

STANDARD FREE ENERGIES OF FORMATION  
CALCULATED FROM DISSOLUTION DATA USING  
SPECIFIC MINERAL ANALYSES

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ABSTRACT

Standard free energies of formation ( $\Delta G_f^\circ$ ) for five primary rock-forming silicate minerals were calculated from their dissolution in aqueous solutions, using (1) the actual mineral formulas derived from the chemical analyses of the specific mineral samples, and (2) the ideal structural formulas of the minerals.  $\Delta G_f^\circ$  (kcal/mole) is:

	<i>Actual Formula</i>	<i>Ideal Formula</i>
Olivine	-461.6	-457.8
Augite	-679.4	
Labradorite	-932.4	-931.1
Microcline	-887.3	-892.6
Muscovite	-1399.9	-1313.8

The standard free energy of formation for  $\text{Fe}(\text{OH})^+$  was also calculated to be -52.58 kcal/mole.

INTRODUCTION

Standard free energies of formation for minerals ( $\Delta G_f^\circ$ ) have been obtained (1) by theoretical calculation from mineral reactions (see Raymahashay, 1968), (2) from calorimetric data (see King *et al.*, 1967), or (3) by calculation from aqueous solubility data (see Reesman and Keller, 1965; Kittrick, 1970). In most cases, however,  $\Delta G_f^\circ$  was calculated on the basis of ideal structural formulas of the minerals rather than the actual mineral formulas as derived from chemical analyses of the specific mineral specimens. Since substitution of ions in crystal structures commonly occurs in minerals, the mineral compositions usually do not conform to ideality. Therefore, free energy data based on ideal formulas may be in error, and misleading when applied to stability diagrams of minerals. The purpose of this paper is (1) to calculate from aqueous solubility data standard free energies of formation for minerals of specific composition from bulk chemical analyses, and (2) to compare the difference in  $\Delta G_f^\circ$  for a mineral calculated from ideal and specific structural formulas.

LABORATORY DISSOLUTION

Freshly fractured fragments of olivine, augite, labradorite, microcline, and muscovite were equilibrated (or at least to an essentially steady state) at room temperature in deionized water for periods of up to 21 days. The detailed experi-

mental procedure and results were given in the paper by Huang and Keller (1970). The results of the laboratory dissolution show that framework cations from minerals were dissolved rapidly in the first 24 hours or so, and then slowed to give near-constant concentrations after 5 or 21 days. Hence, the concentrations of cations at 21 days dissolution may be taken as "apparently" equilibrated concentrations although complete equilibrium can not be established.

## ASSUMPTIONS AND CALCULATIONS

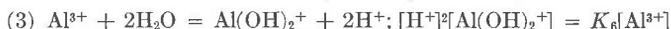
The following steps and certain assumptions must be made to calculate  $\Delta G_f^\circ$  from aqueous dissolution data.

(1) Species in the solution. At 25°C and 1 atmosphere pressure, the distribution of ionic species in water depends primarily on pH and the total concentration of the cations. Since thermodynamic data and stability constants are available for most solution complexed ions (Sillén and Martell, 1964), it is possible to calculate, if chemical equilibrium is assumed, the proportion of each dissolved species of a specific element in the system, assuming that certain species are more likely present in the system. The equilibrium concentration<sup>1</sup> of each species in the system was calculated from (a) the pH of the solution (the activity of  $H^+$  was measured with glass-electrode pH meter) (b) concentration of each cation in the solution in which a total of 144 solutions were analyzed for Si, Al, Fe, Mg, Ca, K, and Na, reported as moles/liter in an earlier paper (Huang and Keller, 1970), using the (c) constants of hydrolysis and dissociation obtained from Sillén and Martell (1964). The validity of the calculation is based on the law of mass action (Butler, 1964). For example, the calculation of each Al species from the dissolution of labradorite is illustrated as follows (Huang, 1969):

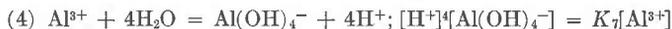
pH = 6.83, total Al =  $0.927 \times 10^{-5}$  moles/liter, then



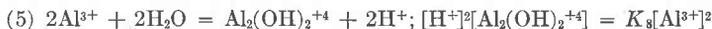
$$K_5 = 10^{-4.89}$$



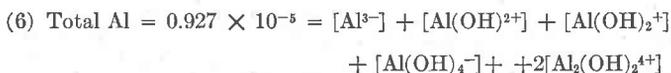
$$K_6 = 10^{-8.56}$$



$$K_7 = 10^{-23.5}$$



$$K_8 = 10^{-6.67}$$



<sup>1</sup>Ideally, the equilibrium constant should be defined in terms of activity. Since the solutions of this study were quite dilute, the equilibrium constants were used for the calculations in terms of concentrations as equivalent to activities.

The results of simultaneous solution of the above six equations using an IBM computer (360/65) are as follows:

$$[\text{Al}^{3+}] = 0.654 \times 10^{-10} \text{ moles/liter}$$

$$[\text{Al}(\text{OH})_2^+] = 0.563 \times 10^{-8} \text{ moles/liter}$$

$$[\text{Al}_2(\text{OH})_2^{4+}] = 0.408 \times 10^{-13} \text{ moles/liter}$$

$$[\text{Al}(\text{OH})_2^+] = 0.804 \times 10^{-5} \text{ moles/liter}$$

$$[\text{Al}(\text{OH})_4^-] = 0.122 \times 10^{-5} \text{ moles/liter}$$

Similar calculations were also made for ferrous iron, ferric iron, Mg, and Ca species in the solutions (Huang, 1969).

(2) The activities of individual ions in the solution were then computed on an IBM 360/65, using the Debye-Hückel method.

(3) The solubility constant ( $K_s$ ) and standard free energies of reaction ( $\Delta G_r^\circ$ ) were calculated. Assuming apparent equilibrium in the systems, the solubility constants ( $K_s$ ) for aqueous dissolution of minerals could be calculated from the most probable chemical reaction of minerals in water (activities of minerals and water are assumed unity). The change in free energy that occurred during the the dissolution of minerals ( $\Delta G_r^\circ$ ) was then calculated from the solubility constants using the Nernst equation:  $\Delta G_r^\circ = -1.364 \log K_s$ , (kcal) (1). Since  $\Delta G_r^\circ = \Sigma \Delta G_f^\circ$  (products) -  $\Sigma \Delta G_f^\circ$  (reactants) (2), standard free energies of formation for the minerals ( $\Delta G_f^\circ$  minerals) can be calculated from (1) and (2), using known standard free energies of formation for other species in equation (2) (Table 1).

## RESULTS AND DISCUSSION

In this paper the actual mineral formula which was derived from the analysis of the specific mineral sample was used, rather than using

Table 1. Standard free energies of formation used  
in calculating  $\Delta G_f^\circ$

Species	$\Delta G_f^\circ$ kcal/mole	Source
$\text{H}_4\text{SiO}_4$	-312.8	Reesman and Keller (1965)
$\text{Al}(\text{OH})_2^+$	-216.1	Reesman et al (1969)
$\text{Al}(\text{OH})_4^-$	-311.3	Reesman et al (1969)
$\text{Fe}^{2+}$	-20.30	Rossini et al (1952)
$\text{Fe}(\text{OH})^+$	-32.58	Calculated from stability constant
$\text{Mg}^{2+}$	-108.76	Langmuir (1968)
$\text{Mg}(\text{OH})^+$	-149.76	Berner (1971)
$\text{Ca}^{2+}$	-132.35	Langmuir (1968)
$\text{Na}^+$	-62.59	Rossini et al (1952)
$\text{K}^+$	-67.47	Rossini et al (1952)
$\text{OH}^-$	-37.63	Reesman and Keller (1965)
$\text{H}_2\text{O}$	-56.72	Wicks and Block (1963)

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Table 2 Analytical data and activities of dissolved species in the dissolution of minerals analyzed.

(I) OLIVINE

pH = 9.55; ionic strength =  $5.26 \times 10^{-4}$

Species	Moles/liter	Activity	Log act.
Mg <sup>2+</sup>	$0.241 \times 10^{-3}$	$0.218 \times 10^{-3}$	-3.66
Mg(OH) <sup>+</sup>	$0.327 \times 10^{-5}$	$0.318 \times 10^{-5}$	-5.50
Ca <sup>2+</sup>	$0.120 \times 10^{-4}$	$0.108 \times 10^{-4}$	-4.92
Al(OH) <sub>4</sub> <sup>-</sup>	$0.371 \times 10^{-6}$	$0.362 \times 10^{-6}$	-6.44
Na <sup>+</sup>	$0.218 \times 10^{-6}$	$0.213 \times 10^{-6}$	-6.66
H <sub>4</sub> SiO <sub>4</sub>	$0.167 \times 10^{-3}$	$0.167 \times 10^{-3}$	-3.78
Fe <sup>2+</sup>	$0.855 \times 10^{-5}$	$0.722 \times 10^{-5}$	-5.11
Fe(OH) <sup>+</sup>	$0.241 \times 10^{-5}$	$0.235 \times 10^{-5}$	-5.63
OH <sup>-</sup>		$10^{-4.45}$	-4.45

(II) AUGITE

pH = 9.62; ionic strength =  $5.11 \times 10^{-4}$

Species	Moles/liter	Activity	Log act.
Mg(OH) <sup>+</sup>	$0.135 \times 10^{-5}$	$0.132 \times 10^{-5}$	-5.88
Mg <sup>2+</sup>	$0.850 \times 10^{-4}$	$0.769 \times 10^{-4}$	-5.11
Fe(OH) <sub>2</sub> <sup>+</sup>	$0.128 \times 10^{-4}$	$0.125 \times 10^{-4}$	-4.90
Al(OH) <sub>4</sub> <sup>-</sup>	$0.371 \times 10^{-5}$	$0.362 \times 10^{-5}$	-5.44
Ca <sup>2+</sup>	$0.162 \times 10^{-3}$	$0.146 \times 10^{-3}$	-3.84
Na <sup>+</sup>	$0.144 \times 10^{-4}$	$0.111 \times 10^{-4}$	-4.96
H <sub>4</sub> SiO <sub>4</sub>	$0.226 \times 10^{-3}$	$0.226 \times 10^{-4}$	-3.65
OH <sup>-</sup>		$10^{-4.38}$	-4.38

(III) LABRADORITE

pH = 6.83; ionic strength =  $5.56 \times 10^{-5}$

Species	Moles/liter	Activity	Log act.
Na <sup>+</sup>	$0.127 \times 10^{-4}$	$0.126 \times 10^{-4}$	-4.90
Ca <sup>2+</sup>	$0.202 \times 10^{-4}$	$0.195 \times 10^{-4}$	-4.71
Mg <sup>2+</sup>	$0.132 \times 10^{-5}$	$0.128 \times 10^{-5}$	-5.89
K <sup>+</sup>	$0.269 \times 10^{-5}$	$0.267 \times 10^{-5}$	-5.57
Al(OH) <sub>2</sub> <sup>+</sup>	$0.804 \times 10^{-5}$	$0.797 \times 10^{-5}$	-5.10
Al(OH) <sub>4</sub> <sup>-</sup>	$0.122 \times 10^{-5}$	$0.121 \times 10^{-5}$	-5.92
Fe(OH) <sub>2</sub> <sup>+</sup>	$0.356 \times 10^{-6}$	$0.353 \times 10^{-6}$	-6.45
H <sub>4</sub> SiO <sub>4</sub>	$0.445 \times 10^{-4}$	$0.445 \times 10^{-4}$	-4.35
OH <sup>-</sup>		$10^{-7.17}$	-7.17

Table 2 (continued)

(IV) MICROCLINE

pH = 8.60; ionic strength =  $1.99 \times 10^{-4}$

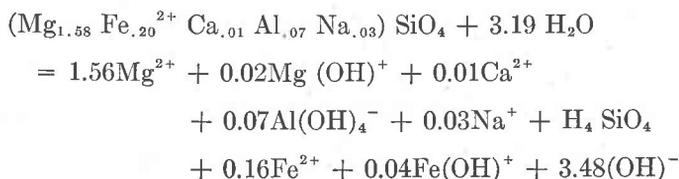
Species	Moles/liter	Activity	Log act.
K <sup>+</sup>	$0.219 \times 10^{-4}$	$0.215 \times 10^{-4}$	-4.67
Na <sup>+</sup>	$0.120 \times 10^{-4}$	$0.118 \times 10^{-4}$	-4.93
Ca <sup>2+</sup>	$0.882 \times 10^{-4}$	$0.827 \times 10^{-4}$	-4.08
Mg <sup>2+</sup>	$0.863 \times 10^{-6}$	$0.809 \times 10^{-6}$	-6.09
Al(OH) <sub>4</sub> <sup>-</sup>	$0.235 \times 10^{-4}$	$0.231 \times 10^{-4}$	-4.64
H <sub>4</sub> SiO <sub>4</sub> <sup>+</sup>	$0.993 \times 10^{-4}$	$0.993 \times 10^{-4}$	-4.03
H <sup>+</sup>		$10^{-8.60}$	-8.60

(V) MUSCOVITE

pH = 7.26; ionic strength =  $1.23 \times 10^{-4}$ 

Species	Moles/liter	Activity	Log act.
K <sup>+</sup>	$0.102 \times 10^{-3}$	$0.101 \times 10^{-3}$	-3.99
Ca <sup>2+</sup>	$0.274 \times 10^{-6}$	$0.260 \times 10^{-6}$	-6.59
Mg <sup>2+</sup>	$0.489 \times 10^{-5}$	$0.465 \times 10^{-5}$	-5.33
Na <sup>+</sup>	$0.109 \times 10^{-4}$	$0.108 \times 10^{-4}$	-4.97
Fe <sup>2+</sup>	$0.483 \times 10^{-5}$	$0.459 \times 10^{-5}$	-5.34
Al(OH) <sub>2</sub> <sup>+</sup>	$0.908 \times 10^{-4}$	$0.896 \times 10^{-4}$	-4.45
Al(OH) <sub>4</sub> <sup>-</sup>	$0.331 \times 10^{-5}$	$0.327 \times 10^{-5}$	-5.49
H <sub>4</sub> SiO <sub>4</sub>	$0.156 \times 10^{-3}$	$0.156 \times 10^{-3}$	-3.81
OH <sup>-</sup>		$10^{-6.74}$	-6.74

an idealized formula for the mineral as previously has been done. The analyses used were published in an earlier paper of ours (Table 1, Huang and Keller, 1970) but each derived formula is presented below in the dissolution reaction of each mineral. Analytical data and calculated activities for the dissolution of the minerals are listed in Table 2.

(I) *Olivine*(A)  $\Delta G_f^\circ$  (olivine) in terms of the specific mineral formula.

$$K_s = [\text{Mg}^{2+}]^{1.56} [\text{Mg}(\text{OH})^+]^{0.02} [\text{Ca}^{2+}]^{0.01} [\text{Al}(\text{OH})_4^-]^{0.07} \\ [\text{Na}^+]^{0.03} [\text{H}_4\text{SiO}_4][\text{Fe}^{2+}]^{0.16} [\text{Fe}(\text{OH})^+]^{0.04} [\text{OH}^-]^{3.48}$$

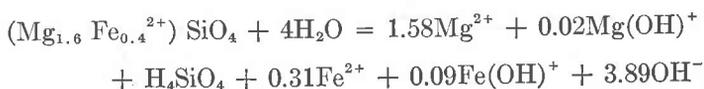
Then,  $\log K_s = -26.91$ .

$$\Delta G_r^\circ = \sum \Delta G_f^\circ (\text{products}) - \sum \Delta G_f^\circ (\text{reactants}) \\ = -424.97 - \Delta G_f^\circ (\text{olivine})$$

$$\Delta G_r^\circ = -1.364 \log K_s$$

$$\Delta G_f^\circ (\text{olivine}) = -424.97 + 1.364 \log K_s \\ = -461.6 \text{ kcal/mole}$$

- (B)  $\Delta G_f^\circ$  of olivine based on the ideal structural formula in which the Mg:Fe ratio was determined by X-ray and optical analyses.



Following the same procedure as in (A),

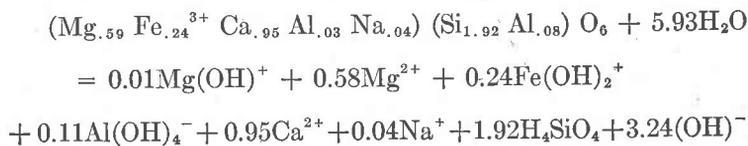
$$\Delta G_f^\circ (\text{olivine}) = -457.8 \text{ kcal/mole; where } K_s = 10^{-29.1}$$

The difference in  $\Delta G_f^\circ$  by the two calculations is 3.8 kcal/mole, which might become significant in determining relative stabilities of minerals in some particular geologic systems.

In Figure 1 are plotted the standard free energies of formation for forsterite ( $\text{Mg}_2\text{SiO}_4$ ) and fayalite ( $\text{Fe}_2\text{SiO}_4$ ) (Robie, 1966; King *et al.*, 1967), and for the olivine of this study. The position of our value, intermediate between the end-member values, adds confidence to  $\Delta G_f^\circ$  values obtained by dissolution.

## (II) Augite

- (A)  $\Delta G_f^\circ$  (augite) in terms of the specific mineral formula.



Following the same procedure as for olivine,

$$\Delta G_f^\circ (\text{augite}) = -679.4 \text{ kcal/mole; where } K_s = 10^{-40.7}$$

The value (-679.4 kcal/mole) just lies between the values for diopside (-725.8 kcal/mole, from Robie, 1966; and

—725 kcal/mole, from Reesman and Keller, 1965) and clinoenstatite (—349.4 kcal/mole, from Robie, 1966; and —350.6 kcal/mole, Reesman and Keller, 1965).

(B) No ideal structural formula is available for augite because of its complex composition.

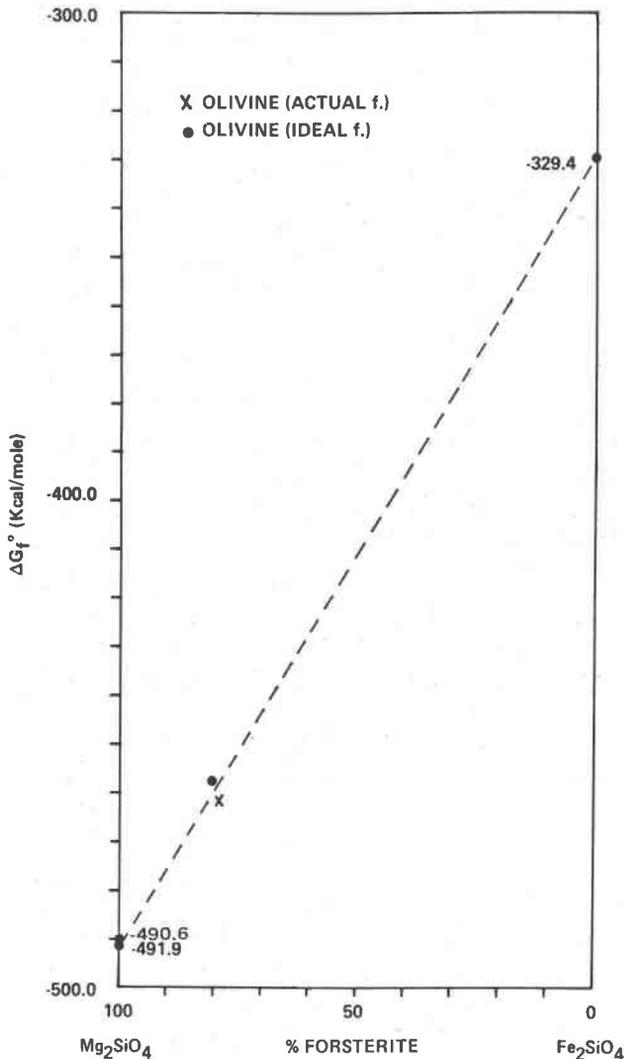
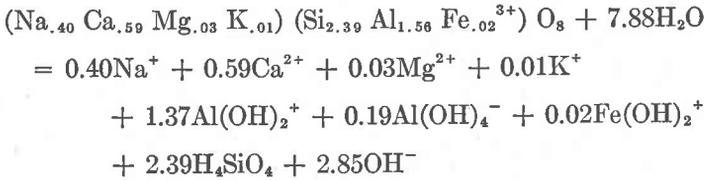
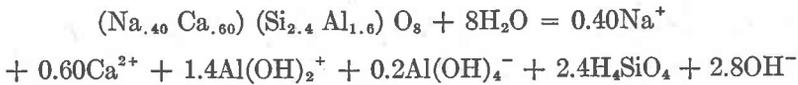
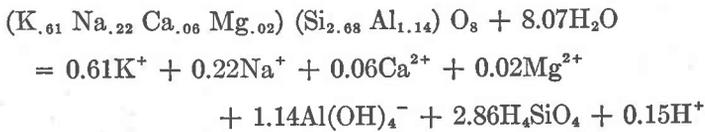
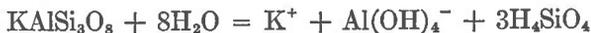
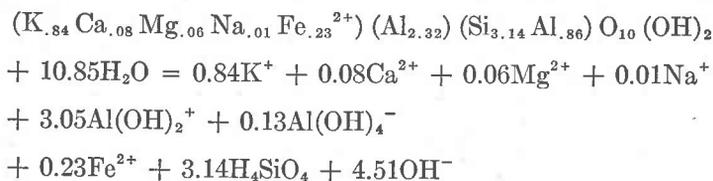


FIG. 1. Standard free energies of formation for the olivine mineral group and those calculated for our sample using ideal and actual formula. The values, —329.4 kcal/mole for Fe<sub>2</sub>SiO<sub>4</sub>, and —491.9 kcal/mole for Mg<sub>2</sub>SiO<sub>4</sub>, are obtained from Robie (1966); —490.6 kcal/mole for Mg<sub>2</sub>SiO<sub>4</sub> from King *et al.* (1967).

(III) *Labradorite*(A)  $\Delta G_f^\circ$  of labradorite from the specific mineral formula.Then,  $\Delta G_f^\circ$  (labradorite) =  $-932.4$  kcal/mole; where  $K_s = 10^{-44.1}$ (B)  $\Delta G_f^\circ$  (labradorite), ideal structural formulaThen,  $\Delta G_f^\circ$  (ideal) =  $-931.1$  kcal/mole; where  $K_s = 10^{-43.6}$ (IV) *Microcline*(A)  $\Delta G_f^\circ$  of microcline from the specific mineral formulaThen,  $\Delta G_f^\circ$  (microcline) =  $-887.3$  kcal/mole; where  $K_s = 10^{-22.4}$ (B)  $\Delta G_f^\circ$  based on the ideal structural formula of microcline.Then,  $\Delta G_f^\circ$  (ideal) =  $-892.6$  kcal/mole; where  $K_s = 10^{-21.40}$ 

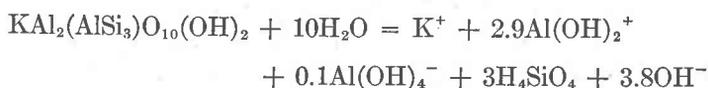
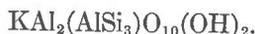
The difference in  $\Delta G_f^\circ$  for microcline by the two calculations is 5.3 kcal/mole. The  $\Delta G_f^\circ$  ( $-892.6$  kcal/mole) for microcline in terms of ideal structural formula, however, agrees with the value of  $\Delta G_f^\circ = 892.8$  kcal/mole for microcline reported by Walbaum (1968).

(V) *Muscovite*(A)  $\Delta G_f^\circ$  of muscovite from the specific mineral formula. This is rather complex since ion substitution occurs extensively in naturally occurring muscovite.



Then,  $\Delta G_f^\circ$  (muscovite) =  $-1399.9$ , where  $K_s = 10^{-62.1}$

(B)  $\Delta G_f^\circ$  based on the ideal structural formula of muscovite,



Then,  $\Delta G_f^\circ$  (ideal) =  $-1313.8$ ; where  $K_s = 10^{-54.5}$

Our data on  $\Delta G_f^\circ$  for muscovite of ideal structural formula ( $-1313.8$  kcal/mole) is higher than the reported data of  $-1330.1$  kcal/mole by Barany (1964) and  $-1328.7$  kcal/mole by Reesman and Keller (1965). Nevertheless, the standard free energy of formation for muscovite of specific mineral formula is a larger negative quantity of  $\Delta G_f^\circ$  than of ideal structural formula. This suggests that actual muscovite, as in this study, is more stable thermodynamically than has been assumed for a muscovite of ideal formula.

#### UNCERTAINTIES AND POSSIBLE ERROR

The values of  $\Delta G_f^\circ$  calculated from dissolution data on specific mineral samples are subject to certain uncertainties and possible sources of error, some of which presumably compensate one another because these values of  $\Delta G_f^\circ$  agree well with those obtained by other methods of determination. First, the bulk chemical analysis of the mineral sample, from which the specific mineral formula is calculated, is subject to analytical errors as were described for G-1 and W-1. Second, the chemical analyses made of the ions dissolved from the minerals during reaction may be in error analogous to those of G-1 and W-1. Third, the standard  $\Delta G_f^\circ$  of ions used in the calculations have uncertainties of  $\pm 0.1$  to  $0.3$  kcal mol $^{-1}$ . Fourth, error in the fractional mols of  $\text{H}_2\text{O}$ , for example, required for reaction of the specific-mineral formula (calculated from its composition) will affect the calculated  $\Delta G_f^\circ$  of the mineral by a factor of 57, the  $\Delta G_f^\circ$  of  $\text{H}_2\text{O}$ .

Thus, if errors from analytical and standard data were, say one per cent, on the plus side for all possible variables, its total effect would

Table 3: A comparison of standard free energies of formation for minerals (kcal/mole)

Mineral	Huang and Keller (1971)	Others
Olivine		
a*	-461.6	-
b*	-455.9	-
Augite		
a	-679.4	-
Labradonite		
a	-932.4	-
b	-931.1	-
Microcline		
a	-887.3	-
b	-892.6	-892.8 (Walbaum, 1968) -891.3 (Reesman and Keller, 1965)
Muscovite		
a	-1299.9	-
b	-1313.8	-1330.1 (Barany, 1964) -1328.7 (Reesman and Keller, 1965)

\* a: calculation based on actual mineral formula from bulk analyses

b: calculation based on ideal mineral formula

be disastrous to the method. The probability, however, of this occurring among all of the multiple data from the bulk analysis, the solution analysis, the computed specific-mineral formula, and the standard values, is exceedingly low. The fact that  $\Delta G_f^\circ$  values calculated from dissolution data agree reasonably well with values determined by other procedures suggests that the possible errors in the dissolution method tend to compensate one another, and add confidence to the validity of the dissolution method.

A summary of the  $\Delta G_f^\circ$  of the minerals examined by us are compared with those of others in Table 3.  $\Delta G_f^\circ$  calculated for microcline and muscovite from specific analyses are larger negatively than those based on ideal structural formulas, although the  $\Delta G_f^\circ$  from ideal structural formula for microcline agrees well with previously reported values.

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