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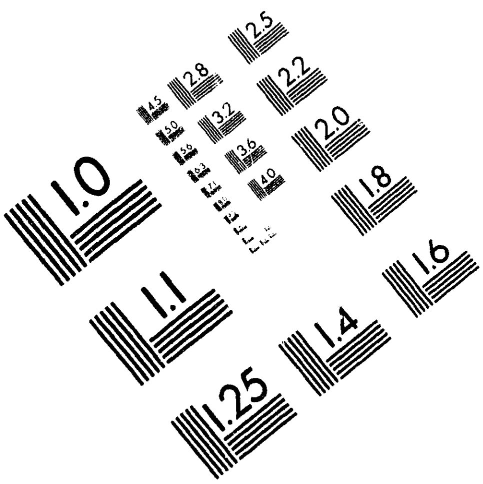

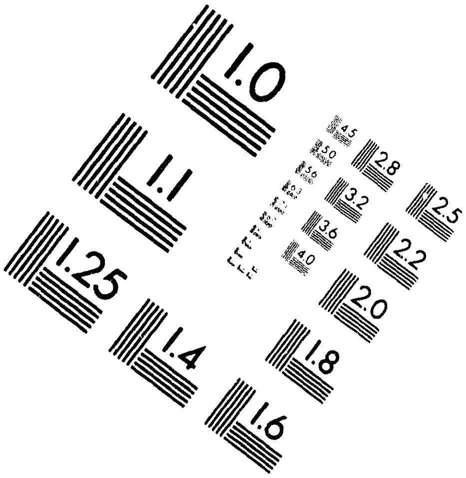

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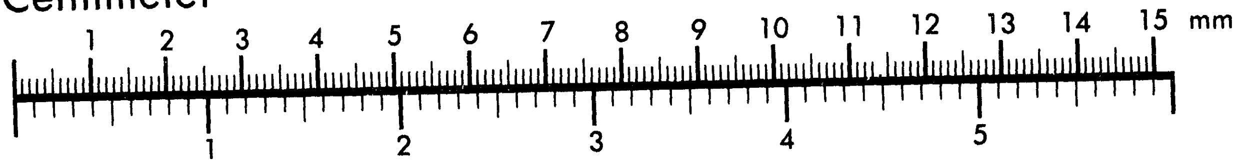

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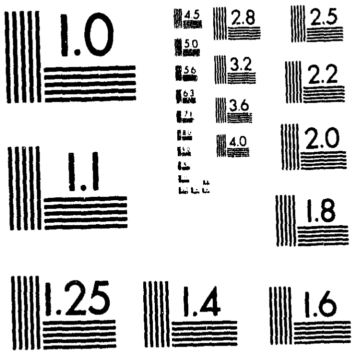

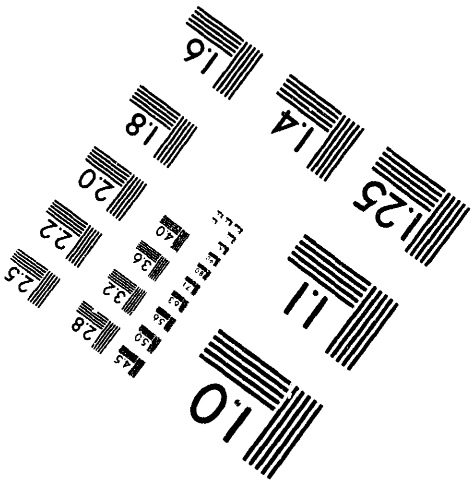

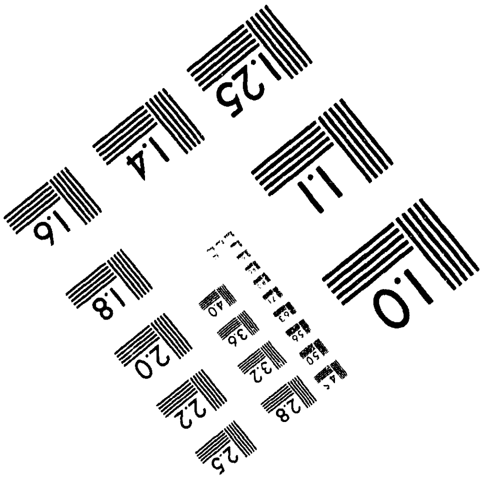

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PROGRESS REPORT

A NOVEL PROCESS FOR METHANOL SYNTHESIS

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Principal Investigators

John W. Tierney

Irving Wender

Chemical and Petroleum Engineering Department
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Pittsburgh, PA 15261.

Prepared for the Department of Energy

Grant No. DE-FG22-89PC89786

June 1, 1993 through September 30, 1993

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Ju-7

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DISCLAIMER

1.0 ABSTRACT

A bench-scale reactor is being used to conduct studies of the conversion of synthesis gas to methanol (MeOH) in the liquid phase by a novel process. In previous reports, we provided evidence for a two step reaction in series, the carbonylation reaction mainly taking place close to the copper chromite surface, and the hydrogenolysis reaction taking place on the surface of the copper chromite. The interaction between the two catalysts enhances the rate of MeOH formation. We have previously shown that, under present operating conditions, the methyl formate (MeF) concentration is not in equilibrium. The presence of H_2O results in a drop in the rate of MeF generation by the carbonylation reaction. In this quarter, preparation of the final report was continued.

2.0 INTRODUCTION

An experimental investigation of a new method for converting synthesis gas to MeOH is being carried out. It was developed in our laboratory and we refer to it as the concurrent synthesis of MeOH. It has advantages over the conventional gas phase synthesis of MeOH in that the recycle of unreacted material can be virtually eliminated and it operates at lower temperatures. The reaction is tolerant to small amounts of CO_2 or H_2O. It has been demonstrated that the reactions proceed with good rates at 150°C and 63 bar pressure. We have previously provided evidence that the formation of MeOH proceeds through methyl formate. However, the nature of the mixed catalyst, comprised of an alkali methoxide (e.g. KOMe) and copper chromite and of the possible intermediates, is not well understood. The thrust of this research program is to obtain a better understanding of the reaction and particularly of the role of the catalyst(s). This information should help make it possible to exploit these reactions commercially.

Four papers have been published or accepted for publication$^{(1-4)}$ reporting our studies. The first paper is a study of the individual consecutive reactions: carbonylation of MeOH to MeF followed by the hydrogenolysis of the MeF. The other papers describe the concurrent reaction in which a carbonylation catalyst (e.g. KOMe or other soluble alkali catalyst) and a hydrogenolysis catalyst (e.g. copper chromite) are used in the same reactor. A United States patent[5] was issued during this quarter. The current work is part of a three and a half-year project which started in September, 1989.

3.0 RESEARCH OBJECTIVES

Principal research objectives are,

1) To determine the effect of various catalysts on the synthesis of MeOH.

2) To determine the nature of the active catalyst in this reaction and the effect of deactivating agents such as CO_2 and H_2O.

3) To determine the rate-limiting step(s) in this reaction. The effect of catalyst loading and reactor volume are of special importance.

4) To develop mathematical models which can be used to predict the rates of reaction and could be useful in eventual scale-up of the reaction.

4.0 CURRENT WORK

Work during this quarter was devoted to preparing the final report.

5.0 FUTURE WORK

This is the last quarterly report for this project. A complete description of the work done will be given in the Final Report.

6.0 REFERENCES

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3) V. Palekar, H. Jung, J. W. Tierney and I. Wender, Slurry Phase Synthesis of Methanol with a Potassium Methoxide/Copper Chromite Catalytic System, in press, *Applied Catalysis*.

4) V. Palekar, J. W. Tierney and I. Wender, Alkali Compounds and Copper Chromite as Catalysts for a Slurry Phase Synthesis of Methanol, in press, *Applied Catalysis*.

5) J.W. Tierney, I. Wender and V.M. Palekar, "Methanol Synthesis Using a Catalyst Combination of Alkali or Alkaline Earth Salts and Reduced Copper Chromite for Methanol Synthesis," U.S. Patent 5,21,562, June 22, 1993.

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