DOE/MT/92017--T2

PROGRESS REPORT

1. GRANT NUMBER: DE-FG22-92MT92017

2. PERIOD COVERED BY REPORT: October 1, 1993 - December 31, 1993

3. TITLE OF PROPOSAL: SIMULTANEOUS SO_2/NO_x ABATEMENT USING ZEOLITE-SUPPORTED COPPER.

4. NAME OF INSTITUTION: Clark Atlanta University

5. AUTHOR(S) OF REPORT: Drs. Mark B. Mitchell and Mark G. White

DISCLAIMER

US/DOE Patent Clearance is not required prior to the publication of this document.

SUMMARY OF RESEARCH RESULTS

Fabrication of Flow Reactor.

A flow reactor is being constructed at Georgia Tech for the NO decomposition reaction. The reactor is partially complete with the construction of a flow meter/controller system and a temperature controller system in place now. The stainless steel reactor tube and the furnace have been fabricated and fit to the sub-assembly containing the temperature control devices.

The reactor sampling system is under fabrication. We plan to purchase a GC-MS from Hewlett-Packard to determine the composition of the exit gas. Additional consideration is given to the gas analytical section and to the gas purification section of the device.

Raman Spectroscopy of Precursors.

We have used Raman spectroscopy to characterize Zeolite A, a cobalt ethylenediamine complex, $Co(en)_3Cl_3$, and Zeolite A impregnated with the cobalt complex. We reported the infrared spectra of this series of samples in an earlier report using diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS). The Raman spectra were recorded by Dr. Manu Chopra in the Department of Chemistry at the Hong Kong University of Science and Technology. The spectra were recorded for a powdered zeolite sample containing about 1 wt% Co; the elemental analyses for C and N in this same sample showed the same stoichiometry as that expected for the metal complex. The DRIFTS spectra showed that the ethylenediamine ligands were intact but that the frequencies of the N-H stretch vibrations were shifted to lower frequencies, which is consistent with a model of the interaction between the complex and the zeolite involving an interaction of the N-H dipole of the complex with dipoles in the zeolite framework.

The Raman spectra from 1700 - 100 cm^{-1} are shown in the attached two figures for Zeolite A, $Co(en)_3Cl_3$, and Zeolite A impregnated with $Co(en)_3Cl_3$. From standard literature on vibrational spectroscopy, it is possible to assign vibrations in the $Co(en)_3Cl_3$ spectrum to specific functional groups. For example, the vibrations at 1585 cm^{-1}, 1559 cm^{-1}, and 1280 cm^{-1} are the NH_2 scissors and wag vibrations. The vibrations at 1467 cm^{-1} and 1367 cm^{-1} are CH_2 antisymmetric and symmetric stretch vibrations.

In the spectrum of $Co(en)_3Cl_3$ impregnated in Zeolite A, it is clear that significant changes have occurred. Many of the changes can be associated with broadening of the infrared absorptions which is consistent with a variety of interaction sites in the zeolite, leading to a distribution of absorption frequencies for many of the vibrations. But most of the vibrations of the complex in the zeolite occur at frequencies similar to those observed for the pure complex. The exception to this is the NH_2 vibrations. These vibrations appear to be shifted by approximately 15 - 30 cm^{-1} to the broad absorption at 1609 cm^{-1}. The low frequency part of the spectrum is remarkably similar to what would be obtained by simply adding the spectrum of the complex to that of the zeolite.

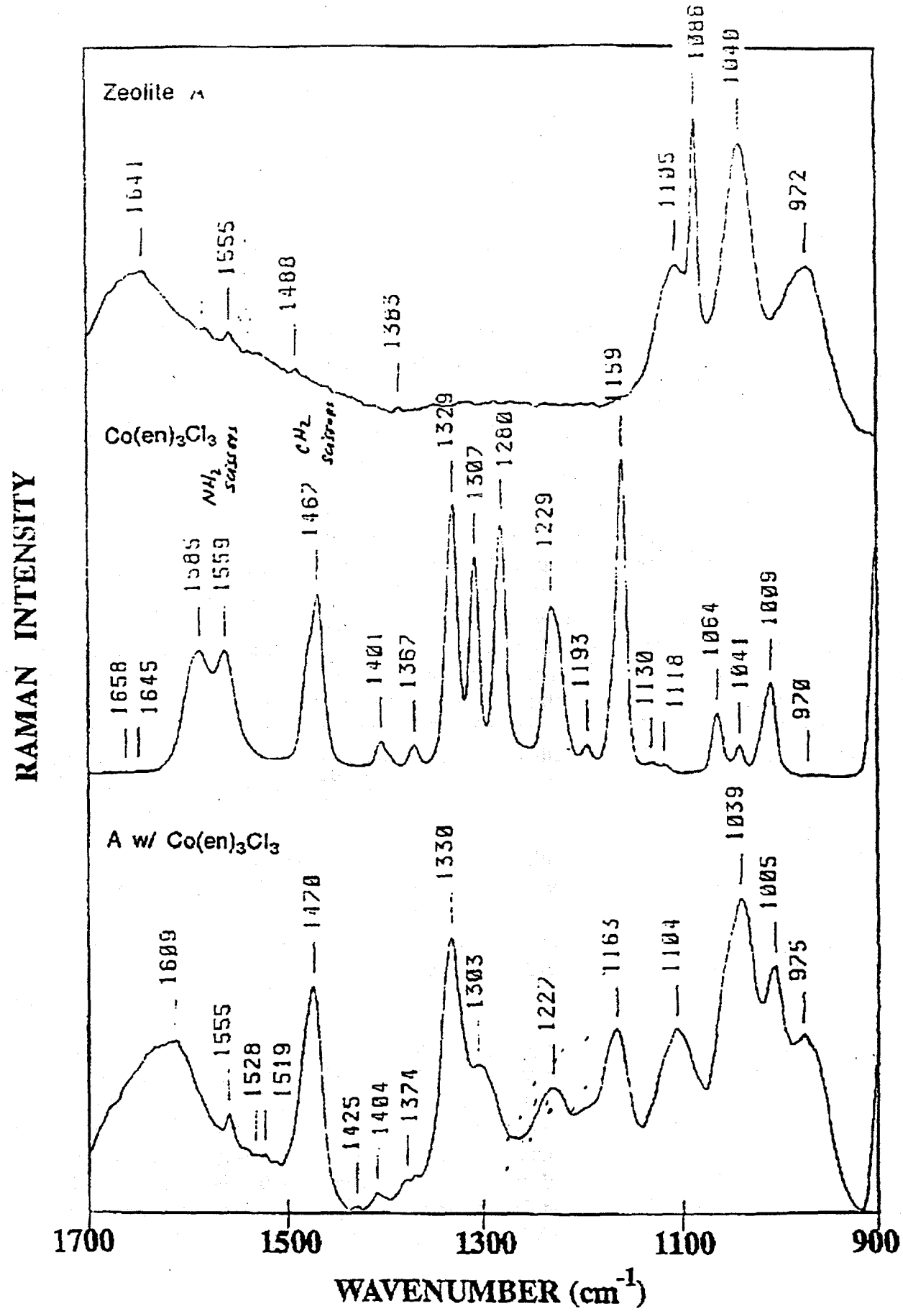
Move to New Building.

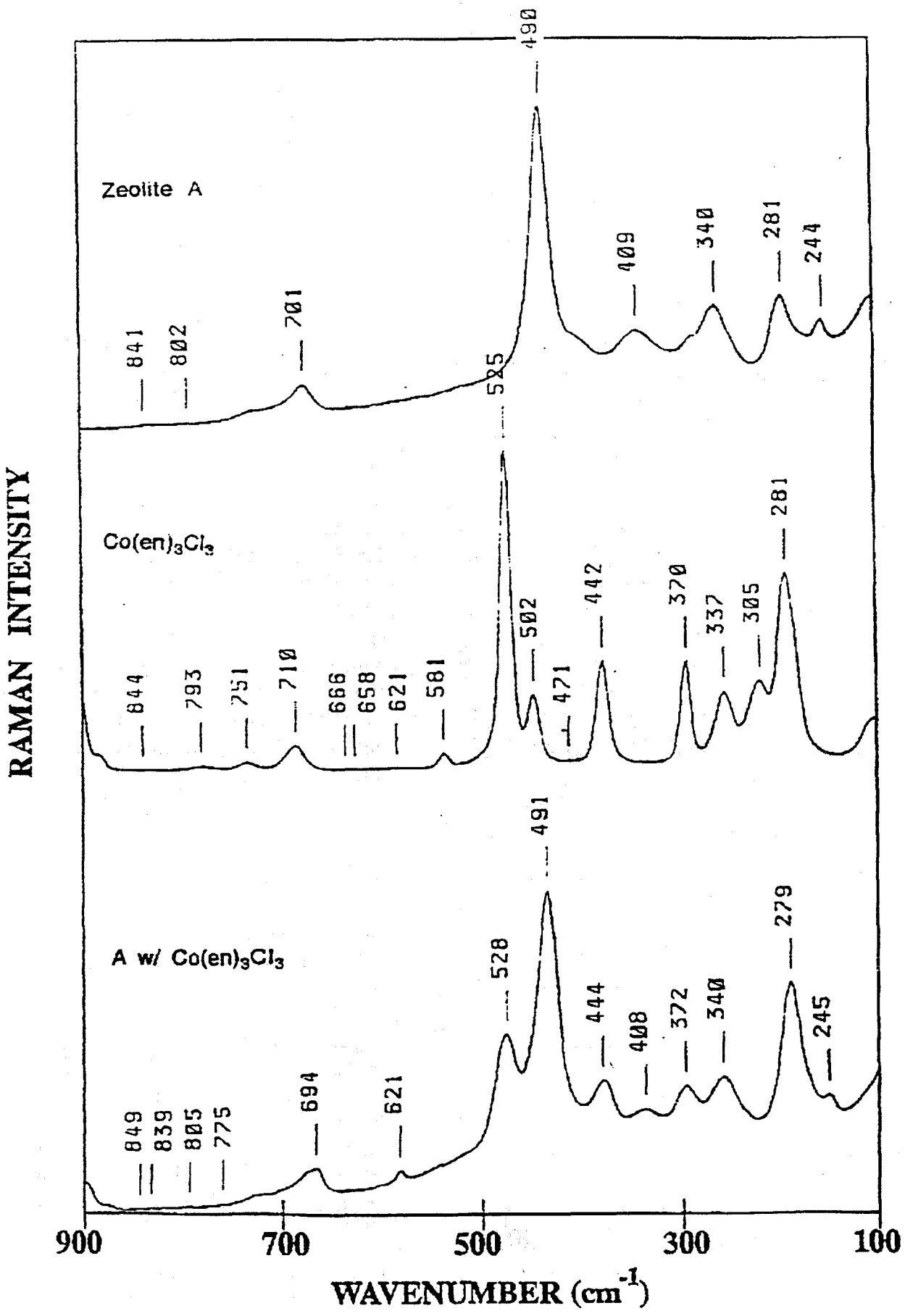
At Clark Atlanta Unviersity, we are beginning the process of packing the laboratory to move to the new Research and Education Center for Science, Engineering, and Technology (RECSET). The move will begin in January, 1994, and will involve moving the laboratory and offices. The new facility will put Dr. Mitchell directly across the hall from the laboratory, which will make his involvement with the day to day operations of the laboratory much more convenient and frequent. In addition, the new laboratory is probably three times larger, has better access to utilities (water and electricity, in particular), and more dependable maintenance personnel.

New Equipment.

As part of the involvement of Dr. Mitchell in the NASA-funded High Performance Polymers and Ceramics Center at CAU, two new pieces of spectroscopic equipment are being purchased and will be installed in the new building. One of the new pieces of equipment is a Nicolet Magna 750 FT-IR which will significantly decrease infrared spectroscopic data acquisition time and increase the data quality due to the inproved signal to noise ratio for this instrument. We will also be installing a Raman spectrometer, utilizing a Coherent Innova 306 Argon-ion laser and a Spex 0.5m monochromator with a CCD detector. This will give us a capability which we do not currently have and does not exist at any of the other research universities in the Atlanta area, Georgia Tech, Emory, or Georgia State. We will have the most comprehensive vibrational spectroscopic capability in the Atlanta area and probably in the Southeastern United States.

We look forward to the move into the new building and starting experiments with the new equipment in the next quarter.


Zeolite A
1641
1555
1488
1363
1159
1135
1086
1040
972
Co(en)3Cl3
NH2 scissors
CH2 scissors
1658
1645
1585
1559
1467
1401
1367
1329
1307
1280
1229
1193
1130
1118
1064
1041
1009
970
A w/ Co(en)3Cl3
1609
1555
1528
1519
1478
1425
1404
1374
1330
1303
1227
1165
1104
1039
1005
975
RAMAN INTENSITY
1700
1500
1300
1100
900
WAVENUMBER (cm-1)


Zeolite A
490
409
340
281
244
841
802
701
525
Co(en)3Cl3
844
793
751
710
666
658
621
581
502
471
442
370
337
305
281
A w/ Co(en)3Cl3
849
839
805
775
694
621
528
491
444
408
372
340
279
245
RAMAN INTENSITY
900
700
500
300
100
WAVENUMBER (cm-1)