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Project Title: IGR NO_x/SO_x
Control Technology

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Project Start: September 25, 1992

Project Completion: September 24, 1997

Approach Changes: None

IGR

Performance Variances,
Accomplishments, or Problems:

SUMMARY

Work this quarter continued on the development of electrocatalyst and electrolyte materials and processing to improve the electrical efficiency required for economical NO_x/SO_x destruction. Efforts focused on two critical areas: first, modifying electrocatalyst composition in order to improve current efficiency, and, second, changing the solid state defect concentration in the solid oxide electrolyte itself.

A. ELECTROCATALYST/ELECTROLYTE PREPARATION

During the past several months IGR has concentrated on making modifications to its electrocatalyst composition in order to improve the current efficiency of the technology. Substantial, although not sufficient, progress has been made in this area. Therefore two separate lines of research were explored; (1) higher conductivity solid oxide electrolytes and (2) modifying the composition of our fully stabilized zirconia with other oxides. In both cases the effort was to provide additional appropriate electronic defect sites for the electrocatalytic destruction of NO_x while simultaneously reducing the amount of current required to carry out the process. Our first approach, using higher

conductivity solid oxide electrolytes with our best electrocatalyst to date, had the paradoxical effect of lowering the amount of NO_x removed for a given condition. In addition, the higher the ionic conductivity of the electrolyte the lower the oxygen tolerance of the system.

The second approach involved the compositional modification of our standard fully yttria stabilized solid electrolyte with oxides. Different preparation procedures were used to modify the defect structure of the solid oxide electrolyte including the use of sol gels. In most cases the removal of NO_x was adversely affected by either the oxide additions or by variations in the preparation procedures. None of the above tests showed electrical efficiency improvements over what was reported previously.

Work with high conductivity solid oxide electrolytes had an inverse effect on NO_x removal, i.e. the higher the conductivity the lower the current efficiency. Therefore, we also tested lower conductivity solid oxide electrolytes in combination with small changes to the electrocatalyst to improve its surface area and efficiency. Two highly refractory solid oxides electrolytes were chosen. To speed up the initial screening process the materials were prepared from sol-gels and sintered onto fully stabilized zirconia substrates before the application of the electrocatalyst. At this time (only) one temperature was used to sinter the sol-gels. For these conditions current efficiency was unexpectedly

degraded at low oxygen levels and no activity at all was noted at high (5.7%) oxygen levels. We believe that this response may be due to the crystallinity of the material and that by altering processing conditions improved results can be obtained.

Alterations in the formulation of the electrocatalyst were made to improve the surface area to allow for increased adsorption of either NO_x or SO_x. Testing failed to show any improvement in the current efficiency implying that the surface area of the electrocatalyst *is not* a controlling factor. Compositional changes to the electrocatalyst in most cases had a detrimental effect on NO_x removal.

Based on our experiments we preliminarily conclude that the desired fundamental mechanism of NO_x and/or SO_x removal is dependent on both the electrocatalyst *and* the electrolyte. The nominal proposed fundamental destruction mechanism can be summarized for the case of NO as follows:

Proposed Fundamental Cathodic Electrode Mechanism

The proposed overall electrode (cathodic) reaction is:

The resulting oxide ions are oxidized at the anode to yield oxygen gas and to complete the electrical circuit.

B. ELECTROLYTE COMPOSITION

Further work involved varying the composition of the electrolyte. The theory behind this approach is that the electronic site(s) for NO_x reduction may be altered by the composition of the phase. A second compositional change involved using a mixed conductor solid oxide electrolyte to modify the surface of standard electrolyte. Our standard electrolyte is a pure, not mixed, oxide electrolytic conductor and as such has a different electronic defect structure.

For these conditions current efficiency was (unexpectedly) degraded at low oxygen levels and no activity was noted at high (5.7%) oxygen levels. It is possible that our processing conditions were insufficient, therefore repeats of several of these tests under varying processing conditions will be run.

Open Items: None

Status Assessment & Forecast:

Based on the above results work will continue on electrocatalyst and electrolyte composition testing.

Key Personnel Staffing Report:

Dr. Arnold Z. Gordon, the president of IGR, has spent during this quarter 172 hours on this contract.

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