TECHNICAL UPDATE: JSC System Using a Solid Electrolytic Cell in a Remote Location to Measure Oxygen Fugacities in CO/CO2 Controlled-Atmosphere Furnaces

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TECHNICAL UPDATE: JSC System Using a Solid Electrolytic Cell in a Remote Location to Measure Oxygen Fugacities in CO/CO\textsubscript{2} Controlled-Atmosphere Furnaces

SUMMARY

The CO/CO\textsubscript{2} controlled-atmosphere furnaces in the Johnson Space Center Experimental Petrology Laboratory have been modified so that only one solid electrolyte zirconia cell\textsuperscript{1,2} is needed for all the furnaces running under standard operating conditions and requiring standard tolerances for deviations in oxygen fugacity. This technique relies on measuring the oxygen fugacity (fO\textsubscript{2}) in each experimental furnace using a zirconia cell in a designated reference furnace. A manifold system diverts the exhaust gas from the experimental furnace containing the sample through the reference furnace. Then, (1) the temperature and fO\textsubscript{2} of the reference furnace are used to calculate the composition of the exhaust gases; (2) given the temperature of the sample furnace and the composition of the exhaust gas, the fO\textsubscript{2} of the sample furnace is obtained from the Deines et al. (1974) tables.\textsuperscript{3} This cumbersome procedure has been computerized to save time and to give added flexibility to the scientist using the system.

Using a remote system for measuring fO\textsubscript{2} saves money by reducing the need for a zirconia cell in each furnace, as well as extending the life of the cells in use. It also widens the range of temperatures and fO\textsubscript{2}'s within which the scientist can run samples. There are, however, additional errors incurred while measuring the gases outside the sample furnace. Calibration procedures can be used to show the size of the additional error for a given furnace configuration and set of experimental conditions.

INTRODUCTION

Williams and Mullins' (1981) NASA Technical Memorandum 58234 describes the basic system used at JSC for performing experiments and materials synthesis under controlled oxygen fugacities.\textsuperscript{2} The system controls the partial pressure of oxygen by passing a gas comprised of a specific mixture of CO and CO\textsubscript{2} gases over the sample slowly enough that they react to produce predictable, equilibrium amounts of CO, CO\textsubscript{2}, and O\textsubscript{2}. Using this method, very small concentrations of O\textsubscript{2} can be controlled very precisely. For example, oxygen partial pressures of 10^{-10} atm (±10^{-0.2} atm) O\textsubscript{2} are obtained routinely.

There are, however, several limitations to the basic system described by Williams and Mullins (1981). That system requires that an electrolytic cell be placed within each of the furnaces, even though the cells are both fragile and expensive. Moreover, the cells are easily contaminated, and they are limited in the range of temperatures and oxygen fugacities at which they will function. So, even as Williams and Mullins (1981) was being published, new modifications were being implemented to make the systems less expensive to maintain, more flexible, and easier to operate.
NEW MODIFICATIONS

The most radical improvement to the JSC system is the implementation of a technique for remotely measuring the oxygen fugacities. The exhaust gas from each furnace is routed through a manifold having a series of T-valves (fig. 1). When all of the valves are closed, the flow of the exhaust gases are unchanged from that of the Williams and Mullins (1981) system. However, to measure fO₂, the exhaust gas for a given furnace is diverted by opening the appropriate valve on the manifold through a second "reference" furnace containing a zirconia cell. The temperature and fO₂ of the reference furnace and the temperature of the sample are then measured. Then, by assuming that the gas mixture is the same in both the specimen furnace and the reference furnace, the fO₂ of the specimen furnace, the ambient oxygen fugacity around the sample can be calculated using the equations of Deines et al., 1981.

Having the cell in a remote furnace has four major advantages. First, only one cell is required to monitor the oxygen partial pressure, or fugacity (fO₂), of gases from each of a whole bank of furnaces. Second, the cells are not subject to contamination by either volatiles from the sample or by actual physical contact from the sample,* so that the life of the individual cell is increased. Third, the cell is not subjected to the heating and cooling cycles encountered while loading or changing samples. Moreover, any change in temperature required during maintenance can be done in a controlled manner; accordingly, the cell does not experience significant thermal cycling or shock, and the life of the cell is increased. Fourth, conditions can be obtained in the sample furnace that normally would be too extreme for monitoring the oxygen fugacity directly. Under extreme conditions, especially for long durations, the zirconia cell begins to be nonlinear and inaccurate. In many cases, the temperature of the reference furnace can be chosen so that, when the exhaust gas from the sample furnace is passed through the reference furnace, the reference fO₂ is within the linear range of the cell. Of course, when using the remote system for this purpose, the experimentalist must assume that the gases within the sample furnace are behaving ideally, as there is no way to measure the partial pressure of oxygen directly.

Because the world is not ideal, the reference system is not a complete panacea. Potential problems are associated with this system or any system that monitors conditions remotely. For example, any leaks of oxygen into the system could swamp the small fO₂s being measured and make the reference fO₂ more oxidizing than the sample conditions would dictate. In theory, a physical leak of O₂ into the manifold system can be circumvented by keeping the gas pressure within the furnaces and lines above one atmosphere. Carbon precipitation from the gas after the gas passes through the hot-spot of the furnace would also make the reference fO₂ appear more oxidizing than that actually in the sample furnace.

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* The electrolytic cell works because the oxygen-defect concentration at a given temperature and fO₂ is predictable; accordingly, even minor contamination may produce significant errors. A reaction between the cell and a sample with which it is in physical contact renders the cell completely inoperative. In the modified JSC system, the sample will clearly never come in contact with the remote solid electrolyte cell. Moreover, most of the volatiles from the sample, such as iron or silica, are likely to precipitate in the cold parts of the sample furnace or in the gas lines leading to the reference furnace. Thus, they will not reach the remote cell.
Several sets of experiments have been run to quantify how large an error is incurred by sampling the gases remotely. In these experiments, the ambient fO2's in the specimen and reference furnaces were measured simultaneously over a variety of conditions.

The first set of these "calibration" experiments was done by R. Williams, O. Mullins, and a then summer intern student, A. Roshko, in 1981-1982 as a part of the initial implementation of the system. Other investigators have since performed similar experiments for their particular furnaces and conditions. In 1990, L. Le performed extensive calibrations over a wide range of fO2's for a variety of furnace configurations. This was followed in 1992 by a recalibration of L. Le's most successful furnace configurations using the smaller, faster-responding Ceramic Oxide Fabricators sensors (Australian Patents #466251 and #513552).

Figure 2 gives the results from one of the latest furnace calibrations. It illustrates an important point concerning the accuracy of the reference system. For the more oxidizing CO/CO2 gas mixtures, the reference system is quite accurate over a wide range of temperatures—the oxygen partial pressures calculated for the sample furnace are within x10±0.2 atmospheres of the actual oxygen content. Although not illustrated here, loss of accuracy can occur when the sample furnace is at a lower temperature. This loss of accuracy is explained if the reaction between the CO and CO2 gas in the sample furnace has not gone to completion. As such, this error can be a function of flow rate. There is also a loss of accuracy for the more reducing CO/CO2 gas mixtures. In these extreme cases, this loss of accuracy is explained if the exhaust gases are unstable in the temperature gradients of either the sample furnace or the reference furnace, or both. The instability could allow carbon to precipitate after the gas has passed the sample and entered the cooler regions of the furnace or exhaust line, causing measurement in the reference furnace to indicate that the sample furnace is significantly more oxidizing than it actually is. In fact, we routinely observe such carbon precipitates under certain operating conditions.

In general, it has been found that the deviation in fO2 between the reference furnace and the sample furnace varies with parameters such as furnace configuration, gas flow rates, and run conditions. However, these deviations are considered acceptably small by the majority of scientists using the system. The restrictions for accurate use of the remote system for all of the controlled-atmosphere furnaces now in use at JSC are

(a) redox conditions well above the graphite saturation surface,
(b) temperatures high enough for the initial gas mixture to react to completion, and
(c) flow rates within the recommended range (cf., ref. 4).

Experimental conditions characterized by CO/CO2 gas mixtures near the graphite saturation surface, low temperatures, or unusual gas flow rates can be attained; however, the scientist must be aware of the potential sources of error.
COMPUTER SOFTWARE

When initially implemented, the remote measuring system saved laboratory costs and promoted flexibility; however, it required that the scientist spend additional time setting up each experiment. First, after a run temperature and fO2 were chosen, the appropriate gas mixture had to be found in the Deines et al. (1974) tables. Then, the gas mixture and the temperature of the reference furnace were used to calculate (a) the fO2 of the reference furnace, and (b) the EMF readout from the zirconia electrolytic cell. Because the Deines et al. (1974) tables were designed for finding a gas mixture given a temperature and fO2, but not the reverse, finding the fO2 given the gas mixture was sometimes a time-consuming process.

For several years after the implementation of the reference system, many scientists used approximations to make the calculation process less cumbersome. One shortcut was to notice that in T-fO2 space, the CO/CO2 mixing curves were similar in trajectory to standard buffer curves (fig. 3). Then, for small changes in temperature, the buffer curve was assumed to be parallel to the CO/CO2 mixing curve on a plot of T versus fO2. If the fO2 of the reference furnace were set relative to that of a standard buffer, the fO2 inside the sample furnace could be computed analytically by assuming it to have the same offset from that standard buffer, although at a different temperature. If, at a later date, the assumption of parallel trajectories were deemed insufficient, then the actual fO2 to which the sample was exposed could be calculated.

Similarly, when a sample was set at a given temperature and fO2, and then the temperature was ramped, it was generally assumed that the fO2 stayed constant relative to a standard buffer curve (cf., notation of fig. 2). This approximation was adequate for small changes in temperature. However, large changes in temperature at a given gas mixture produced large changes in fO2, which had to be tediously calculated from the Deines et al. (1974) tables.3

In 1990, A. J. G. Jurewicz developed a computer program to use with the remote measuring system based upon a preexisting BASIC program.5 The central algorithm is based upon the computations of Deines et al. (1974). This new program

(a) calculates the fO2 in the reference furnace and EMF measured by the zirconia cell given the T, fO2 of the sample furnace and the temperature of the reference furnace

(b) back-calculates the fO2 in the sample furnace given the T and fO2 (or EMF measured by the zirconia cell) in the reference furnace

(c) calculates the fO2 as a function of temperature for a given gas mixture, the situation that occurs when the furnace conditions are set, and then the temperature is ramped up or down during the run.

In addition, the program calculates (1) changes in EMF with changes in both temperature and fO2, (2) changes in fO2 with small fluctuations in the gas composition, as well as (3) whether or not carbon will precipitate in the hot spot of the furnace. A sample printout from version 2.2 is shown in figure 4.
CONCLUDING REMARKS

Both the network of JSC controlled-atmosphere furnaces and the applicable computer software constitute a dynamic system. Physical modifications that may increase the reliability and the ease of using the furnaces are being tested continuously. Similarly, the computer program has been modified several times as needs arise, and it will continue to grow and provide more flexibility and efficiency for the scientist.

REFERENCES


Figure 1. Schematic of the manifold system used to divert the exhaust gases from the sample furnace to the reference furnace. Each valve controls the exhaust gas of a different furnace. When a valve is closed, the exhaust is unchanged from that of Williams and Mullins (1981). When a valve is opened, the exhaust gas is diverted through the reference furnace. Care must be taken not to open more than one valve at a time. Also, it is recommended that the valves on the manifold be check valves, although any gas-tight, two-way valve is adequate.
Figure 2. The difference between (1) the oxygen content of the sample furnace as measured directly and (2) the oxygen content of the sample furnace calculated from measurements in the reference furnace (e.g., \( \log f_O^2 \) sample - \( \log f_O^2 \) reference). Standard flow rates of CO/CO\(_2\) gasses were used; the zirconia cells were calibrated for any cell-specific nonlinearities; and, prior to the calibration, the sample furnace was determined to be free of gas leaks (air leaking into the system AFTER the hot spot of the sample furnace would distort the results). For this calibration, the reference furnace was set to near 1200°C (1205°C±5°C): a temperature high enough to ensure rapid gas-phase reactions. The CO/CO\(_2\) gas mixtures were chosen so that they produced oxygen partial pressures, which can be related to that of the Iron-Wustite (IW) standard buffer assemblage. For example, IW+1 refers to one log unit above the oxygen partial pressure produced by the IW buffer (i.e., 10 times the oxygen content), while IW-1 refers to conditions one log unit below the oxygen partial pressure produced by the IW buffer (i.e., the oxygen content divided by 10).
Figure 3. A graph in T-fO₂ space showing how lines of constant gas mixtures are subparallel to solid buffers.
### Example Run Conditions

**Specimen Furnace**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>T(°C)</td>
<td>1180</td>
</tr>
<tr>
<td>log(fO2/atm)</td>
<td>-13.16</td>
</tr>
<tr>
<td>%CO2</td>
<td>10.10</td>
</tr>
<tr>
<td>dlogfO2/d%CO2</td>
<td>+0.015</td>
</tr>
<tr>
<td>EMF (mV)*</td>
<td>+0.015</td>
</tr>
<tr>
<td>dEMF/dT (mV)</td>
<td>+0.015</td>
</tr>
<tr>
<td>dEMF/dfO2 (mV/0.1)</td>
<td>+0.015</td>
</tr>
<tr>
<td>Buffer+offset</td>
<td>IW-1.00</td>
</tr>
<tr>
<td>C in H.S.?</td>
<td>NO</td>
</tr>
</tbody>
</table>

**Reference Furnace**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>T(°C)</td>
<td>1110</td>
</tr>
<tr>
<td>log(fO2/atm)</td>
<td>-14.18</td>
</tr>
<tr>
<td>%CO2</td>
<td>10.10</td>
</tr>
<tr>
<td>dlogfO2/d%CO2</td>
<td>+0.015</td>
</tr>
<tr>
<td>EMF (mV)*</td>
<td>-953.9</td>
</tr>
<tr>
<td>dEMF/dT (mV)</td>
<td>+0.47</td>
</tr>
<tr>
<td>dEMF/dfO2 (mV/0.1)</td>
<td>+6.86</td>
</tr>
<tr>
<td>Buffer+offset</td>
<td>IW-1.00</td>
</tr>
<tr>
<td>C in H.S.?</td>
<td>NO</td>
</tr>
</tbody>
</table>

*This number is 19 mV from the ideal.

Figure 4. Printout from the BASIC program, which is used to relate the measurements from the reference furnace to conditions in the sample furnace. The information printed includes the temperatures of both furnaces (T(°C)); the oxygen content of the gas in each furnace in log units (log(fO2/atm)); the initial CO₂/(CO+CO₂) ratio of the gas (%CO₂); the voltage in millivolts measured across the electrolytic cell in the reference furnace (EMF); estimates of the changes in EMF with changes in temperature (dEMF/dT) and oxygen content (dEMF/dfO₂); the relationship of the specimen fO₂ to a standard buffer (buffer+offset); whether or not carbon will precipitate in the hot spots of the furnaces (C in H.S.); and, the correction for the intrinsic error of that particular zirconia cell (*).
APPENDIX

Software Used along with the Reference System of Determining Furnace Conditions.

This software actually consists of three separate GWBASIC programs. The programs are: GAS.BAS, used for selecting gas mixtures to achieve specific oxygen fugacities given ideal sample furnace temperatures; GASREV2.BAS, used for calculating the true oxygen fugacity in the sample furnace, given the sample temperature and the measured conditions in the reference furnace; and RAMP.BAS, used for seeing how conditions change in the sample furnace at a given gas mix when the temperature is ramped. All three applications have been linked using the CHAIN function, and are organized by a menu program (MENU.BAS), used for selecting applications. A fourth application program, CALIB.BAS, used for calculating the ideal EMF of a zirconia cell given a chemical analysis of a calibration gas and the measured furnace temperature, is still in progress. The software is based upon the BASIC program by A. Jurewicz (1986).
List of Variables.

A. Branching and String Variables

A$ = general branch variable.
B$ = YES if carbon will precipitate in the hot spot of the furnace and NO otherwise
BUFFERS = the label of the buffer ( = NONE if Z$=N)
CHANGE% = branch variable in menu.
CHOICE% = branch variable in menu.
C$ = general branch variable.
DECIDES = to continue or exit if carbon precipitation.
FURNACES = sample or reference
ICOUNTER = for indicating sample or reference
J, J1 = counting variables for print-out in ramping program.
LABEL$ = title for calculation &/or print-out.
P$ = branch, asking program to end loop.
Q$ = branch variable.
ROUND% = for checking if printer to use (Laser or Epson) has been defined.
STEPFO2 = counter for iteratively calculating oxygen fugacity, stepwise.
STEPT% = temperature steps over which to calculate oxygen fugacity at a
constant gas mixture.
V$ = checks for INKEY to end pause.
Y = number of loops the program has run (Y>0 means heading has already been
printed).
X$, XX$ = simple branches.
Z = counter for H= +1 or -1 (see Q(Z)).
Z$, Y = fO 2 corresponds to a buffer; N = fO 2 does not correspond to a buffer.

Constants.

Numeric Variables.

A = function of K1 and LOGFO2 as listed.
AA = new A for shift in temperature (calculating DELEMFDELT).
CORREMF = the systematic offset of the EMF of the reference furnace measured
during calibration.
DELEMFDELF02 = the change in ideal EMF with a small change in furnace fO2.
DELEMFDELT = the change of EMF with small changes in temperature.
EMF(T,K) = EMF at temperature T (celcius) and oxygen fugacity K (log units).
FCO = the partial pressure of carbon monoxide.
FCO2 = the partial pressure of carbon dioxide.
FO2OFFSET = deviation of oxygen fugacity from that of a standard, reference
buffer in sample furnace.
FO2OFFSET2 = deviation of oxygen fugacity from that of a standard, reference
buffer in reference furnace.
GG = new Gibbs free energy for calculating DELEMFDELT.
G1 = Gibbs free energy for the reaction, CO+O2 to CO2 from Deines et al (1974).
G2 = Gibbs free energy for the reaction, C+O2 to CO from Deines et al (1974).
H = incremental change in temperature (DELEMFDELT).
IDEALEMF = EMF calculated from the Nernst Equation.
K1 = equilibrium constant for the reaction CO+O2 to CO2 from Deines et al (1974).
K2 = equilibrium constant for the reaction, C+O2 to CO from Deines et al (1974).
KK = variable equilibrium constant (for calculating DELEMFDELT).
LOGFO2 = the natural logarithm of the oxygen fugacity of interest (atmospheres).
MIXRATIO% = %CO2 in gas mix for both furnaces.
DELRATIO = difference in gas mix between wanted (MIXRATIO) and calculated for calculating oxygen fugacity of reference furnace.
NEWFO2 = LOGFO2 after changing temperature.
Q(Z) = equilibrium constants used for checking stability of oxygen fugacity with temperature and gas mix.
REAL EMF = EMF including systematic offset.
REFFO2 = oxygen fugacity of reference furnace.
SPECFO2 = "specific" oxygen fugacity wanted in the sample furnace during an experiment; the specimen fO2.
TC = sample furnace temperature; initial temperature during ramp.
TREF = reference furnace temperature.
TFINAL = final temperature during ramp.
VOLCO2 = the gas mixture in volume% CO2.
W = TC+H for calculating stability of oxygen fugacity with temperature and gas mix.
This sub-program segment links all of the sub-programs together using the CHAIN command. Essentially, it acts like a menu for selecting applications (the other sub-programs).

Selects either Epson printer or Laser printer (auto form feed). See line 204.

1 ROUND%=-1:PRINTER$="EPSON"

Initial Title for Program

```
2 DIM REFFO2(100), FO2OFFSET2(100)
5 DEFDBL G,A,K
10 COLOR 15,14,14:CLS
20 PRINT:PRINT:PRINT:PRINT
30 PRINT "CO/CO2 GAS MIXING UTILITY PROGRAM"
32 PRINT "version 3.0":PRINT:PRINT
34 PRINT "See A.J.G Jurewicz, if you have any questions"
40 PRINT:PRINT:PRINT
41 COLOR 14,15,15
42 PRINT "Press any key to continue ..."
50 V$=INKEY$
60 IF (LEN(V$)=0) GOTO 50
```

Program Menu

```
70 COLOR 15,9,9:CLS:PRINT:PRINT:PRINT:PRINT
75 ROUND% = ROUND% + 1
80 PRINT "Do you want to:"
90 PRINT:PRINT " 1. Take known sample temperature and oxygen fugacities"
100 PRINT " and determine the %CO2 of the gas, and the "
102 PRINT " fO2, & EMF of reference furnace"
110 PRINT:PRINT
120 PRINT " 2. Take EMF (or setpoint) and T of reference furnace and"
130 PRINT " determine fO2 of the sample"
140 PRINT:PRINT
150 PRINT " 3. Determine the range of fO2's experienced by a sample"
160 PRINT " with a fixed gas mixture, but where the temperature was"
170 PRINT " ramped"
180 PRINT:PRINT:PRINT " 4. Exit program"
185 COLOR 1,15,15
190 PRINT:PRINT:PRINT "Enter choice of 1, 2, 3, or 4. ":;INPUT "",CHOICE%
200 IF (CHOICE%<1) OR (CHOICE%>4) GOTO 190
201 IF (CHOICE% = 4) GOTO 211
202 IF ROUND%>0 GOTO 210
```

Selects either Epson printer or Laser printer. The only difference for this program is that the Laser printer requires an automatic form feed after printing.

```
204 COLOR 14,15,15:PRINT:PRINT:PRINT "Do you have a laser printer (Y/N)?":
206 Q$=INPUT$(1):IF (Q$="y") OR (Q$="Y") GOTO 209
207 IF (Q$="n") OR (Q$="n") GOTO 210
208 PRINT "HUH?":GOTO 204
209 PRINTER$="LASER"
```
Branch to other sub-programs, or Exit, as selected on menu.

210 COLOR 31,1,1:CLS:PRINT:PRINT:PRINT:PRINT "LOADING NEXT SEGMENT"
211 ON CHOICE% GOTO 220,230,240,250
220 CHAIN "GAS.BAS",10,ALL
230 CHAIN "GASREV2",10,ALL
240 CHAIN "RAMP",10,ALL
250 COLOR 17,15,15
252 PRINT:PRINT:PRINT "Are you sure that you want to exit (Y/N)?":;
260 Q$=INPUT$(1):IF (Q$="y") OR (Q$="Y") GOTO 290
270 IF (Q$="n") OR (Q$="n") GOTO 70
280 PRINT "HUH?":GOTO 240
290 PRINT "O.K.";COLOR 15,0,0:CLS
300 PRINT:PRINT:PRINT
305 PRINT "At the OK prompt, type SYSTEM"
310 PRINT "To return to the operating system":PRINT:PRINT:END
GAS.BAS
This sub-program takes the sample temperature and oxygen fugacity which the experimentalist desires, and calculates what the gas mixture should be. Then, given the temperature of the reference furnace, calculates the oxygen fugacity of the reference furnace and the EMF registered by the zirconia cell.

Sets User-defined functions as well as main menu.

10 REM
20 REM THIS VERSION INCLUDES CHANGING REFERENCE GAS TO OXYGEN
30 DEF FNEMF(T,K) = .0496055*(T+273)*(K+0)
40 REM DEFDBL G,K,A
50 DEF FN RM(X,Y)=(X-3*X*(10^Y)-
2*((10^Y)^3(2)))/(2*X*(10^Y)+(10^Y)+((10^Y)^3(2))+SQR(10^Y))
52 COLOR 15,1,1:CLS
60 PRINT:PRINT
62 PRINT " PROGRAM TO CALCULATE PARAMETERS TO BE USED WITH"
70 PRINT " NASA CO/CO2 GAS-MIXING FURNACES"
72 PRINT
73 PRINT " REMINDER .... "
80 PRINT " FOR EXTREME RANGES (OTHER THAN 1000C<T<1400,
-6<logfO2<16)"
90 PRINT " THE ZIRCONIA CELL MAY BECOME NON-IDEAL"
100 COLOR 6,15,15
127 PRINT:PRINT "Do you wish to continue, or return to the main menu (C/R)?"
128 Q$=INPUT$ (1)
129 IF (Q$="c") OR (Q$="C") GOTO 150
130 IF (Q$="r") OR (Q$="R") GOTO 140
135 PRINT:PRINT "HUH?":PRINT:GOTO 127
140 CHAIN "MENU.BAS",70,ALL

Selecting and changing variables.

150 Y=0: R=1.98726E-03
200 REM VARIABLES USED ARE:
202 REM MORE VARIABLES USED ARE: IDEALEMF
205 PRINT:PRINT:PRINT
210 TC=1400:TREF=1065:FO2OFFSET2=-1:BUFFER$="IW":SPECFO2=-10.69719
215 CORREMF=26:BUFFER$="IW"
220 COLOR 15,3,3:CLS:PRINT:PRINT:PRINT "The current conditions are:"
225 B$(1)="NO":B$(2)="NO":DECIDE$="YES"
230 PRINT:PRINT " 1. Sample temperature is ";TC," degrees C"
250 PRINT " 2. Sample oxygen fugacity is ";SPECFO2," log (/atm)"
255 IF (FO2OFFSET2=0) GOTO 265
260 PRINT " --- which is ";BUFFER$," offset ";FO2OFFSET2," log units":GOTO 270
265 PRINT " --- which is ";BUFFER$" 3. Reference temperature is ";TREF," degrees C"
280 PRINT " 4. The correction to the zirconia cell is ";CORREMF," mV"
300 PRINT " and 5. The reference buffer is ";BUFFER$"
320 PRINT:PRINT:PRINT  
380 COLOR 15,9,9:PRINT "Enter the number you wish to change (1 - 5, 6 to continue; 7 to exit). ":;  
390 INPUT ",CHANGE%  
400 IF (CHANGE%>0) AND (CHANGE%<8) GOTO 420  
410 COLOR 31,9,9:PRINT "Please enter a number, 1 - 7. ":;COLOR 15,9,9:GOTO 390  
420 CLS:ON CHANGE% GOTO 430,580,440,450,487,1000,140  

Changing sample temperature at constant fO2.  
430 INPUT "Enter the new sample temperature in degrees C";TC  
431 IF (BUFFER$="IW") GOTO 850  
432 IF (BUFFER$="WM") GOTO 851  
433 IF (BUFFER$="MH") GOTO 852  
434 IF (BUFFER$="QFM") GOTO 853  
435 IF (BUFFER$="NNO") GOTO 854  
436 PRINT "ERROR IN PROGRAM/NO BUFFER":STOP  

Changing reference furnace temperature.  
440 INPUT "Enter the new reference temperature in degrees C";TREF:CLS:GOTO 220  

Changing the deviation of zirconia cell from the ideal (systematic offset determined by calibration at reference temperature).  
450 INPUT "Enter the new correction to the zirconia cell";CORREMF:CLS:GOTO 220  

Changing the oxygen fugacity of the sample furnace, as well as the reference buffer (see also 580).  
460 CLS:PRINT:PRINT:PRINT  
470 PRINT "The sample fO2 data will be given as relative to a buffer, as well"  
480 PRINT "as in log(fO2/atm) units. Your choice of 'reference buffers' are:"  
485 GOTO 490  
487 CLS:PRINT:PRINT:PRINT:PRINT "Your choice of reference buffers are:"  
490 PRINT:PRINT "  1. IW"  
500 PRINT "  2. WM"  
510 PRINT "  3. MH"  
520 PRINT "  4. QFM"  
530 PRINT "  5. NNO"  
535 PRINT:PRINT "(Your current choice is )":BUFFER$  
540 PRINT:PRINT "Enter the number of your selection."  
550 INPUT ",CHANGE%  
560 IF (CHANGE%<1) OR (CHANGE%>5) GOTO 460  
570 ON CHANGE% GOTO 571,572,573,574,575  
571 BUFFER$="IW":GOTO 850  
572 BUFFER$="WM":GOTO 851  
573 BUFFER$="MH":GOTO 852  
574 BUFFER$="QFM":GOTO 853  
575 BUFFER$="NNO":GOTO 854
580 CLS
590 PRINT:PRINT:PRINT
600 PRINT "The sample furnace fO2 can be expressed as any of three ways:"
610 PRINT:PRINT "1. as a standard buffer"
620 PRINT "2. as offset from a standard buffer"
630 PRINT "3. as a specific fO2 (independent of any buffer)"
650 PRINT:PRINT "How would you like to express your new fO2"
660 PRINT "(Enter 1-3, or 4 for unchanged)."
670 INPUT "",CHANGE%
680 IF (CHANGE%<1) OR (CHANGE%>4) GOTO 590
690 ON CHANGE% GOTO 700,700,820,692
692 CLS:GOTO 220
700 PRINT:PRINT "Your choices of standard reference buffers are:":PRINT
701 PRINT "1. IW"
702 PRINT "2. WM"
703 PRINT "3. MH"
704 PRINT "4. QFM"
705 PRINT "5. NNO"
706 PRINT "6. none above. Return to previous menu."
710 PRINT:PRINT "Enter the number of your selection. "":;
720 INPUT "",CHOICE%
730 IF (CHOICE%<1) OR (CHOICE%>6) GOTO 710
740 FO2OFFSET2=0:IF (CHANGE%=1) GOTO 760
750 INPUT "Enter the offset from the buffer, in log (fO2/atm) units.",FO2OFFSET2
760 ON CHOICE% GOTO 770,780,790,800,810,820
770 SPECFO2 = 6.57 - 27215/(TC + 273) + FO2OFFSET2:BUFFER$="IW":CLS:GOTO 220
780 SPECFO2 = 13.12 - 32730/(TC + 273) + FO2OFFSET2:BUFFER$="WM":CLS:GOTO 220
790 SPECFO2 = 13.966 - 24634/(TC + 273) + FO2OFFSET2:BUFFER$="MH":CLS:GOTO 220
800 SPECFO2 = 9.1 - 25738/(TC + 273) + FO2OFFSET2:BUFFER$="QFM":CLS:GOTO 220
820 CLS:PRINT:PRINT "The current log(fO2) is ";SPECFO2
830 PRINT:PRINT "What is the log of the new fO2 that you want? "":;
840 INPUT SPECFO2:GOTO 431
850 FO2OFFSET2=SPECFO2-(6.57 - 27215/(TC + 273)):CLS:GOTO 220
851 FO2OFFSET2=SPECFO2-(13.12 - 32730/(TC + 273)):CLS:GOTO 220
852 FO2OFFSET2=SPECFO2-(13.966 - 24634/(TC + 273)):CLS:GOTO 220
853 FO2OFFSET2=SPECFO2-(9.1 - 25738/(TC + 273)):CLS:GOTO 220
854 FO2OFFSET2=SPECFO2-(9.359999 - 24930/(TC + 273)):CLS:GOTO 220

Program branches to calculate the parameters of interest: the gas mix needed for the sample furnace, the oxygen fugacity of the reference furnace, the EMF of the zirconia cell in the reference furnace, and the effect of small changes in the gas mixture on the oxygen fugacity in the sample furnace and the EMF of the reference furnace.

1000 COLOR 15,0,0
1001 REM NOW FO2 HAS BEEN INPUT
1010 REM SUBROUTINE AT 1750 CALCULATES GASMIXTURUES, 1830 = C STABILITY
1020 REM SUBROUTINE AT 1100 CALCULATES EMF'S
1030 REM SUBROUTINE AT 1380 CALCULATES DELEMFDDEL; 1680 = DEMFDO2
1031 REM SUBROUTINE AT 1530 ITERATES REFERENCE FURNACE FO2
1035 CLS:PRINT:PRINT:PRINT "WORKING .... "
1040 LOGFO2=SPECFO2:T=TC:GOSUB 1760
1050 MIXRATIO=VOLCO2:FURNACE$="SAMPLE":ICOUNTER=1:GOSUB 1830
1060 IF (VOLCO2>100) GOTO 1290
1070 IF (DECIDE$="NO") GOTO 220
1080 GOSUB 1530
1090 FURNACE$="REFERENCE":ICOUNTER=2:GOSUB 1830
1100 IF (DECIDE$="NO") GOTO 220
1110 GOSUB 1680
1120 GOSUB 1380
1121 REM CALCULATING APPROX. DCO2/DFO2
1125 DELFO2DELCO2=0.1/(DVOLCO2)

Printing routine for printing finished results to the screen.

1130 PRINT:PRINT:PRINT "Enter a label for your run conditions: ";:INPUT LABELS
1140 CLS
1150 PRINT LABELS
1160 PRINT:PRINT:PRINT SPC(15);"SPECIMEN FURNACE ";"REFERENCE FURNACE"
1170 PRINT USING ";########";"T( C)";TC;TREF
1180 PRINT USING ";+_##.##+_##.##";"log(FO2/atm)";SPECFO2;REFFO2
1190 PRINT USING "
###.###","%CO2";MIXRATIO;(MIXRATIO-DELRATIO)
1195 PRINT USING "
###.###","dlogFO2/d%CO2";DELFO2DELCO2
1200 PRINT USING "
###.###","EMF(mV)";REALEMF
1210 PRINT USING "
###.###","dEMF/dT(mV)";DELEMFDDEL
1220 PRINT USING "
###.###","dEMF/dFO2(mV/.1)";DELEMFDDELFO2
1225 IF (FO2OFFSET2=0) GOTO 1240
1230 PRINT USING "
###.###","buffer + offset";BUFFER$;FO2OFFSET2:GOTO 1260
1240 PRINT USING "
###.###","buffer";BUFFER$
1250 PRINT USING "C in H.S. ? 
###.###","B$(1);B$(2)
1270 PRINT:PRINT "This number is offset ";CORREMF;" mV from the ideal. "
1272 PRINT:PRINT "Do you wish to Print the results, Continue with this program, "
1273 PRINT " or Return to the main menu? (P/C/R)"
1274 P$= INPUTS (1)
1275 IF (P$="r") OR (P$="R") GOTO 1360
1276 IF (P$="c") OR (P$="C") GOTO 1350
1277 IF (P$="p") OR (P$="P") GOTO 1279
1278 PRINT:PRINT "Huh?":PRINT:GOTO 1272
1279 GOSUB 2150
1280 GOTO 1300
1290 PRINT "The gasmix is unrealistic (eg.,";VOLCO2;"%). Try again."
1300 PRINT:PRINT "Do you wish to continue with this program, "
1301 PRINT " or return to the main menu? (C/R)"
1310 A$= INPUTS (1)
1320 IF (A$="r") OR (A$="R") GOTO 1360
1330 IF (A$="c") OR (A$="C") GOTO 1350
Sub-routine for calculating the deviations in oxygen fugacity with temperature.

```basic
1380 Z = 1
1390 RATIO = FCO2 /((1-FCO2-10*REFFO2)
1400 AA = (1 - 2*RATIO*(100 / MIXRATIO - 1))/(1 + 2*(100 / MIXRATIO - 1))
1410 PART = LOG(1 - AA) - LOG(100 / MIXRATIO - 1)
1420 FOR I = 1 TO -1 STEP -2
1430 H = I
1440 W = T + H
1450 GG = 62.110326# -.02144446#*W + 4.720326E-07*(W^2)+(-4.5574288#)*(10^-12)*(W^3)- 7.343018200000001 #(10^-15)*(W^4)
1460 KK = EXP(- GG/(R*(W + 273.18)))
1470 Q(Z) = KK
1480 Z = Z + 1
1490 NEXT I
1500 NEWFO21 = LOG10*.5*(LOG(Q(1)) + PART):NEWFO22 = LOG10*.5*(LOG(Q(2)) + PART)
1510 DELEMFDELT = (FN EMF(TREF + 1,NEWFO21) - FN EMF(TREF - 1,NEWFO22))/2
1520 RETURN
```

Given a gas mixture and a temperature, this sub-routine calculates the oxygen fugacity of the furnace, as well as the expected EMF of the zirconia cell.

```basic
1530 REM SUBROUTINE TO ITERATIVELY CALCULATE THE FO2 OF THE REFERENCE FURNACE
1540 STEPFO2=1:IF (TC>TREF) GOTO 1570
1550 IF (TC=TREF) GOTO 1570
1560 STEPFO2=-STEPFO2
1570 T=TREF
1580 LOGFO2=LOGFO2-STEPFO2:GOSUB 1760
1590 DELRATIO=MIXRATIO-VOLCO2
1600 IF ABS(DELRATIO)<.001 GOTO 1660
1610 IF (TREF>TC) GOTO 1640
1620 IF (DELRATIO<0) GOTO 1580
1630 LOGFO2=LOGFO2+STEPFO2:STEPFO2=STEPFO2/2:GOTO 1580
1640 IF (DELRATIO>0) GOTO 1580
1650 LOGFO2=LOGFO2+STEPFO2:STEPFO2=STEPFO2/2:GOTO 1580
1660 REFFO2=LOGFO2
1665 IDEALEMF=FN EMF(TREF,REFFO2):REALEMF=IDEALEMF+CORREMF
1670 RETURN
```

Sub-routine calculates the EMF of the zirconia cell in the reference furnace, as well as the change of EMF with small changes in temperature and oxygen fugacity.

```basic
1680 REM NOW THAT WE HAVE TEMPERATURE AND LOGFO2, WE NEED TO CALCULATE THE EMF'S AND DELTA-EMF'S
1690 IDEALEMF = FN EMF (TREF,REFFO2)
```

A-10
Sub-routine for taking the temperature of a furnace and computing the CO/CO2 gas mixture needed to obtain a specific oxygen fugacity.

Sub-routine for checking whether carbon will precipitate in the hot-spot of the furnace.

Routine for printing the results to paper.
2230 LPRINT USING "\ \ \ \ \ +.##";"buffer +
offset";BUFFER$;FO2OFFSET2:GOTO 2260
2240 LPRINT USING \ \ \ \ ;"buffer";BUFFER$
2260 LPRINT USING "C in H.S.? \ \ \ \ B$(1);B$(2)
2270 LPRINT:LPRINT "* This number is offset ";CORREM;" mV from the ideal. "
2275 IF (PRINTER$="EPSON") GOTO 2278
2276 LPRINT CHR$(12);:GOTO 2280
2278 LPRINT:LPRINT
2280 CLS
2290 RETURN
GASREV2.BAS

This sub-program takes old data, for which (1) the reference conditions (temperature, EMF) and (2) the sample temperature are known and calculates what the ambient oxygen fugacity was in the sample furnace.

Sets User-defined functions, as well as main menu.

1 REM VARIABLES INCLUDE: MIXRATIO, VOLCO2, G1, G2, (ALL VARIABLES
2 REM FROM DEFAULTS.BAS), DVOLCO2, FC02, R, K1, K2, A, B$(1), B$(2).
3 REM OPTIONS%
4 REM T, LOGFO2, DC02DFO2REF
10 DEF FN RM(X,Y)=(X-3*X*(10^Y)-2*((10^Y)^((3/2))))/(2*X*(10^Y)+(10^Y)+((10^Y)^((3/2)))+SQR(10^Y))
15 COLOR 15,14,14:CLS:PRINT:PRINT:PRINT
20 PRINT "PROGRAM TO TAKE DATA FROM REFERENCE
25 PRINT " AND TO CALCULATE THE CONDITIONS IN THE SAMPLE
30 PRINT "FURNACE";
35 PRINT "FOR CO/CO2 GAS MIXING SYSTEMS"
40 PRINT:PRINT:PRINT
50 COLOR 15,1,1 :PRINT "Do you wish to continue, or return to the main menu
55 PRINT "(C/R)?"
60 Q$=INPUT$(1)
70 IF (Q$="c") OR (Q$="C") GOTO 110
80 IF (Q$="R") OR (Q$="r") GOTO 100
85 PRINT "HUH?":GOTO 50
100 CHAIN "MENU.BAS",70,ALL

Sets default variables, and allows them to be changed using a menu-driven system.

110 CLS
113 TC=1400:TREF=1065:FO2OFFSET=0:BUFFER$="IW":REFFO2=-
114 IDEALEMF=-913.9486:CORREMF=26:BUFFER$="IW"

200 REM VARIABLES USED ARE:
201 REM T, TC, TREF, VOLCO2, G1, G2, (ALL VARIABLES
202 REM FROM DEFAULTS.BAS), DVOLCO2, FC02, R, K1, K2, A, B$(1), B$(2).
203 REM OPTIONS%
204 REM T, LOGFO2, DC02DFO2REF
205 REM IDEALEMF, CORREMF, REFFO2
206 REM IDEALEMF+CORREMF
207 REM IDEALEMF+CORREMF
208 REM IDEALEMF+CORREMF
209 REM IDEALEMF+CORREMF
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330 REM IDEALLEMF+CORREMF

A-13
380 PRINT "Enter the number you wish to change (or 6 to continue; 7 to exit)";:
390 INPUT ", CHANGE% 
400 IF (CHANGE%>0) AND (CHANGE%<8) GOTO 420 
410 COLOR 31,1,1:PRINT "Please enter a number, 1 - 7.";COLOR 15,1,1:GOTO 390 
420 ON CHANGE% GOTO 430,440,580,450,487,1000,100

Changes sample temperature at constant.

430 INPUT "Enter the new sample temperature in degrees C";TC:CLS:GOTO 205

Changes reference temperature.

440 INPUT "Enter the new reference temperature in degrees C";TREF:CLS:GOTO 845

Changes systematic deviation in the zirconia cell.

450 INPUT "Enter the new correction to the zirconia cell";CORREMF:CLS:GOTO 205

Changes the standard reference buffer used to describe the oxygen fugacity.

460 CLS:PRINT:PRINT:PRINT
470 PRINT "The sample fO2 data will be given as relative to a buffer, as well"
480 PRINT "as in log(fO2/atm) units. Your choice of 'reference buffers' are:");
485 GOTO 490
487 CLS:PRINT:PRINT:PRINT:PRINT "Your choice of buffers are:" 
490 PRINT:PRINT " 1. IW"
500 PRINT " 2. WM"
510 PRINT " 3. MH"
520 PRINT " 4. QFM"
530 PRINT " 5. NNO"
535 PRINT:PRINT "(Your current choice is )";BUFFER$ 
540 PRINT:PRINT "Enter the number of your selection.";:
550 INPUT " ", CHANGE% 
560 IF (CHANGE%<1) OR (CHANGE%>5) GOTO 460 
570 ON CHANGE% GOTO 571,572,573,574,575 
571 BUFFER$="IW":GOTO 850 
572 BUFFER$="WM":GOTO 850 
573 BUFFER$="MH":GOTO 850 
574 BUFFER$="QFM":GOTO 850 
575 BUFFER$="NNO":GOTO 850

Changes the oxygen fugacity of the reference furnace. This section allows the oxygen fugacity to be described either as a standard buffer, as an offset from a standard buffer, or in atmospheres (log units).

580 CLS 
590 PRINT:PRINT:PRINT 
600 PRINT "The reference furnace fO2 can be expressed as any of several ways:" 
610 PRINT:PRINT " 1. as a standard buffer 
620 PRINT " 2. as offset from a standard buffer" 
630 PRINT " 3. as a specific fO2 (independent of any buffer) "

A-14
4. as the EMF (in mV) indicated by the zirconia cell

How would you like to express your new fO2

Enter 1-4 (or 5 to continue; 6 to exit).

IF (CHANGE% < 1) OR (CHANGE% > 6) GOTO
ON CHANGE% GOTO 700,700,820,870,205,100

Your choices of standard reference buffers are:

1. IW
2. WM
3. MH
4. QFM
5. NNO
6. none above. Return to previous menu.

Enter the number of your selection.

IF (CHOICE% < 1) OR (CHOICE% > 6) GOTO 710
FO2OFFSET = 0: IF (CHANGE% = 1) GOTO 760
INPUT "Enter the offset from the buffer, in log (fO2/atm) units.",FO2OFFSET
ON CHOICE% GOTO 770,780,790,800,810,580
REFFO2 = 6.57 - 27215/(TREF + 273) + FO2OFFSET:
IDEALEMF = 0.0496055*(TREF+273)*REFFO2:
BUFFER$ = "IW":CLS:GOTO 205
REFFO2 = 13.12 - 32730/(TREF + 273) + FO2OFFSET:
IDEALEMF = 0.0496055*(TREF+273)*REFFO2:
BUFFER$ = "WM":CLS:GOTO 205
REFFO2 = 13.966 - 24634/(TREF + 273) + FO2OFFSET:
IDEALEMF = 0.0496055*(TREF+273)*REFFO2:
BUFFER$ = "MH":CLS:GOTO 205
REFFO2 = 9.1 - 25738/(TREF + 273) + FO2OFFSET:
IDEALEMF = 0.0496055*(TREF+273)*REFFO2:
BUFFER$ = "QFM":CLS:GOTO 205
REFFO2 = 9.359999 - 24930/(TREF + 273) + FO2OFFSET:
IDEALEMF = 0.0496055*(TREF+273)*REFFO2:
BUFFER$ = "NNO":CLS:GOTO 205

What is the log of the fO2 you want?

IF (BUFFER$ = "IW") GOTO 860
IF (BUFFER$ = "WM") GOTO 862
IF (BUFFER$ = "MH") GOTO 864
IF (BUFFER$ = "QFM") GOTO 866
IF (BUFFER$ = "NNO") GOTO 868
PRINT "THERE IS A PROBLEM WITH THE PROGRAM!!!!!!!":STOP
FO2OFFSET = REFFO2 - 6.57 - 27215/(TREF + 273):GOTO 205
FO2OFFSET = REFFO2 - 13.12 - 32730/(TREF + 273):GOTO 205
FO2OFFSET = REFFO2 - 13.966 - 24634/(TREF + 273):GOTO 205
FO2OFFSET = REFFO2 - 9.1 - 25738/(TREF + 273):GOTO 205
FO2OFFSET = REFFO2 - 9.359999 - 24930/(TREF + 273):GOTO 205

CLS
PRINT:PRINT:PRINT
At this point, the temperature and oxygen fugacity of the reference furnace is known, as is the temperature of the sample furnace. Therefore, the program needs to calculate the gas mixture. Once the gas mixture is known, it can be used to calculate the oxygen fugacity in the sample furnace.

Calculates gas mixture via subroutines.

```
1000 REM CALCULATE THE GAS MIXTURE
1005 COLOR 20,15,0:CLS:PRINT "WORKING ... PLEASE BE PATIENT"
1010 T=TREF:LOGFO2=REFFO2:ICOUNTER=2
1020 B$(2)="NO":GOSUB 2000
1240 MIXRATIO=VOLCO2:DCO2DFO2REF=DVOLCO2
1241 IF (MIXRATIO<100.0) GOTO 1250
1242 CLS:PRINT:PRINT:PRINT:PRINT:PRINT:COLOR 4,15,0:PRINT "The gas mixture is unrealistic"
1243 PRINT:PRINT:PRINT:PRINT "Do you want to:
1244 PRINT:PRINT:PRINT "1. see your defaults"
1245 PRINT " 2. continue with this unrealistic mixture, or"
1246 PRINT " 3. return to the main menu":PRINT "Please enter your choice (1, 2, or 3)";
1248 INPUT OPTIONS%:IF (OPTIONS%<1) OR (OPTIONS%>3) GOTO 1242
1249 ON OPTIONS% GOTO 1250,1250,100
1250 IF ((10^REFFO2)>(K2*FCO2)) GOTO 1300
1255 CLS:B$(2)="YES"
1260 PRINT:PRINT:COLOR 4,15,0:PRINT "Carbon HAS precipitated in the reference furnace."
1270 PRINT "Accordingly, the fO2 is not what the zirconia cell suggests."
1280 PRINT "Do you wish to continue using these same parameters? (Y/N)"
1282 Q$=INPUT$(1)
1283 IF (Q$="Y") OR (Q$="y") GOTO 1300
1285 IF (Q$="n") OR (Q$="n") GOTO 220
1287 PRINT "HUH?":GOTO 1280
```

Calculates oxygen fugacity of the sample furnace, given the sample temperature and the gas mixture via sub-routines. This section of the program also checks to see if carbon would have precipitated in the hot spot of the sample furnace. If so, the conditions are flagged because it is certain that the calculation of oxygen fugacity is not valid.
1300 REM WE NOW HAVE TC, VOLCO2=MIXRATIO, WE NEED FO2 OF
SAMPLE FURNACE
1310 ICOUNTER=1
1320 COLOR 15,1,1:B$(1)="NO";GOSUB 2300
1330 GOSUB 2000
1340 DCO2DFO2SPEC=DVOLCO2
1350 IF ((10^ SPECFO2)>(K2*FCO2)) GOTO 1390
1355 CLS:B$(1)="YES"
1360 PRINT:PRINT "Carbon HAS precipitated in the
specimen furnace."
1370 PRINT "Accordingly, the fO2 is not what the zirconia cell suggests."
1380 PRINT "Do you wish to continue using these same parameters? (Y/N)
1382 Q$=INPUT$(1)
1383 IF (Q$="Y") OR (Q$="y") GOTO 1400
1385 IF (Q$="n") OR (Q$="N") GOTO 220
1387 PRINT "HUH?";GOTO 1380
1390 GOSUB 2800

Prints results to the screen.

1400 REM EVERYTHING IS CALCULATED -- JUST NEED TO PRINT
1580 CLS: COLOR 15,4,4:PRINT:PRINT:PRINT "Enter a label for your run
conditions:";INPUT LABEL$
1582 CLS
1585 PRINT LABEL$
1590 PRINT:PRINT:PRINT SPC(15);"SPECIMEN FURNACE ";=" REFERENCE
FURNACE"
1595 PRINT USING " \\ \\ \\ \\ ";"T( C)";TC;TREF
1600 PRINT USING " \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ \\ ";"log(fO2/atm)";SPECFO2;REFFO2
1605 PRINT USING " \\ \\ \\ \\ ";";"EMF(mV)";REALEMF
1610 PRINT USING " \\ \\ \\ \\ \\ \\ \\ \\ ";"buffer +
offset";BUFFER$;FO2OFFSET2;BUFFER$;FO2OFFSET
1625 PRINT USING "C in H.S.? \\ \\ \\ \\ \\ \\ \\ \\ \\ ";B$(1);B$(2)
1630 PRINT:PRINT "This number is offset ";CORREMF; mV from the ideal."
1640 COLOR 15,1,1:PRINT:PRINT "Do you wish to:";
1645 PRINT " 1. print the results
1650 PRINT " 2. change the variables & continue, or 
1660 PRINT " 3. exit to main menu"
1670 PRINT:PRINT "Enter your choice, 1,2, or 3:";INPUT OPTIONS$
1680 IF (OPTIONS%<1) OR (OPTIONS%>3) GOTO 1640
1690 ON OPTIONS% GOTO 1700,205,100
1700 GOSUB 3000
1710 STOP

Sub-routine which calculates the gas mix from the reference furnace parameters.

2000 REM ROUTINE TO TAKE REFERENCE FURNACE T, FO2 AND
CALCULATE THE GAS MIX
2180 G1 = 62.110326# + T^*(.02144446#)+(T^2)*(4.720326)*(10^(- 7))+(T^3)*(-
4.5574288#)*(10^(- 12))+(T^4)*(- 7.343018200000001#)*(10^(- 15))
2190 \( G_2 = 94.25770200000001 + T^* (7.321945) -(10^{-4}) -(T^2) *(10^{-7}) -(3.416474) + (T^3) *(4.7858617) *(10^{-11}) \)

2200 \( K_1 = \exp((-1/((R^*(T + 273.18))))); K_2 = \exp((- G_2 /((R^*(T + 273.18)))) \)

2210 \( A = (K_1 -(\sqrt{10^\log F_{O_2}}) )*\text{FN RM}(K_1, \log F_{O_2})/(K_1 + \sqrt{10^\log F_{O_2}}) \)

2220 \( F_{CO_2} = 2(1 - A)/(2 + A + 2*\text{FN RM}(K_1, \log F_{O_2}) \)

2230 \( VOL_{CO_2} = 100/(1 + \text{FN RM}(K_1, \log F_{O_2}) \)

2240 \( DVOL_{CO_2} = 100/(1 + \text{FN RM}(K_1, \log F_{O_2} + 1)) - 100/(1 + \text{FN RM}(K_1, \log F_{O_2} - 1)) \)

2240 \( \text{RETURN} \)

Sub-routine which calculates the oxygen fugacity seen by the sample (specimen) given the gas mix calculated for the reference furnace and the temperature of the sample furnace.

2300 \( \text{REM SUBROUTINE TO TAKE TC, GAS MIX AND CALCULATE SPECFO2} \)

2305 \( \text{STEPFO2}=0 \text{ IF (TREF}>TC) \text{ OR (TREF} = TC) \text{ GOTO 2385} \)

2310 \( \text{STEPFO2}=-\text{STEPFO2} \)

2315 \( T=TC \)

2320 \( \text{LOGFO2}=\text{LOGFO2} - \text{STEPFO2} \)

2325 \( \text{GOSUB 2000} \)

2330 \( \text{DELRATIO} = \text{MIXRATIO} - \text{VOLCO2} \)

2335 \( \text{IF (ABS(DELRATIO)}<.0001) \text{ GOTO 2490} \)

2340 \( \text{IF (TC>TREF) GOTO 2425} \)

2345 \( \text{IF (DELRATIO>0) GOTO 2390} \)

2350 \( \text{LOGFO2}=\text{LOGFO2} + \text{STEPFO2} \)

2355 \( \text{STEPFO2} = \text{STEPFO2} / 2 \)

2360 \( \text{GOTO 2390} \)

2365 \( \text{IF (DELRATIO<0) GOTO 2390} \)

2370 \( \text{LOGFO2}=\text{LOGFO2} + \text{STEPFO2} \)

2375 \( \text{STEPFO2} = \text{STEPFO2} / 2 \)

2380 \( \text{GOTO 2390} \)

2385 \( \text{SPECFO2}=\text{LOGFO2} \)

2390 \( \text{RETURN} \)

Sub-routine for printing the final results to paper.

3000 \( \text{LPRINT LABELS} \)

3010 \( \text{LPRINT:LPRINT:LPRINT SPC(15);"SPECIMEN FURNACE ";" REFERENCE FURNACE"} \)

3015 \( \text{LPRINT USING " \\

### \\
### "T(C);TC;TREF} \)

3020 \( \text{LPRINT USING " \\

### \\
### \\
### \\
### "log(FO2/atm);SPECFO2;REFFO2} \)

3025 \( \text{LPRINT USING " \\

### \\
### \\
### \\
### \\
### "%CO2;MIXRATIO;MIXRATIO-DELRATIO} \)

3030 \( \text{LPRINT USING " \\

### \\
### \\
### \\
### "EMF(mV)*;REALMF} \)

A-18
3119 LPRINT USING "\ \ \ +#.## \ \ +#.##":"buffer +
offset";BUFFER$;FO2OFFSET2;BUFFER$;FO2OFFSET
3125 LPRINT USING "C in H. S.? \ \ \ \ \ ";B$(1);B$(2)
3130 LPRINT:LPRINT "";CORREMF;" mV from the ideal. "
3135 IF (PRINTER$="EPSON") GOTO 3138
3137 LPRINT CHR$(12);GOTO 3140
3138 LPRINT:LPRINT
3140 CLS
3150 RETURN 1640
RAMP.BAS

This sub-program ramps the temperature of a furnace at a constant gas mixture, and calculates the oxygen fugacity at each temperature. This option is a useful tool for designing cooling-rate experiments.

Sets User-defined functions and presents main menu.

10 REM DIM REFFO2(100), FO2OFFSET2(100)
20 REM THIS VERSION INCLUDES CHANGING REFERENCE GAS TO OXYGEN
30 DEF FNEMF(T,K) = .0496055*(T+273)*(K+0)
40 REM DEFDBL G,K,A
50 DEF FN RM(X,Y)=(X-3*X*(10^Y) - 2*((10^Y)^(3/2)))/(2*X*(10^Y)+( 10^Y)+ (( 10 ^Y) ^(3/2))+SQR(10^Y))
52 COLOR 15,6,6:CLS
60 PRINT:PRINT:PRINT "PROGRAM THAT CALCULATES FO2'S"
70 PRINT:PRINT "WHEN THE TEMPERATURE IS RAMPED IN A FURNACE WITH A SET GAS MIX"
72 PRINT:PRINT
100 COLOR 6,15,15
127 PRINT:PRINT "Do you wish to continue, or return to the main menu (C/R)?"
128 Q$=INPUT$(1)
129 IF (Q$="c") OR (Q$="C") GOTO 150
130 IF (Q$="r") OR (Q$="R") GOTO 140
135 PRINT:PRINT "HUH?":PRINT:GOTO 127
140 CHAIN "MENU.BAS",70,ALL

Sets default parameters, and allows them to be changed using a menu-driven system.

150 COLOR 15,14,14:CLS:PRINT:PRINT "If you have just run options 1 or 2 from the main menu,"
151 PRINT "you can pick up your last results for this program."
152 PRINT "Do you want to pick up the new results, or set new defaults (P/D)?";
153 C$=INPUT$(1)
154 IF (C$="p") OR (C$="P") GOTO 157
155 IF (C$="D") OR (C$="d") GOTO 157
156 PRINT:PRINT "HUH?":PRINT:GOTO 153
157 Y=0: R=1.98726E-03
200 REM VARIABLES USED ARE:
202 REM MORE VARIABLES USED ARE: IDEALEMF
205 PRINT:PRINT
207 IF (C$="p") OR (C$="P") GOTO 219
210 TC=1400:FO2OFFSET2=-1:BUFFER$="IW":SPECFO2=-10.69719
215 BUFFER$="IW"
219 TFINAL=TC-100:STEPT%=50
220 COLOR 15,3,3:CLS:PRINT:PRINT "The current conditions are:"t
225 B$$(1)="NO":"B$$(2)="NO":DECIDE$="YES"
230 PRINT " 1. The original sample temperature is ";TC;" degrees C"
250 PRINT " 2. The original sample oxygen fugacity is ";SPECFO2;" log (/atm)"
255 IF (FO2OFFSET2=0) GOTO 265
260 PRINT " --- which is ";BUFFER$;" offset ";FO2OFFSET2;" log units":GOTO 270

A-20
265 PRINT " --- which is "; BUFFERS$  
270 PRINT " 3. Final temperature is "
275 PRINT ";TFINAL;" "degrees C"  
280 PRINT " --- and fO2's will be printed for every ";STEPT%;" "degrees"  
300 PRINT " and 4. The reference buffer is ";BUFFERS$  
320 PRINT:PRINT:PRINT  
380 COLOR 15,9,9:PRINT "Please enter a number: 1 - 4 for changes; 5 to continue;  
6 to exit."::  
390 INPUT " ";CHANGE%  
400 IF (CHANGE%>0) AND (CHANGE%<7) GOTO 420  
410 PRINT:COLOR 31,9,9:PRINT ****:GOTO 380  
420 CLS:ON CHANGE% GOTO 430,580,440,487,1000,140  
430 INPUT "Enter the new sample temperature in degrees C";TC  
431 IF (BUFFER$="IW") GOTO 850  
432 IF (BUFFER$="WM") GOTO 851  
433 IF (BUFFER$="MH") GOTO 852  
434 IF (BUFFER$="QFM") GOTO 853  
435 IF (BUFFER$="NNO") GOTO 854  
436 PRINT "ERROR IN PROGRAM/NO BUFFER":STOP  
440 INPUT "Enter the final temperature in degrees C";TFINAL  
450 INPUT "Enter the interval for which you want fO2's printed 
";STEPT%;CLS:GOTO 220  
460 CLS:PRINT:PRINT:PRINT  
470 PRINT "The sample fO2 data will be given as relative to a buffer, as well"  
480 PRINT "as in log(fO2/atm) units. Your choice of 'reference buffers' are:"::  
487 CLS:PRINT:PRINT:PRINT:PRINT "Your choice of reference buffers are:"  
490 PRINT:PRINT" 1. IW"  
500 PRINT " 2. WM"  
510 PRINT " 3. MH"  
520 PRINT " 4. QFM"  
530 PRINT " 5. NNO"  
535 PRINT "(Your current choice is )";BUFFER$  
540 PRINT:PRINT "Enter the number of your selection."  
550 INPUT " ";CHANGE%  
560 IF (CHANGE%<1) OR (CHANGE%>5) GOTO 460  
570 ON CHANGE% GOTO 571,572,573,574,575  
571 BUFFERS$="IW":GOTO 850  
572 BUFFERS$="WM":GOTO 851  
573 BUFFERS$="MH":GOTO 852  
574 BUFFERS$="QFM":GOTO 853  
575 BUFFERS$="NNO":GOTO 854  
580 CLS  
590 PRINT:PRINT:PRINT  
600 PRINT "The sample furnace fO2 can be expressed as any of three ways:"  
610 PRINT:PRINT " 1. as a standard buffer "  
620 PRINT " 2. as offset from a standard buffer"  
630 PRINT " 3. as a specific fO2 (independent of any buffer) "  
650 PRINT:PRINT "How would you like to express your new fO2"  
660 PRINT " (Enter 1-3, or 4 for unchanged)."  
670 INPUT " ";CHANGE%  
680 IF (CHANGE%<1) OR (CHANGE%>4) GOTO 590  
690 ON CHANGE% GOTO 700,700,820,692  
692 CLS:GOTO 220  
700 PRINT:PRINT "Your choices of standard reference buffers are:";PRINT
Given the initial conditions in the sample furnace, the cooling rate, and the temperature of the reference furnace, the other parameters of interest can be calculated using sub-routines.
1075 FOR l=TC TO TFINAL STEP STEPT%
1078 TREF=i1 :i1=i1+1 :GOSUB 1530
1079 GOSUB 1830
1080 PRINT USING "### +##.## \
#.##";TREF;REFFO2;BUFFER$;FO2OFFSET2
1085 FO2OFFSET2(I1)=FO2OFFSET2:REFFO2(I1)=REFFO2
1090 NEXT I
1100 PRINT:PRINT "Do you wish to Print the results, Continue with this program, "
1101 PRINT " or Return to the main menu? (P/C/R)"
1110 P$= INPUT$ (1)
1120 IF (P$="r") OR (P$="R") GOTO 1360
1130 IF (P$="c") OR (P$="C") GOTO 1350
1135 IF (P$="P") OR (P$="p") GOTO 1150
1140 PRINT:PRINT:PRINT "Huh?":PRINT:GOTO 1100
1150 GOSUB 2000
1200 GOTO 1300
1290 PRINT "The gasmix is unrealistic (eg.,";VOLCO2;"%). Try again."
1300 PRINT:PRINT "Do you wish to continue with this program, "
1301 PRINT " or return to the main menu? (C/R)"
1310 P$= INPUT$ (1)
1320 IF (P$="r") OR (P$="R") GOTO 1360
1330 IF (P$="c") OR (P$="C") GOTO 1350
1340 PRINT:PRINT:PRINT "Huh?":PRINT:GOTO 1300
1350 CLS:GOTO 220
1360 CLS
1365 CHAIN "MENU.BAS",70,ALL
1370 END

Sub-routine for calculating the change in oxygen fugacity with change in temperature for small changes in temperature.

1380 Z = 1
1390 RATIO = FCO2 / (1 - FCO2 - 10^REFFO2)
1400 AA = (1 - 2*RATIO*(100 / MIXRATIO - 1))/(1 + 2*(100 / MIXRATIO - 1))
1410 PART = LOG(1 - AA) - LOG(100 / MIXRATIO - 1)
1420 FOR I= 1 TO- 1 STEP -2
1430 H = I
1440 W -- T + H
1450 GG = 62.110326# - .02144446#*W + 4.720326E-07*(W ^ 2)+(-4.5574288)*(10^-(- 12))*(W ^ 3)- 7.343018200000001*(10^-(- 15))*(W ^ 4)
1460 KK = EXP(- GG /(R*(W + 273.18)))
1470 Q(Z)= KK
1480 Z = Z + 1
1490 NEXT I
1500 NEWFO21 = LOG(10)*.5*(LOG(Q(1))+ PART):NEWFO22 = LOG(10)*.5*(LOG(Q(2))+ PART)
1510 DELEMFDELT =(FN EMF (TREF + 1,NEWFO21) - FN EMF (TREF - 1,NEWFO22))/2
1520 RETURN

Sub-routine to calculate the oxygen fugacity for the reference furnace.

1530 REM SUBROUTINE TO ITERATIVELY CALCULATE THE FO2 OF THE REFERENCE FURNACE

A-23
Sub-routine to calculate the EMF of the zirconia cell, and how it changes with small fluctuations in temperature and oxygen fugacity.

```
1680 REM NOW THAT WE HAVE TEMPERATURE AND LOGFO2, WE NEED TO CALCULATE THE EMF'S AND DELTA-EMF'S
1690 IDEALEMF = FN EMF (TREF,REFFO2)
1700 DELEMFDELFO2 = (FN EMF (TREF,(REFFO2 + .1))- FN EMF (TREF,(REFFO2 -.1)))/2
1740 RETURN
```

Sub-routine for calculating the gas mixture.

```
1750 REM NOW WE WANT TO TAKE THE T, FO2 CONDITIONS & COMPUTE THE %CO2
1760 G1 = 62.110326 + T*(-.02144446)+(T^2)*(4.720326)*(10^-7)+(T^3)*(-4.5574288)*(10^-12)+(T*4)*(-7.343018200000001)*(10^-15)
1770 G2 = 94.25770200000001 + T*(7.321945)*(10^4)-(T^2)*(3.416474)*10^-7-(T^3)*(4.7858617)^*(10^-11)
1780 K1 = EXP(-G1/(R*(T + 273.18))): K2 = EXP(-G2/(R*(T + 273.18))
1790 A = (K1 -(SQR(10ALOGFO2)))*FN RM(K1,LOGFO2))/((K1 + SQR(10^LOGFO2))
1800 FCO2 = 2*(1 - A)/(2 + A + 2*FN RM(K1,LOGFO2))
1810 VOLCO2 = 100/(1 + FN RM(K1,LOGFO2)): DVOLCO2 = 100/(1 + FN RM(K1,LOGFO2 + .1))-(100/(1 + FN RM(K1,LOGFO2-.1)))
1820 RETURN
```

Sub-routine for comparing the oxygen fugacities in the sample and reference furnaces with a standard buffer.

```
1830 REM CALCULATING FO2OFFSET
1831 IF (BUFFERS$="IW") GOTO 1850
1832 IF (BUFFERS$="WM") GOTO 1851
1833 IF (BUFFERS$="MH") GOTO 1852
1834 IF (BUFFERS$="QFM") GOTO 1853
1835 IF (BUFFERS$="NNO") GOTO 1854
1850 FO2OFFSET2 = REFFO2-(6.57 - 27215 /(TREF + 273)):
1851 FO2OFFSET2 = REFFO2-(13.12 - 32730 /(TREF + 273)):
1852 FO2OFFSET2 = REFFO2-(13.966 - 246344 /(TREF + 273)):
1860 RETURN
```

A-24
1853 FO2OFFSET2=REFFO2-(9! - 25738 / (TREF + 273))\GOTO 1860
1854 FO2OFFSET2=REFFO2-(9.359999 - 24930 / (TREF + 273))
1860 RETURN

Sub-routine for printing the results to paper.

2000 REM PRINTING SUBROUTINE
2035 LPRINT LABEL$:LPRINT:LPRINT "(at constant gas mix of ");LPRINT USING
"###.##","MIXRATIO":LPRINT ","LPRINT:LPRINT "T":LPRINT(SPC(5)), "Buffer + offset"
2040 J1=0
2050 FOR J=TC TO TFINAL STEP STEPT%
2060 TPRINT=J:JI=JI+I
2080 LPRINT USING "#### +##.##
+### \\ +###
",TPRINT;REFFO2(J1);BUFFER$;FO2OFFSET2(J1)
2100 NEXT J
2110 IF (PRINTER$="EPSON") GOTO 2125
2120 LPRINT CHR$(12);:GOTO 2127
2125 LPRINT:LPRINT
2127 CLS
2130 RETURN

A-25
Details are given for the design and application of a (one atmosphere) redox-control system. This system differs from that given in NASA Technical Memorandum 58234 in that it uses a single solid-electrolytic cell in a remote location to measure the oxygen fugacities of multiple CO/CO₂ controlled-atmosphere furnaces. This remote measurement (1) extends the range of sample-furnace conditions that can be measured using a solid-electrolytic cell, and (2) cuts costs by extending the life of the sensors and by minimizing the number of sensors in use. The system consists of a reference furnace and an exhaust-gas manifold. The reference furnace is designed according to the redox control system of NASA Technical Memorandum 58234, and any number of CO/CO₂ controlled-atmosphere furnaces can be attached to the exhaust-gas manifold. Using the manifold, the exhaust gas from individual CO/CO₂ controlled-atmosphere furnaces can be diverted through the reference furnace, where a solid-electrolyte cell is used to read the ambient oxygen fugacity. The oxygen fugacity measured in the reference furnace can then be used to calculate the oxygen fugacity in the individual CO/CO₂ controlled-atmosphere furnace. A BASIC computer program was developed to expedite this calculation and is available from the first author.