

## Comparison of alkali interdiffusion rates for cryptoperthites

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### ABSTRACT

Lamellar homogenization rates were used to determine “average” interdiffusion coefficients ( $\bar{D}$ ) for seven largely disordered cryptoperthites with relatively straight, coherent lamellae and < 1.6 mol% An and for two highly ordered samples with diagonally associated lamellae and about 2 and 5 mol% An. The average  $\bar{D}$  values for the disordered samples at 600°C agree within a factor of ten with the  $\bar{D}$  values calculated from independent tracer diffusion data using Margules parameters for the coherent solvus to estimate activity of Or. The ordered samples did not homogenize, even at 1000°C, but their average  $\bar{D}$  values at 780°C agree with the calculated  $\bar{D}$  values using the same tracer diffusion data and an activity of Or based on their high-temperature solvi. The results suggest that alkali self-diffusion rates are similar for many natural feldspars and the difference in their interdiffusion rates is due primarily to a difference in their thermodynamic properties (activities).

### INTRODUCTION

Tracer diffusion coefficients for Na and K in alkali feldspars have been determined in numerous previous studies (e.g., see review by Yund, 1983a), and the data for orthoclase (Foland, 1974) and albite (Kasper, 1975) are consistent with interdiffusion coefficients determined directly from conventional bicrystal diffusion profiles (Christoffersen et al., 1983) and from lamellar homogenization data (Brady and Yund, 1983). The tracer diffusion coefficients were determined at 2-kbar water pressure, whereas the interdiffusion data were determined with nominally dry crystals at 1 atm to 15 kbar. The good agreement of these data argues that the presence of water in the feldspar structure does not affect the rate of alkali diffusion. This is in contrast to the rates of oxygen diffusion (Yund and Anderson, 1974; Giletti et al., 1978), Al-Si disordering (e.g., Yund and Tullis, 1980), and NaSi-CaAl interdiffusion in peristerite (Yund, 1986), which are significantly enhanced by water in a crystal.

Although the defect caused by water in the feldspar structure does not appear to affect alkali diffusion rates, it is not known whether defects associated with other impurities or nonstoichiometry affect these rates (e.g., Lasaga, 1981). In order to apply experimental diffusion data, or lamellar coarsening rate data which depend on diffusion rates, to mineralogical and geochemical problems, it is important to evaluate the variability in the diffusion rates for natural samples.

In alkali feldspars, Na and K diffusion are thought to occur, at least in part, by a vacancy mechanism (Petrović, 1974). Thus an increase in the alkali vacancy concentration, such as through the introduction of the common

aliovalent ions of Ca<sup>2+</sup> or Ba<sup>2+</sup>, might enhance the mobilities of the alkali ions. On the other hand, a divalent cation in the feldspar alkali site may form a defect pair with an adjacent alkali vacancy in order to preserve local charge balance. If the divalent cation diffuses slowly, the alkali interdiffusion rate might be unchanged with increasing concentration of these aliovalent impurities.

The purpose of this study was to determine “average” interdiffusion coefficients ( $\bar{D}$ ) using the lamellar homogenization rate method for a number of cryptoperthites of different compositions and from different cooling environments. The interdiffusion rate for a given composition is dependent on the tracer diffusion rates of Na and K and on the activity of Or in the solid solution. The latter can be estimated from Margules parameters for the solvus of the cryptoperthite (Brady and Yund, 1983). This method allows any observed difference in the “average” interdiffusion rates between samples to be correlated with either a difference in the self-diffusion rates or the thermodynamic activities of Or in the samples. A limitation of this method is that it provides an “average” interdiffusion value for the compositional interval between the lamellae, and the value is only for the direction normal to the lamellae.

### EXPERIMENTAL PROCEDURES

#### Sample description

The feldspars used in this study include cryptoperthites from the Bishop Tuff (BiT), California; Bandelier Tuff (BaT), New Mexico; Rabb Canyon porphyry dike (RCD), New Mexico; Big Bend rhyolite dike (BBD), Texas; Lake Amont rhyolite plug (LAP), Texas; two disordered, ion-exchanged feldspars (IF-1, IF-2); and two samples from the Klokken layered syenite-gabbro (KP-1 and KP-2), Greenland. The Or content of these samples ranges from about 27 to 64 mol%, and the An content from 0 to 5 mol%. Compositional data, the degree of Al-Si order, and the lamellar

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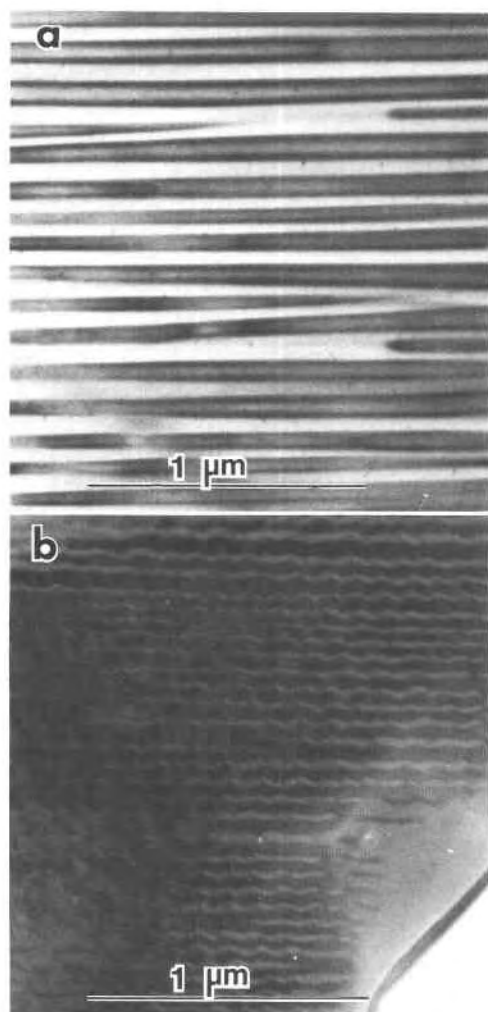


Fig. 1. TEM micrographs typical of (a) the disordered samples (Rabb Canyon) and (b) the ordered Klokken samples (KP-1). Both bright-field micrographs are approximately (001) sections.

spacings are listed in Table 1. The samples are divided into two groups; the first group includes essentially disordered cryptoperthites (BiT, BaT, RCD, BBD, LAP, and IF) which have long, bifurcating lamellae approximately parallel to  $(\bar{6}01)$ . A representative transmission-electron micrograph of one of these samples (BaT) is shown in Figure 1a. These coherent lamellar structures probably formed by spinodal decomposition below the coherent spinodal (e.g., Yund, 1983b). The second group comprises two samples (KP-1 and KP-2) from the Klokken layered series described by Brown et al. (1983) and Brown and Parsons (1984). Our samples are characterized by zig-zag, Albite-twinned, low albite and diagonally associated maximum microcline lamellae, with their interface parallel to  $(\bar{6}\bar{6}1)$ . See Brown and Parsons (1984) for further details. A typical microstructure for our samples is shown in Figure 1b.

Bulk compositions for these ordered crystals are somewhat variable as noted and discussed by Brown et al. (1983). The average An contents of our samples KP-1 and KP-2 are about 2.0 and 5.0 mol%, respectively. See Table 1 for details. In addition to the fine-scale microstructure in these samples which was described above, they also contain opaque patches that are coarser, two-phase intergrowths. This is the "deuteric" perthite

Table 1. Data for the cryptoperthite samples

Sample*	Or (mol%)	An (mol%)	Ba (wt%)	Fe (wt%)	Ti (wt%)	Mn (wt%)	Lamellar half spacing (Å)
1. Disordered samples with straight, coherent lamellae							
BiT	64.3	0.8					1600 ± 100
BaT	38.0	1.1	0.02	0.10	0.01	0.01	1150 ± 60
RCD	61.0	0.6					850 ± 50
BBD	40.7	0.0					550 ± 30
LAP	38.0	1.6					172 ± 20
IF-1	48.0	0.0					430 ± 45
IF-2	27.0	0.0					172 ± 20
2. Ordered samples with zig-zag, semicoherent lamellae							
KP-1	32	2.0	0.08	0.06	0.03	0.06	916 ± 142
KP-2	35	5.0	0.15	0.03	0.07	0.10	750 ± 70

\* BiT—Bishop Tuff (BT-9 of Snow and Yund, 1985). BaT—Bandelier Tuff. RCD—Rabb Canyon dike. BBD—Big Bend dike (from dike margin; see Christoffersen and Schedl (1980). LAP—Lake Amont plug (homogenized and then exsolved at 525°C for 1150 h. IF-1—ion-exchanged. IF-2—ion-exchanged. KP-1—Klokken Syenite (GGU140130 of Brown et al., 1983). KP-2—Klokken Syenite (GGU140194 of Brown et al., 1983).

referred to by Brown et al. (1983). They found no significant difference in the bulk compositions of patchy and nonpatchy areas. If the volume fraction of the coarser intergrowth is large, its presence can be recognized in X-ray single-crystal photographs because the phases in the coarse intergrowth are noncoherent. For the homogenization experiments, we chose crystals that gave reasonably sharp, single  $0k0$  reflections, indicating the coherent, fine-scale intergrowth. The  $h00$  reflections are sharp doublets indicating essentially uniform compositions for the two phases.

In addition to the natural samples in the first group, two synthetic, disordered cryptoperthites (IF-1 and IF-2) with bulk compositions of Or<sub>27</sub> and Or<sub>48</sub> were prepared by heat treatment and ion exchange (Sipling and Yund, 1976) in order to have samples with lower and intermediate Or contents. The An content of these crystals is essentially zero.

#### Homogenization experiments

Lamellar compositions of all samples before and after annealing were determined from the reciprocal cell edge  $a^*$ . These measurements were corrected for coherency strain using the data of Yund and Tullis (1983). The ordered phases have their lamellar interfaces parallel to  $(\bar{6}\bar{6}1)$  whereas the compositional corrections assume coherency parallel to  $(\bar{6}01)$ . However, the compositional corrections are only about 3 mol% for these and the disordered samples and not really significant for this study.

All disordered cryptoperthites were annealed in air at  $600 \pm 5^\circ\text{C}$  until completely homogenized. (The ordered samples did not homogenize even at  $1000^\circ\text{C}$ .) The progression of homogenization of the lamellae was monitored by periodically removing a crystal from the furnace and taking an  $(hk0)$  photograph from which the compositions of the two sets of lamellae could be determined. With continued heating, the  $a^*$  reflections for the two lamellae began to merge. When the compositional difference was less than about 20 mol%, the reflections from the K- and Na-rich lamellae became elongated and were not clearly resolved. The compositional difference was then estimated from the length of the elongated reflection. Complete homogenization was marked by a sharp  $a^*$  reflection that was often resolved into the  $K\alpha_1$  and  $K\alpha_2$  doublet.

## RESULTS AND DISCUSSION

### Disordered cryptoperthites

The compositional difference between the lamellae ( $\Delta C$ ) decreased with increased annealing time for the disor-

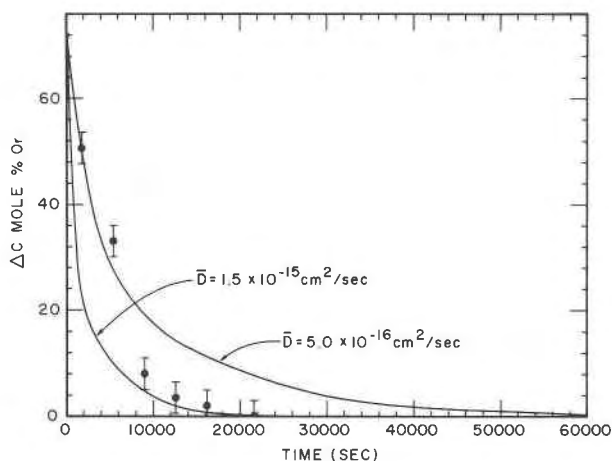


Fig. 2. Representative homogenization data for the Bishop Tuff (BiT) sample at 600°C. The ordinate is the compositional difference of the lamellae ( $\Delta C$ ). The experimental points are shown with error bars and the two lines are calculated curves assuming constant  $\bar{D}$  values as shown. The "average"  $\bar{D}$  for the sample is bracketed by the  $\bar{D}$  values from the two curves.

dered samples. Representative homogenization data for the Bishop Tuff sample (BiT) are shown in Figure 2. These data can be used to calculate an "average"  $\bar{D}$  for the compositional interval between the lamellae. The curves shown in Figure 2 represent the predicted change in lamellar composition as a function of time assuming the constant value for  $\bar{D}$  shown on each curve. These curves were calculated using an infinite series solution to the equation for finite, one-dimensional diffusion (Crank, 1975, p. 63). The calculated curves bracket the experimental data points, and the "average"  $\bar{D}$  is the mean of these values.  $\bar{D}$  values determined in this manner typically have an uncertainty of two or three times their value.

The data and procedure used to calculate the curves on Figure 3 are discussed by Brady and Yund (1983). The calculated alkali interdiffusion coefficients are based on the following relation (Manning, 1968, p. 21; Brady, 1975), where the Na and K tracer diffusion coefficients ( $D^*$ ) are from Foland (1974) and Kasper (1975) and it is assumed that the self-diffusion and tracer diffusion coefficients are equal (correlation coefficient is 1.0):

$$\bar{D}_{K-Na} = \frac{D_K^* D_{Na}^*}{X_{Or} D_K^* + X_{Ab} D_{Na}^*} \cdot \frac{d \ln a_{Or}}{d \ln X_{Or}}, \quad (1)$$

where  $X$  is the mole fraction of albite (Ab) or orthoclase (Or) and  $a_{Or}$  is the molar activity. A log-linear extrapolation of the near endmember tracer diffusion coefficients was used to estimate  $D^*$  for intermediate compositions. In addition to the errors in the original tracer data, this extrapolation introduces an unknown error for intermediate  $D^*$  and hence for intermediate  $\bar{D}$  values. The predicted curve assuming ideal mixing is shown by the dash-dot curve in Figure 3. Because Na and K mixing is nonideal at 600°C, the thermodynamic term in Equation 1 must

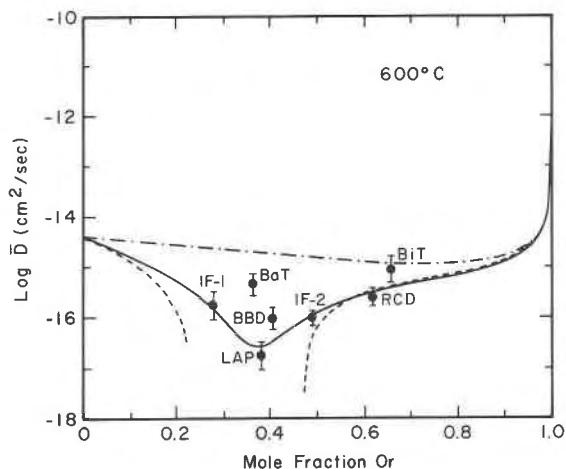


Fig. 3. Comparison of experimental "average"  $\bar{D}$  values (points with error bars) and the calculated  $\bar{D}$  curves from Eq. 1. The solid curve is based on the disordered, coherent solvus which most closely approximates the solvus for these samples. For comparison, the calculated  $\bar{D}$  for an ideal solid solution (dash-dot curve) and for the disordered, strain-free solvus (dash curve) are shown. The samples are described in Table 1.

be evaluated. This was done following the procedure outlined in Thompson and Waldbaum (1969) to obtain Margules parameters for the disordered, coherent solvus (Sipling and Yund, 1976). This solvus is expected to most closely correspond to that for these disordered samples, and the  $\bar{D}$  curve is shown by the solid line in Figure 3. For comparison the short dashed curve is based on an activity of Or derived from the strain-free solvus. The same values for the self-diffusion coefficients were used to calculate all of these curves.

The "average"  $\bar{D}$  values are plotted as a function of the bulk composition of the cryptoperthite on Figure 3. This is somewhat arbitrary because the "average"  $\bar{D}$  value is a time average of values that are in turn a function of the compositional interval between the lamellae. Brady and Yund (1983) reported that the "average"  $\bar{D}$  value corresponds approximately with the minimum in the calculated curve (solid line on Fig. 3). Correlation of the "average"  $\bar{D}$  with either the minimum or the bulk composition is not completely justified, but using the bulk composition is a convenient way to compare the data for the different samples. Six of the seven samples plot on or near the calculated interdiffusion curve within the estimated uncertainties. (The error in the calculated intermediate  $\bar{D}$  values is not known but must be at least plus or minus half an order of magnitude.) Because the correlation of the "average"  $\bar{D}$  with bulk composition is only approximate, the comparison between the experimental data and the calculated curves should not be over interpreted.

Only the "average"  $\bar{D}$  value for the Bandelier Tuff (BaT) plots more than half an order of magnitude from the calculated curve. This sample has a K/Na ratio similar to that of the Lake Amont (LAP) and Big Bend (BBD) samples and an intermediate An content, 1.1 mol% compared

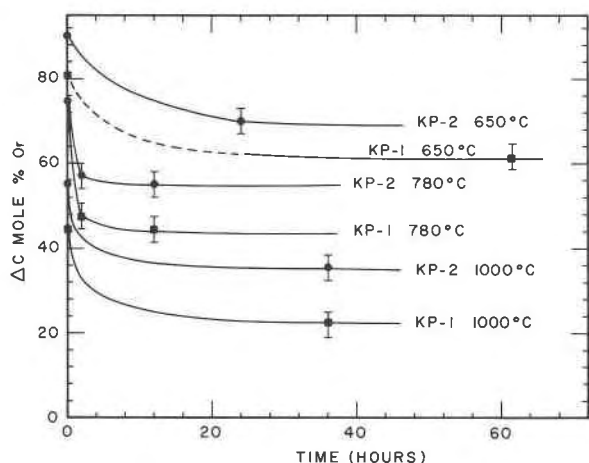


Fig. 4. Annealing data for the ordered samples at three temperatures. The initial  $\Delta C$  values are not the same for all crystals of a given sample because of their compositional variability. The crystals annealed at 1000°C were first annealed at 780°C. The shapes of the curves are only approximate.

to 1.6 and 0.0 mol% for LAP and BBD, respectively. Thus the difference between the "average" and calculated  $\bar{D}$  values for this sample does not correlate with any known difference in its An content.

Although all of these samples are approximately disordered, have a low An content, and have a high degree of coherency, it is possible that they may not all correspond exactly to the disordered, coherent solvus for experimental samples (Sipling and Yund, 1976). Thus their thermodynamic factors in Equation 1 may not be exactly equal. Partial loss of coherency, an increase in the Al-Si ordering, or an increase in the An content (Smith, 1978) would raise the solvus, and this would decrease the value of the thermodynamic term and lower the calculated  $\bar{D}$  curve shown on Figure 3. Thus a slightly elevated solvus does not appear to explain the difference in the calculated and "average"  $\bar{D}$  values for the Bandelier Tuff sample. If the difference is real, it seems more likely that it is due to a difference in the self-diffusion coefficients that might in turn be due to a difference in the concentration of point defects.

One experiment was carried out in order to determine whether water would affect the alkali diffusion rate in cryptoperthites. Crystals (100–200- $\mu\text{m}$  diameter) from the Rabb Canyon sample were heated at 400°C for 12 h at 2-kbar water pressure to allow water to enter the crystal if possible. At these conditions, lamellar homogenization or a change in Al-Si ordering does not occur. It is not known how much water entered the crystals during this treatment. After heating at 400°C, the crystals were annealed in a hydrothermal experiment at 600°C, 2kbar, for 1 h. The degree of homogenization was the same as when this sample was annealed in air at the same temperature for the same time. Thus the presence of water did not alter the diffusion rate. The result is consistent with previous experimental observations that water in feldspar

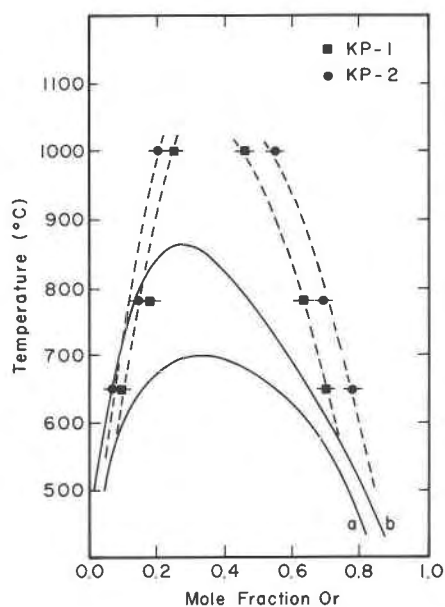


Fig. 5. The steady-state compositions (symbols) for the ordered samples from Fig. 4 shown as a function of annealing temperature. The data approximately outline two solvi that differ in the An content of the crystals. The ordered, coherent solvus (a) from Yund (1974) and the ordered strain-free solvus (b) from Bachinski and Müller (1971) are shown for comparison.

does not affect the rate of alkali diffusion (e.g., Yund, 1983a; Brady and Yund, 1983; Christoffersen et al., 1983).

#### Ordered cryptoperthites

Changes in lamellar compositions with annealing time for KP-1 and KP-2 are shown in Figure 4. Because of the large compositional variation of crystals in these samples, the initial compositional difference ( $\Delta C$ ) was not the same for all crystals. In addition, the crystals used for the 1000°C homogenization experiment were from the previous experiments at 780°C and hence their lower initial  $\Delta C$  values.

None of these crystals homogenized even after annealing at 1000°C for 2 d, although the lamellae reached steady-state compositions within that time. These steady-state values are plotted on a temperature-composition diagram in Figure 5. The steady-state compositional difference of the lamellae decreases with increasing temperature, and  $\Delta C$  at a given temperature is larger for the sample (KP-2) with the higher An content. Although these experiments have not been reversed, they appear to define solvi that lie above the ordered coherent and strain-free solvi in  $T$ - $X$  space (see Fig. 5). TEM examination of the lamellar microstructures of these crystals after annealing at 780°C shows that the lamellae still have a high degree of coherency. The degree of Al-Si order is also not altered significantly after two days at 1000°C. Therefore the data in Figure 5 represent the approximate positions of the metastable solvi for these two samples.

The fact that these solvi are wider and higher than for

the ordered strain-free solvus is probably due in part to the high An content of KP-2 (~5 mol% An). Smith (1978) reported that an increase in An content from 2 to 10 mol% raises and widens the solvus for disordered, strain-free feldspars by several hundred degrees. Although the solvus for KP-2 is higher than for KP-1, the An content of KP-1 (~2.0 mol% An) appears too low to account for the elevated position of its solvus. Thus there may be other reasons for the elevated positions of the solvi for these samples.

Average  $\bar{D}$  values calculated for KP-1 and KP-2 at 780°C were obtained using the relation (Brady and Yund, 1983)

$$\bar{D}t/L^2 = 0.5, \quad (2)$$

where  $t$  is the time required for homogenization of the lamellae or for their compositions to reach steady state, and  $L$  is half the lamellar spacing—the distance from center to center of adjacent (unlike) lamellae. (This method of estimating “average”  $\bar{D}$  values is as accurate as the method used for the disordered samples if  $t$  is accurately determined.) Because the lamellae never homogenized, the interface remains sharp and must move as the lamellae exchange Na and K unless the compositional difference between their initial and steady-state values is equal for both Na- and K-rich lamellae. Thus we assumed that the mobility of this interface is not rate limiting in order to use the  $\Delta C$  versus  $t$  data to calculate “average”  $\bar{D}$  values. (See discussion in Brady and McCallister, 1983.) These values are shown in Figure 6 along with the curves calculated from Equation 1 using Margules parameters based on the solvi for KP-1 and KP-2 (Fig. 5). (The same values for the self-diffusion coefficients were used for these calculations as were used for the disordered samples shown in Fig. 3.) The agreement between the “average” and calculated  $\bar{D}$  values is very good and supports the assumption that movement of the lamellar interface is not rate limiting. For comparison, the calculated  $\bar{D}$  values at 780°C for the disordered, coherent solvus are shown in Figure 6 by the upper curve labeled (a). For the steady-state lamellar compositions of KP-1 and KP-2, the difference between these curves is about two orders of magnitude. Thus the difference in the activity of Or for the ordered and disordered samples accounts for the difference in their interdiffusion rates.

### CONCLUSIONS

The results presented here indicate that there is only a small difference in the alkali self-diffusion coefficients for all natural and synthetic feldspars investigated to date, including the samples used for previous tracer and interdiffusion studies. Perhaps the only exception is the Banderier Tuff sample; however, the approximately one order of magnitude difference for this sample is hardly outside the combined experimental errors.

The similarity of the self-diffusion coefficients for these feldspars is somewhat surprising and suggests that either alkali diffusion does not involve a vacancy mechanism, and hence the rate is insensitive to whether there is a

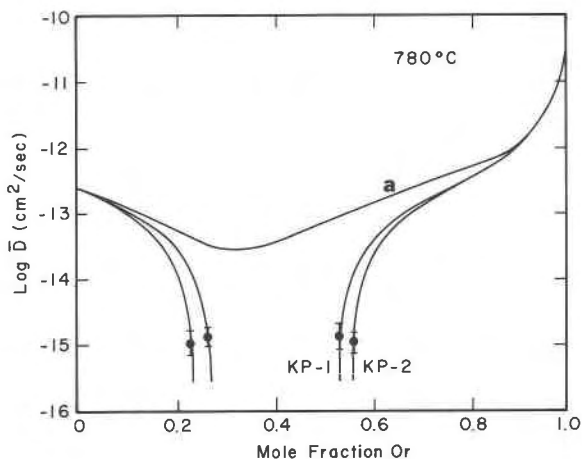


Fig. 6. Comparison of experimental “average”  $\bar{D}$  values for the ordered samples (dots with error bars) at 780°C with the calculated  $\bar{D}$  values using their solvus data shown in Fig. 5. The final lamellae compositions were used to plot the four experimental data points. There is good agreement between the experimental and calculated values. For comparison, the upper curve (a) is the calculated  $\bar{D}$  values based on the disordered, coherent solvus.

variation in the vacancy concentration due to impurities, or if the mechanism does involve vacancies, the concentration of unpaired vacancies does not vary appreciably in these samples. The latter suggests that although diffusion may be extrinsically controlled, the number of unpaired vacancies may be a maximum in all these samples, perhaps because the total concentration of impurities is large. On the other hand for a mineral such as olivine that contains a major ion with a variable oxidation state, there is a large difference in the number of vacancies and in the diffusion rate as the  $\text{Fe}^{3+}/\text{Fe}^{2+}$  ratio is varied (Buening and Buseck, 1973; Stocker, 1978). Clearly it is necessary to evaluate the importance of impurities and defects for controlling diffusion rates in other minerals.

Regardless of the reason for the minimum variation in the tracer diffusion rates of these samples, it appears that the tracer diffusion data are applicable to mineralogical and geochemical problems without undue concern for differences in the kind or level of impurities or degree of non-stoichiometry, if any, of the samples. Inasmuch as our samples came from diverse localities and geologic environments, their diffusion rates should be representative of alkali feldspars in general.

In spite of the fact that the *self-diffusion* coefficients are similar for many alkali feldspars, their alkali *interdiffusion* rates will not be equal unless their mixing properties (activity of Or) are the same. This is especially true at temperatures where the departure from ideal mixing becomes significant. Margules parameters for the appropriate solvus can be used to estimate the activity and, together with the self-diffusion coefficients, can be used to estimate the interdiffusion coefficients. This is important for estimating the rate of lamellar coarsening in samples

that have cooled slowly and are thus partially ordered, or those that have a high An content. In addition, the "average" interdiffusion rate depends on bulk composition; hence cryptoperthites with Na or K-rich bulk compositions would coarsen faster than those with intermediate bulk compositions.

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