High pressure reaction cell and transfer mechanism for ultrahigh vacuum spectroscopic chambers

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Citation: Review of Scientific Instruments **71**, 2471 (2000);
View online: https://doi.org/10.1063/1.1150637
View Table of Contents: http://aip.scitation.org/toc/rsi/71/6
Published by the American Institute of Physics

High pressure reaction cell and transfer mechanism for ultrahigh vacuum spectroscopic chambers

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(Received 29 December 1999; accepted for publication 15 February 2000)

A novel high pressure reaction cell and sample transfer mechanism for ultrahigh vacuum (UHV) spectroscopic chambers is described. The design employs a unique modification of a commercial load-lock transfer system to emulate a tractable microreactor. The reaction cell has an operating pressure range of $<1\times10^{-4}$ to 1000 Torr and can be evacuated to UHV conditions to enable sample transfer into the spectroscopic chamber. Additionally, a newly designed sample holder equipped with electrical and thermocouple contacts is described. The sample holder is capable of resistive specimen heating to 400 and 800 °C with current requirements of 14 A (2 V) and 25 A (3.5 V), respectively. The design enables thorough material science characterization of catalytic reactions and the surface chemistry of catalytic materials without exposing the specimen to atmospheric contaminants. The system is constructed primarily from readily available commercial equipment allowing its rapid implementation into existing laboratories. © *2000 American Institute of Physics.* [S0034-6748(00)01906-7]

I. INTRODUCTION

The difficulty associated with ultrahigh vacuum (UHV) characterization of model catalytic systems is correcting for the pressure difference between actual catalyst operating conditions and UHV pressures.[1] This pressure difference, or pressure gap, presents a substantial obstacle for correlating UHV characteristics with actual catalyst mechanisms and attributes. A common approach to circumvent the pressure gap is to combine an UHV spectroscopic chamber with an elevated pressure reactor and sample transfer system.[1] The combination of techniques facilitates a wide range of catalyst treatment conditions followed by the insertion of the material into an UHV chamber for spectroscopic analysis.[1,2] The procedure allows the material to be treated under realistic conditions while collecting valuable information in the UHV chamber. Several combined UHV compatible high pressure reaction cell and transfer system designs have been published in the literature.[1–10] The reported designs incorporate a variety of desirable attributes for specific analytical systems and arrangements. The ultimate design of a high pressure reaction cell and sample transfer mechanism depends on the ease of sample transfer from reaction conditions to an UHV environment and the specific sample electrical connections and orientation requirements.[1,2] In many instances, the systems are complex and require substantial equipment design and construction and often require a significant capital expense.

We have designed and constructed a simplistic and inexpensive reaction cell and sample transfer mechanism for implementation with standard UHV spectroscopic chambers. The initial design of the equipment is the result of a collaborative research effort with the Ford Motor Company.[11] The system design incorporates several distinct advantages based on the unique combination of the transfer mechanism and the elevated pressure cell. A transfer mechanism, or load-lock, is typically employed only for introducing samples into UHV conditions from atmospheric pressure. Through extensive design, the load-lock has been modified to provide electrical and thermocouple connections for specimen heating and temperature measurement. In order to optimize the system capabilities, a custom sample stage has also been designed. The sample stage facilitates specimen heating and temperature measurement in both the reaction cell and spectroscopic chamber. The specimen stage can be resistively heated in the reaction cell through the translation of a custom electrical and thermocouple linear motion feedthrough or in the main UHV chamber with a stationary electrical and thermocouple contact head mounted on a *xyz*-rotary manipulator. As a result, samples can either be treated in the reaction cell and inserted into the main UHV chamber for analysis or treated and characterized entirely within the main UHV chamber. The straightforward system design eliminates redundant equipment and unnecessary expenses while providing a versatile analytical technique.

II. UHV SYSTEM DESCRIPTION

The reaction cell and transfer mechanism is designed for use with a Physical Electronics (PHI) Model 545 Scanning Auger Microprobe system. However, the design could be readily implemented to any existing system with a 2.75 in. flange on a stainless steel bell jar. Samples are mounted on a *xyz*-rotary motion manipulator in the UHV chamber and translated into position for each specific analytical technique. The chamber is equipped with a PHI 15-110 Auger electron spectrometer (AES), capable of both point and scanning

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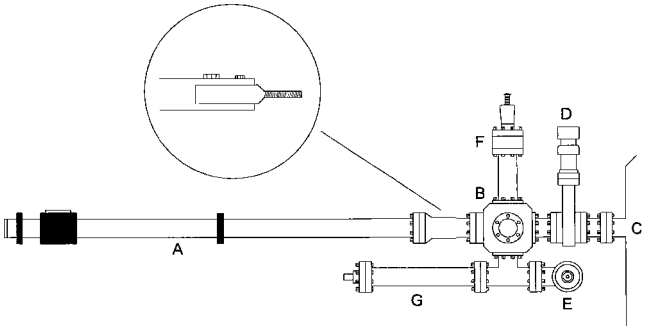

FIG. 1. Diagram illustrating the configuration of the reaction cell and load-lock transfer mechanism. The system is composed of the transfer mechanism (A) and a six-way cross (B). The load-lock mechanism is secured to the main UHV chamber (C) on a 2.75 in. flange and is separated from the chamber with a manual gate valve (D). A right angle valve (E) on the bottom of the six-way cross is connected to a turbomolecular pump and the final port is equipped with a custom electrical feedthrough (F). A mass spectrometer (G) may also be added to the system to analyze reaction gases.

mode Auger analysis. The unit is operated as a cylindrical mirror analyzer with an incident electron beam energy of 2.0 kV and 2.0 mA. The analyzer electronics are interfaced with a 386-based personal computer operating a customized LabVIEW® data collection program. The system is also equipped with a Leybold Inficon Transpector H200M quadrupole mass spectrometer for temperature programmed desorption (TPD) experiments. The mass spectrometer can also be mounted on the reaction cell to analyze reaction gasses. Mass spectral data are collected with Leybold Inficon SQX-Single Sensor QX software capable of collecting 12 mass-to-charge ratios simultaneously. Ultrahigh vacuum conditions are achieved with a PHI 200 ℓ/s ion pump and a Ultek Boostivac Combination Control titanium sublimination pump.

III. REACTION CELL DESIGN

The design of the reaction cell is centered around a commercial 2.75 in. load-lock sample transfer mechanism (MDC 665081) with 24 in. linear travel and full 360° rotation (Fig. 1). The load-lock transfer mechanism (A) is equipped with a 2.75 in. six-way cross (B) sample chamber with an additional 1.33 in. venting port. The six-way cross serves as the primary reaction cell and is equipped with several components. Three of the ports are occupied by the load-lock constituents, including the transfer rod, quartz viewport, and a Viton® sealed quick access door. The load-lock is mounted to the main UHV system (C) through a 2.75 in. radial flange and is separated from the chamber with a 1.5 in. manual gate valve (D). A 2.75 in. bakeable right angle valve (Thermionics AMV-150-22) is mounted on the sample chamber (E) and separates the system from a turbomolecular pump (Leybold TMP-151), which controls the reaction cell pressure. The final port (F) is equipped with a custom electrical feedthrough (ISI 0950201) mounted on a high temperature linear motion feedthrough (MDC 665515). The operating pressure of the reaction cell is measured with a HPS convection gauge tube controlled with a Terranova Model 926 (Duniway Stockroom TERRA-926) dual convection gauge controller.

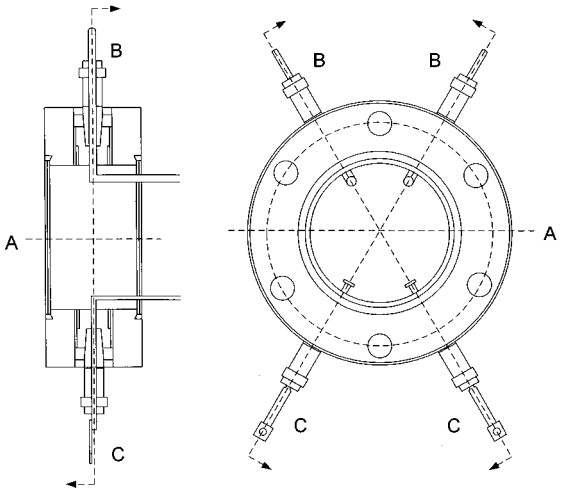

FIG. 2. Schematic view of the custom power/thermocouple feedthrough. The feedthroughs are mounted in a 2.75 in. flange (A). The power feedthrough pair (B) are 30 A/5 kV and the thermocouple pair (C) are type *K* screw type connectors.

The linear motion feedthrough assembly facilitates simple electrical and thermocouple connections to the sample stage with the added ability to be completely withdrawn from the path of the sample stage. The design also involves the inclusion of a custom electrical and thermocouple feedthrough. The custom electrical feedthrough (Fig. 2) is constructed using a double sided 2.75 in. flange (A) with an i.d. of 1.5 in. and an overall thickness of 1 in. Two feedthroughs are 30 A/5 kV (ISI 9421123) power terminals (B) to supply current for resistive sample heating and the other feedthrough pair are type *K* single leg thermocouples (ISI 9311035) with screw terminal connectors (C).

The electrical and thermocouple feedthroughs are connected to a custom probe terminal mounted on the linear motion feedthrough (Fig. 3). The power feedthrough pair is connected to 14 AWG wire braid (A) and insulated with ceramic beads (Omega FS-200-14-1000). Similarly, the thermocouple wires (0.010 in.) are insulated with ceramic beads (Omega FS-110-20-1000) and connected to the thermocouple feedthrough terminals. The power and thermocouple leads are soldered onto four 0.125 in. diameter copper pins mounted in a machined MACOR® (Accuratus Ceramic Corporation) ceramic holder (B). The MACOR® head is attached to the linear motion feedthrough (C) with a machined stainless steel holder (D). The entire assembly is mounted in a 2.75 in. nipple (E) and secured to the reaction cell. The position of the MACOR® terminal is adjusted with the linear feedthrough until the pins contact the specimen stage. Electrical current is supplied to the power feedthroughs (F) to resistively heat the specimen and the temperature is monitored through the thermocouple feedthrough (G) with a thermocouple gauge.

A similar electrical and thermocouple probe terminal (Fig. 4) is mounted on a *xyz*-rotary motion sample manipulator in the main UHV chamber. Power and thermocouple connections are supplied through a standard feedthrough (ISI 9392015) mounted on the main UHV chamber with the same current and voltage specifications as the reaction cell. The

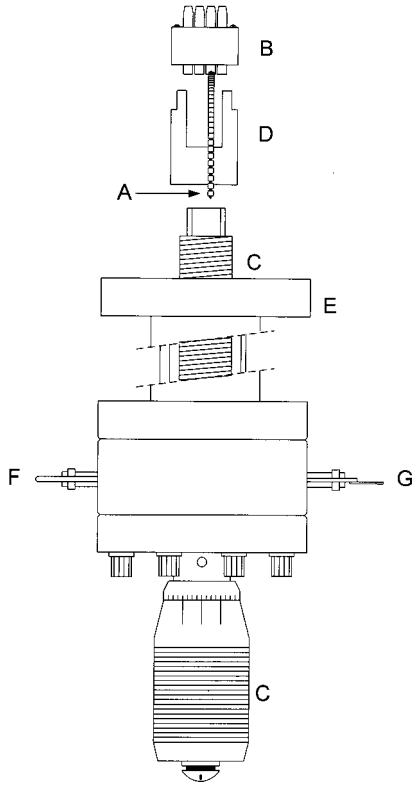

FIG. 3. Diagram illustrating the reaction cell linear motion feedthrough assembly. The electrical and thermocouple wires are insulated with ceramic beads (A) and soldered to copper pins mounted in a MACOR® head assembly (B). The head is attached to a linear motion feedthrough (C) with a stainless steel holder (D). The feedthrough is mounted on a 2.75 in. nipple (E). External connections are achieved with the electrical (F), and thermocouple (G) feedthrough pairs.

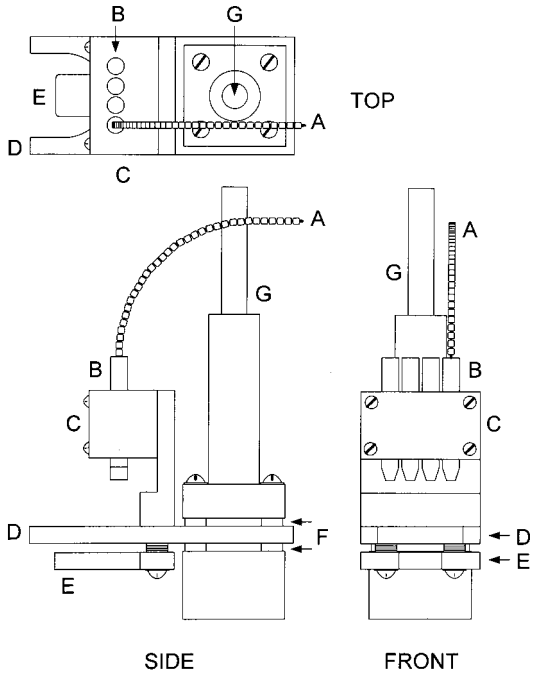

FIG. 4. Scaled drawing (top, side, front) of the probe terminal mounted on the *xyz*-rotary manipulator in the main UHV chamber. Insulated power and thermocouple wires (A) are secured to copper contacts (B) mounted in a MACOR® terminal (C). The MACOR® is attached to a sample fork (D) which includes an additional alignment tongue (E). The fork is insulated from the main UHV chamber with ceramic spacers (F) and secured to the *xyz*-rotary manipulator with a machined shaft (G).

power and thermocouple wires are insulated with ceramic beads (A) and soldered onto four 0.125 in. diameter copper pins (B) mounted in a machined MACOR® ceramic holder (C). The probe head is secured to a stainless steel holder using 2-56 slot head stainless steel screws. The head assembly is attached to a platen fork (D) with 4-40 stainless slot head screws inserted from the underside of the fork. The primary function of the fork is to accept the grove of the sample stage and ensure the correct height alignment of the sample holder. The fork also includes an alignment tab (E) to prevent the round stage from rotating and misaligning the electrical and thermocouple contacts during sample transfer. The sample fork is insulated from the main chamber through the use of ceramic spacers (F) (Kimball Physics Al_2O_3-SP-C-050) between the fork and manipulator shaft. The complete assembly is secured to the *xyz*-rotary manipulator with a round shaft (G) and socket cap set screw.

The probe head is similar to the feedthrough terminal located in the reaction cell, except the MACOR® head is fixed to the manipulator fork. Instead of advancing the MACOR® probe head into position, the sample holder is advanced and electrical continuity is achieved with spring contacts located on the sample holder. The manipulator and alignment tongue prevent sample stage misalignment and provide the necessary guidance to ensure secure thermocouple and electrical connections.

IV. SPECIMEN HOLDER DESIGN

While commercial sample holders are readily available, a custom sample stage is required to maximize the potential of the reaction cell and UHV system. The specimen holder (Fig. 5) is designed to resistively heat the sample while facilitating rapid and simplistic sample transfer. The electrical and thermocouple contacts (A) are created from 0.010 in. copper shim stock mounted on 0.125 in. by 0.0625 in. copper terminal bars (B). The shim stock is secured to the copper bars with two 0-80 socket cap screws over a 0.75 in. length and are bent on one end to serve as spring contacts. The electrical and thermocouple contacts are completely isolated through the use of ceramic spacers and machined MACOR®. The copper electrical bars are mounted to a 0.0625 in. thick sheet of tapped MACOR® (C) with 0-80 socket cap screws and are separated from the MACOR® with 0.05 in. ceramic spacers (D) (Kimball Physics Al_2O_3-SP-B-050). The tapped MACOR® sheet provides the necessary electrical insulation between the individual contacts and the sample base. The MACOR® sheet is machined with two additional 0-80 flat head screw recesses to secure the assembly to the specimen base. The MACOR® is subsequently secured to the brass specimen holder base (E) with 0-80 recessed flat head screws and separated with an additional set of 0.025 in. ceramic spacers (F) (Kimball Physics Al_2O_3-SP-C-025). Ceramic spacers serve to develop two vacuum levels which minimizes thermal contact and heat transfer facilitating rapid specimen heating profiles and reduced electrical current requirements.

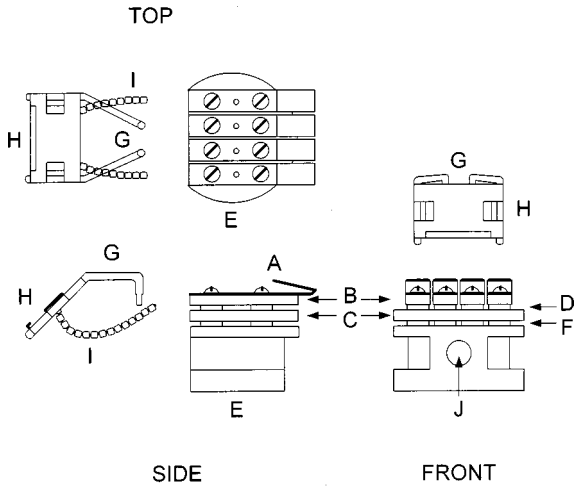

FIG. 5. Scaled drawing (top, side, front) of the sample holder stage. Electrical and thermocouple spring contacts (A) are mounted to copper terminal bars (B). The terminal bars are mounted to a sheet of MACOR® (C) and insulated with ceramic spacers (D). The MACOR® is attached to the specimen base (E) with 0-80 recessed flat head screws and insulated with an additional set of ceramic spacers (F). The specimen mount is fashioned from copper terminals (G) and titanium foil (H). A type *K* thermocouple (I) is spot welded to the back of the foil and the assembly is secured to the sample stage. The stage includes an 8-32 tapped hole (J) to secure the base to the transfer rod and a 0.1875 in. grove for positioning in the main UHV chamber.

The actual specimen mount is fashioned from copper terminals and titanium foil. A pair of round copper terminals (12 AWG) are secured (G) to the inner specimen stage contacts to mount the sample. The terminals are threaded into 0-80 tapped holes in the center of the stage contacts and curved to develop a uniform spacing of 0.5 in. at a 45° angle. A 0.005 in. thick sheet titanium foil (H) is spot welded on the terminals to hold the sample and provide the resistive heating element. The titanium foil is equipped with three tabs to secure the specimen from the bottom and each side. A type *K* thermocouple is spot welded to the back of the titanium holder and secured to the outer contacts on the specimen stage. The thermocouple wires (I) are insulated from the stage with ceramic beads and secured to the contacts with socket cap stainless steel screws.

The base is secured to the transfer rod through an 8-32 tapped hole (J) located in the front of the stage. A corresponding 8-32 threaded terminal is mounted to the transfer rod. The sample stage is also machined with a 0.1825 in. channel for transfer to the fork mounted on the *xyz*-rotary manipulator in the UHV chamber. The two distinct mounting schemes allow the specimen to be transferred from the reaction cell to the UHV chamber on a uniform *z*-directional height, minimizing analytical inconsistencies which arise from inaccurate sample repositioning.

V. OPERATION

Specimens are mounted to the sample stage using the titanium foil and secured with the tabs mounted on the sides and bottom. The sample holder is subsequently introduced into the reaction cell at atmospheric pressure through a quick-access door. The sample holder is secured to the transfer rod terminal by rotating the transfer rod and screwing the terminal threads into the sample stage base. The right angle valve is opened and the pressure of the reaction cell is reduced to $<1\times10^{-4}$ Torr with the turbomolecular pump. Samples are allowed to degas under vacuum for a predetermined amount of time prior to analysis. The stage is positioned under the electrical and thermocouple probe terminal and the MACOR® head is lowered into position by adjusting the linear motion feedthrough until the continuity of each terminal is verified with a multimeter. The right angle valve to the turbomolecular pump is closed and the desired gasses are introduced into the cell. When the predetermined pressure is achieved, current is supplied to the specimen and the temperature is monitored using the linear motion feedthrough assembly. Electrical current is supplied with an a Hewlett-Packard 6259B direct current power supply capable of delivering 0–100 A at 0–20 V. The actual current load will vary with end temperature and sample size. When the treatment is complete, the probe terminal is withdrawn and the specimen is allowed to cool. The pressure of the reaction cell is reduced and the isolation gate valve to the UHV chamber is opened.

The stage is introduced into the UHV chamber by advancing the transfer rod towards the manipulator fork assembly on the *xyz*-rotary manipulator. The channel on the sample stage is aligned with the manipulator fork and the transfer rod is slowly advanced to engage sample stage grove with the fork and alignment tongue. The electrical and thermocouple spring contacts on the specimen stage engage the stationary probe terminal facilitating contact with the copper pins. Upon verification of continuity with the sample stage, the transfer rod is carefully rotated to disengage the 8-32 threaded terminal and the transfer rod is completely withdrawn from the chamber. The isolation gate valve is closed and the sample can them be moved into the desired position using the *xyz*-rotary manipulator. Electrical current is supplied to the specimen stage through the stationary contact pins and the specimen is heated in the main UHV chamber, similar to the operation in the reaction cell. The sample is treated and analyzed in the main UHV chamber and removed from the system with the transfer mechanism.

VI. DISCUSSION

The reaction cell and transfer mechanism has been successfully used to characterize model catalytic materials during the past several months. Samples have been degassed under vacuum at temperatures in excess of 800 °C without exceeding the current requirements for the electrical feedthroughs. For 100 mg metal oxide samples, temperatures of 400 and 800 °C have been maintained for 1 h with current requirements of 14 A (2 V) and 25 A (3.5 V), respectively. In addition, initial catalyst deactivation experiments have been performed using varying quantities of sulfur dioxide on model automotive emissions control catalysts. The model catalysts were treated in the sulfur dioxide environments and surface sulfur was quantified with Auger electron spectros-

copy in the UHV chamber. The amount of surface sulfur was correlated to sample composition, catalyst temperature, and exposure to sulfur dioxide.

With the exception of the sample holder, the entire system was constructed primarily from commercial equipment. Although inexpensive to construct, the sample holder requires the craftsmanship of a qualified machinist. A commercial sample holder could be used in place of the designed holder, however, performance can not be guaranteed. The simple design of our system allows its implementation into a variety of UHV chambers. The inexpensive nature due to the availability of the commercial components ensures its usefulness in both industrial and academic research facilities.

ACKNOWLEDGMENTS

The authors gratefully acknowledge financial support from the Ford Motor Company and George W. Graham of the Ford Scientific Laboratories for providing the conceptualization and insights in the construction of the sample holder and transfer mechanism. The authors would also like to acknowledge Jerry Norkol at Michigan Technological University for his technical suggestions and material fabrication.

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