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# DEVELOPMENT OF WORKING FLUID THERMODYNAMIC PROPERTIES INFORMATION FOR GEOTHERMAL CYCLES - PHASE I

MASTER

Semiannual Report

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# ABSTRACT

During the first half of Phase I of this research program, the following elements of research have been performed: (1) the collection and processing of data for pure components which is 80% complete as to substance, (2) the evaluation of the generalized MBWR equation of state for halogenated hydrocarbon saturated thermodynamic properties, (3) the determination of the pure component parameters of the MBWR for four polar fluids, and (4) the investigation of modifications of the MBWR for improved prediction of properties of polar fluids. A primary conclusion of the Phase I work is that the MBWR will predict properties for polar fluids with small to moderate dipole moments (less than 1.6 Debyes) but needs modification for fluids with larger moments.

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## 1. Summary and Overview

In the first year, the project thrust is development of thermodynamic property correlations for pure component halogenated hydrocarbons, water, and ammonia. This is being accomplished by the acquisition of data and processing to a data bank, the testing of the GMBWR equation of state for fluids using acentric factors determined from vapor pressure, the use of multiproperty analysis for the determination of pure component MBWR equation parameters, and the modification of the MBWR equation for better property prediction. The pure component data collection is 80% complete as to substance. The testing of the GMBWR is complete while other phases are in progress.

## 2. Literature Survey and Data Collection

Data have been collected for and processed into the data base for 30 pure components. This includes five halogenated hydrocarbons, water, ammonia, eighteen hydrocarbons, and five miscellaneous fluids. Data for 13 other halogenated hydrocarbons are being processed into the data base. These 6000 data points already processed include information on density, vapor pressure and enthalpy departure.

## 3. Use of the GMBWR Equation for Halogenated Hydrocarbons

The GMBWR equation was used to predict thermodynamic properties for 18 pure component halogenated hydrocarbons. An optimum acentric factor was determined which could be used in the correlation previously used for light hydrocarbons. The acentric factor was determined from vapor pressure data and then was applied to predict other saturated properties. For 12 components, with dipole moments less than 1.6 Debyes, the overall property average absolute percentage deviation for vapor pressures = 1.6, liquid density = 2.48, and vapor specific volume = 2.38. For other thermodynamic properties, the average absolute deviations were for liquid enthalpy = 1.75 Btu/lbm, vapor enthalpy = 1.21 Btu/lbm, liquid entropy = 0.05 Btu/lbm°R and vapor entropy = 0.05 Btu/lbm°R.

For 6 other halogenated hydrocarbons, the dipole moment was too large for the nonpolar correlation or good tests were not possible because of unreliable or unavailable data. This work is summarized in Appendix A.

## 4. Pure Component MBWR Correlations

The MBWR is being applied to the vapor phase and saturated region for halogenated hydrocarbons, water, and ammonia. The results for halogenated hydrocarbons, R-11, R-12, and R-13 and water are in Appendix B. The density is predicted to less than 1%, vapor pressures to less than 1%, and enthalpy departures to 1.5 Btu/lbm. For water, the results with this desired accuracy were obtainable by restricting the pressure to under 2000 psia. This does not put a severe restriction the correlation's use for many design applications. Work is in progress to determine MBWR parameters for other halogenated hydrocarbons and ammonia.

## 5. Modification of the MBWR for Improved Predictions

The MBWR is not able to represent as wide a range of conditions for a polar fluid such as water as has been possible with hydrocarbons. For water, covering the region from freezing to 2600°R and pressures to 15000 psia, density was predicted to only 2.5%. Plots of isochores and isotherms seem to indicate there is trouble in the representation of the low temperature, high density region. In supporting this, the error in density was decreased to 0.8% by the addition of the term $\rho_r{}^3/T_r{}^3$ to the compressibility equation. Research is continuing on modifications needed for better property predictions.

## 6. Conclusions and Plans

In the first half year significant progress has been made toward the goal of using one method for prediction of the thermodynamic properties of hydrocarbons, halogenated hydrocarbons, water, and ammonia. The feasibility of using the MBWR equation for fluids with small to moderate dipole moments has been shown.

With this established, the determination of parameters for pure components is proceeding. Research is being carried out to improve the prediction for the more polar fluids such as water and ammonia. With the results of this work, the goal of the first year, i.e., the prediction of working fluid thermodynamic properties for geothermal cycles for pure components, will be accomplished.

# APPENDIX A

# Use of a Generalized Modified BWR Equation of State for Halogenated Hydrocarbon Saturated Thermodyanmic Properties

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## ABSTRACT

A generalized modified BWR equation of state developed from normal paraffin hydrocarbon data has been tested for prediction of the thermodynamic properties of eight halogenated hydrocarbons. Average absolute deviations of predicted saturated properties from tabulated values were 1.61% for vapor pressure, 2.49% for liquid density, 2.38% for vapor specific volume, 1.75 Btu/lb for liquid enthalpy, 1.21 Btu/lb for vapor enthalpy, and 0.05 Btu/lb - °R for both liquid and vapor entropy.

## Introduction

A generalized modified BWR equation of state (1) developed primarily for hydrocarbon thermodynamic properties predictions was tested in this study for prediction of halogenated hydrocarbon thermodynamic properties. The major impetus for this study was provided by needs in the geothermal power industry, although other industries also can utilize the results. The MBWR (Modified Benedict-Webb-Rubin) equation is presently being used in the geothermal power industry for convenient prediction of power cycle working fluid properties for hydrocarbons (2). Although the Martin-Hou equation has been used for halogenated hydrocarbons (3), difficulties in its use have been encountered (2,4), probably because the Martin-Hou equation was not originally intended for use in the compressed liquid region at low reduced temperatures. The present study should be considered to be a feasibility study of use of the MBWR equation for halogenated hydrocarbons. Only generalized equation of state parameters have been used herein. Obviously, determination of specific parameter values for each halogenated hydrocarbon would yield considerably improved predictions. The determination of specific MBWR parameters and/or development of an improved equation of state will be carried out in subsequent work.

## The GMBWR Equation

The GMBWR (Generalized Modified Benedict-Webb-Rubin) equation of state, the related thermodynamic functions and methodology for computations are well documented (1) and therefore, only a summary discussion of the GMBWR equation is presented here. The MBWR equation for the absolute pressure, P, is the following function of absolute temperature, T, and molar density, ρ,

$$P = \rho RT + \left(B_o RT - A_o - \frac{C_o}{T} + \frac{D_o}{T^3} - \frac{E_o}{T^4}\right)\rho^2 + \left(bRT - a - \frac{d}{T}\right)\rho^3 + \alpha\left(a + \frac{d}{T}\right)\rho^6 + \frac{c\rho^3}{T^2}\left(1 + \gamma\rho^2\right)\exp(-\gamma\rho^2) \qquad (1)$$

where R is the universal gas constant and $B_o$, $A_o$, etc. are the eleven MBWR equation of state parameters. The GMBWR results when the following generalized relations are used for the MBWR parameters of a substance, where $T_c$, $\rho_c$, and ω are, respectively, the critical temperature, critical density and acentric factor of the substance and $A_j$ and $B_j$ (j = 1, 2, . . . 11) are universal parameters given in Table 1.

$$\rho_c B_o = A_1 + B_1\omega \qquad (2)$$

$$\frac{\rho_c A_o}{RT_c} = A_2 + B_2\omega \qquad (3)$$

$$\frac{\rho_c C_o}{RT_c^{\,2}} = A_3 + B_3\omega \qquad (4)$$

$$\rho_c^2\gamma = A_4 + B_4\omega \qquad (5)$$

$$\rho_c^2 b = A_5 + B_5\omega \qquad (6)$$

$$\frac{\rho_c^2 a}{RT_c} = A_6 + B_6\,\omega \qquad (7)$$

$$\rho_c^2\alpha = A_7 + B_7\omega \qquad (8)$$

$$\frac{\rho_c^2 C}{RT_c^3} = A_8 + B_8\omega \qquad (9)$$

$$\frac{\rho_c D_o}{RT_c^4} = A_9 + B_9\omega \qquad (10)$$

$$\frac{\rho_c^2 d}{RT^2} = A_{10} + B_{10}\omega \qquad (11)$$

$$\frac{\rho_c E_o}{RT_c^5} = A_{11} + B_{11}\omega \quad \exp(-3.8\omega) \qquad (12)$$

Predictions of the thermodynamic properties of a given nonpolar substance (such as a hydrocarbon) using the GMBWR equation require only specification of the critical temperature, $T_c$, the critical density, $\rho_c$, and the acentric factor $\omega$, for the substances, the parameter $\omega$ can be treated as a pseudo acentric factor, that is, as a characterization parameter to be determined from experimental data such as vapor pressure data, rather than requiring that the value of $\omega$ be determined from the defining relation for the acentric factor,

$$\omega = -\log_{10} (P_r^o)_{T_r=0.7} - 1 \qquad (13)$$

where $P_r^o$ is the reduced vapor pressure ($P^o/P_c$) and $T_r$ is the reduced temperature ($T/T_c$).

## GMBWR Characterization Parameters for Halogenated Hydrocarbons

The chemical formulas, molecular weights, critical constants, and boiling points of the twelve halogenated hydrocarbons considered in this study are given in Table 2. All property values used in this work were taken from ASHRAE tabulations (5). Given the values of the critical temperature and critical density in Table 2, the only task to allow use of the GMBWR equation for halogenated hydrocarbons was determination of the pseudo acentric factor for each substance.

To determine the optional value of the pseudo acentric factor for a given fluid, an iterative search method was used in which the trial value of the pseudo acentric factor was varied until the average absolute deviation of predicted vapor pressures from reported values was minimized. Optimal values of the pseudo acentric factor were determined from vapor pressure values for three regions of temperature (1) 0°F and below, (2) 0°F and above, and (3) overall (total range of values). Figure 1 shows the behavior of the average absolute deviation of predicted from experimental vapor pressures for these temperature regions versus trial values of the pseudo acentric factor for trichlorofluoromethane. Optimal values of the pseudo acentric factor determined in this manner are given in Table 3. For most of the fluids studied, the optimal value of the pseudo acentric factor is virtually independent of the temperature range of the vapor pressure data and is within the range of values of acentric factors reported in the literature (6,7) and values obtained using estimation methods (6).

## Comparison of Predicted and Reported Saturated Properties

Evaluation of the capability of the GMBWR equation for predictions of the saturated thermodynamic properties of halogenated hydrocarbons was performed by comparing predicted and reported property values at the predicted vapor pressure at tabulated saturation temperatures. A summary of the results of these comparisons is given in Table 3. The results are very reasonable for most of the fluids studied. It can be noted in Table 3 that the overall average absolute deviations of predicted saturated properties from reported values for the twelve halogenated hydrocarbons were 1.61% for vapor pressure, 2.49% for liquid density, 2.38% for vapor specific volume, 1.75 Btu/lb for liquid enthalpy, 1.21 Btu/lb for vapor enthalpy, and 0.05 Btu/lb-°R for both liquid and vapor entropy. although these deviations are somewhat larger than for hydrocarbons (1), the GMBWR equation is sufficiently accurate for many engineering calculations involving these halogenated hydrocarbons.

## Conclusions

The feasibility of using the GMBWR equation of state for prediction of the thermodynamic properties of halogenated hydrocarbons has been demonstrated. It was found that with the use of the three characterization parameters, critical temperature, critical density, and pseudo acentric factor, the saturated thermodynamic properties of eight halogenated hydrocarbons could be predicted with accuracies sufficient for many engineering calculations. The methods used for determination of the pseudo acentric factor is simple and can be used for other halogenated hydrocarbons (with dipole moments less than 1.6 Debyes).

## Acknowledgment

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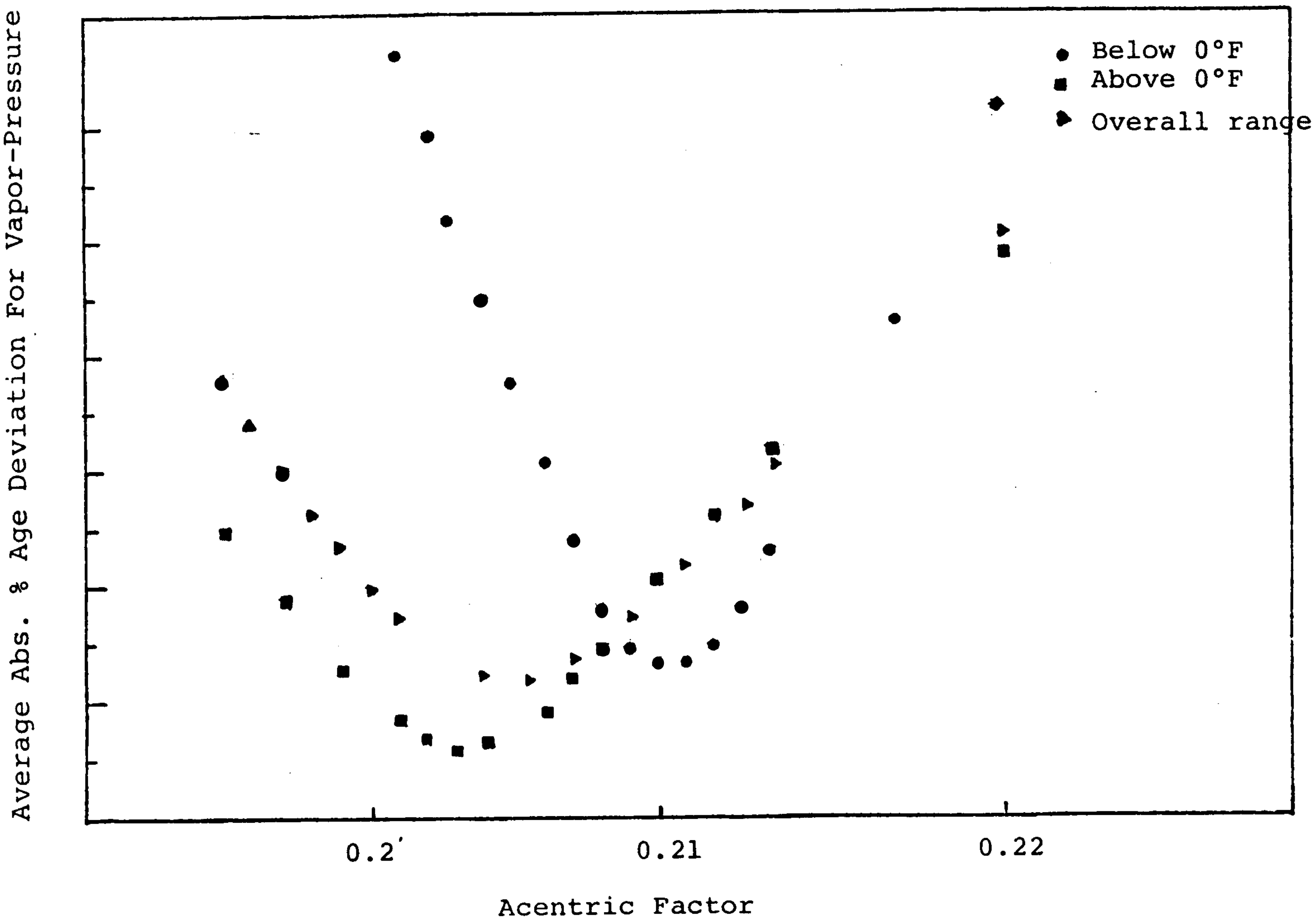

Fig. 1 Optimum Acentric Factor Estimation

Table 1. Values of Generalized Parameters $A_j$ and $B_j$ for Use with Generalized Equation of State.

| Parameter Subscript(j) | Parameter Value | |
|---|---|---|
| | $A_j$ | $B_j$ |
| 1 | 0.443690 | 0.115449 |
| 2 | 1.28438 | -0.920731 |
| 3 | 0.356306 | 1.70871 |
| 4 | 0.544979 | -0.270896 |
| 5 | 0.528629 | 0.349261 |
| 6 | 0.484011 | 0.754130 |
| 7 | 0.0705233 | -0.044448 |
| 8 | 0.504087 | 1.32245 |
| 9 | 0.0307452 | 0.179433 |
| 10 | 0.0732828 | 0.463492 |
| 11 | 0.006450 | -0.022143 |

Table 2. Physical Properties of Halogenated Hydrocarbons (Refrigerants) Included in this Study.

| Refrig. No. | Chemical Name | Chemical Formula | Molecular Weight | Critical Pressure (Psia) | Critical Density (Lb mole/cu.ft) | Critical Temperature (°F) | Boiling Point at 1 atm (°F) |
|---|---|---|---|---|---|---|---|
| 11 | Trichlorofluoromethane | $CC\ell_3F$ | 137.38 | 639.5 | 0.251872 | 388.4 | 74.87 |
| 12 | Dichlorodifluoromethane | $CC\ell_2F_2$ | 120.93 | 596.9 | 0.288127 | 233.6 | -21.62 |
| 13 | Chlorotrifluoromethane | $CC\ell F_3$ | 104.47 | 561.0 | 0.345564 | 83.9 | -114.6 |
| 14 | Carbon Tetrafluoride | $CF_4$ | 88.01 | 543.0 | 0.443842 | -50.2 | -198.3 |
| 22 | Chlorodifluoromethane | $CHC\ell F_2$ | 86.48 | 721.9 | 0.379127 | 204.8 | -41.36 |
| 23 | Trifluoromethane | $CHF_3$ | 70.02 | 701.4 | 0.459217 | 78.1 | -115.7 |
| 113 | Trichlorotrifluoroethane | $CC\ell_2FCC\ell F_2$ | 187.39 | 498.9 | 0.191959 | 417.4 | 117.63 |
| 114 | Dichlorotetrafluoroethane | $CC\ell F_2CC\ell F_2$ | 170.94 | 473.0 | 0.212728 | 294.3 | 38.8 |

## Table 3. Summary of Results of Application of the MBWR Equation for Prediction of Halogenated Hydrocarbon Saturated Properties

| Freon No. | No. of Points | Temp. Range (°F) | Optimum Acentric Factor | P (%) | $\rho_\ell$ (%) | $V_g$ (%) | $h_\ell$ (Btu/lb) | $h_g$ (Btu/lb) | $S_\ell$ (Btu/lb-°R) | $S_g$ (Btu/lb-°R) |
|---|---|---|---|---|---|---|---|---|---|---|
| F-11 | 40 | -85 to 0 | 0.1862 | 1.07 | 0.68 | 1.03 | 0.25 | 0.92 | 0.010 | 0.009 |
| | | 0 to 388.4 | 0.1822 | 0.90 | 1.36 | 2.71 | 0.98 | 1.86 | 0.063 | 0.064 |
| | | -85 to 388.4 | 0.1842 | 1.4 | 1.18 | 2.73 | 0.75 | 1.51 | 0.05 | 0.050 |
| F-12 | 40 | -152 to 0 | 0.178 | 1.67 | 0.65 | 1.89 | 0.40 | 0.43 | 0.024 | 0.020 |
| | | 0 to 230 | 0.162 | 0.60 | 2.90 | 1.07 | 1.95 | 1.51 | 0.047 | 0.048 |
| | | -152 to 230 | 0.176 | 1.89 | 1.79 | 2.76 | 0.98 | 0.46 | 0.037 | 0.035 |
| F-13 | 34 | -200 to 0 | 0.169 | 1.62 | 0.60 | 1.81 | 0.66 | 0.16 | 0.036 | 0.030 |
| | | 0 to 83.93 | 0.165 | 0.53 | 4.85 | 3.37 | 1.87 | 0.82 | 0.026 | 0.028 |
| | | -200 to 83.93 | 0.169 | 1.26 | 2.92 | 2.43 | 1.09 | 0.34 | 0.032 | 0.029 |
| F-14 | 19 | -230 to -52 | 0.170 | 1.11 | 3.69 | 2.16 | 1.42 | 0.97 | 0.093 | 0.096 |
| F-22 | 37 | -150 to 0 | 0.2260 | 0.26 | 0.65 | 0.69 | 0.35 | 0.21 | 0.041 | 0.040 |
| | | 0 to 204.81 | 0.2270 | 0.97 | 1.53 | 1.98 | 1.11 | 1.00 | 0.064 | 0.062 |
| | | -150 to 204.8 | 0.2254 | 0.74 | 1.88 | 1.97 | 1.68 | 1.13 | 0.067 | 0.066 |
| F-23 | 28 | -190 to 0 | 0.264 | 1.07 | 1.89 | 1.13 | 2.10 | 1.60 | 0.046 | 0.042 |
| | | 0 to 78.66 | 0.278 | 1.74 | 2.25 | 2.11 | 3.94 | 2.11 | 0.025 | 0.023 |
| | | -190 to 78.66 | 0.264 | 1.45 | 3.16 | 3.55 | 4.80 | 3.49 | 0.040 | 0.037 |
| F-113 | 27 | -30 to 0 | 0.256 | 0.31 | 0.32 | 0.37 | 0.10 | 0.92 | 0.009 | 0.010 |
| | | 0 to 417.4 | 0.250 | 1.68 | 1.55 | 2.87 | 1.24 | 2.19 | 0.049 | 0.051 |
| | | -30 to 417.4 | 0.250 | 1.92 | 1.35 | 2.93 | 1.07 | 2.06 | 0.043 | 0.045 |
| F-114 | 42 | -135 to 0 | 0.2495 | 0.39 | 0.70 | 0.24 | 0.18 | 0.41 | 0.017 | 0.014 |
| | | 0 to 290 | 0.2455 | 0.90 | 2.21 | 1.39 | 0.92 | 1.50 | 0.042 | 0.044 |
| | | -135 to 290 | 0.2495 | 1.10 | 1.69 | 1.83 | 0.62 | 0.93 | 0.033 | 0.033 |
| OVERALL | | | | 1.61 | 2.48 | 2.37 | 1.74 | 1.21 | 0.05 | 0.05 |

# APPENDIX B

# Parameters for the MBWR Equation for Four Polar Fluids

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## ABSTRACT

Parameters for the MBWR equation of state, which was developed from normal paraffin hydrocarbon data, have been determined for four polar fluids; water and the halogenated hydrocarbons, R-11, R-12, and R-13. Average absolute deviations from data were within 1% for densities, within 1% for vapor pressures, and with 1.5 Btu/lbm for enthalpy departures.

## Introduction

The MBWR (modified Benedict-Webb-Rubin) equation of state, used previously for hydrocarbons [5] has been applied to the vapor phase and saturated liquid for four polar fluids; water, R-11, R-12, and R-13. The parameters were determined by multiproperty analysis of density, enthalpy departure, and vapor pressure data. The results represent the behavior of the fluids with accuracy acceptable for engineering calculations in the region of prime interest in geothermal energy conversion processes.

## Data

The physical constants for the fluids are given in Table I. The number of points, the range of the variables and the data references are given in Table II.

## Equation of State

The reduced form of the MBWR equation used is

$$\begin{aligned} Z = 1 &+ (A_1 - A_2/T_r - A_3/T_r^3 + A_9/T_r^4 - A_{11}/T_r^5)\rho_r \\ &+ (A_5 - A_6/T_r - A_{10}/T_r^2)\rho_r^2 + A_7(A_6/T_r + A_{10}/T_r^2)\rho_r^5 \\ &+ A_8\rho_r^2/T_r^3(1 + A_4\rho_r^2)\exp(-A_4\rho_r^2) \end{aligned}$$

where Z is the compressibility and the reduced variables are $T_r = T/T_c$, and $\rho_r = \rho/\rho_c$. The parameters determined from the regression are given in Table III.

## Discussion of Results

The results, which are summarized in Table II, show that the

MBWR equation can be used to predict the properties of some polar fluids in the vapor phase and liquid phase. It is able to predict properties for fluids with low dipole moments, such as the halogenated hydrocarbons shown, with good accuracy. For fluids with stronger dipole moments such as water, by restricting the range of the variables, good results can also be obtained. For water, restricting the pressure to under 2000 psia does not put a severe limitation on the equation for use in design work in geothermal energy conversion applications. Typically the MBWR equation can predict densities within 1%, vapor pressures within 1%, and enthalpies departures within 1.5 Btu/lbm.

## TABLE I

### Physical Constants of Fluids in This Study

| Substance | Formula | Molecular Weight | Critical Temperature (°R) | Critical Pressure (psia) | Critical Density (lbmole/ft$^3$) |
|---|---|---|---|---|---|
| Water | $H_2O$ | 18.015 | 1165.3 | 3208. | 1.1102 |
| R-11 | $CCl_3F$ | 137.38 | 848.07 | 639.5 | 0.251872 |
| R-12 | $CCl_2F_2$ | 120.93 | 693.57 | 596.9 | 0.288127 |
| R-13 | $CClF_3$ | 104.47 | 543.57 | 561.0 | 0.345564 |

# TABLE II

## Summary of Results of Regression

| Substance | Temperature Range (°R) | Pressure Range (psia) | Data Type | Number Points | A.A.D.* | Data Reference |
|---|---|---|---|---|---|---|
| $H_2O$ | $491<T<2751$ | $.089<P<2476.8$ | $\rho$ | 260 | 0.57% | 3,4 |
| | | | ΔH | 260 | 1.39 Btu/lbm** | 2 |
| | | | VP | 60 | 0.51% | 2 |
| R-11 | $375<T<930$ | $.13<P<639.5$ | $\rho$ | 138 | 0.19% | 1 |
| | | | ΔH | 57 | 0.22 Btu/lbm | 1 |
| | | | VP | 25 | 0.11% | 1 |
| R-12 | $300<T<920$ | $0.14<P<440$ | $\rho$ | 128 | 0.44% | 1 |
| | | | ΔH | 113 | 0.19 Btu/lbm | 1 |
| | | | VP | 39 | 1.04% | 1 |
| R-13 | $260<T<920$ | $0.5<P<535$ | $\rho$ | 128 | 0.27% | 1 |
| | | | ΔH | 102 | 0.17 Btu/lbm | 1 |
| | | | VP | 33 | 0.73% | 1 |

*A.A.D. = Average absolute deviation = $\Sigma\ |\text{dev.}|\ /N$

**For vapor phase, 0.83 Btu/lbm; for liquid phase, 0.35%.

# TABLE III

## Parameters for the MBWR Equation for Four Polar Fluids

| Reduced Parameter | Reducing Equation | Reduced Parameter Value | | | |
|---|---|---|---|---|---|
| | | Water | R-11 | R-12 | R-13 |
| $A_1$ | $B_o\rho_c$ | 0.505447 | 0.522637 | 0.460523 | 0.421120 |
| $A_2$ | $\rho_c A_o/RT_c$ | 1.10376 | 1.12761 | 1.11758 | 1.03330 |
| $A_3$ | $\rho_c C_o/RT_c^3$ | 0.802531 | 0.675384 | 0.660287 | 0.641215 |
| $A_4$ | $\rho_c\gamma$ | 0.273071 | 0.474721 | 0.496065 | 0.455649 |
| $A_5$ | $\rho_c^2 b$ | 0.257467 | 0.564544 | 0.590491 | 0.649375 |
| $A_6$ | $\rho_c^2 a/RT_c$ | 0.0507135 | 0.647511 | 0.622282 | 0.737947 |
| $A_7$ | $\rho_c^3\alpha$ | 0.0490011 | 0.0619089 | 0.0630347 | 0.0561039 |
| $A_8$ | $\rho_c C/RT_c^3$ | 0.683023 | 0.714059 | 0.739536 | 0.722251 |
| $A_9$ | $\rho_c^2 D_o/RT_c^4$ | 0.0382764 | 0.0640857 | 0.0611971 | 0.0415828 |
| $A_{10}$ | $\rho_c^2 d/RT_c^2$ | 0.333881 | 0.113973 | 0.148282 | 0.140216 |
| $A_{11}$ | $\rho_c E_o/RT_c^5$ | 0.00108719 | 0.00389269 | 0.00464252 | 0.00192894 |

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# APPENDIX C

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