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ABSTRACT

January 1969

The effect of the limitation of Griffith flaws, introduced in an abraded surface of glass, on the strength of glass-tungsten composites was investigated for small sizes on Griffith flaws. Hasselman and Fulrath's postulate was found to extend to small flaw sizes.

It was found that the use of tungsten spheres with two particle sizes or a wide distribution about one average size as the dispersed phase in a glass matrix decreased the average mean free path in the matrix phase considerably more than could be achieved with a single particle size dispersion.

Experimentally determined average mean free paths by statistical techniques were in good agreement with that calculated by Fullman's equation. This technique was successfully used when the dispersed phase had two particle sizes or a wide particle size distribution. The strong effect of internal stresses due to a mismatch of thermal expansion of the two phases in reducing the strength of a composite was demonstrated.

This work was done under the auspices of the United States Atomic Energy Commission.

The authors are graduate student and professor, Lawrence Radiation Laboratory, and Department of Materials Science and Engineering, University of California, Berkeley, California 94720.
I. INTRODUCTION

In 1966 Hasselman and Fulrath\(^1\) developed a theory for the strengthening of glass containing a dispersed phase. This hypothesis was based on the premise that the dispersed phase could limit the average Griffith flaw size which would be produced in the surface of a severely abraded composite. When alumina microspheres were dispersed in a glass matrix it was found that the theory could be experimentally confirmed. In their work they used a relation developed by Fullman\(^2\) to calculate the average mean free path between dispersed phase particles. This average mean free path was then considered to be the average Griffith flaw size and would therefore determine the composite strength. Because different abrasion techniques would produce different flaw sizes in the glass matrix, the relation relating strength to flaw size was not followed when the calculated mean free path was greater than a given value for a specific system. Therefore, a plot of the composite strength vs the reciprocal square root of the average mean free path as determined by the Fullman equation showed two regions.

In the first region, the strength of the composite was relatively independent of the calculated mean free path. In the second region, the predicted relation was followed.

The use of the Fullman equation requires that the size and volume fraction of spherical dispersed phase particles be known. Therefore, the systems which can be experimentally investigated are limited to composites with a single particle size as the dispersed phase. This limitation and the forming process (vacuum hot pressing) used, limited the volume fraction of dispersed phase material that could be incorporated
into a glass matrix to approximately 50 v/o.

This study was undertaken to, first, experimentally confirm the Hasselman and Fulrath postulate to smaller Griffith flaw sizes and second, to achieve higher volume fractions of the dispersed phase in a glassy matrix than previously reported. Both of these objectives should be attained if the dispersed phase consists of two or more particle sizes.
II. EXPERIMENTAL PROCEDURE

A. Materials

Three glass compositions were used in this study. All were in the soda borosilicate system and were chosen because of their thermal expansion coefficients. Table I lists the three compositions used, along with the measured density and thermal expansion coefficient. The glasses were compounded using sodium carbonate, boric acid and silica flour. The compositions were mixed, melted in platinum, and ground to -400 mesh.

Tungsten was chosen for the dispersed phase because microspheres in a wide size range are commercially available. Size fractions were obtained by screening or air elutration. The average particle sizes of the separated fractions were measured by optical microscopy.

B. Specimen Fabrication

The powdered glass and tungsten microspheres were weighed to the desired volume composition, mixed, and loaded into graphite dies. The composites were vacuum hot pressed in equipment previously described. Both surfaces of the fabricated composite (one inch diameter by 1/4 inch thick) were ground flat with 400 grit SiC. The compact density was measured by immersion in ethyl alcohol.

The disks were then cut into bars approximately 0.050 inches thick by 1/4 inch wide on a precision diamond saw. The as-sawn surface was stressed in tension on a 4-point loading device with a span of 0.75 inches. The average modulus of rupture was calculated from the results of eight or nine specimens of each composition.
C. Determination of Mean Free Path

Because the Fullman expression is not applicable when a composite contains two or more size fractions, the determination of the mean free path in the glassy matrix was done by statistical methods.

Random sections were cut through the composite system. The surface was then ground and polished. A photomicrograph was then taken and the photograph enlarged to eight inches by ten inches. Following the techniques outlined by Underwood et al., random lines of a given length were drawn on the photograph. The number of spherical particles intercepted by each line was counted. The average value of \( n \), the number of particles intercepted per line of length, \( L \), was determined from the average of 30 to 40 lines on sections through each composition.

The average mean free path, \( \lambda \), was determined from the relation

\[
\lambda = \frac{1-V_v}{N_L} \tag{1}
\]

where

- \( V_v \) = the volume fraction of the dispersed phase
- \( N_L \) = average number of particles intersected by unit length of line.

A typical photograph used is shown in Fig. 1. Where spherical cavities existed due to particle pullout during grinding and polishing, the cavity was assumed to be a particle.
III. RESULTS AND DISCUSSION

A. Relation of Calculated to Measured Mean Free Paths

In the previous work of Hasselman and Fulrath,\(^1\) all mean free paths in the matrix of the composite were calculated from the Fullman equation. To compare the Fullman equation with Eq. (1), a single size fraction varying volume fraction series was fabricated and the average mean free path calculated and experimentally determined by Eq. (1). The results are given in Table II. There appears to be good agreement between the two values, with the experimentally determined mean free path always less than that calculated. As the vol fraction of dispersed phase is increased, the difference between the two values of mean free path decreases. This is probably due to better statistics being achieved in the experimental method when \(N_L\) increases with either increased volume fraction or decreased particle size. Further, the Fullman equation assumes one particle size whereas in any real system there exists a size distribution. This leads to an uncertainty in the calculated average mean free path.

Because this study was to concentrate on increased volume fraction loadings of a dispersed phase and smaller average mean free paths than previously reported, it was concluded that the statistical method reported by Underwood\(^4\) could be used successfully for the average mean free path.

B. Density of Glass-Tungsten Composites

In previously reported work\(^1,5,6\) on glass matrix composites formed by vacuum hot pressing, porosity was introduced into the system at approximately 50 v/o dispersed phase. All previous systems used one dispersed phase particle size. This is in agreement with Sohn's\(^7\)
findings that high packing densities of powders cannot be achieved unless special vibration techniques are used for packing. Two methods are available to increase particle packing density. First is to use mixed size fractions and second, use one size with a large size distribution. Systems of both types were fabricated and the results shown in Fig. 2. (Dotted curve is predicted from Hasselman's work for a particle size of 15μ or less.) As shown in the figure, increased loadings can be achieved in either case. However, the maximum volume fraction of dispersed phase that can be incorporated into a glass matrix is far less than the maximum packing density that can be achieved and has been shown in particle packing studies. In vacuum hot pressing with no deformation of the crystalline dispersed phase and starting with a glass powder, the techniques adopted in particle packing studies to achieve high packing density cannot be used. Therefore, the maximum densities reported in this study are limited by fabrication techniques.

C. Strength of Glass-Tungsten Composites

The average mean free path of each of the compositions of Ny glass-tungsten shown in Table III was determined by the method described in Section II, C. The strength is plotted in Fig. 3 according to the postulate of Hasselman and Fulrath.1

In the figure, only those composites with less than 3 v/o porosity are plotted. Those composites with greater than 3 v/o porosity were drastically weakened as was found by Bertolotti and Fulrath.9 The behavior of these composite systems with closely matching thermal expansion is exactly the same as that observed for the Al2O3-D glass composites reported previously. However, this study has extended the average mean
free path to lower values.

The separation between Regions I and II occurred at a measured mean free path of 50µ. Using the elastic modulus of glass as 8.7 x 10.6 psi (as measured by frequency resonance method\(^1\)) and the dynamic surface energy of 2500 ergs/cm\(^2\) in the Griffith equation\(^1\) gives a Griffith flaw of approximately 57µ.

The slope of the line in Region II can be used as shown by Hasselman and Fulrath to calculate the approximate dynamic surface energy. Again using the appropriate elastic modulus as given by Hasselman and Fulrath for tungsten-glass composites,\(^1\) the dynamic surface energy for fracture can be calculated as 2300 ergs/cm\(^2\). These results correlate remarkably well for a brittle matrix composite system.

Using glasses with thermal expansion coefficients differing from that of tungsten, the composites fabricated showed the same relative mechanical behavior. It is apparent that strengthening is considerably reduced in these systems. The strength of composites with high and low expansion glasses relative to tungsten are given in Table III and plotted in Figs. 4 and 5 as strength vs square root of calculated mean free path, d, for single particle size dispersion. When the same volume fraction and particle sizes of tungsten are dispersed in each of the three glasses and the resultant strength plotted against the thermal expansion of the matrix phase (Fig. 6), it is apparent that internal stress is contributing to lower the strength as reported by Fulrath.\(^1\) The behavior of different glasses with tungsten dispersion is similar to that shown by D. B. Bins\(^1\) for glass-alumina and glass-zircon composites.
IV. SUMMARY AND CONCLUSIONS

As a result of this work, it has been shown that the experimental
determination of the average mean free path in a dispersed phase com­
posite system agrees well with that calculated by the Pullman\textsuperscript{2} equation.
Further, this method can be used when the dispersed phase consists of
two particle sizes or a wide distribution about one average size.

The systems studied experimentally extended the postulate of
Hasselman and Fulrath\textsuperscript{1} to smaller Griffith flaw sizes. By using three
different glass compositions, the strong effect of internal stresses in
reducing the strength of a composite has been demonstrated.
ACKNOWLEDGMENTS

Thanks are extended to M. A. Stett for helpful discussions and to G. Dahl for technical assistance.

This work was done under the auspices of the United States Atomic Energy Commission.
REFERENCES


## TABLE I

Properties and Compositions of Materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Composition (wt %)</th>
<th>Thermal expansion* (in/in°C x 10⁶)</th>
<th>Density (gm/cm³)</th>
<th>Hot pressing Temp. (°C)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ny glass</td>
<td>SiO₂:65.0</td>
<td>4.9</td>
<td>2.20</td>
<td>690</td>
</tr>
<tr>
<td></td>
<td>Na₂O:8.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>B₂O₃:26.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>D glass</td>
<td>SiO₂:70.0</td>
<td>7.7</td>
<td>2.46</td>
<td>660</td>
</tr>
<tr>
<td></td>
<td>Na₂O:16.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>B₂O₃:14.0</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>N₄ glass</td>
<td>SiO₂:67.0</td>
<td>3.25</td>
<td>2.16</td>
<td>640</td>
</tr>
<tr>
<td></td>
<td>Na₂O:4.5</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>B₂O₃:28.4</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Tungsten</td>
<td></td>
<td>4.6</td>
<td>19.3</td>
<td></td>
</tr>
</tbody>
</table>

*All the values of thermal expansion are given in the range of room temperature to 450°C.*
### TABLE II

Calculated and Measured Average Mean Free Paths

<table>
<thead>
<tr>
<th>Composite Composition</th>
<th>Average Particle Size $\mu$</th>
<th>$\lambda$ Cal. $\mu$</th>
<th>$\lambda$ Meas. $\mu$</th>
<th>Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>5 v/o Al$_2$O$_3$ *</td>
<td>82</td>
<td>1038</td>
<td>800</td>
<td>22%</td>
</tr>
<tr>
<td>10 v/o Nickel *</td>
<td>95</td>
<td>570</td>
<td>472</td>
<td>20%</td>
</tr>
<tr>
<td>20 v/o Al$_2$O$_3$ *</td>
<td>78</td>
<td>208</td>
<td>170</td>
<td>18%</td>
</tr>
<tr>
<td>30 v/o Al$_2$O$_3$ *</td>
<td>78</td>
<td>121</td>
<td>104</td>
<td>14%</td>
</tr>
<tr>
<td>30 v/o Tungsten</td>
<td>75</td>
<td>116</td>
<td>100</td>
<td>13.5%</td>
</tr>
<tr>
<td>30 v/o Tungsten</td>
<td>50</td>
<td>78</td>
<td>70</td>
<td>10%</td>
</tr>
<tr>
<td>40 v/o Tungsten</td>
<td>50</td>
<td>50</td>
<td>46</td>
<td>8%</td>
</tr>
<tr>
<td>50 v/o Tungsten</td>
<td>50</td>
<td>33</td>
<td>30.5</td>
<td>7.5%</td>
</tr>
</tbody>
</table>

* Composites from previously reported work.
### TABLE III

Cross-Bending Strength of Sodium Borosilicate Glass Matrices Containing Dispersed Spherical Tungsten Particles

<table>
<thead>
<tr>
<th>Tungsten Particle Size (μ)</th>
<th>Volume Fraction W (φ)</th>
<th>Matrix</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>50</td>
<td>7900</td>
<td>8700</td>
</tr>
<tr>
<td></td>
<td>(4.3)</td>
<td>(8.2)</td>
</tr>
<tr>
<td>70 + 10</td>
<td>8650</td>
<td>9725</td>
</tr>
<tr>
<td></td>
<td>(6.2)</td>
<td>(8)</td>
</tr>
<tr>
<td>50 + 6</td>
<td>10,600</td>
<td>12,150</td>
</tr>
<tr>
<td></td>
<td>(5.5)</td>
<td>(4.2)</td>
</tr>
<tr>
<td>3 to 13</td>
<td>16,900</td>
<td>18,800</td>
</tr>
<tr>
<td></td>
<td>(4.5)</td>
<td>(5.5)</td>
</tr>
<tr>
<td>50</td>
<td>7120</td>
<td>7050</td>
</tr>
<tr>
<td></td>
<td>(6.3)</td>
<td>(5.3)</td>
</tr>
<tr>
<td>20</td>
<td>8300</td>
<td>9247</td>
</tr>
<tr>
<td></td>
<td>(9.5)</td>
<td>(3.3)</td>
</tr>
<tr>
<td>50</td>
<td>2700</td>
<td>2725</td>
</tr>
<tr>
<td></td>
<td>(4.9)</td>
<td>(12.5)</td>
</tr>
<tr>
<td>20</td>
<td>7480</td>
<td>7000</td>
</tr>
<tr>
<td></td>
<td>(7.6)</td>
<td>(13)</td>
</tr>
</tbody>
</table>

Matrix:
- Ny glass (Strength = 8250 psi)
- N4 glass (Strength = 7900 psi)
- D glass (Strength = 7050 psi)

All bend strengths are given in psi. Numbers in parenthesis represent the percentage standard deviation. The strengths were determined from 7 to 9 breakings per sample.
Fig. 1. A typical photomicrograph of a glass and tungsten composite, containing 63.2\% tungsten spheres of sizes 70 and 10\mu (72\% coarse + 28\% fine). Black spheres are pull outs during polishing.
PARTICLE SIZE DISTRIBUTION (3 TO 13μ)

TWO PARTICLE SIZES OF TUNGSTEN (50μ + 7μ)

TWO PARTICLE SIZES OF TUNGSTEN (70μ + 10μ)

FIG. 2 MEASURED DENSITY OF THE Ny GLASS AND TUNGSTEN COMPOSITES PLOTTED AS % OF THEORETICAL DENSITY AGAINST VOLUME FRACTION OF TUNGSTEN.
FIG. 3 EXPERIMENTAL DATA FOR THE STRENGTH OF NY GLASS AND TUNGSTEN COMPOSITES PLOTTED AGAINST RECIPROCAL SQUARE ROOT OF MEASURED MEAN FREE PATH.
FIG. 4 EXPERIMENTAL DATA FOR THE STRENGTH OF D GLASS AND TUNGSTEN COMPOSITES PLOTTED AGAINST RECIPROCAL SQUARE ROOT OF MEAN FREE PATH.
FIG. 5 EXPERIMENTAL DATA FOR THE STRENGTH OF N₄ GLASS AND TUNGSTEN COMPOSITES PLOTTED AGAINST RECIPROCAL SQUARE ROOT OF MEAN FREE PATH.
FIG. 6 EFFECT OF THERMAL EXPANSION OF THE GLASS ON THE STRENGTH OF COMPOSITES CONTAINING 40% TUNGSTEN SPHERES.
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