be required to fully ascertain the similarity of these materials, but these preliminary results coupled with the PVD results suggest that the carbonaceous pyropolymer structure is neither destroyed nor substantially altered by either the leaching, Pt impregnation or electrode fabrication processes.
Figure 3
Differential Pore Volume Distribution of Kocite Electrocatalyst 3289-127A
Figure 4
Ratio of Differential Pore Volume Distributions of Kocite Electrocatalyst 3289-127A and Kocite Material 3289-120

The histogram plots the ratio of the differential pore volume at each pore diameter. The dashed line indicates the expected ratio just considering the weight changes caused by leaching and impregnation with 17.3 wt-% platinum.
IV. ELECTRODE FABRICATION AND TESTING

4.1 Electrode Fabrication

During the last nine month period, ERC has prepared Kocite electrodes by two separate techniques, 1) the standard ERC machine calendaring process used previously as described in earlier reports, and 2) an experimental process utilizing a sheet mold technique. Machine calendared (rolled) electrodes are fabricated by rolling a discrete catalyst layer and then pressure bonding this layer to a carbon fiber paper electrode substrate. In contrast, in the sheet mold process a catalyst-PTFE mixture is deposited directly onto the carbon fiber paper. The density of the sheet mold catalyst layer is equivalent to the medium density machine calendared catalyst layer used during the previous contract period. Sintering conditions of both types of catalyst layers are similar to those described in previous reports.

Teflon contents of both types of catalyst layers are typically between 40 and 50 weight percent. The lower Teflon content was emphasized in order to obtain peak electrode performance quickly at the risk of later electrode flooding. Electrode performance is expected to have superior long term stability at the higher Teflon loading, which is equivalent to the optimal Teflon loadings used during the previous contract period.

4.2 Cell Testing

Details of cell testing techniques and apparatus have been discussed in earlier reports. No changes in cell components have been made during the present period. All cells were operated at 180°C with an ERC supplied Kynol matrix and employed, with two exceptions, a 2 mg/cm² Pt black counter electrode. The other two cells were operated with Kocite electrodes as both cathode and anode.

During the previous contract period, the best cell performance was obtained with an experimental fibrous silicon carbide matrix prepared by ERC. This matrix could not be used during the present contract period because of both reproducibility and materials availability problems. Other ERC silicon carbide matrix fabrication techniques were not judged by ERC to be sufficiently developed to warrant their use with Kocite electrodes.

The most important change in cell testing procedures made during the present contract period was the assembly of a number of cells at ERC where the Kocite electrode was not prewetted or float-filled in phosphoric acid. Deletion of this step was possible with Kocite electrocatalysts made from leached Kocite materials, in part, because of the improved electrocatalyst utilization characteristics ERC believes are obtainable with the sheet mold electrode.
V. RESULTS AND DISCUSSION

5.1 Introduction

In this section, the characteristics and performance of Kocite electrodes tested at UOP and ERC during the last nine month contract period are presented. Where possible, the cell performance is correlated with Kocite electrocatalyst and Kocite electrode properties. The results obtained during the last nine month period are summarized in section 5.3.

5.2 Cell Performance Results

The performance results of model phosphoric acid electrolyte fuel cells with a Kocite electrode used as the cathode are reported in this section. The characteristics of the Kocite electrodes and cell builds tested at ERC and UOP are listed in Tables VIII and IX respectively. A total of 47 cells were assembled and tested. The cell build characteristics and performance levels are reported only for those cells which reached the performance goals of the previous contract period, 0.6 volts at 100 mA/cm² and 180°C with a 0.5 mg/cm² Pt-loaded Kocite cathode. Over 50% of cells assembled reached this level. The remaining cells failed to reach this performance level either as a result of cross gas leaks, electrode flooding, cell hardware problems, or problems associated with the reproducibility of sheet mold electrodes.

Three Kocite electrocatalysts were tested in electrodes at ERC, 3289-127A, 3576-61A and 3576-61B. As indicated in Tables VI and VII, these electrocatalysts had the highest Pt dispersion achieved for the Pt-impregnation of leached Kocite materials by the respective Pt-impregnation techniques. Most of the cells tested used sheet mold Kocite electrodes with a 40 wt-% Teflon content as shown in Table VIII. Kocite electrode Pt-loadings were normally between 0.50 and 0.60 mg/cm² for the 2" x 2" electrodes. A substantial number of the cells tested at ERC were assembled with no Kocite electrode prewetting.

At UOP, cell testing activity was reduced from that of the previous contract period because of the emphasis on Kocite electrocatalyst production by various Pt-impregnation techniques. As a result, many of the Kocite electrode batches produced by ERC were tested only at ERC. Testing at UOP was primarily limited to electrodes from Kocite electrocatalyst batch 3289-127A. From this batch both machine calendared and sheet mold electrodes were tested. A few electrodes were tested from other electrocatalyst batches without notable success.

Summaries of full cell performance results at 180°C obtained at both ERC and UOP are presented in Tables X and XI. The most significant result presented in these tables is that for four cell builds, one of which is an all Kocite electrode cell, the peak full cell terminal voltage reached or exceeded the performance goal of 0.60 volts terminal at 200 mA/cm² set for this contract period.
<table>
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<th>Cell Build Number</th>
<th>Kocite Electrode Batch</th>
<th>Platinum Concentration (Pt/Pt+C) (Wt-%)</th>
<th>Electro catalyst Loading (mg/cm²)</th>
<th>Platinum Loading (mg/cm²)</th>
<th>PTFE Content (wt-%)</th>
<th>Fabrication Method</th>
<th>Electrode Prewetting (days)</th>
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<tr>
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<td>0.56</td>
<td>40</td>
<td>Sheet Mold</td>
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*Kocite electrodes used as cathode and anode.
<table>
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<tr>
<th>Cell Build Number</th>
<th>Platinum Concentration (wt-%)</th>
<th>Electrolyte Loading (mg/cm²)</th>
<th>Platinum Loading (mg/cm²)</th>
<th>PTFE Content (wt-%)</th>
<th>Fabrication Method</th>
<th>Electrode Prewetting (days)</th>
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<td>Cell Build Number</td>
<td>Kocite Electrocatlyst Batch</td>
<td>Platinum Loading (mg/cm²)</td>
<td>Terminal Voltage (V) @ 100 ma/cm²</td>
<td>Terminal Voltage (V) @ 200 ma/cm²</td>
<td>Days Tested</td>
<td>Peak Performance</td>
</tr>
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<td>-------------------</td>
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<td>---------------------------</td>
<td>-----------------------------------</td>
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<td>ERC-113</td>
<td>3289-127A</td>
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<td>0.670 0.725</td>
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<td>0.65</td>
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<td>3289-127A</td>
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<td>ERC-122</td>
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<td>6 days</td>
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<td></td>
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<td>0.640 0.700</td>
<td>0.570 0.635</td>
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<td>5 days</td>
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<td>ERC-127</td>
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<td>1 day</td>
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<td>0.600 0.670</td>
<td></td>
<td>64</td>
<td>2 days</td>
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<td>ERC-138</td>
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<td>0.60</td>
<td>0.680 0.740</td>
<td>0.620 0.685</td>
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<td>22 days</td>
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<td>3576-61B</td>
<td>0.60</td>
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<td>ERC-140b</td>
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<td>0.60</td>
<td>0.670 0.730</td>
<td>0.600 0.670</td>
<td>7</td>
<td>7 days</td>
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*a* Cell voltage obtained with Kocite cathode and 2 mg/cm² Pt-black counter electrode; cell voltage measured at current collector plates.

*b* Kocite electrodes used as cathode and anode.
### Table XI

**Summary of UOP Cell Performance Results at 180°C**

<table>
<thead>
<tr>
<th>Cell Build Number</th>
<th>Platinum Loading (mg/cm²)</th>
<th>Terminal Voltage (V) @ 100 ma/cm²</th>
<th>Days Tested</th>
<th>Peak Performance</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>3289-127A-1</td>
<td>0.58</td>
<td>Air: 0.604, Oxygen: 0.674</td>
<td>10</td>
<td>3 days</td>
<td>Gas crossover</td>
</tr>
<tr>
<td>3289-127A-2</td>
<td>0.58</td>
<td>Air: 0.600, Oxygen: 0.672</td>
<td>7</td>
<td>3 days</td>
<td>Gas crossover</td>
</tr>
<tr>
<td>3289-127A-3</td>
<td>0.58</td>
<td>Air: 0.620, Oxygen: 0.692</td>
<td>9</td>
<td>2 days</td>
<td>Gas crossover</td>
</tr>
<tr>
<td>3289-127A-4</td>
<td>0.58</td>
<td>Air: 0.610, Oxygen: 0.681</td>
<td>9</td>
<td>2 days</td>
<td>Gas crossover</td>
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<td>3289-127A-5</td>
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<td>Air: 0.629, Oxygen: 0.687</td>
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<td>31 days</td>
<td>Gas crossover</td>
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<tr>
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<td>0.58</td>
<td>Air: 0.650, Oxygen: 0.716</td>
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<td>28 days</td>
<td>Reverse polarized</td>
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<tr>
<td>3289-127A-7</td>
<td>0.58</td>
<td>Air: 0.622, Oxygen: 0.669</td>
<td>14</td>
<td>8 days</td>
<td>Gas crossover</td>
</tr>
<tr>
<td>3289-127A-8</td>
<td>0.58</td>
<td>Air: 0.618, Oxygen: 0.674</td>
<td>23</td>
<td>18 days</td>
<td>Decayed</td>
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<tr>
<td>3289-127A-13</td>
<td>0.47</td>
<td>Air: 0.618, Oxygen: 0.702</td>
<td>36</td>
<td>3 days</td>
<td>Reverse polarized</td>
</tr>
<tr>
<td>3289-127A-14</td>
<td>0.47</td>
<td>Air: 0.644, Oxygen: 0.712</td>
<td>72</td>
<td>52 days</td>
<td>Continuing with decay</td>
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*Cell voltage obtained with Kocite cathode and 2 mg/cm² Pt-black counter electrode; cell voltage measured at current collector plates.*
Two of these cells, ERC-113 and ERC-128, will be discussed in more detail later in this section. Comparing the results of cell ERC-138 with the best results obtained during the previous contract period, listed in Table II, shows that for cells with a Kynol matrix, cell performance improvements of 30 mV at 100 ma/cm² and 55 mV at 200 ma/cm² were obtained.

Inspection of Tables X and XI indicates that low oxygen gain figures are obtainable with Kocite electrodes, which is indicative of low diffusion losses. The average O₂ gain at 200 ma/cm² for the three cells achieving the performance goal is only 67 mV.

The current contract performance goal has been met with Kocite electrodes fabricated from both Kocite electrocatalyst batches 3289-127A and 3289-61B with both machine calendared and sheet mold electrodes. Cells containing Kocite electrodes with 50 wt-% Teflon content generally did not reach peak performance until several days after startup. In the case of cell ERC-113 the peak cell performance was not reached until the 73rd day. However, at 40 wt-% Teflon content, used for many of the later Kocite electrodes, peak performance was normally reached much sooner. At this Teflon content many, but not all, cells showed electrode flooding problems so endurance testing was generally not performed. From the results indicated in Tables X and XI, a Kocite electrode Teflon composition of 50 wt-% appears to be desirable for long term electrode stability.

Cells assembled using Kocite electrodes fabricated from electrocatalyst batch 3576-61A, prepared by the PDD impregnation technique, did not perform as well as expected based on the X-ray diffraction determination of Pt crystallite size (shown in Table VII). While the testing of this electrocatalyst was not extensive, only one cell, ERC-134, reached 0.60 volts at 100 ma/cm². The reasons for the relatively poor performance of electrodes fabricated from this electrocatalyst batch are not presently understood.

The best cell performance reached at UOP was on cell 3289-127A-6, which was stable until it was accidentally reversed polarized by the power supply driving it at 200 ma/cm². A large number of cells developed gas crossover problems early during cell testing and were subsequently terminated. It is speculated that this problem arose from the use of a particular Kynol matrix batch. With the limited testing performed at UOP, no cell reached the specified performance goals. The results obtained at ERC were not reproduced at UOP.

Both cells ERC-113 and ERC-138, which reached the performance goal, have been under test for more than 500 hrs., as indicated in Table X. To further characterize these cells, the oxygen and air polarization curve of cell ERC-113 at its performance peak and its performance as a function of time are shown in Figures 6 and 7, respectively. Similar curves are shown for cell ERC-138 in Figures 8 and 9. In the case of cell ERC-138, the polarization curves were extended to 250 ma/cm². At this current a terminal voltage of 0.595
Figure 6

Polarization Curve for Kocite 3289-127A Electrode-ERC Cell-113 as a Cathode at 180°C. Platinum Loading 0.55 mg/cm².

Terminal Voltage

- Air Cathode
- Oxygen Cathode

Potential (Volts, Terminal)

0.9
0.8
0.7
0.6
0.5
0.4
0

Current Density (mA/cm²)

50
100
150
200
250
Figure 1

Lifetime Test for Kocite 3289-127A
180°C. Electrode-ERC Cell 113 as a Cathode at 100 ma/cm².
Terminal Voltage: Current Density

- Air
- O₂
- Current Density
  - 100 ma/cm²
  - 200 ma/cm²
Polarization Curve for Kocate 3576-61B Electrode-ERC Cell-138 as a Cathode at 180°C. Platinum Loading is 0.60 mg/cm².

Terminal Voltage
- ● Air Cathode
- ▲ Oxygen Cathode
volts on air was achieved. As shown in Figures 7 and 9, both cells have good long term stability with cell ERC-113 operating more than 4000 hours and cell ERC-138 more than 1250 hours. The data in these figures emphasize the low oxygen gain figures of these cells, with cell ERC-138 having an oxygen gain of only 75 mV at 250 ma/cm².

To determine if improvements in cell performance might be obtained by using a SiC matrix rather than a Kynol matrix, ERC has analyzed the various contributions to the polarization losses observed in increasing the cell current from 100 to 200 ma/cm². Activation polarization losses are estimated to be 27 mV assuming a Tafel slope of 90 mV per decade. Ionic polarization losses, caused mainly by the resistance in the cell matrix, are estimated to be in the range of 30 to 40 mV for each 100 ma/cm² with the presently used Kynol matrix. Diffusion polarization losses on air are believed to be low at 100 ma/cm² with properly wet-proofed Kocite electrodes having small catalyst particle sizes, but probably are a factor at higher current densities. A diffusion loss of 5 mV from 100 to 200 ma/cm² is thought by ERC to be a reasonable estimate for a good Kocite electrode. Thus a doubling of the cell current from 100 to 200 ma/cm² should give a minimum polarization loss of 60 to 70 mV assuming that anode polarization losses may be neglected. As indicated in Table II, the best cell performance results obtained during the previous contract period exceed this estimate by 20 to 30 mV indicating somewhat larger diffusion losses. Inspection of the cell performance results listed in Tables X and XI shows that for many of the best cells tested during the present contract period, the polarization loss is equal to the estimated polarization loss for a good electrode. This suggests that diffusion losses are quite small and further improvement in cell performance will require improved Pt utilization in the Kocite electrode and a lower ionic resistance of the cell matrix.

5.3 Summary of Results

The most important results obtained during this contract period are summarized below.

(A) Production of Leached Kocite Materials

A variety of leaching conditions have been investigated to determine the preferred conditions under which the alumina substrate is leached from Kocite particles by H₃PO₄ to produce a high-surface-area pyropolymer structure serving as the electrically conductive support for highly dispersed Pt crystallites. Depending on the initial substrate surface area and carbon content of the Kocite particles, surface areas up to 1000 m²/g can now be routinely produced. To accomplish this, Kocite particles are leached by the H₃PO₄ for 24 hours at 140°C.
Figure 9

Lifetime Test for Kocite 3576-61B Electrode-ERC Cell 138 as a Cathode at 180°C.

Platinum Loading is 0.60 mg/cm².

Terminal Voltage Current Density (ma/cm²)

- O2
- 100 ma/cm²
- 200 ma/cm²

Time (hours)
(B) Kocite Electrocatalyst Production

i. Impregnation of Leached Kocite Materials

Kocite electrocatalysts have been produced by the Pt impregnation of leached Kocite materials using chloroplatinic acid, platinum diaminedinitrite, and platinum acetylacetonate. The highest Pt dispersions were obtained using platinum diaminedinitrite, although, the best fuel cell performance was obtained using electrocatalysts prepared by chloroplatinic acid impregnation.

ii. Kocite Electrocatalyst Structure

Initial efforts directed at determining Kocite electrocatalyst pore structure and particle shape were made using pore volume distribution measurements and high resolution Scanning Electron Microscopy. These results indicate that

a. Kocite structure is not destroyed by either leaching the substrate from the Kocite particles nor the catalyization process.

b. Currently used Kocite electrocatalysts have a platelet shape.

c. The maximum in the pore volume distribution is shifted from pore diameters of \(~4.0\) nm for a typical Kocite material to \(~7.0\) nm for the resultant electrocatalyst with the substrate materials and process conditions employed currently.

(C) Cell Performance

i. Cell Performance Level

Cells with Kocite cathodes fabricated from different Kocite electrocatalysts and by both machine calendering and sheet mold techniques have reached or exceeded the contract performance goals of 0.60 volts at 200 mA/cm\(^2\). These cells were operated at 180°C with \(~0.5\) mg/cm\(^2\) Pt-loaded Kocite electrodes and used a Kynol matrix. The maximum full cell performance achieved was 0.62 volts at 200 mA/cm\(^2\) with an oxygen gain at this current of 65 mV. The improvement in cell performance during the current and previous contract periods is shown in Figure 10. Of two cells constructed with Kocite electrodes as both anode and cathode, one reached the contract performance goal with no prewetting of either electrode.

ii. Endurance Testing

Endurance testing on cells achieving the performance goal were performed. The longest running cell has operated stably in excess of 4000 hours.
Figure 10

Increases in the Terminal Voltage of Cells Containing a Kocite Electrode as the Cathode.

Current Density

- 100 ma/cm²
- 200 ma/cm²

Temperature 180°C
iii. Electrode Structure

The best results of this contract period were achieved with a sheet mold electrode having a 40 wt-% Teflon content. Results with other cells indicate that an electrode with 50 wt-% Teflon should have improved long term stability by being more resistant to electrode flooding.

iv. Electrode Prewetting

Cells assembled with sheet mold electrodes, with no electrode prewetting, achieved the best performance during this contract period.

VI. CONCLUSIONS

Based on the results obtained in the last nine months in the areas of Kocite particle substrate leaching, Kocite electrocatalyst preparation, improved Kocite electrode performance, and new electrode structures, the following conclusions concerning low cost fuel cell electrodes fabricated from Kocite electrocatalysts are warranted.

(A) Leached Kocite materials can be routinely prepared to produce high-surface-area (≥1000 m²/g) electrocatalyst supports with a narrow pore volume distribution. The structure appears to be stable both through the leaching and catalyzation process.

(B) The best Kocite electrocatalysts prepared during this contract period were made by Pt-impregnation of leached Kocite material with chloroplatinic acid.

(C) Full cell performance levels of 0.68 V at 100 ma/cm² and 0.62 V at 200 ma/cm² can be achieved by cells with a ~0.5 mg/cm² Pt-loaded Kocite cathode operated at 180°C with a Kynol matrix.

(D) Optimal fuel cell performance can be obtained without pre-wetting the Kocite electrode with H₃PO₄.

(E) Improvements in cell performance may be achieved by

i. Achieving and maintaining improved Pt dispersions.

ii. Producing the electrocatalyst surface chemistry and pore volume distribution to maintain the optimum three-phase boundary.

iii. Increasing Pt utilization with improved electrode structures.

iv. Using a matrix (probably SiC) with reduced ionic resistance.

(F) Substantially increased efforts must be made in cell testing to establish cell reliability and reproducibility at the performance levels attained during the present contract period.
Variations in performance between cells observed during the present contract period are believed to arise from differences in the Kocite electrocatalysts with different impregnation techniques, lack of optimization of electrode structure for leached Kocite electrocatalysts, and the experimental nature of the sheet mold electrodes. Further experimentation in these areas should lead to more consistent cell performance levels.

VII. RECOMMENDATIONS

To further improve the performance of Kocite electrocatalysts and Kocite electrodes, and continue their development in H3PO4 electrolyte fuel cells, the following recommendations are made.

(A) Continued efforts must be made to achieve higher Pt dispersions (smaller Pt crystallite size) on leached Kocite supports.

(B) The structural analysis of Kocite electrocatalysts should be continued to determine the effect of process conditions and impregnation technique on both pore and particle structures.

(C) The structural parameters of sheet mold Kocite electrodes should be varied and optimized to obtain increased Pt utilization and reliable wet proofing. To allow this, 50 g electrocatalyst batch should normally be produced.

(D) Kocite electrodes should be tested in sufficient quantities and with uniform cell hardware and matrix material to establish electrode reliability and reproducibility.

(E) Extended lifetime testing (5000 hrs. or more) of Kocite electrodes should be expanded.
VIII. REFERENCES


3. Pictures taken at Scanning Electron Microscope Users' Laboratory at the University of Chicago. The lab is operated on a resource grant from the National Institutes of Health.
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