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THE QUARTZ- AND Na/K GEOTHERMOMETERS. I. NEW THERMODYNAMIC CALIBRATION

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ABSTRACT

New temperature equations are proposed for the quartz and Na/K geothermometers. They are based on revision of experimental data on quartz solubility in water and of the thermodynamic properties of albite and K-feldspar, respectively. The equations are:

$$t°C = -55.3 + 0.3659 \cdot S - 5.3954 \cdot 10^{-4} \cdot S^2 + 5.5132 \cdot 10^{-7} \cdot S^3 + 74.360 \cdot \log S$$

and

$$t°C = 733.6 - 770.551 \cdot Y + 378.189 \cdot Y^2 - 95.753 \cdot Y^3 + 9.544 \cdot Y^4$$

where S stands for unionized aqueous silica concentrations as SiO_2 in ppm and Y for the molal Na^+/K^+ activity ratio, respectively. This ratio can be taken to be equal to the Na/K concentration ratio for dilute to moderately saline waters below 300°C. For saline waters and waters above 300°C activity ratios, which can be retrieved from the analytical data with the aid of aqueous speciation programs, should be used. Estimation of quartz equilibrium temperatures from analysed silica concentrations in water samples from well discharges with "excess" enthalpy requires information about the processes that give rise to the "excess" enthalpy.

1. INTRODUCTION

The first geothermometers developed to estimate underground temperatures in geothermal systems from data on the chemical composition of thermal spring waters and shallow drillholes discharges were the quartz and the Na/K geothermometers (Fournier and Rowe, 1966; Mahon, 1966; Truesdell, 1976). Since originally developed, the calibration of the quartz geothermometer has only been slightly revised whereas many geochemical calibrations have been proposed for the Na/K geothermometer (see e.g. Fournier, 1991). A new calibration, which rests on thermdynamic data, is proposed here for both of these geothermometers. In the case of quartz, it is based on revision of experimental data on quartz solubility by Gunnarsson and Arnórsson (2000). For the Na/K geothermometer it is based on review of experimental solution calorimetric data on albite and microcline (the stable K-feldspar at low temperatures) by Arnórsson and Stefánsson (1999). Today, these geothermometers are not only used to estimate underground temperatures in geothermal systems but also, and probably more importantly, to monitor the response of exploited geothermal reservoirs to the production load with respect to cooling and recharge. During prolonged production from geothermal reservoirs, various processes come into play that can affect the extent to which the aquifer water vaporizes from undisturbed pressure conditions to wellhead. These processes affect the relation beween the silica concentration in the aquifer water and in a water sample collected at a particular pressure at the surface. To estimate aquifer temperatures by the quartz geothermometer, these boiling processes must be assessed. On the other hand, solute concentration ratios, like Na/K ratios, are not affected by these processes and, therefore, not calculated Na/K geothermometry temperatures.

2. NEW CALIBRATIONS

Gunnarsson and Arnórsson (2000) revised experimental data on quartz solubility in water (Fig. 1). According to them, the temperature dependence of quartz solubility for the reaction

$$SiO_{2\,aq} + 2H_2O \rightleftharpoons H_4SiO_4^\circ \quad (1)$$

is:

$$\log(SiO_2) = -34.188 + 197.47/T - 5.851 \cdot 10^{-6} \cdot T^2 + 12.245 \cdot \log T \quad (2)$$

where SiO_2 stands for unionized silica concentration ($H_4SiO_4^\circ$) in moles/kg H_2O. This relationship between quartz solubility (aqueous silica concentration) and temperature is accurately described numerically by:

$$t°C = -55.3 + 0.3659 \cdot S - 5.3954 \cdot 10^{-4} \cdot S^2 + 5.5132 \cdot 10^{-7} \cdot S^3 + 74.360 \cdot \log S \quad (3)$$

In equation (3) S stands for unionized aqueous silica concentrations as SiO_2 in ppm. Both equations are valid in the range 0-350°C at 1 bar below 100°C and at vapour saturation pressures at higher temperatures. In terms of temperature the difference between the two equations is ≤1.6°C except above 340°C where it is larger and increases with rising temperature being 5°C at 350°C. In the range 150-270°C equation (3) yields temperatures that differ by less than 10°C from from the equation given Fournier and Potter (1982) (Fig. 2), which has generally been used for geothermometry purposes to date. The difference is larger below 150°C and above 270°C reaching a maximum of 35°C at 350°C (Fig. 2).

Very many geochemical calibrations have been proposed for the Na/K geothermometer (see e.g. Fournier, 1991, Fig. 3). They are based on correlation between Na/K

concentration ratios and aquifer temperature in wells. Only one thermodynamic calibration has been proposed so far (Giggenbach, 1988). It is based on simultaneous equilibrium between solution, pure low-albite ($NaAlSi_3O_8$) and pure microcline ($KAlSi_3O_8$, the stable K-feldspar at low temperatures) according to the reaction:

$$NaAlSi_3O_8 + K^+ = KAlSi_3O_8 + Na^+ \qquad (4)$$

Both feldspars are taken to be fully ordered with respect to Al and Si. The equilibrium constant for this reaction is equal to the Na^+/K^+ activity ratio. Thus, strictly speaking, the thermodynamic calibration requires the use of Na^+/K^+ activity ratios, rather than aqueous Na/K concentration ratios. However, for waters of low to moderate salinity (ionic strength <0.1-0.2) and below 300°C it is a good approximation to take analysed Na/K concentration ratios to be equal to Na^+/K^+ activity ratios when calculating Na/K temperatures. With increasing temperature above 300°C and increasing water salinity these two ratios depart progressively more and more due to different degree of complexing of the Na^+ and K^+ ions, in particular with Cl^- to form $NaCl^\circ$ and KCl° ion pairs and differences in the values taken by the activity coefficients for Na^+ and K^+ as salinity and temperature increase.

In retrieving a geothermometry equation based on the reaction according to equation (4) above, Giggenbach (1988) used the thermodynamic data selected by Bowers et al. (1984) and chose an equation of the form of the v'ant Hoff equation (the logarithm of the equilibrium constant varies linearly with the reciprocal of the absolute temperature) to describe the temperature dependence of the Na^+/K^+ activity ratio, i.e. the standard heat capacity of the reaction is taken to be zero. The new thermodynamic calibration proposed here is consistent with the reaction according to equation (4) above. It is based on the assessment of Arnórsson and Stefánsson (1999) on the thermodynamic properties of low-albite and microcline and the thermodynamic properties of Na^+ and K^+ as given by Shock and Helgeson (1988). According to Arnórsson and Stefánsson (1999) the temperature variation in the $\log(Na^+/K^+)$ activity ratio for the alkali feldspar/solution equilibrium is:

$$\log(Na/K) = -17.673 - 6{,}427/T^2 + 2{,}083.35/T - 0.811\cdot10^{-6}\cdot T^2 + 5.482\cdot\log T \qquad (5)$$

where T is in K and Na/K represents the activity ratio of Na^+ over K^+. This equation yields a log(Na/K)-temperature curve that lies slightly below calculated Na^+/K^+ activity ratios for selected drillhole discharges with aquifer temperatures above 200°C. It is based on expermental data of Waldbaum and Robie (1971) on the difference in the heat of solution between low-albite and microcline. By selecing the difference, as given by Hovis (1988), a better match is obtained with the drillhole data, leading to the following equation:

$$\log(Na/K) = -17.673 - 6{,}427/T^2 + 2{,}109.46/T - 0.811\cdot10^{-6}\cdot T^2 + 5.482\cdot\log T \qquad (6)$$

The results of Waldbaum and Robie (1971) and Hovis (1988) correspond to an enthalpy difference of 500 J/mole. It is anticipated that this difference is well within the limit of measurement error of the calorimetric experiments used to calculate the standard Gibbs energy of formation values for both feldspars and the equilibrium constant (equal to the Na^+/K^+ activity ratio) according to the reaction of equation (4) above. The selection of the standard enthalpy value sets the value for the standard Gibbs energy for the reaction at a particular temperature. Variation in the standard Gibbs energy of the reaction with temperature, which is obtained from standard heat capacity data, is, on the other hand, quite accurate.

The K-feldspar reported as hydrothermal mineral in active geothermal systems is adularia. The heat of solution experiments carried out by Waldbaum and Robie (1971) and Hovis (1988) indicate that the standard enthalpy of formation from the elements of adularia and microcline are very similar and, at a given temperature, yield practically the same Na^+/K^+ activity ratios at equilibrium with low-albite and solution.

The following relationship between temperature and Na^+/K^+ activity ratios yields results which are numerically very similar to those obtained from equation (6):

$$t°C = 733.6 - 770.551\cdot Y + 378.189\cdot Y^2 - 95.753\cdot Y^3 + 9.544\cdot Y^4 \qquad (7)$$

Y represents the logarithm of the activity ratio of Na^+ over K^+ in moles/kg H_2O. In terms of temperature the difference between equations (6) and (7) is <1°C except at 0° and >340°C where it reaches a maximum of 1.4°C. A plot of $\log(Na^+/K^+)$ against 1/T according to equation (7) does not yield a straight line except in the range 25-175°C (Fig. 4). Therefore, a geothermometry equation of the form of the v'ant Hoff equation does not describe the thermodynamic data as accurately as equation (7).

The thermodynamic calibration of the Na/K geothermometer, proposed here (equation (7)), is rather similar to the geochemical calibration of this geothermometer as given by Fournier (1979). At 0°C the difference is 5°C, increasing to 16°C at 180°C but decreasing again to 1°C at 350°C (Fig. 5). Except below 15°C, the calibration of Fournier (1979) always yields higher Na/K-temperatures for any particular aqueous Na/K ratio than the thermodynamically derived equation presented here. Other geothermometry equations show similar or larger differences (Fig. 5).

3. CALCULATION OF QUARTZ EQUILIBRIUM TEMPERATURES

Although silica forms various aqueous species, the $H_4SiO_4^\circ$ species is predominant in all geothermal waters at

temperatures in excess of about 180°C. For this reason it is a satisfactory approximation to take analysed silica concentration to be equal to $H_4SiO_4^o$. It is customary to represent $H_4SiO_4^o$ as $SiO_{2,aq}$. This has been done in the present study.

Samples of water from wet-steam wells are often collected after the water has flashed to atmospheric pressure. Subsequently, the temperature corresponding to equilibrium with quartz - the quartz equilibrium temperature - is calculated from the analysed silica concentration in the water sample by assuming adiabatic boiling from the quartz equilibrium temperature to atmospheric pressure. This is indeed a good approximation, if the fluid can be treated, in thermodynamic terms, as an isolated system, which can be assumed to be valid for wells with liquid enthalpy (the enthalpy of the well discharge is the same as that of liquid water at the aquifer temperature). It may also be valid for "excess" enthalpy wells, if the "excess" enthalpy is due to the presence of steam in the aquifer. The term "exess" enthalpy is used here according to common practice. It means that the enthalpy of a well discharge is in excess of that of steam saturated water at the respective aquifer temperature. In other words, the steam to water ratio in the discharge at any particular pressure is higher than can be accounted for by depressurization of the aquifer water.

If the fluid cannot be regarded as an isolated system, but either as closed (heat is not conserved) or open (neither heat nor mass are conserved), it is not valid to take the boiling to be adiabatic. By assuming boiling to be adiabatic, when it is not, may lead to considerable error in the calculated quartz equilibrium temperature, particularily, when the discharge enthalpy is approaching that of dry steam. In a closed system the "excess" enthalpy would be produced by conductive heat flow from the aquifer rock to the fluid, but in the case of an open system, by segregation of the flowing water and steam in the aquifer.

When boiling is adiabatic and the discharge enthalpy equals that of liquid water at the aquifer temperature, the concentration of silica in the aquifer water is related to the silica concentration in a sample collected at the surface by:

$$SiO_{2,aq} = (1 - \frac{h_d - h_s}{L_s})SiO_{2,s} \qquad (8)$$

$SiO_{2,aq}$ and $SiO_{2,s}$ in equation (8) designate silica concentrations in the aquifer water and in a water sample collected at the surface at pressure s, respectively. h_d is the enthalpy of steam saturated water at the respective aquifer temperature, and h_s and L_s the enthalpy of steam saturated water at the sampling pressure and its latent heat of vaporization, respectively. To calculate $SiO_{2,aq}$ knowledge of the aquifer temperature is required. The following equation, which is consistent with equation (1), can be used to calculate the quartz equilibrium temperature, if the sample is of water that had boiled adiabatically to atmospheric pressure (100°C):

$$t°C = -66.9 + 0.1378 \cdot S_1 - 4.9727 \cdot 10^{-5} \cdot S_1^2 + 1.0468 \cdot 10^{-8} \cdot S_1^3 + 87.841 \cdot \log S_1 \qquad (9)$$

i.e. equation (9) represents silica concentrations (S_1) as SiO_2 in ppm in water initially in equilibrium with quartz after adiabatic boiling to 100°C.

If aquifer steam is present and the boiling is adiabatic, the silica concentration in the reservoir water can be obtained from:

$$SiO_{2,aq} = SiO_{2,s} \frac{1 - X_s}{1 - Y} \qquad (10)$$

where X_s and Y represent the steam fractions in the discharge at the sampling pressure and in the aquifer, respectively, given by:

$$X_s = \frac{h_t - h_s}{L_s} \qquad (11)$$

and

$$Y = \frac{h_t - h_d}{L_d} \qquad (12)$$

where h_t represents the discharge enthalpy (in this case equal to the aquifer fluid enthalpy) and L_d the latent heat of vaporization of the aquifer water. Other symbols have the same signature as in equations (8) and (10). Having calculated $SiO_{2,aq}$ the quartz equilibrium temperature can be obtained from equation (3). To make equation (12) consistent with the derived quartz equilibrium temperature an iterative calculation procedure is required involving estimation of the aquifer temperature until it is consistent with equations (3), (12) and (10).

There are two possibilities (models) to calculate the aquifer silica concentrations, and therefore the quartz equilibrium temperatures, for "excess" enthalpy wells, when the "excess" enthalpy is not due to the presence of aquifer steam. The first assumes that the total well discharge is representative of the aquifer water (closed system). By this model, the silica concentration in the aquifer water is the same as that of the total discharge. From measurement of discharge enthalpy and anaysed silica concentration in a water sample ($SiO_{2,s}$), the silica concentration of the aquifer water can be obtained from

$$SiO_{2,aq} = (1 - \frac{h_t - h_s}{L_s})SiO_{2,s} \qquad (13)$$

Subsequently, the quartz equilibrium temperature can be derived from equation (3). Symbols in equation (13) have the same signature as in equations (10) and (11).

The second model that can be used to estimate quartz equilibrium temperatures in the producing aquifer of

"excess" enthalpy wells assumes that the system is open. A special case of an open system is one in which heat transfer is linked to mass transfer, i.e. there is no heat transfer by conduction. By this model "excess" discharge enthalpy can be produced by separating partly the steam and water that flow towards the well. Calculation of quartz equilibrium temperatures by this model is summarized in Table 1. The extent to which the water vaporizes by depressurization by this model depends on the temperature at which segregation occurred because the enthalpy of saturated steam varies with temperature (Fig. 6). This enthalpy is about constant in the range 180-270°C (equivalent to 10-55 bars abs. vapour pressure) but at higher and lower temperatures it falls. The steam to water ratio of geothermal fluid flowing through aquifers and wells affects little the vaporization of the water in response to pressure drop in the range 10-55 bars abs. because the steam enthalpy is about constant in this range but at higher and lower pressures it does. Specifically, below 10 bars abs. vapour pressure "excess" steam enhances the vaporization of the water as pressure drops.

4. DISCUSSION

The calibration of the quartz geothermometer proposed here does not differ significantly from that recommended by Fournier and Potter (1982) at temperatures below 300°C. The difference is, however, significant at higher temperatures. Field data do not provide evidence which of the two calibrations describes more accurately quartz solubility above 300°C. Na/K ratios according to the present, and other Na/K-geothermometry calibration curves, vary little with temperature above 200°C and especially above 300°C. Thus, this ratio only decreases by 38% in going from 300° to 350°C. For this reason it is not satisfactorily accurate to use directly analytical data on total sodium and total potassium to calculate Na/K-geothermometry temperatures, when above about 300°C, in particular for waters of high saliniity. It is necessary to use activity ratios of the respective ions. Ion pairing, especially with Cl^- as well as the values taken by the activity coefficicents of Na^+ and K^+ cause Na^+/K^+ activity ratios to depart significantly from Na/K concentration ratios. Reliable estimation of Na/K temperatures, when above about 300°C, also requires precise analysis of Na and K.

Quartz equilibrium temperatures are frequently lower than Na/K temperatures, both for hot spring and well discharges. This has been attributed to faster re-equilibration with quartz, as compared to the alkali-feldspars during cooling of the water either in the upflow or in the depressurization zone around discharging wells. It is, however, observed that Na/K-temperatures are sometimes lower than quartz equilibrium temperatures, both for hot spring and well discharges. Geochemical monitoring studies of producing wet-steam well discharges have also indicated that quartz- and Na/K-temperature frequently decrease with time and by about the same amount. Discrepancy between the quartz- and Na/K geothermometers depends on how closely equilibrium has been approached between solution and quartz, on one hand, and albite and K-feldspar, on the other. Such discrepancy also depends on the accuracy by which these two geothermometers are calibrated, analytical precision, as well as boiling processes in the case of "excess" enthalpy wells. Further, the discrepancy depends on whether the fluid discharged is mixed or not. Permeability in most geothermal systems is little doubt anisotropic. When this is the case, the fluid entering a discharging well or issuing from a hot spring is inevitably a mixture of fluids that have travelled different distances from their source areas to the point of well inflow. If all these source areas have similar temperature, which is the case if temperature gradients are small across relatively large parts of the system, the discharged fluid can be regarded as being unmixed if local equilibrium has been attained in all of the source areas.

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TABLE 1. Calculation of quartz equilibrium temperature for "excess" enthalpy well discharges.

1. Sample collected from the weirbox or a Webre separator downstream from a wellhead steam separator with pressures < 10 bars abs.

A. Calculate the silica concentration in the water of the steam separator from:

$$SiO_{2,sep} = (1 - \frac{h_{sep}-h_s}{L_s})SiO_{2,s} \qquad (1.1)$$

where $SiO_{2,sep}$ and h_{sep} stand for the silica concentration in the wellhead steam separator water and its enthalpy of vaporization, respectively. See equation (6) for other symbols.

B. Calculate the silica concentration in steam saturated water at 10 bars abs. vapour pressure (180°C) with the aid of

$$SiO_{2,10ba}(1 - \frac{h_t-h_{10ba}}{L_{10ba}}) = (1 - \frac{h_t-h_{sep}}{L_{sep}})SiO_{2,sep} \qquad (1.2)$$

where the 10ba subscript designates the 10 bar abs. vapour pressure, h_t discharge enthalpy, and h_{sep} and L_{sep} the enthalpy of steam saturated water and its latent heat of vaporization at the pressure in the steam separator.

C. Calculate the quartz equilibrium temperature from

$$t°C = -143.5 - 0.0136 \cdot S_{10} + 1.1659 \cdot 10^{-4} \cdot S_{10}^2 - 5.2013 \cdot 10^{-8} \cdot S_{10}^3 + 136.954 \cdot \log S_{10} \qquad (1.3)$$

where S_{10} represents the silica concentration in steam saturated water at 10 bars abs. vapour pressure. Equation (1.3) decribes silica concentrations in water initially in equilibrium with quartz after adiabatic boiling to 180°C (10 bars abs. vapour pressure).

2. Water sample collected from a steam separator, which has a lower pressure than 10 bars abs.

A. Calculate silica concentrations in water at 10 bar abs. steam pressure with the aid of equation (1.2)

B. Insert the silica concentration so calculated into equation (1.3) to obtain a value for the quartz equilibrium temperature.

3. Water sample collected at pressure above 10 bars abs.

A. Calculate $SiO_{2,10ba}$ using equation (1.2) and insert the value so obtained into equation (1.3) to get a value for the quartz equilibrium temperature. In this case the subscript sep in equation (1.2) stands for sampling pressure.

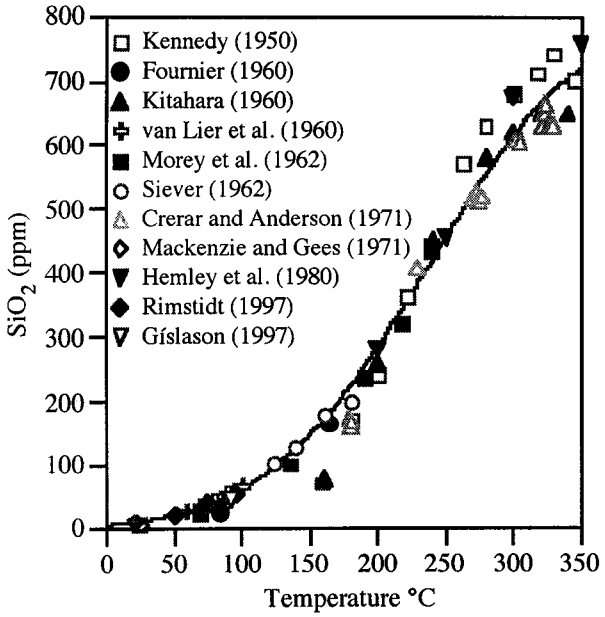


Figure 1. Experimental data on the solubility of quartz in pure water. From Gunnarsson and Arnórsson (2000).

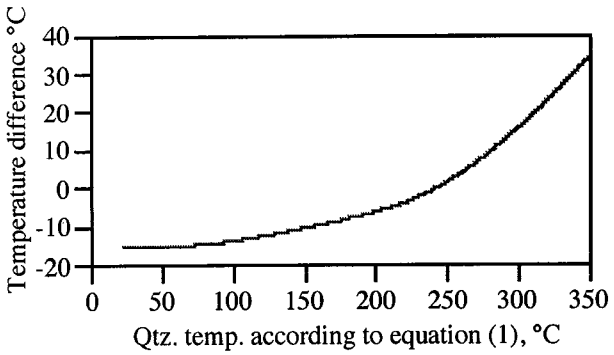


Figure 2. Difference in temperature calculated from a certain aqueous silica concentration using equation (1) and the quartz solubility equation of Fournier and Potter (1982). Negative difference indicates that equation (1) yields a lower temperature and vica versa.

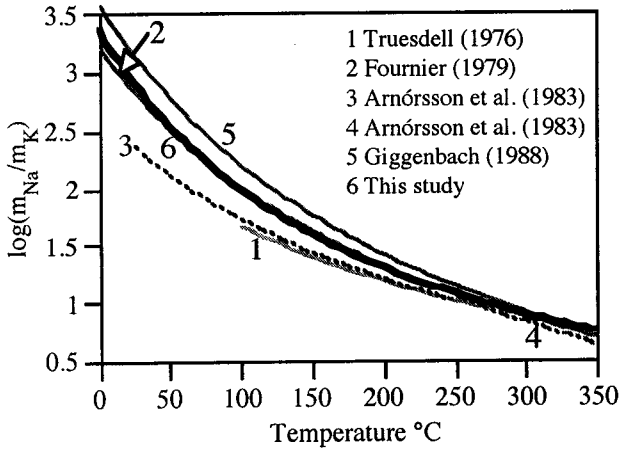


Figure 3. Some proposed Na/K geothermometry curves.

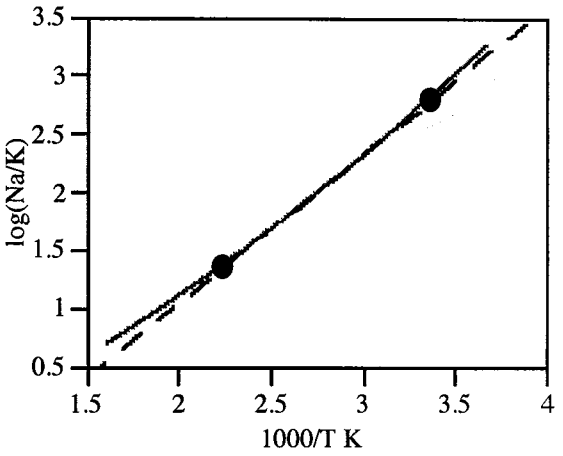


Figure 4. Na and K ion activity ratios according to equation (7) (solid curve) versus 1000/T Kelvin. It is seen that log(Na/K) varies only linearly (straight broken line) with the reciprocal of the absolute temperature in the range 25-175°C (marked by the dots).

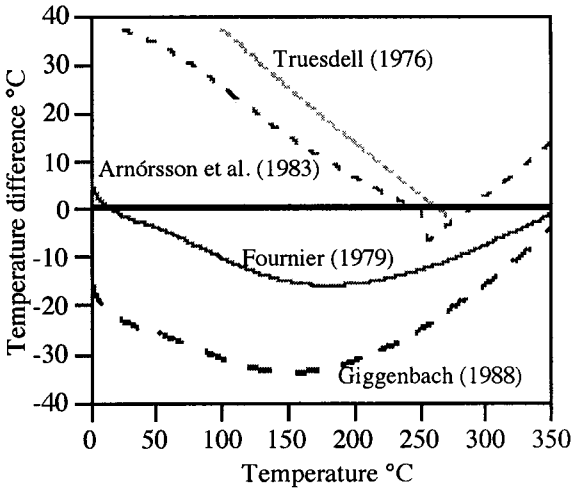


Figure 5. Difference in temperature as calculated from a given aqueous Na/K ratio using equation (7) and various other Na/K-geothermometry equations as indicated. Negative difference indicates that equation (7) yields lower temperature and vica versa.

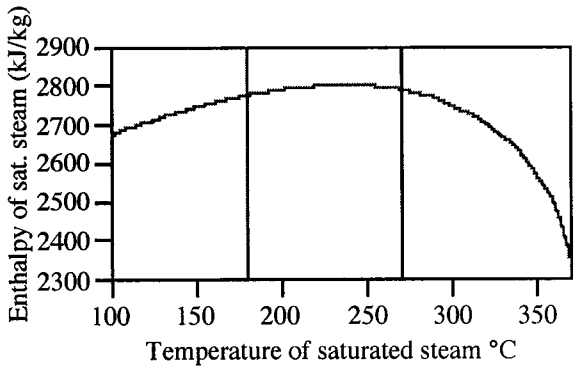


Figure 6. Enthalpy of saturated steam versus temperature.