An improved test fixture for biaxial-tension strength testing of ceramics featuring uniform pressure loading of disks was developed and qualified. Biaxial data were obtained for an alumina ceramic, along with comparable uniaxial data from three- and four-point flexure tests. Weibull statistics provided a good description of the size effect on data from the two uniaxial tests, but underestimated the effect of stress biaxiality. The biaxial strengthening observed in the alumina ceramic is consistent with that observed previously in a glass-ceramic.

I. Introduction

Precise evaluation and correlation of strengths in different stress states are prerequisites for developing a sound structural-design technology for brittle materials. This paper is one in a series evaluating a number of strength-testing schemes that generate different principal-stress ratios. The resulting data are being evaluated as part of a study of multiaxial-stress effects on ceramic strengths. Literature data are often conflicting and do not provide simple, consistent rules for predicting biaxial strengths from uniaxial strengths. Often, strength values for the same stress state obtained by different test techniques do not agree; this was noted, for example, by Petrovic and Stout in their recent survey of ceramic-strength data in tension-compression stress states. Leaving aside experimental inadequacies, which may well be responsible for some of the conflicts in data, two factors may influence the relation between biaxial and uniaxial strengths of ceramics:

1. The effective flaw populations subject to stress may be different in uniaxial- and biaxial-strength tests, and this condition can lead to differences in strengths due to statistical effects (Weibull explanation).

2. Critical flaws in ceramics may not be ideal Griffith cracks, in which case the flaw severity may depend on the loading geometry.

Among recent studies that have examined biaxial and uniaxial strengths of ceramics, Giovan and Stein found that strength of an alumina ceramic in biaxial tension was described adequately by a statistical fracture theory that used a shear-sensitive-fracture criterion. Petrovic and Stout, in combined tension-torsion studies of Al₂O₃ rods, noted an increase in the tensile principal failure stress with increasing compressive principal-stress component, in qualitative agreement with the Weibull theory. The magnitude of this increase was, however, greater than the Weibull prediction. The present authors, in an experimental study of a glass-ceramic that did not exhibit size dependence of strengths in uniaxial flexure, obtained strengths in balanced biaxial tension that were greater than uniaxial-flexure strengths. The magnitude of this strength difference decreased with increasing slow crack growth associated with the strength tests. These results were consistent with the idea of strength control by cracks associated with blunt surface flaws.

In the present paper, fracture strengths of an alumina ceramic in biaxial tension are examined. Biaxial tension is conveniently obtained in flexure schemes that subject disk specimens to lateral, radially symmetrical bending forces. In the biaxial fracture study of the glass-ceramic, a ball-loading scheme was evaluated in detail. Although the ball-on-ring test has several advantages, chief among them being its minimal requirements for test fixtures, the ball loading generates steep stress gradients parallel to the specimen face and stresses only a small area of the disk specimen. For this reason, a hydraulic-pressure-loading fixture was constructed to provide a biaxial-tension-strength test in which the effective stressed area of the specimen is comparable to the conventional uniaxial-flexure tests, such as four-point bend tests. This paper discusses development of the test apparatus, analysis of bending stresses, formulations of fracture statistics based on both Weibull methodology and its variations, and some preliminary strength results obtained on a commercial, sintered alumina.

II. Uniform-Pressure-on-Disk Test

The hydraulic pressure loading test was apparently first used in the British glass industry for strength testing of plate glass. More recently, it has been used to evaluate residual strengths of glass and ceramic specimens following impact with liquid jets. In essence, the test consists of loading a disk specimen, which is supported along a concentric line support near its periphery, with lateral uniform pressure. A self-contained hydraulic system was designed and constructed for this purpose.

(1) Test Apparatus

Figure 1 is a schematic diagram of the cross section of the hydraulic test cell designed for uniform pressure loading of disk specimens. The test specimen (31.75 mm in diam. by 2.5 mm thick) was supported on 31 ball bearings, 2.4 mm in diam., placed along a circle 25.4 mm in diam. The ball bearings were attached to each other by a flexible polymer web. This support arrangement was required in order to eliminate friction that usually is encountered in rigid-ring supports. Constant-velocity motion of a plunger applied uniformly rising pressure over the lower face of the specimen through a spring-steel diaphragm, 0.25-mm thick, by direct compression of mercury in the cell. Mercury was chosen as the test fluid because of its low compressibility. The pressure rise was monitored through a pressure transducer. The test cell was designed for a maximum pressure of 30 MPa, which would impose a tensile stress of about 750 MPa for the specimen geometry used in this test. A filling tube and valve (not shown in the figure) were also connected to the mercury chamber. The test-fixture assembly was designed for mounting on the crosshead of a universal testing machine so that a constant displacement rate of the plunger and, therefore, a constant pressure-rise rate was obtained by the crosshead motion. A maximum crosshead-displacement rate of 50 mm/min resulted in a pressure-rise rate of 10 MPa/s.
(2) Stress Distributions

For disk specimens with ratios of thickness, \( t \), to diameter, \( d \), in the range \( 0.01 < (t/d) < 0.1 \), stresses in the specimen are adequately described by simple-plate theory. The stress distribution is axisymmetric, and the following relations apply for the radial, \( \sigma_r \), and tangential, \( \sigma_\theta \), stress components on the tension surface bounded by the support circle:

\[
\sigma_r = \frac{3p r^2}{8t^2} \left[ 2(1-\nu) + (1+3\nu) \left( \frac{r_2}{r_1} \right)^2 - 4(1+\nu) \left( \frac{r_2}{r_1} \right)^2 \ln \left( \frac{r_2}{r_1} \right) \right] + \sigma_0 \tag{1}
\]

and

\[
\sigma_\theta = \frac{3p r^2}{8t^2} \left[ 2(1-\nu) + (1+3\nu) \left( \frac{r_2}{r_1} \right)^2 - 4(1+\nu) \left( \frac{r_2}{r_1} \right)^2 \ln \left( \frac{r_2}{r_1} \right) \right] + \sigma_0 \tag{2}
\]

where \( p \) is the uniform pressure applied on the specimen surface, \( r_1 \) the radius of the support circle, \( r_2 \) the disk-specimen radius, \( r \) the radial position where the stresses are evaluated, and \( \nu \) Poisson's ratio for the specimen material; \( \sigma_0 \) is a correction for finite plate thickness given by Timoshenko and Woinowsky-Krieger as

\[
\sigma_0 = \left[ (3+\nu)p/4(1-\nu) \right] \tag{3}
\]

It results from a correction to the plate thickness as given by the simple-plate-bending theory by considering the effects of shearing stresses and lateral pressures on the plate deflection.

The accuracy of the analytical solutions was examined by comparing the stresses predicted by Eqs. (1) and (2) with those measured by experiments using strain gages. For this purpose, an AISI 4340 steel disk specimen (31.75 mm in diam. by 2.5 mm thick) heat-treated for high yield strength (1400 MPa) was strain-gaged at five radial locations to read both radial and tangential strains. Measured strains, both radial and tangential, varied linearly with pressure for maximum strains up to 0.0015, a range that covers fracture strains of most ceramics. Measured strains per unit pressure, obtained from least-squares linear regression of strain-pressure data, were converted to radial and tangential stresses per unit pressure using the elastic properties of the steel, Young's modulus, \( E = 203.4 \) GPa, and Poisson's ratio, \( \nu = 0.3 \). Figure 2 shows plots of the normalized stresses as functions of normalized radial distance \( (r/r_1) \). The solid lines are from Eqs. (1) and (2), whereas the points are from experiments. The agreement is within 3% at all locations, which is considered to be sufficient verification of the applicability of Eqs. (1) and (2) for describing the stresses on the uniform-pressure-loaded disk specimen. Analysis of stresses by finite-element methods and strain-gage measurements on ceramic specimens are currently under way; they will be reported in a future paper.

Figure 2 shows that the tangential and radial stresses are within 3% of one another and within 5% of the maximum within a circular area whose radius is 20% of the support circle.

III. Fracture Statistics

Most brittle materials exhibit a dispersion in fracture stresses of nominally identical test specimens due to the variability in the severity of the strength-controlling flaws. Weibull,\(^1\) in his statistical theory of the strength of materials, described this dispersion in terms of a cumulative distribution function:

\[
F = 1 - \exp \left[ -B \right] \tag{4}
\]

where \( F \) is cumulative probability of fracture and \( B \) risk of rupture.

Weibull's formulation of the risk of rupture for uniaxial stress is straightforward and generally well accepted. But his formulation for multiaxial-stress states is not readily accepted, and several variations have been suggested.\(^{15,18}\) To analyze the data obtained by pressurized-disk loading, the original Weibull treatment and some of the variations suggested since were examined.

(1) Weibull Analysis

For multiaxial-stress states, Weibull defined the risk of rupture at any point in the stressed body as

\[
\frac{dB}{d\omega} = n(\sigma_\omega) d\omega \tag{5}
\]

where \( n(\sigma_\omega) \) is a characteristic material function; \( \sigma_\omega \) normal tensile stress at an arbitrary angle relative to the principal stresses \( \sigma_1, \sigma_2, \) and \( \sigma_3 \); and \( d\omega \) an elemental area on a unit solid sphere. The geometric variables used to describe \( \sigma_\omega \) and \( d\omega \) are defined in Fig. 3. In terms of the angular coordinates, \( \phi \) and \( \psi \), \( \sigma_\omega \) and \( d\omega \) are given by

\[
\sigma_\omega = \cos^2 \phi (\sigma_1 \cos^2 \psi + \sigma_2 \sin^2 \psi + \sigma_3 \sin^2 \phi) \tag{6}
\]

and

\[
d\omega = \cos \phi d\phi d\psi \tag{7}
\]
those portions of the unit-sphere surface where

Weibull assumed that the risk of rupture was zero for orientations both a two-parameter and a three-parameter form for the material function, \( r^2=15.0 \). For a three-point-flexure specimen, the equations that describe the risk of rupture, \( B_f \), and the loading factor, \( L_f \), are

\[
B_f = (bl+lt)(\sigma_f/\sigma_0)^a L_f
\]

(13)

and

\[
L_f = \frac{b(m^2+1)(m+1)+lt(m_l^2+1)}{(m+1)^2(bl+lt)}
\]

(14)

where \( a \) is loading span and \( \sigma_f \), maximum outer-fiber stress.

For the uniform-pressure-on-disk specimen, the stress state is biaxial tension, with \( \sigma_1=\sigma_r=\) radial stress, \( \sigma_2=\sigma_t=\) tangential stress, and \( \sigma_3=0 \); the risk of rupture, \( B_r \), for failure from surface flaws, Eq. (9), takes the form

\[
B_r = (bl+lt)(\sigma_f/\sigma_0)^a L_r
\]

(10)

The variation of the radial and tangential stress, given by Eqs. (1) and (2) and shown in Fig. 2, are of the form

\[
\sigma_r = \sigma_0 (1 - \alpha(r/r_1)^2)
\]

(16)

and

\[
\sigma_t = \sigma_0 (1 - \beta(r/r_1)^2)
\]

(17)

where \( \sigma_0 \) is maximum stress at the center of the disk specimen (Eqs. (1) and (2) for \( r=0 \))

\[
\alpha = \frac{3\sigma_0}{r_1^2} \nonumber
\]

(18)

\[
\beta = \frac{3\sigma_0}{r_1^2} \nonumber
\]

(19)

Substituting Eqs. (16) and (17) in Eq. (15) leads to the following solutions for the risk of rupture, \( B_{ai} \), and the loading factor, \( L_{ai} \), for the uniform-pressure-on-disk specimen:

\[
B_{ai} = (\sigma_0^2)(\sigma_0/\sigma_0)^a L_{ai}
\]

(20)

and

\[
L_{ai} = \frac{2(2m^2+1)}{\pi} \int_{x=0}^{x=\pi/2} \int_{\phi=0}^{\phi=\pi/2} \cos^{m+1} \phi d\phi \phi d\phi \]

(21)

where \( x = (r/r_1) \).

(2) Barnett-Freudenthal Approximation

Weibull's method of treating the risk of rupture in multiaxial-stress states is not based on a specific, theoretical rationale, and yet it involves rather tedious calculations, especially when dealing with practical structures with continuously varying stress states and stress gradients.\(^{17}\) Barnett et al.\(^{11}\) and Freudenthal\(^{12}\) suggested an alternative, simple approximation for handling multiaxial fracture statistics. In this approach, referred to in the literature as the Barnett-Freudenthal approximation,\(^{12}\) the principal stresses are assumed to act independently. i.e., the survival probability of the test material under multiaxial-stress state is the product of the probabilities of survival under the principal stresses applied independently. This assumption leads to the following equation for the risk of rupture in biaxial tension in the uniform-pressure-on-disk test specimen for surface-flaw failures.
B_{w} = 2\pi \left[ \int_{r_{a}}^{r_{b}} (\sigma_{r}/\sigma_{0})^{m} r dr + 2\pi \int_{r_{a}}^{r_{b}} (\sigma_{\theta}/\sigma_{0})^{n} r dr \right] (22)

Unlike Eqs. (15) and (21) that require numerical integration, Eq. (22) can be integrated in closed form giving

\[ B_{w} = (\pi r_{m}^{2})(\sigma_{r}/\sigma_{0})^{m} L_{w2} \] (23)

where

\[ L_{w2} = \frac{\alpha + \beta}{(m+1) \alpha \beta} \] (24)

The fracture statistics for the different strength tests can be compared in a number of ways. The loading factor is one convenient parameter for this purpose, because it incorporates the stress-state and stress-gradient effects on the fracture statistics, while separating the absolute specimen size effects. Figure 4 shows plots of the loading factors for the uniform-pressure-on-disk specimen and the three-point-flexure specimen as a function of the Weibull modulus, m. Loading factors for volume-flaw failures also are included in this figure. Development of the equations for the risks of rupture and the loading factors for volume-flaw failure is similar to the formulations given above for the surface-flaw failures. The stress gradients through the thickness of the test specimens were assumed to be linear in every case; final equations obtained for volume-flaw failures are presented in the Appendix.

For clarity, the loading factors for the four-point-flexure specimens are not included. It should also be noted that, for the biaxial test specimen, loading factors were calculated using both the Weibull formulation (Eqs. (21) and A–6) and the Barnett-Freudenthal approximation (Eqs. (24) and A–8).

A loading factor of unity corresponds to a direct-tension test specimen. As is seen in Fig. 4, loading factors for both uniaxial- and biaxial-flexure specimens decrease with increasing Weibull modulus, reflecting the influence of stress gradients. The biaxial-flexure specimen, of course, has higher loading factors than does the uniaxial three-point specimen. Also, higher loading factors result from the Weibull formulation than from the Barnett-Freudenthal approximation. This behavior results from the fact that the calculation of risk of rupture in the Weibull formulation involves an additional integration over the unit hemispherical surface. Thus, the Weibull treatment results in a more conservative prediction of biaxial strengths than does the Barnett-Freudenthal approximation. The volume-loading factors are, of course, smaller than the surface-loading factors because of the additional stress gradient in the thickness direction.

### IV. Fracture Strengths of Alumina

The uniform-pressure-on-disk test and the three-point and four-point uniaxial-flexure tests were used to evaluate and correlate strengths of a commercially available sintered alumina. This ceramic was chosen for study because an earlier investigation designed to limit errors from spurious stresses to less than 1% of the bending-formula stresses. Strength values from the three-point tests were expected to be $=2\%$ greater than the actual values because of the wedging effect. The uniform-pressure-on-disk tests were conducted on disks 31.8 mm in diam. by 2.5 mm thick. All tests, uniaxial and biaxial, were conducted in a dry N₂ environment at a stress rate of 250 MPa/s, conditions chosen to minimize slow crack growth effects on strengths.

#### (2) Strength Tests

Uniaxial-flexure tests were conducted on 2.5 by 5.0 by 38 mm specimens. The specimen support span was 31.8 mm and the loading span in the four-point test was 19 mm. The fixtures used in these tests have been described in the literature. They were designed to limit errors from spurious stresses to less than 1% of the bending-formula stresses. Strength values from the three-point tests were expected to be $=2\%$ greater than the actual values because of the wedging effect.

#### (3) Fracture Morphology and Fractography

Fracture surfaces of all strength-tested specimens were examined microscopically. In each case, fracture initiated at the surface and cursory fractography revealed no apparent differences in origin regions among the different specimens. In the uniaxial tests, fracture origins were randomly distributed on the tension face. No preferential edge failures were observed. A typical fracture pattern in a uniform-pressure-on-disk test specimen of alumina is shown in Fig. 5. The arrow in the figure indicates the grinding direction on
the tension surface. Fracture generally initiated in an annular region near the center, reflecting a statistical area distribution of the strength-controlling, "worst" flaws. Fracture initially followed a plane; after a characteristic "jog" was formed, multiple crack branching occurred, resulting in the fracture pattern shown in Fig. 5. This fracture morphology is similar to that observed in our earlier investigation of the glass-ceramic in the ball-on-ring test. The number of crack branches was greater in the uniform-pressure-on-disk test, presumably because of the greater amount of strain energy stored in this test specimen for the same nominal maximum stress. Note that the plane of the jog was at an angle to the grinding direction. This behavior indicates that the critical flaws were not preferentially oriented along the grinding direction. The jogs in different specimens were oriented at different angles with respect to the grinding direction.

Figure 6 shows the fracture surface of the test specimen shown in Fig. 5, as viewed normal to the jog. The jog length on the tension face is indicated by the arrows. The fracture origin is at the surface, approximately in the center of the jog. Uniformly distributed, grain-boundary pores also were observed on the fracture surface.

(4) Statistical Analysis

Figure 7 shows Weibull plots of fracture stresses, i.e., \( \ln(1/1-F) \) vs \( \ln \sigma \), where \( F \), the failure probability, was defined by the relation

\[
F = (i-0.5)/N
\]

(25)

where \( i \) is ranking number of a specimen in a sample size \( N \) in increasing order of fracture stress, \( \sigma \).

The fracture stress plotted in Fig. 7 was the maximum stress at failure calculated from the conventional bending formulas for the uniaxial-flexure test specimens and Eq. (1) for the biaxial test specimens. The linearity of these plots indicates that the two-parameter form of the Weibull distribution function is adequate to describe the observed dispersions of fracture stress.

The slopes of the Weibull plots, i.e., the values of the Weibull modulus, \( m \), determined from linear regression by the least-squares method, are nearly identical — \( m = 23.8 \) for the four-point-flexure specimens, 23.4 for the three-point-flexure specimens, and 22.0 for the biaxial-flexure specimens. Since the fracture stresses of the longitudinal and transverse specimens were very close, the isotropy of fracture behavior was verified. Those two observations confirmed that a single flaw population was sampled in all three tests. Mean strengths were 349, 400, and 376 MPa, respectively, for four-point, three-point, and biaxial-flexure test specimens.

The critical test of any statistical theory is its ability to predict both specimen-size and stress-state effects on strength. For this purpose, the two Weibull parameters, \( m = 23.8 \) and \( \sigma_0 = 242 \) MPa, obtained from the four-point-flexure-strength data and Eqs. (13), (14), and (4), were used to predict three-point-flexure strengths (size effect) and biaxial-flexure strengths (stress-state effect). The predicted strength distributions are indicated by the solid lines in Fig. 7 and are identified by the risk-of-rupture formulations used to derive them. The predicted three-point-flexure strengths (identified by \( B_f \)) were \( 2 \% \) less than the measured values and this difference could well have been due to the above-noted wedging effect. Therefore, the statistical theory adequately described the specimen-size effect. The measured biaxial strengths were, however, significantly greater than the statistically predicted values. The measured median strength was 12% greater than the prediction based on the Weibull formulation (identified by \( B_n \)) and 5% greater than the prediction based on the Barnett-Freudenthal approximation (\( B_B \)).

V. Discussion

The objectives of this paper were to report on the development and qualification of a biaxial-flexure-strength test, discuss the application of Weibull statistics to treat the biaxial-strength data, and present a preliminary examination of the adequacy of Weibull statistics to account for both size and stress-state effects on the strengths of a sintered alumina.

The uniform-pressure-on-disk test described in this paper, although developed for the specific purpose of examining the relations between uniaxial and biaxial strengths, has several attractive features that make it a convenient test for routine evaluation of ceramic strengths. These features may be itemized as follows:

(1) Edge failures, common in direct-tensile and uniaxial-bend tests, are eliminated because stresses are nearly zero along the disk-specimen periphery.

(2) Hydraulic loading used in the test eliminates stress concentrations usually encountered in mechanical-loading schemes, such as ring-on-ring loading.

(3) The stress distribution in the disk specimen can be described accurately by simple-plate-bending-theory equations.

(4) Weibull statistics are easily formulated for this biaxial-strength test because of the simple form of the stress variations in the disk specimen.

(5) The principal stresses in the specimen surface are nearly equal over a large area near the center of the disk. Thus, the probability of failure at a certain flaw is not a sensitive function of its orientation to the applied stress field.

(6) The test procedure is simple and rapid, allowing testing of the many specimens that are usually required for statistical analyses.

(7) Environment and stressing rate, two variables that can significantly influence ceramic strengths, are easily controlled.

The statistical analyses of the preliminary strength data obtained for the alumina ceramic indicated that Weibull statistics adequately accounted for the size effect on strength in uniaxial tension, but underestimated strength in biaxial tension. This result can be compared with those obtained by Giovan and Sines and Petrovic and Stout, the two recent studies of biaxial-fracture strengths of alumina ceramics. Giovan and Sines compared four-point-bend strengths and strengths obtained in biaxial-flexure by ring-on-ring loading. They compared the measured biaxial-flexure strengths with those predicted from fracture statistics using four-point-bend-strength data as the baseline. The Weibull formulation underestimated the biaxial strengths; this result is consistent with the present finding. The Barnett-Freudenthal approximation overestimated their biaxial-fracture strengths, in contrast to the present finding. They obtained best agreement when a shear-sensitive fracture criterion was used to replace the normal stress (Griffith) criterion in their fracture statistics.

Petrovic and Stout compared maximum, principal tensile stresses at failure, measured for alumina rods under combined tension and torsion, with predictions of the Weibull theory. Weibull theory underestimated the increase in the principal failure stress with increasing torsion loading.

One common conclusion is apparent in all three studies: Weibull's formulation of fracture statistics for biaxial-stress states overestimates the risk of rupture and failure probability or underestimates the biaxial-fracture stress for a given failure probability. A number of explanations are possible for this discrepancy between the Weibull theory and the experimental results. Weibull's
rationale for extending the fracture statistics to multiaxial-stress states may not be valid or the failure criterion implicitly assumed in the Weibull theory, i.e. a normal-stress failure criterion, may be inadequate. Giovan and Sines' invoked these arguments to explain their strength results. But these arguments cannot completely explain our results. Statistical fracture theories that employ a shear-sensitive fracture criterion would definitely be an improvement over Weibull theory, but they would still underestimate the biaxial strengths observed in the present study. The present authors, in a study of a glass-ceramic that did not exhibit size dependence of strengths in uniaxial tension (i.e. the material did not exhibit scatter of strengths in the Weibull sense), rationalized the increased strengths in biaxial tension in terms of a biaxial-stress-field effect on the flaw severity. The stress intensity of strength-controlling surface flaws can be sensitive to in-plane stress fields under certain conditions of geometrical configuration of these flaws. The present results on alumina are consistent with the glass-ceramic results. Petrovic and Stout suggested that a material's intrinsic resistance to fracture may change in different stress states because of possible influence of the applied stress fields on such energy-dissipating processes as microcracking and process-zone formation. Duckworth and Rosenfield examined this explanation quantitatively and showed that it can lead to higher strengths in tension-compression stress states but generally predicts weakening in the tension-tension stress states.

It is clear from the above discussion that a number of factors can enter into the relations between uniaxial and biaxial strengths of ceramics. Precise evaluation of ceramic strengths in different stress states and critical examination of the strength data in the light of the predictions of fracture statistics and mechanics theories are important steps in sorting these factors. It is hoped that the strength test described in this paper will significantly contribute to generating a data base for such evaluations.

**APPENDIX**

The following equations for risk of rupture and loading factors for volume-flaw failures can be derived following the procedures discussed in the text for surface flaws.

**Three-Point Flexure**

\[ B_3 = \frac{b_d t}{2} \left( \frac{\sigma}{\sigma_0} \right)^m L_3 \]  
\[ L_3 = \frac{1}{(m+1)^2} \]  
\[ (A-1) \]

**Four-Point Flexure**

\[ B_4 = \frac{b_d t}{2} \left( \frac{\sigma}{\sigma_0} \right)^m L_4 \]  
\[ (A-3) \]

\[ L_4 = \left( \frac{a^2 + 1}{m+1} \right)^2 \]

\[ (A-4) \]

**Uniform-Pressure-on-Disk**

\[ B_{u1} = (\pi r^2/2)(\sigma/\sigma_0)^m L_{u1} \]
\[ (A-5) \]

where

\[ L_{u1} = \frac{2(2m+1)}{\pi(m+1)} \int_{\phi=0}^{\phi=\frac{\pi}{2}} \cos^{2m+1}{\phi} \int_{\phi=-\frac{\pi}{2}}^{\phi=\frac{\pi}{2}} (1 - \alpha x^2 \cos^2 \phi - \beta x^2 \sin^2 \phi)^{m+1} dx \]
\[ \left( \frac{a}{b} \right)^2 \frac{1}{\sigma_0} \alpha \beta \left( \frac{a}{b} \right)^2 \frac{1}{\sigma_0} \alpha \beta \]
\[ (A-6) \]

\[ B_{u2} = (\pi r^2/2)(\sigma/\sigma_0)^m L_{u2} \]
\[ (A-7) \]

where

\[ L_{u2} = \frac{1}{(m+1)^2} \frac{d\sigma_f}{d\beta} \]
\[ (A-8) \]

**Acknowledgments:** The authors thank W. Schaezenberg and the staff of the machine shop at Battelle's Columbus Laboratories for their assistance in the design and construction of the test apparatus and S. Sampath and C. Tsay for helpful discussions on the stress analysis of the disk specimen.

**References**

Electrical Conduction in Co_{1-x}Mg_{x}O

KUNIHITO KOUMOTO,* KEIKO YAMAYOSHI, and HIROAKI YANAGIDA*

Department of Industrial Chemistry, Faculty of Engineering, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo, Japan

113

Electrical conductivity and thermoelectric power were measured on Co_{1-x}Mg_{x}O (0.16<x<0.87) at 900°C to 1200°C. Electrical conductivity \( \sigma \) decreased and the activation energy increased with increasing MgO content. Seebeck coefficient \( \alpha \) appeared to be positive for all \( x \). The difference in temperature dependence of \( \sigma \) and \( \alpha \) was 0.2 to 0.5 eV and the hole mobility decreased with increasing MgO content. The results were interpreted as showing that electron holes were localized to cation clusters and their thermally activated hopping conduction was operative in the solid solution. The dominant lattice defect changed from singly to doubly ionized cobalt vacancy with increasing MgO content.

I. Introduction

The defect structure of nonstoichiometric compounds and the mechanism of valence control by doping with small amounts of additives have been extensively investigated. In the present study, CoO and MgO were chosen as a typical \( p \)-type semiconductor and an insulator, respectively, to investigate the electrical conduction mechanism for the solid solution Co_{1-x}Mg_{x}O (0.16<x<0.87). This solid solution material is reported to be a candidate for an oxygen sensor and a thermionic fuel for an isotopic power source.

Table I. Characteristics of Sintered Specimens Used for Electrical Measurements and Apparent Activation Energy for Electrical Conduction

<table>
<thead>
<tr>
<th>( x ) in Co_{1-x}Mg_{x}O</th>
<th>Lattice constant (nm)</th>
<th>Relative density (%)</th>
<th>Average grain size (( \mu m ))</th>
<th>Single crystal*</th>
<th>Apparent activation energy</th>
<th>Polycrystal*</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.16</td>
<td>0.4254</td>
<td>80</td>
<td>20</td>
<td>0.53</td>
<td>0.55±0.01</td>
<td></td>
</tr>
<tr>
<td>0.25</td>
<td>0.4249</td>
<td>86</td>
<td>16</td>
<td>0.62</td>
<td>0.60±0.01</td>
<td></td>
</tr>
<tr>
<td>0.43</td>
<td>0.4241</td>
<td>93</td>
<td>12</td>
<td>0.73</td>
<td>0.60±0.02</td>
<td></td>
</tr>
<tr>
<td>0.65</td>
<td>0.4230</td>
<td>94</td>
<td>10</td>
<td>1.05</td>
<td>0.82±0.02</td>
<td></td>
</tr>
<tr>
<td>0.87</td>
<td>0.4220</td>
<td>95</td>
<td>7</td>
<td>1.26</td>
<td>1.10±0.01</td>
<td></td>
</tr>
</tbody>
</table>

*Evaluated from \( \Delta H_f \) (enthalpy of defect formation) and \( \Delta H_a \) (hopping energy) reported in Ref. 19. *Present study.

There have been a number of studies on pure and doped CoO\(^{4-8}\) and MgO\(^{9-16}\). For the solid solution between the two oxides, the defect structure and the conduction mechanism have been discussed by several investigators. Zintl\(^{17}\) determined the nonstoichiometric deviation for the composition \( x=0.6 \). Schier and coworkers\(^{18,19}\) concluded from the electrical conductivity and tensivolumetric measurements that an activated hopping conduction of electron holes was operative for all the compositions. Park and Logothetis\(^{20}\) measured the electrical conductivity and thermoelectric power and proposed a mechanism changing from band to hopping conduction with increasing content of MgO. The reason for this discrepancy is not clear, since such conflicting results for pure CoO have also been obtained by many workers.\(^{21,22}\) However, the present measurements on conductivity and thermoelectric power revealed that an activated hopping process was dominant in the solid solution, which is consistent with the results obtained by Schier and coworkers. The defect structure of the solid solution was also clarified from the oxygen partial pressure dependences of both conductivity and Seebeck coefficient.

II. Experimental Procedure

Magnesium oxide (99.999% pure) and CoCO\(_3\)\(_6\)H\(_2\)O (99.999% pure) were used as starting powders. They were mixed in a desired proportion and heated in a platinum crucible at 1000°C for 2 h, crushed, mixed with ethanol in an alumina mortar, and calcined further at 1000°C for 2 h. The calcined powders were again crushed and cold-pressed into a rectangular bar (5 by 5 by 15 mm) under 225 MPa pressure. The pressed body was covered with