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Equilibrium Solubilities of $LiFeO_2$ and $(Li,K)_2CrO_4$ in Molten Alkali Carbonates at 650°C

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FOR THE UNITED STATES
DEPARTMENT OF ENERGY

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22161
NTIS price codes—Printed Copy: A03; Microfiche A01

ORNL/TM-10142

METALS AND CERAMICS DIVISION

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Date Published - August 1986

Research sponsored by
the U.S. Department of Energy,
Advanced Research and Technology Development
Fossil Energy Materials Program

Prepared by the
OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee 37831
operated by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under Contract No. DE-AC05-84OR21400

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H. S. Hsu, J. H. DeVan, and M. Howell

ABSTRACT

The equilibrium solubilities of $LiFeO_2$ and $(Li,K)_2CrO_4$ in 62% Li_2CO_3–38% K_2CO_3 were quantitatively measured at 650°C under simulated oxidizing gas environments of molten carbonate fuel cells. Both materials in the carbonate mixture were dissolved according to a physical dissolution process, where the solubilities of $LiFeO_2$ and $(Li,K)_2CrO_4$ are independent of partial pressures of CO_2 and O_2. Equilibrium solubilities of $LiFeO_2$ and $(Li,K)_2CrO_4$ in the carbonate mixture were determined to be 77 wppm of iron and 13.7 wt % of chromium, respectively.

INTRODUCTION

The intensive development of molten carbonate fuel cells (MCFCs) as high-efficiency systems for directly converting gasified coal or natural gas into electricity has been under way for the last decade. One of the critical problems with regard to lifetimes of MCFCs is corrosion of current collectors. The current collectors, which contact both alkali carbonate and reactant gas, are degraded gradually. As a result, cell performance declines due to the increase of internal resistance and the contamination of carbonate electrolyte.

In order to define and study the corrosion mechanisms of current collector materials in MCFCs, our previous studies were designed to provide a fundamental understanding of the thermodynamics of metal-alkali carbonate systems and of the corrosion behavior of pure metals in molten carbonate environments. Phase stability diagrams of Cr-Li-K-C-O and Fe-Li-K-C-O systems at 650°C were established by thermochemical calculations,[1-3] and the corrosion of chromium and iron with carbonate

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*Research sponsored by the U.S. Department of Energy, AR&TD Fossil Energy Materials Program [DOE/FE AA 15 10 10 0, Work Breakdown Structure Element ORNL-3(D)], under contract DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc.

deposits was investigated under fuel cell operating environments.[4,5] Many theoretical mechanisms have been proposed for this type of molten salt corrosion. The most widely discussed mechanism is the oxide fluxing process wherein the corrosion of metals or alloys is enhanced by the dissolution/reprecipitation process of metal oxides in the molten salt.[6] The oxide fluxing process is caused by the variation of metal oxide solubility with salt chemistry (basicity or acidity). This phenomenon has been reported when NiO was used as a cathode in the MCFC.[7] It showed that significant NiO dissolution occurred at the cathode, while metallic nickel precipitates within the carbonate electrolyte layer. Thus, a knowledge of the solubilities of metal oxides as a function of salt chemistry is essential in understanding the corrosion of current collectors in MCFCs.

Since nickel and NiO were used as anode and cathode materials, respectively, in the prototypical MCFC system, the solubility of NiO in alkali carbonates has received most attention and has been investigated thoroughly.[8,9] However, the corrosion products formed on types 310 and 316 stainless steel, which are frequently used as current collector materials, are primarily iron- and chromium-containing oxides.[7] Only limited results on the solubilities of $LiFeO_2$ and chromate in alkali carbonates were reported by Baumgartner et al.[10,11] The purpose of this paper is to present results of solubility measurements on $LiFeO_2$ and $(Li,K)_2CrO_4$ in 62% Li_2CO_3–38% K_2CO_3 under oxidizing gas environments of MCFC systems at 650°C.

EXPERIMENTAL PROCEDURE

The Li_2CO_3 and K_2CO_3 powders used in this study were reagent-grade chemicals. The carbonate powders were dried in air at 200°C for 1/2 h. The eutectic composition of 62% Li_2CO_3–38% K_2CO_3 was obtained by mixing the dried powders in appropriate proportions. The mixtures were then hermetically stored with a desiccant until required for use.

The experimental assembly is shown in Fig. 1. About 0.15 g of Fe_2O_3 powder (99.9% purity) and 40 to 45 g of the carbonate mixture were mixed and contained in a gold crucible, which was located inside a closed-end cylindrical quartz chamber. The quartz chamber was sealed with a brass

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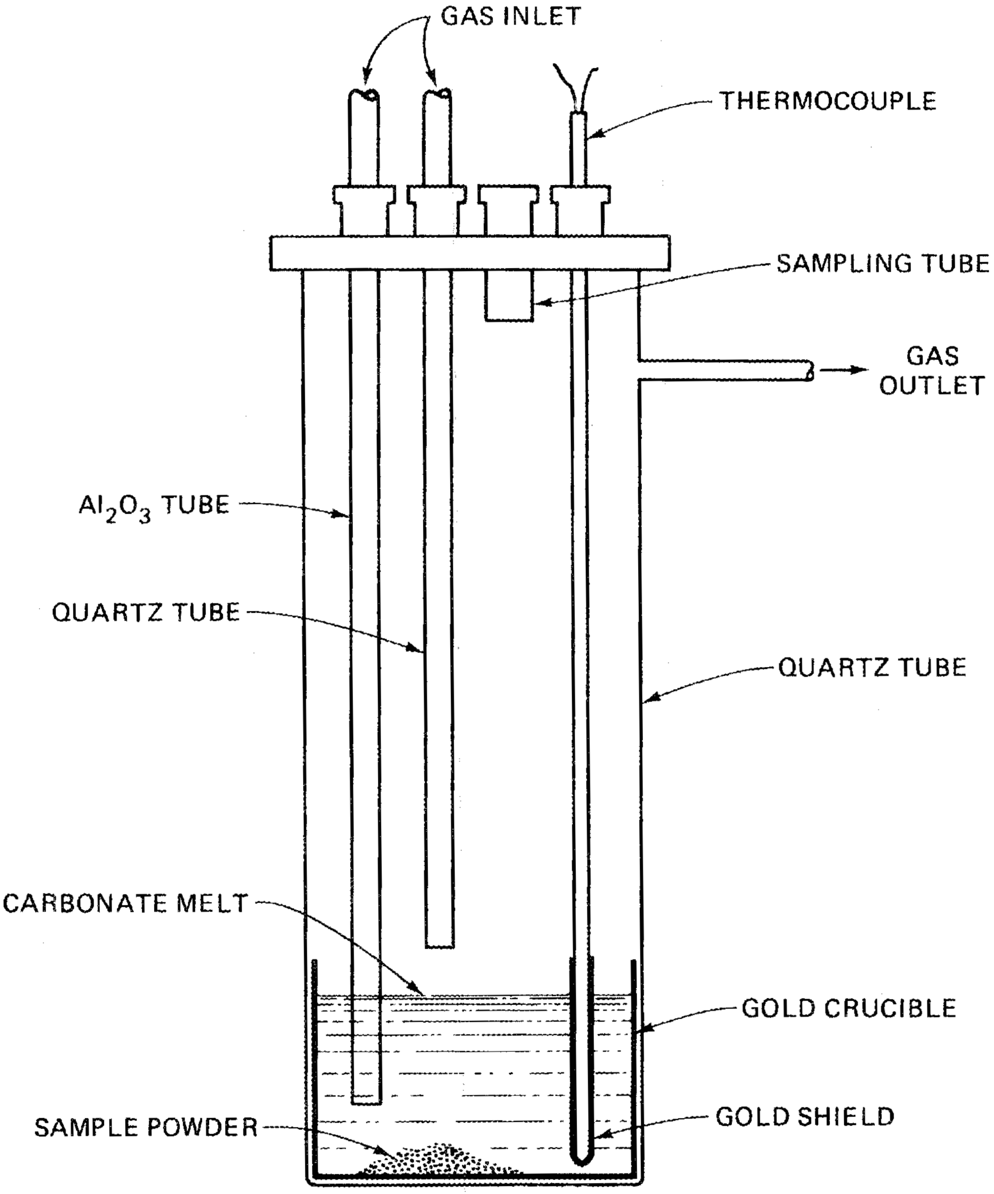

Fig. 1. Experimental setup for solubility measurements.

flange and cap. A gold-shielded Pt-Pt/10% Rh thermocouple was positioned within the test salt to obtain in-situ temperature measurements.

The gas atmosphere was established by using premixed gases containing CO_2, O_2, and Ar supplied by Matheson Gas Company. All gas mixtures were dried by passing them through Drierite ($CaSO_4$). One test gas stream was passed over the carbonate melt surface while another gas stream of the same composition was bubbled through the melt to assure the equilibrium between gas and melt and accelerate dissolution of the metal oxides. The carbonate melt was first heated to 750°C for one day to accelerate dissolution. Then the melt was initially maintained at 650°C for one week, and dissolved iron was sampled every other day for an additional one to two weeks.

Samples of the carbonate were taken with a gold tube and solidified on a gold plate. The gas stream bubbling through the melt was stopped for about 10 min before sampling. The tip of the gold sample tube was kept just below the surface of the melt so that no $LiFeO_2$ powder was included along with the salt sample. Each salt sample was about 0.5 to 1.0 g. The samples were weighed and dissolved in distilled water, and the contents of soluble $LiFeO_2$ were determined by using an atomic absorption spectrophotometer.

The solubility study of chromate in the carbonate mixture followed the same procedure described above. However, the equilibrium solubility of chromate in the carbonate mixture was unexpectedly high. Once the initial charge of Cr_2O_3 powder in the carbonate mixture was completely dissolved, additional Cr_2O_3 powder was added repeatedly to the carbonate melt until the solubility measurement reached a steady state.

RESULTS AND DISCUSSION

Figure 2 is the phase stability diagram for the Fe-Li-K-C-O system under CO_2-O_2-Ar gas mixtures at 650°C (ref. 5). The diagram was constructed for a ratio of Li:K = 6:4 and a ratio of (Li+K):Fe = 100:1. It shows that a mixture of $(Li,K)_2CO_3$ and $LiFeO_2$ is stable at $-4.76 < \log(P_{CO_2}) < 0.06$ under oxidizing environments, while a mixture of $(Li,K)_2CO_3$ and $Li_2Fe_3O_5$ is stable under the similar partial pressures of CO_2 and $\log(P_{O_2}) < \sim -14$. The phase relationships vary at different

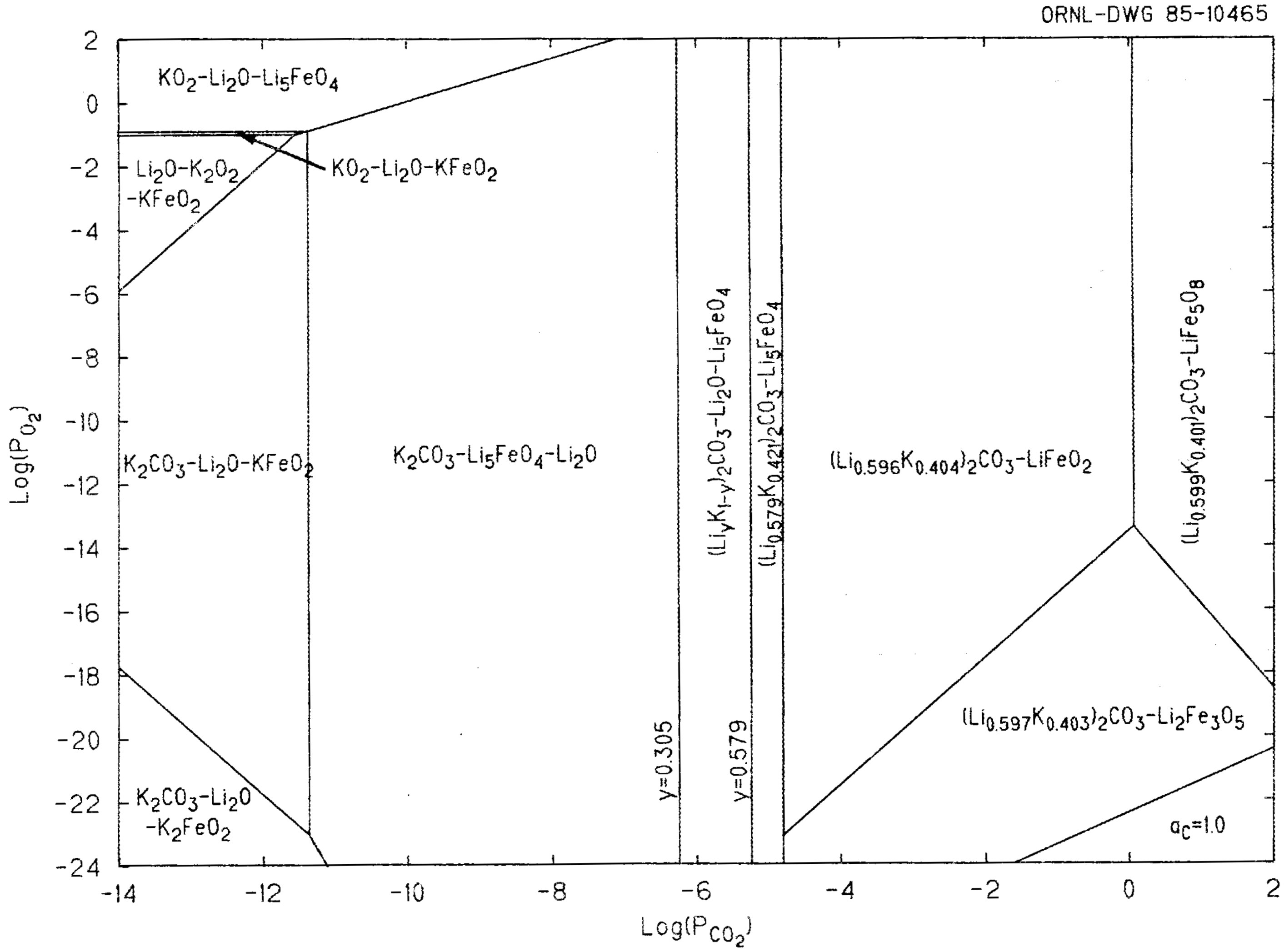

Fig. 2. Phase stability diagram of the Fe-Li-K-C-O system under CO_2-O_2-Ar gas mixtures at 650°C. The ratio of Li:K is 6:4, and the ratio of (Li+K):Fe is 100:1.

ratios of (Li+K):Fe, but Fig. 2 is generally suitable for use with melts where the ratio of (Li+K):Fe > 100:1, although small deviations in locations of the phase boundaries are to be expected. In this study, Fe_2O_3 powder mixed with $(Li_{0.62}K_{0.38})_2CO_3$ was fixed at the ratio of (Li+K):Fe = 450:1. This concentration of iron was sufficient to saturate the carbonate mixture with dissolved $LiFeO_2$ and to form solid $LiFeO_2$ in equilibrium with the carbonate solution. Meanwhile, the iron content was low enough that the concentration of carbonate did not deviate appreciably from its eutectic composition, $(Li_{0.62}K_{0.38})_2CO_3$, when $LiFeO_2$ formed.

Results of solubility measurements for $LiFeO_2$ as a function of P_{CO_2} in $(Li_{0.62}K_{0.38})_2CO_3$ at 650°C with P_{O_2} of 2×10^4 and 10^2 Pa (0.2 and 0.001 atm) are shown in Fig. 3. The plot shows no trend in the dependence of $LiFeO_2$ solubility in the carbonate mixture on P_{CO_2} and P_{O_2}.

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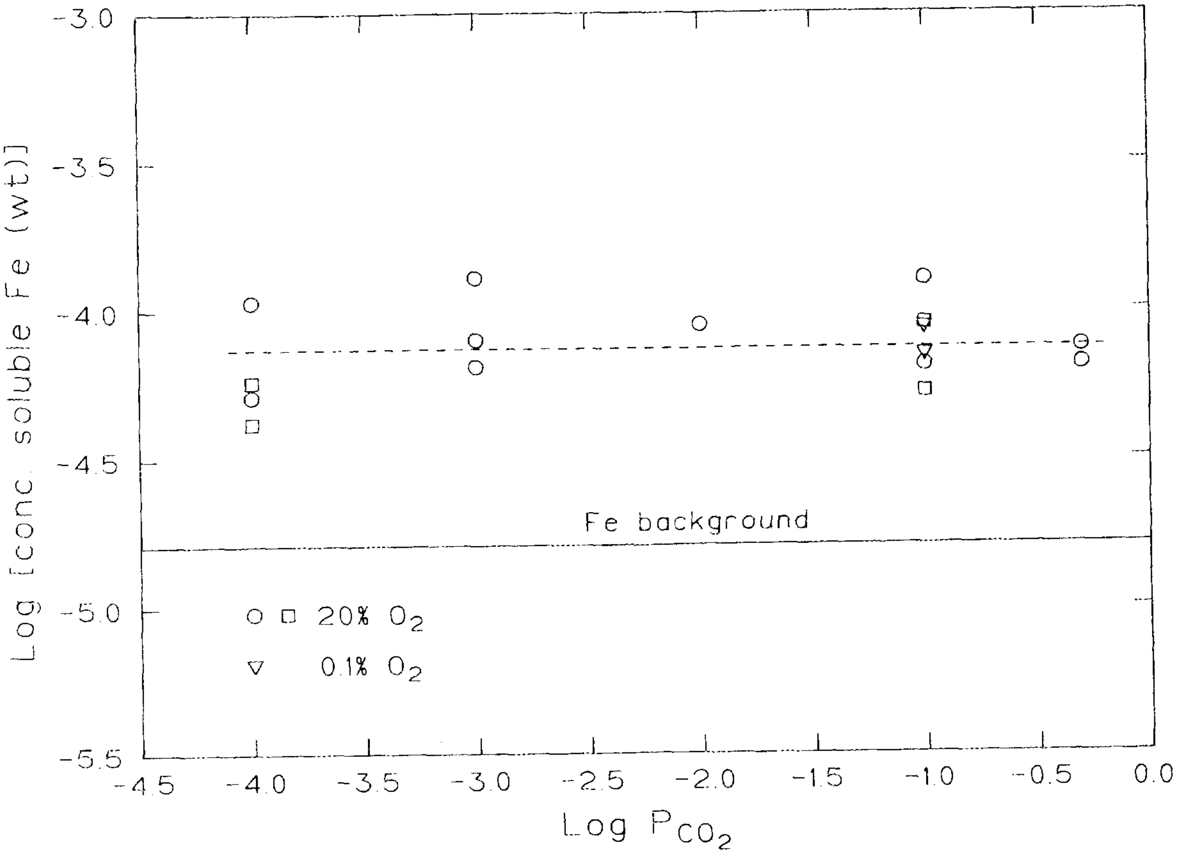

Fig. 3. Equilibrium solubility of $LiFeO_2$ in $(Li_{0.62}K_{0.38})_2CO_3$ at 650°C, where open circles and open squares represent two separate experiments under $P_{O_2} = 2 \times 10^4$ Pa (0.2 atm).

The average solubility of $LiFeO_2$ in $(Li_{0.62}K_{0.38})_2CO_3$ in the range of 5×10^4 and 10 Pa (0.5 to 10^{-4} atm) of P_{CO_2} at both of the above oxygen partial pressures is 78 wppm of iron with ±26 wppm standard deviation. The impurity level of iron in the carbonate mixture was measured to be 16 wppm. Although Baumgartner et al. have reported $LiFeO_2$ solubilities in the carbonate mixture between 550 and 850°C under 29% CO_2-14% O_2-3% H_2O-N_2 (ref. 10), it is difficult to compare the present results with their data because of the different experimental systems and procedures for the respective solubility studies. Under their experimental conditions, solute concentration in the carbonate mixture did not decrease with time up to 500 h, even though the solubility of $LiFeO_2$ (extrapolated from results at temperatures above 700°C) was predicted to be lower than the iron impurity concentration in the nascent carbonate. It was concluded that the system was not at equilibrium. Therefore, we believe that some agitation of the carbonate melts and gas bubbling through the melts are necessary to assure the equilibrium for the solubility study.

According to the phase stability diagram of the Fe-Li-K-C-O system at 650°C, three potential processes for the dissolution of $LiFeO_2$ can be expressed as follows:

Acidic dissolution

$$5\ LiFeO_2(s) + 2\ CO_2 \rightarrow \underline{LiFe_5O_8} + 2\ Li_2CO_3\ ; \qquad (1)$$

Basic dissolution

$$LiFeO_2(s) + 2\ Li_2CO_3 \rightarrow \underline{Li_5FeO_4} + 2\ CO_2\ ; \qquad (2)$$

Physical dissolution

$$LiFeO_2(s) \rightarrow \underline{LiFeO_2}\ . \qquad (3)$$

Since both acidic and basic dissolution processes are P_{CO_2} dependent, the solubility of $LiFeO_2$, under conditions where it is controlled by either the acidic or basic dissolution process or by both acidic and basic dissolution processes in the carbonate mixture, must also be P_{CO_2} dependent. Similar arguments have been used to explain the solubilities of Fe_2O_3 and Cr_2O_2 as a function of melt basicity in fused Na_2SO_4 at 1200°C (refs. 12 and 13). However, the solubility of $LiFeO_2$, as shown in Fig. 3, is independent of P_{CO_2} and P_{O_2}. The dissolution of $LiFeO_2$ in the carbonate mixture at 650°C must, therefore, be controlled by the physical dissolution process. The solid $LiFeO_2(s)$ is dissolved as $\underline{LiFeO_2}$ molecules or complexes in the carbonate mixture, with no chemical reaction between $LiFeO_2(s)$ and $(Li,K)_2CO_3$. According to the criterion for hot corrosion fluxing suggested by Rapp and Goto,[6] a gradient in the solubility across the carbonate film on $LiFeO_2$ leads to the dissolution and reprecipitation of iron or iron oxides and accelerates the corrosion reaction. Since the solubility of $LiFeO_2$ in the carbonate mixture is independent of P_{CO_2} and P_{O_2}, the chemical stability of the $LiFeO_2$-$(Li,K)_2CO_3$ mixture at 650°C under the indicated partial pressures of CO_2 and O_2 reduces the tendency for hot corrosion to occur.

From the phase stability diagram, $Li_2Fe_3O_5$ is stable under reducing gas environments. In other words, under fuel cell operating conditions, $LiFeO_2$ dissolved in the carbonate electrolyte under cathode gas environments does not reduce to highly conductive metallic iron under anode gas environments. Thus, $LiFeO_2$ remains a good candidate for a MCFC cathode material. However, the solubility of $Li_2Fe_3O_5$ in the carbonate mixture is expected to be different from that of $LiFeO_2$, since some fluxing of dissolved iron-containing species in the carbonate electrolyte can occur when $LiFeO_2$ is used as cathode material in the MCFC system.

According to the Li_2CrO_4-K_2CrO_4 phase diagram,[14] liquid $(Li,K)_2CrO_4$ is stable at 650°C when the mole fraction of K_2CrO_4 is less than 0.77. The Li_2CrO_4-K_2CrO_4 system was treated as a Henrian solution in the liquid state during the thermochemical calculations for establishing phase relationships in the Cr-Li-K-C-O system. In addition, the mutual solubility between $(Li,K)_2CO_3$ and $(Li,K)_2CrO_4$ was assumed to be negligible. The calculated phase stability diagrams of the Cr-Li-K-C-O system at 650°C are shown in Figs. 4 and 5. In both figures the ratio of Li:K is 6:4 and the ratios of (Li+K):Cr are 10:1 and 1:1, respectively.[4] According to these phase stability diagrams, a mixture of solid K_2CrO_4 and liquid $(Li,K)_2CO_3$ or a mixture of liquid $(Li,K)_2CrO_4$ and liquid $(Li,K)_2CO_3$ is stable under oxidizing gas environments, while a mixture of $LiCrO_2$ and $(Li,K)_2CO_3$ is predominant in reducing gas environments. Although the composition of $(Li,K)_2CrO_4$ that

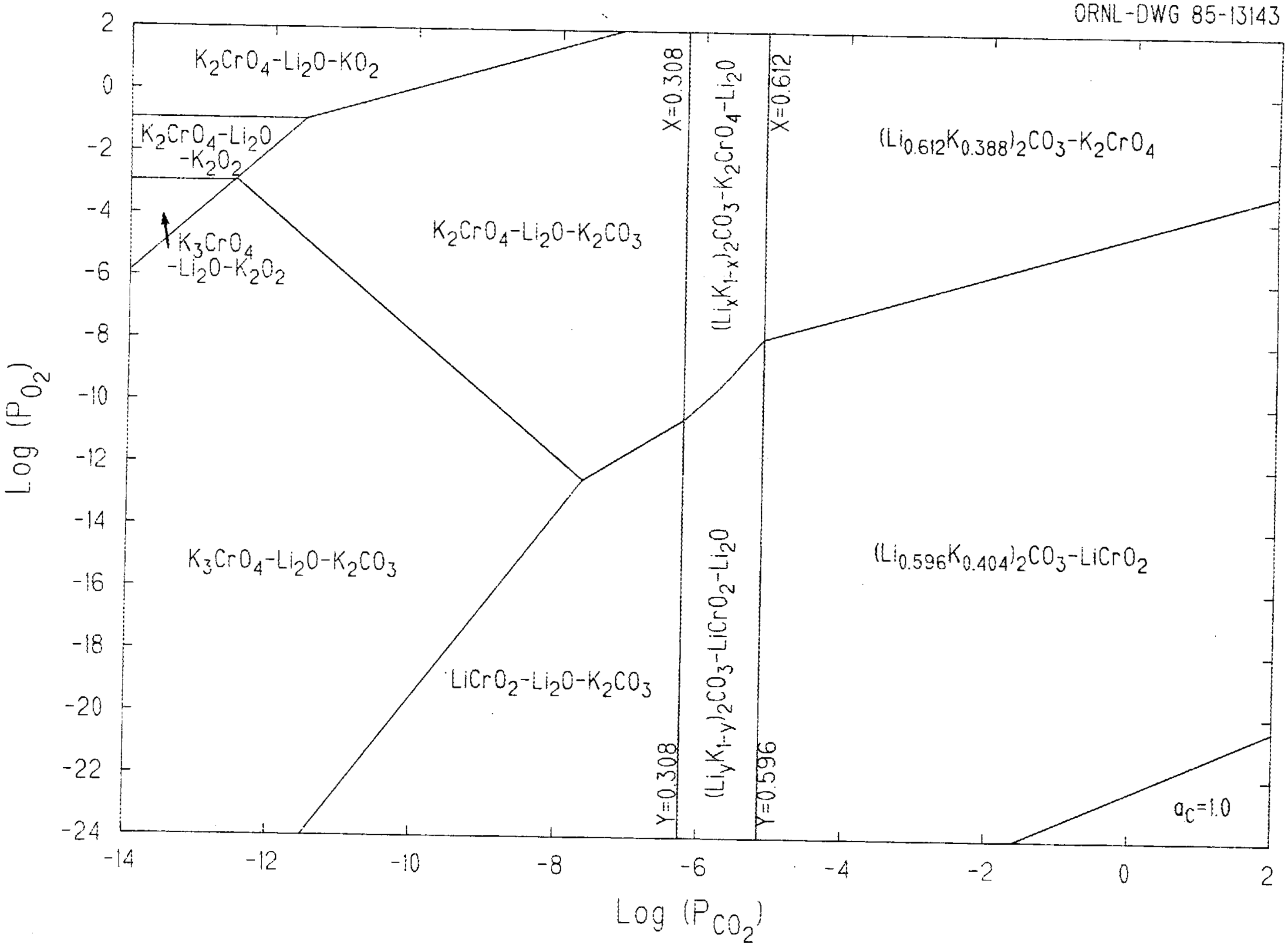

Fig. 4. Phase stability diagram of the Cr-Li-K-C-O system under CO_2-O_2-Ar gas mixtures at 650°C. The ratio of Li:K is 6:4 and the ratio of (Li+K):Cr is 10:1.

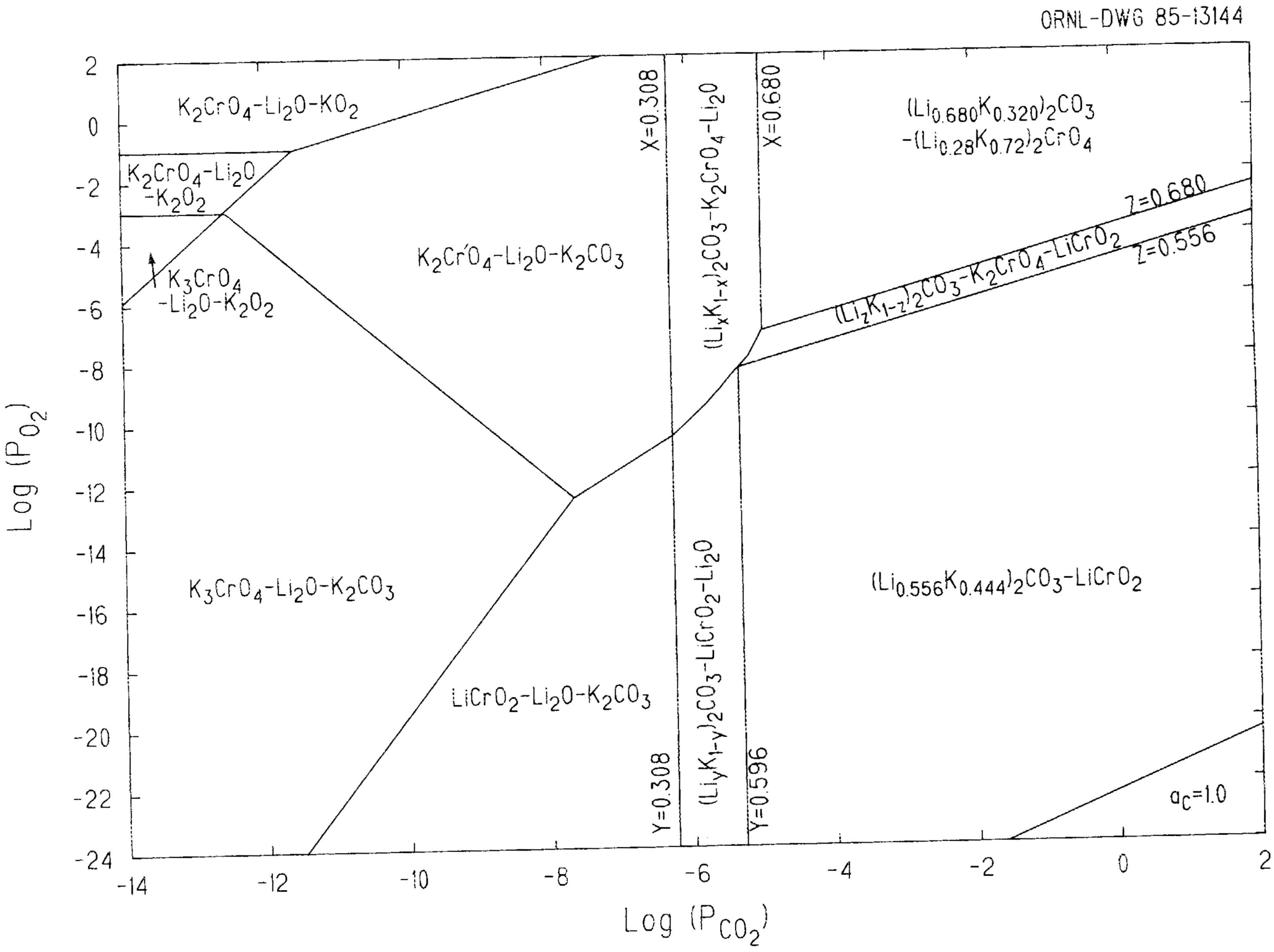

Fig. 5. Phase stability diagram of the Cr–Li–K–C–O system under CO_2–O_2–Ar gas mixtures at 650°C. The ratio of Li:K is 6:4 and the ratio of (Li+K):Cr is 1:1.

equilibrates with $(Li_{0.62}K_{0.38})_2CO_3$ is still uncertain, the formation of the liquid phase of $(Li,K)_2CrO_4$ from the reaction of Cr_2O_3 with $(Li,K)_2CO_3$ has been confirmed from our previous work.[3]

The concentration of dissolved chromium in $(Li,K)_2CO_3$ determined under oxidizing gas environments at 650°C increased linearly as the reaction time increased. Some results of the concentration of dissolved chromium as a function of reaction time are shown in Fig. 6. Since a study of the dissolution kinetics of chromate into the carbonate melt was not included in the scope of research, no attempt was made to control gas flow rate, surface area of the carbonate melt, etc., which would have had some effects on the dissolution rate of $(Li,K)_2CrO_4$ in $(Li,K)_2CO_3$. Although gas streams with the same composition were passed over the melt surface and bubbled through the melt to increase the rate of the dissolution reaction, the measurements of chromate in solution at 650°C did not reach equilibrium over 35 days.

To avoid the long periods of time that were required to achieve equilibrium solubility at 650°C, a mixture of Cr_2O_3 powder and $(Li,K)_2CrO_4$ was held at 750°C for one week and then at 650°C for an additional two to three weeks. The carbonate was sampled, and the concentration of dissolved chromium in $(Li,K)_2CO_3$ was measured. Then, additional Cr_2O_3 powder was added to the carbonate melt. The carbonate melt was again subjected to the heating procedure described above. The equilibrium solubility of $(Li,K)_2CrO_4$ in $(Li,K)_2CO_3$ was determined to have been attained when the concentration of chromium measured in the carbonate did not increase with additional Cr_2O_3 powder. Results of equilibrium solubility of $(Li,K)_2CrO_4$ in $(Li,K)_2CO_3$ under 20% O_2–(0.01–10%) CO_2–Ar at 650°C are shown in Fig. 7. The equilibrium solubility of $(Li,K)_2CrO_4$ in the carbonate melt is 13.7 wt % of chromium [or 30.4 mole % of $(Li_{0.62}K_{0.38})_2CrO_4$ in $(Li_{0.62}K_{0.38})_2CO_3$], which is independent of partial pressures of CO_2 at $P_{O_2} = 2 \times 10^4$ Pa (0.2 atm) with a 10^5-Pa (1.0-atm) total pressure.

According to the phase stability diagram of the Cr-Li-K-C-O system at 650°C, two potential dissolution processes for $(Li,K)_2CrO_4$ in $(Li,K)_2CO_3$ can be expressed as follows:

Acidic dissolution

$$2\ Li_2CrO_4 + CO_2 \rightarrow 2\ \underline{LiCrO_2} + Li_2CO_3 + 3/2\ O_2\ , \tag{4}$$

$$2\ K_2CrO_4 + Li_2CO_3 + CO_2 \rightarrow 2\ \underline{LiCrO_2} + 2\ K_2CO_3 + 3/2\ O_2\ \ ; \tag{5}$$

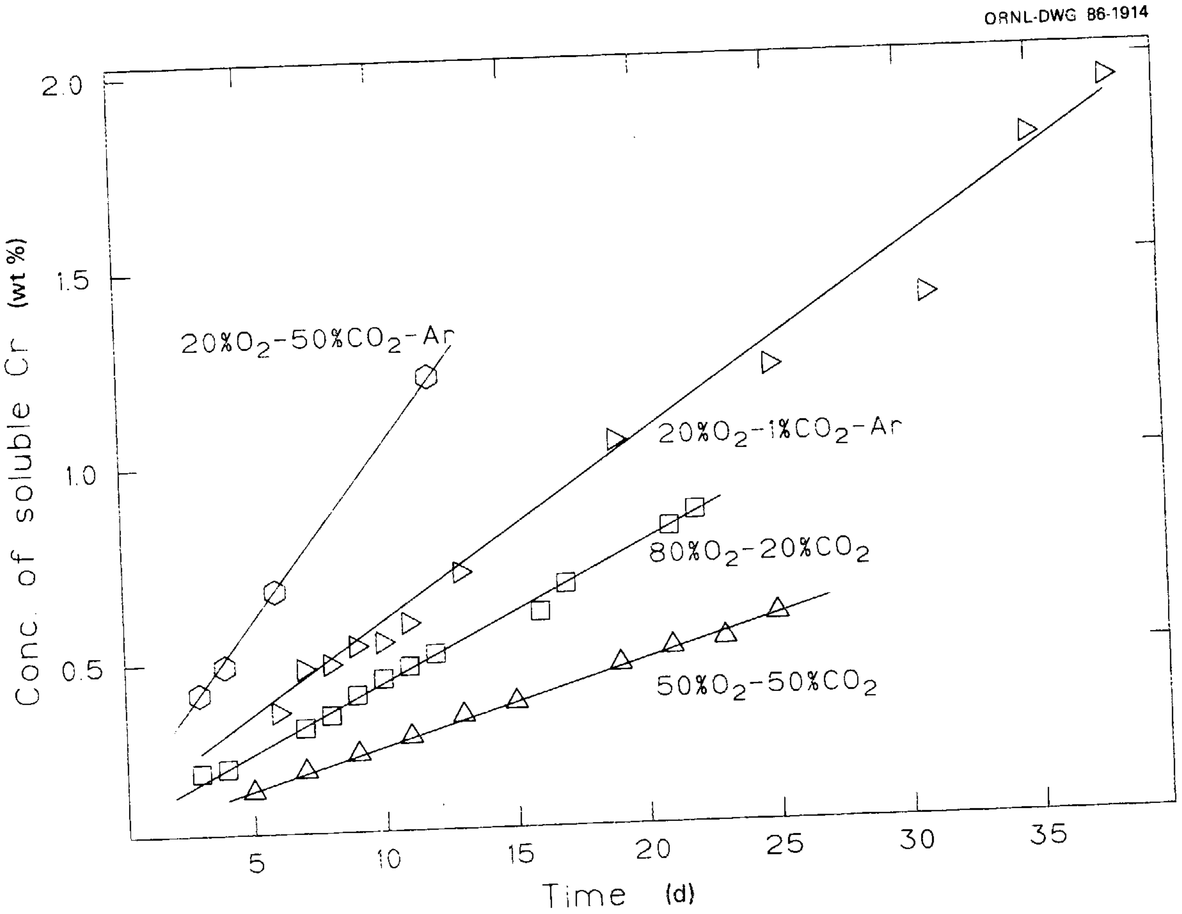

Fig. 6. Solubility of $(Li,K)_2CrO_4$ in $(Li_{0.62}K_{0.38})_2CO_3$ at 650°C.

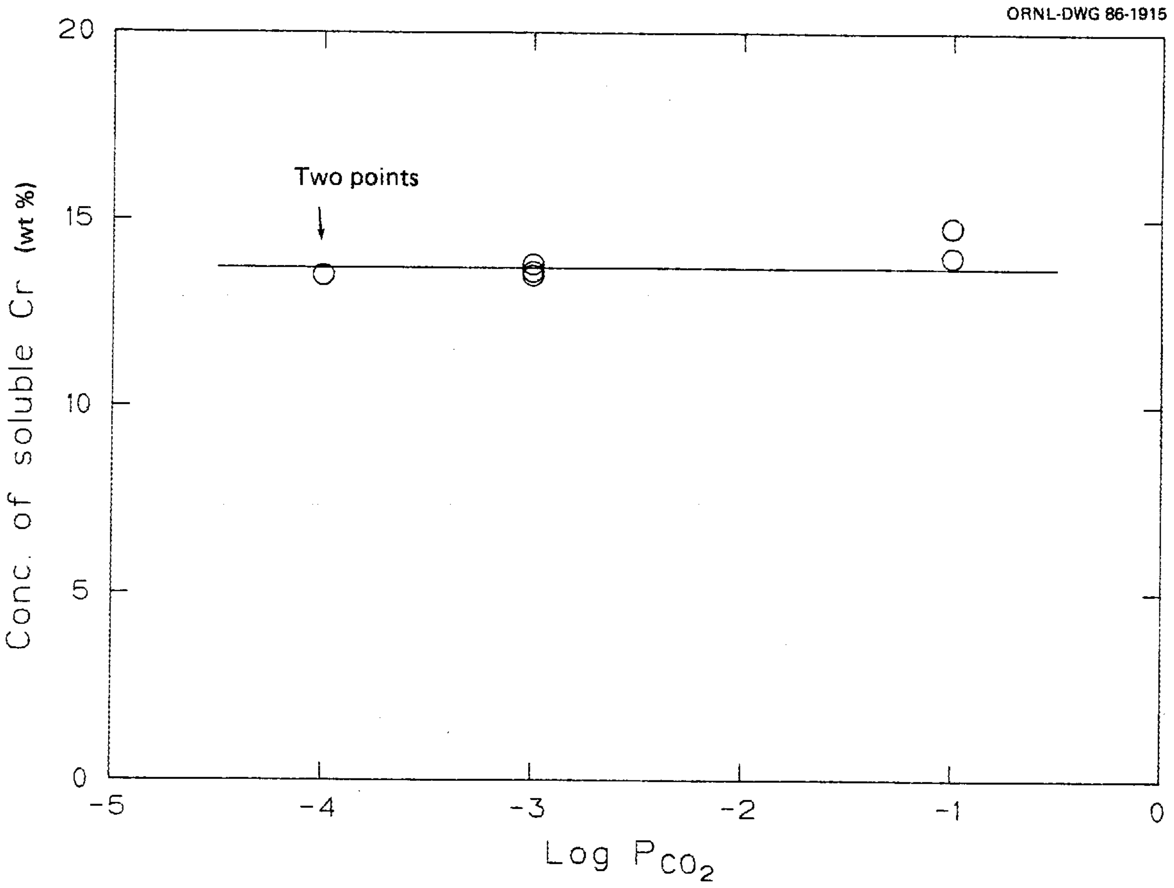

Fig. 7. Equilibrium solubility of $(Li,K)_2CrO_4$ in $(Li_{0.62}K_{0.38})_2CO_3$ at 650°C and $P_{O_2} = 2 \times 10^4$ Pa (0.2 atm).

Physical dissolution

$$(Li,K)_2CrO_4 \rightarrow \underline{(Li,K)_2CrO_4} \quad . \qquad (6)$$

Since the acidic dissolution process is (P_{CO_2} − P_{O_2}) dependent, the dissolution of $(Li,K)_2CrO_4$ in $(Li,K)_2CO_3$ must proceed according to a physical dissolution process as shown in Eq. (6). In addition, the oxidation state of dissolved chromium in the carbonate mixture determined by a colorimetric absorption spectrophotometer was +6, which is only consistent with the oxidation state of chromium in $\underline{(Li,K)_2CrO_4}$.

Although the solubility of $(Li,K)_2CrO_4$ in $(Li,K)_2CO_3$ under oxidizing gas atmospheres was high, the solubility of $LiCrO_2$ under reducing gas atmospheres was very low. The solubility of $LiCrO_2$ in the carbonate mixture under 50% CO_2–1.8% CO–Ar ($\log P_{O_2} = -20$) was below 10 wppm chromium at 650°C. Thus, pure chromium cladded on type 310 stainless steel was used as the anode current collector material in the prototypical fuel cell. Nevertheless, under oxidizing gas environments chromium was continuously leached from chromium-containing materials.[10] Because of the extremely high solubility of $(Li,K)_2CrO_4$ in the carbonate melt, chromium from chromium-containing material or chromium reaction products from the corrosion of chromium-containing alloys should be excluded from direct contact with the alkali carbonates in MCFCs under oxidizing gas environments.

SUMMARY

According to phase stability diagrams of Cr-Li-K-C-O and Fe-Li-K-C-O systems at 650°C, both $LiFeO_2$ and $(Li,K)_2CrO_4$ are stable phases resulting from the reaction of $(Li_{0.62}K_{0.38})_2CO_3$ and metal oxides under oxidizing gas environments of MCFCs. The equilibrium solubilities of $LiFeO_2$ and $(Li,K)_2CrO_4$ in $(Li_{0.62}K_{0.38})_2CO_3$ were determined to be 77 wppm of iron and 13.7 wt % of chromium, respectively. Because the dissolution of both $LiFeO_2$ and $(Li,K)_2CrO_4$ in the carbonate mixture is independent of P_{CO_2} and P_{O_2}, the fluxing mechanism proposed for the molten salt corrosion will not occur during the corrosion of iron and chromium with carbonate deposits under oxidizing gas environments. From this solubility study, $LiFeO_2$

remains a good candidate for a MCFC cathode material. However, chromium from chromium-containing materials or its corrosion products should be prevented from directly contacting the carbonate electrolyte of MCFCs under oxidizing gas environments.

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