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STRENGTH OF INTERNALLY STRAINED BRITTLE MATRIX COMPOSITES

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ABSTRACT

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The effect of internal stresses on the mechanical strength of brittle matrix composites where a good chemical bond existed between the matrix and dispersed phase was investigated. By dispersing spherical Al_2O_3 particles in glasses of varying thermal expansion coefficients, a range of internal stresses was set up in the composites. Uniaxial strengths of the crystal-glass bodies were approximately all the same. Therefore, it is concluded that internal stresses do not affect the strength of well bonded, brittle-matrix composites.

I. INTRODUCTION

Composite materials are of considerable technological importance as structural materials. Fiber-reinforced composites have many industrial applications because of their high strength-to-weight ratios. Glass-ceramics, a new development in the materials field, can be classified as brittle-matrix composites. They are being used in numerous applications because they can be formed economically by conventional glass forming techniques.

For a number of years, brittle-matrix composites have been studied at this laboratory in order to understand better the mechanical properties of multiphase bodies. A model system consisting of a continuous glass matrix with a metal or oxide phase dispersed throughout the matrix has been used in all investigations. Such a system can be fabricated by vacuum hot pressing, utilizing the viscous nature of glass, to give a dense composite. The main parameters which have been found to affect the strength of such systems are: (1) the volume fraction and size of the dispersed phase, (2) the micromechanical stress concentrations developed upon loading, and (3) the degree of chemical bonding between the glass and dispersed phase.

One parameter which has not been resolved as to its effect on strength is internal stress. This stress is set up in the brittle-matrix composite upon cooling from the fabrication temperature, when there is a difference in thermal expansion coefficient of the dispersed and continuous phase. Figure 1 is a simplified model of the internal stress produced in the matrix when a sphere is introduced into a continuous phase. In case I, where the thermal expansion coefficient of the matrix

is less than that of sphere, radial tensile and tangential compressive stresses are set up in the matrix at the interface. In case II, the tensile and compressive stresses are reversed. These stresses can be calculated by an equation developed by Selsing[1] which applies for a crystal of a high modulus of elasticity embedded in a low modulus glass:

$$P = \frac{\Delta\alpha\Delta T}{\frac{1+\nu_m}{2E_m} + \frac{1-2\nu_D}{E_D}} \qquad (1)$$

$$P_r = 2P_t = -P.\frac{R^3}{r^3} \qquad (2)$$

where P = external hydrostatic pressure exerted on spherical particle

P_r = radial stress in matrix

P_t = tangential stress in matrix

R = radius of particle

r = distance from a point in matrix to center of sphere

$\Delta\alpha$ = difference in thermal expansion between matrix and particle

ΔT = cooling range over which matrix can support a stress

ν_m = Poisson's ratio of matrix phase

ν_D = Poisson's ratio of dispersed phase

E_m = Young's modulus of matrix phase

E_D = Young's modulus of dispersed phase

Fulrath[2] was the first to measure internal stresses in brittle-matrix composites by X-ray diffraction techniques. He dispersed 2 to 5 μm plately Al_2O_3 crystals in glasses of different coefficients of thermal expansion, and determined the internal strains produced in the (330)

plane of the embedded crystal. The average stress on the Al_2O_3 particles was determined to be as high as 30,000 psi. Fulrath also measured the strengths of his specimens and found that there was a slight decrease in strength with increasing internal stress. Unfortunately, there was some porosity in the one system of maximum internal strain. The actual effect of internal stress on strength, therefore, was not completely resolved.

Hasselman and Fulrath[3] proposed a hypothesis for the strengthening mechanism in brittle-matrix composites, which stated that the average interparticle spacing of the dispersed phase would limit the size of the Griffith flaw. Due to a mechanical abrasion, the surface of the glass had an average flaw size. A strengthening of a composite occurred when the average interparticle spacing which was predicted by the Fullman[4] equation was less than the average flaw size. Hasselman experimentally verified this hypothesis by dispersing Al_2O_3 microspheres in a glass whose coefficient of thermal expansion matched that of Al_2O_3 and measuring composite strengths.

Nivas[5] investigated the strength of composites consisting of spherical tungsten dispersed in three soda-borosilicate glasses whose thermal expansion coefficients were respectively higher, equal, and lower to that of tungsten. He measured the average interparticle spacing by using a statistical line intercept technique. Nivas' results of composite strengths were in good agreement with Hasselman's hypothesis; however, the strengths were lower for the composites with the glasses of higher and lower thermal expansions. Nivas then plotted the strength of composites at a constant size and volume fraction of the dispersed phase as a function of the coefficient of thermal expansion of the glass matrix.

There was a significant decrease in strength when the thermal expansion of the glass did not match that of tungsten. This result was interpreted as a consequence of internal stress set up in the body.

Later on, however, Stett[6] showed the degree of chemical bonding between a glass-metal composite greatly affected the mechanical strength. The question then arose as to whether good bonding occurred in Nivas' two-phase systems. His photomicrographs of polished sections revealed many pullouts of spherical tungsten, which could be explained by poor bonding between the metal and matrix phase.

Therefore, the purpose of this investigation was to determine what effect internal stresses had on the mechanical strength of brittle-matrix composites when all significant parameters were defined. A constant size and volume fraction of spherical, oxide crystals was dispersed in glasses of varying thermal expansion coefficients. Therefore, the micromechanical stress concentrations would be constant in all composites. Finally, the system selected was experimentally known to have good chemical bonding between the dispersed and matrix phase.

II. EXPERIMENTAL PROCEDURE

(1) Materials

For this investigation, seven soda-borosilicate glasses were made from silica flour, sodium carbonate, anhydrous borax and/or boric acid. The composition of each glass is given in Table I. The raw materials were dry mixed and then melted and fined in a platinum crucible in air at 1375°C for 2 h. Linear coefficients of thermal expansion of annealed glasses were measured with a quartz dilatometer furnace which had a 2.5°C/min heating rate. Densities were measured on the cast glasses

using an immersion technique with ethyl alcohol. The glasses were then crushed and ground dry in an alumina-lined ball mill until the average particle size was below 15 μm.

The Al_2O_3 powder was spheroidized using a F-R induction-coupled plasma generator, and the resulting powder separated into various particle sizes by sonic screening. The average particle size range of the Al_2O_3 used in this investigation was 25 μm. The average density of the powder was measured with an air picnometer. There was some porosity in the spheroidized powder.

(2) Specimen Preparation

Composite specimens were prepared by intimately dry mixing 40 vol % of Al_2O_3 microspheres with the glass powder. No mixing was required for the glass specimens. The sample powder was then placed in a 2 in. diameter graphite die, lined with Grafoil,* and cold pressed at 1000 psi. Vacuum hot pressing of the powder was done at 2000 psi for 10 min at the temperatures given in Table I. The hot-pressed specimen which had a final thickness of 1/5 in. was furnace cooled. The densities and linear coefficients of thermal expansion are given in Table I.

Each hot-pressed disk was mounted on a graphite block and cut at a constant sawing rate using a 270 grit diamond blade into approximately .080 in. wide bars. These bars were prepared for uniaxial strength measurements by carefully abrading them in order to obtain a constant surface condition for all specimens. This was accomplished by hand grinding each bar the same way using 240 SiC grit and kerosene on a

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* Product of Union Carbide.

glass plate.

(3) Testing Procedure

Uniaxial strengths were measured using a four-point loading device with a .75 in. overall span. Specimens were loaded such that the abraded surface was stressed. Time to failure was approximately 20 to 30 sec for the glasses and 45 to 60 sec for the composites. Fractured surfaces were studied with a scanning electron microscope by depositing a layer of Pt-Pd on the surfaces.

III. RESULTS AND DISCUSSION

An X-ray diffraction analysis of the composites showed that no other phases crystallized during hot pressing. Also, an electron microprobe analysis revealed that there was negligible diffusion of aluminum into the glass phase for all composites. Therefore, the composites consisted of only the Al_2O_3 and glass phases.

The uniaxial strengths of the vacuum hot-pressed glasses and composites are given in Table II. In Figure 2, the lower set of points is a plot of glass strength versus the coefficient of thermal expansion of each glass. All values lie within a relatively narrow range between 8,000 and 10,000 psi. The composite data, which are the upper set of points, show significant increases in strengths compared to the original strengths of the glasses. As the graph shows, the composite strengths also lie within a narrow band rather than decreasing sharply with increasing stresses set up in the composites.

A statistical line intercept technique similar to the one used by Nivas[5] showed that the average inter particle spacing, λ, varied slightly in the different composites. Since the strength is proportional

to the inverse of the square root of λ, from Hasselman's hypothesis, the strength, σ, multiplied by the square root of λ for each composite will give a normalized curve. Figure 3 is a plot of the composite strength with the correction factor versus the coefficient of thermal expansion of the glass matrix.

The Selsing equation was used to calculate the internal stress in each system. The average coefficient of thermal expansion of Al_2O_3 was taken to be 8.0×10^{-6} in./in/°C and those of the glasses were the experimentally measured values. Poisson's ratio and Young's modulus of Al_2O_3 were respectively .26 and 60×10^6 psi. No elastic properties were measured on the glasses so Poisson's ratio and Young's modulus of all glasses were assumed to be respectively .20 and 10×10^6 psi. Although these values were only for D glass, it was believed that they could be used for all of the soda-borosilicate glasses in order to determine the stress by the Selsing equation. The cooling range over which the matrix can support a stress was to be that from the annealing point to room temperature. The annealing point was considered to be the transition point which was determined from the thermal expansion curve of each glass. The maximum matrix radial stress, P_r, at the interface was then calculated for all composites and the resulting values plotted in Fig. 4.

Figure 4 shows that internal stresses did not play an important role in this system, even though these stresses were as high as 2.5 times the macroscopic strengths. All strength data were for severely abraded composites whose surfaces, it was believed, had a constant flaw density and size. This certainly suggests that the length of the Griffith flaw

is limited by the average interparticle spacing of the dispersed phase, even if internal stresses are set up in the brittle-matrix composites. Therefore, the internal stress cannot cause the existing microscopic flaws to extend statistically beyond the average interparticle distance. This occurs only if there is a good chemical bond between the dispersed and matrix phase. Also, the composites must be in the strengthening region of Hasselman's hypothesis, which means that the average interparticle spacing is less than the Griffith flaw size in the original glass. Binns[7] and Davidge and Green[8] also studied two-phase crystal-glass materials where a good chemical bond existed and found decreases in strength when there was a difference in thermal expansion between the dispersed and matrix phases. However, the interparticle spacing between the dispersed particles in their composites were always greater than the original Griffith flaw size. Their composites were not in the strengthening region of Hasselman's hypothesis, as were the composites of this study.

A basic premise of this study was that good chemical bonding existed between the Al_2O_3 and glass. The most conclusive evidence of this was from a study of fractured surfaces of composites with a scanning electron microscope (SEM). This technique of studying the path of fracture as a function of the degree of chemical bonding and thermal expansion difference of the two phases was first employed by Stett.[6] In a chemically bonded composite where the thermal expansion coefficient of the glass phase is less than that of the dispersed phase, Stett found that the fracture propagated around the sphere at a finite distance from it. Figure 5(a) which is a SEM picture of the composite with the lowest

expanding glass shows this type of fracture and thus indicates that good bonding is occurring. When the spherical particles were introduced into a glass of higher thermal expansion than that of the dispersed phase, Stett found the fracture propagated directly to the sphere and around it. Figure 5(b) is a SEM picture of the composite with the highest expanding glass. In some cases, the fracture propagated directly through the sphere, which could only occur if there was a strong chemical bond at the interface of the two phases.

IV. SUMMARY

The effect of internal stresses on the mechanical strength of brittle-matrix composites was studied by varying the thermal expansion of the matrix phase. Uniaxial strengths of the crystal-glass materials did not vary significantly with the thermal expansion coefficient of the glass phase. Therefore, internal stresses did not play an important role in the strength of composites when a good chemical bond between the dispersed and matrix phase existed.

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Table I. Properties and Compositions of Materials

Material	Composition (wt.%)			Thermal Expansion Coefficient (in/in/°Cx10^6) (RT - 450°C)	Density (gm/cm^3)	Hot Pressing Temp. (°C)
	SiO_2	B_2O_3	Na_2O			
L-1 glass	67.0	28.5	4.5	3.1	2.83	690
L-2 glass	65.0	26.5	8.5	4.4	2.89	670
L-3 glass	66.1	21.1	12.7	6.3	3.00	660
D glass	70.0	14.0	16.0	7.8	3.01	660
H-3 glass	67.0	12.8	20.2	9.0	3.03	640
H-2 glass	65.5	10.1	24.4	10.6	3.03	600
H-1 glass	61.6	8.3	30.1	12.5	3.04	580
Alumina	Al_2O_3			8.0	3.83	

Table II. Crossbending strength data of glasses and composites

Glass	Glass Data			Composite Data		
	Average Strength (psi)	No. of Samples	Standard Deviation (% of aver.)	Average Strength (psi)	No. of Samples	Standard Deviation (% of aver.)
L-1	8,600	27	10.6	12,600	30	5.2
L-2	9,280	25	9.2	12,500	31	4.8
L-3	8,220	28	13.1	12,600	21	5.6
D	9,040	19	11.9	13,800	29	5.7
H-3	10,180	26	13.8	13,000	29	4.0
H-2	8,840	25	13.3	13,000	22	6.1
H-1	9,970	27	12.6	14,500	31	5.0

FIGURE CAPTIONS

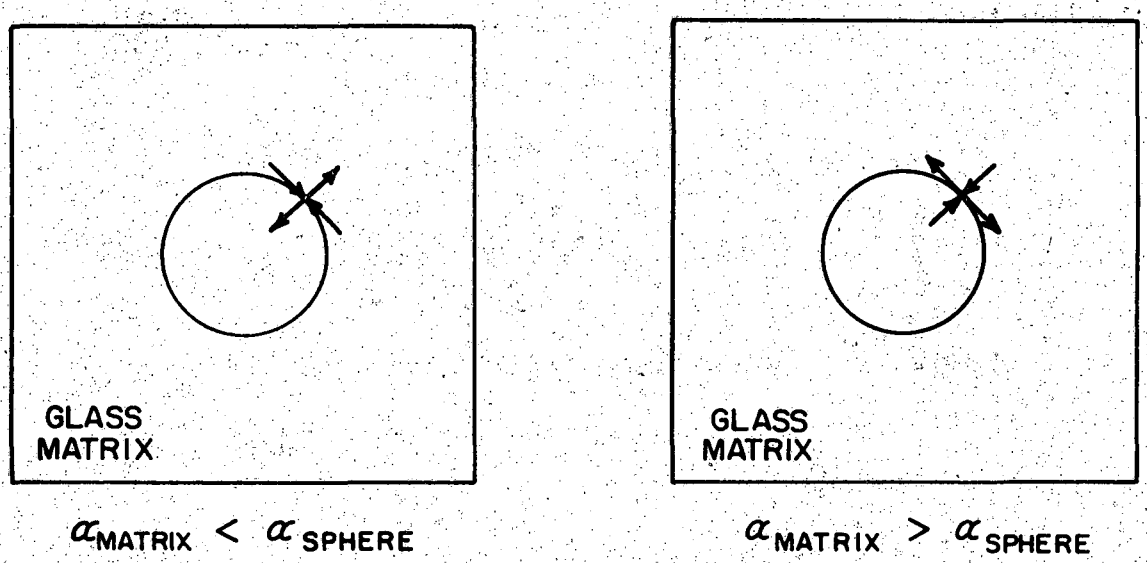
Figure 1. Internal stresses developed in matrix at the interface when the thermal expansion coefficient is either higher or lower than that of the dispersed phase.

Figure 2. Uniaxial strength of glasses and composites as a function of the coefficient of thermal expansion of glass phase.

Figure 3. Uniaxial strength corrected for small variations of interparticle spacing of dispersed phase as a function of thermal expansion coefficient of matrix.

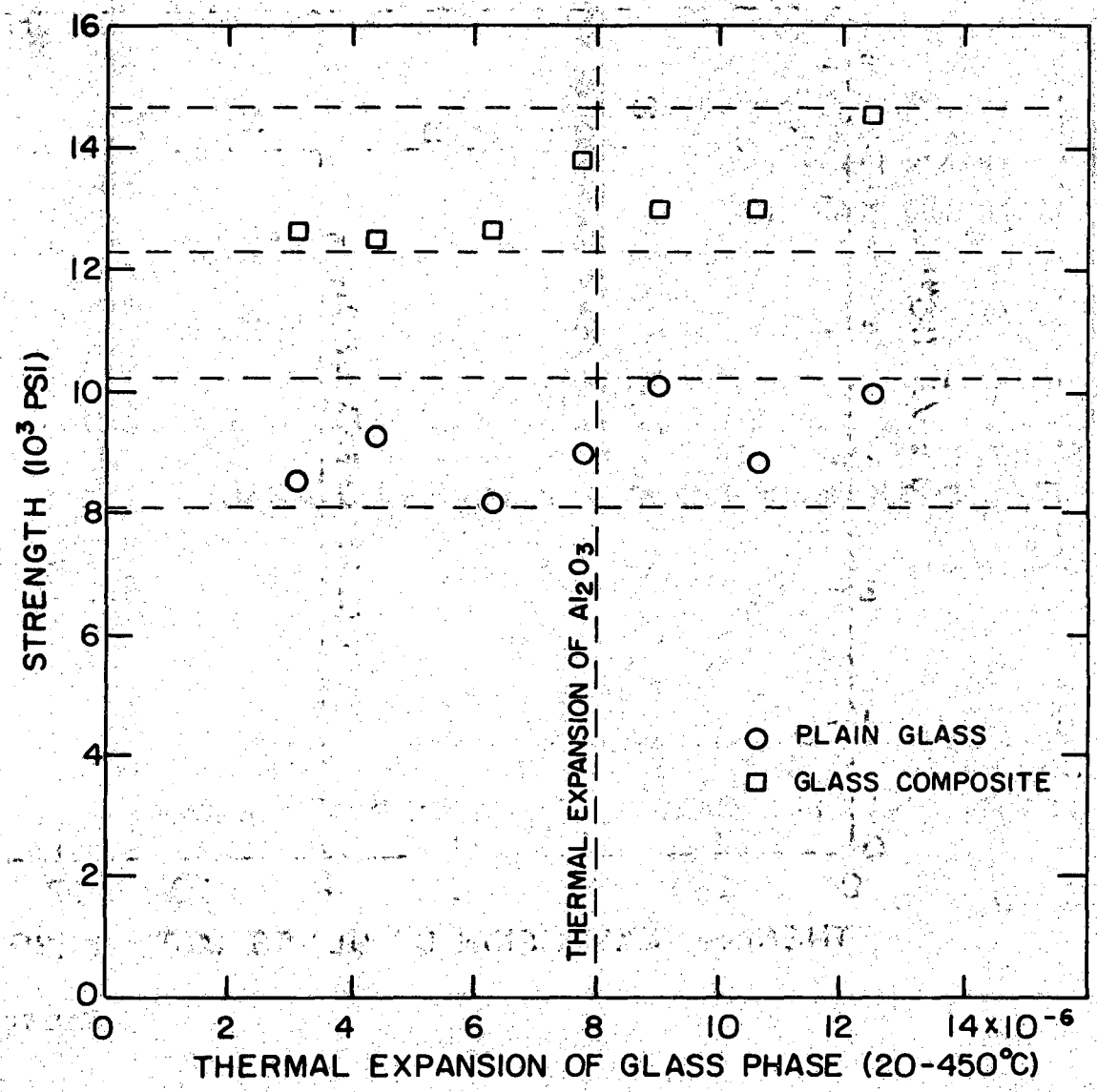
Figure 4. Internal radial stress in each composite matrix at the interface as a function of the thermal expansion coefficient of matrix.

Figure 5. SEM pictures at 1000X of fractured surfaces: (a) composite with glass of lowest thermal expansion coefficient (b) composite with glass of highest coefficient.



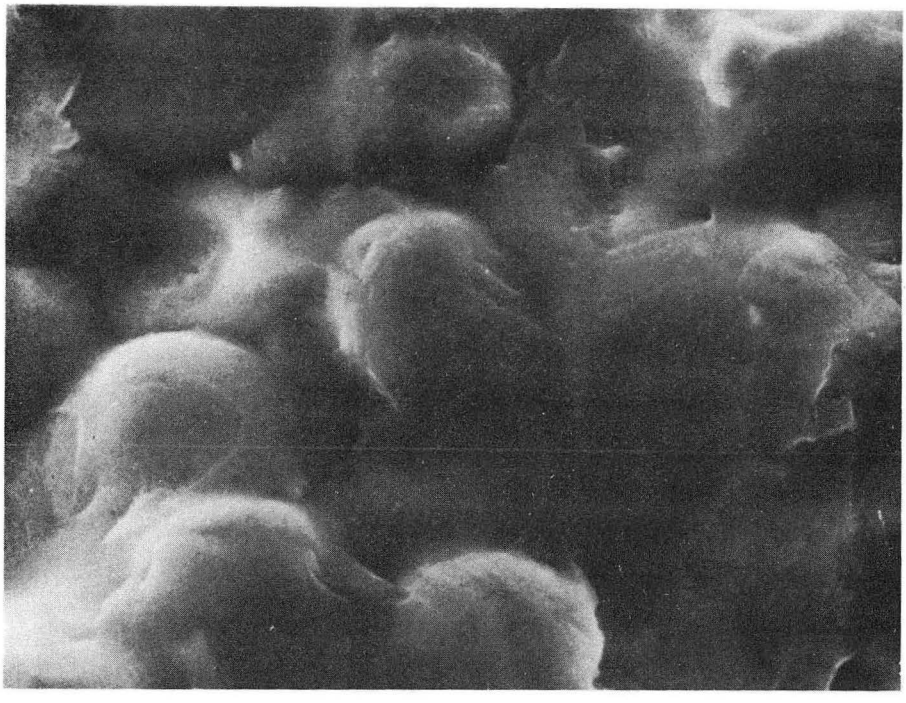
XBL 7010-6722

Fig. 1

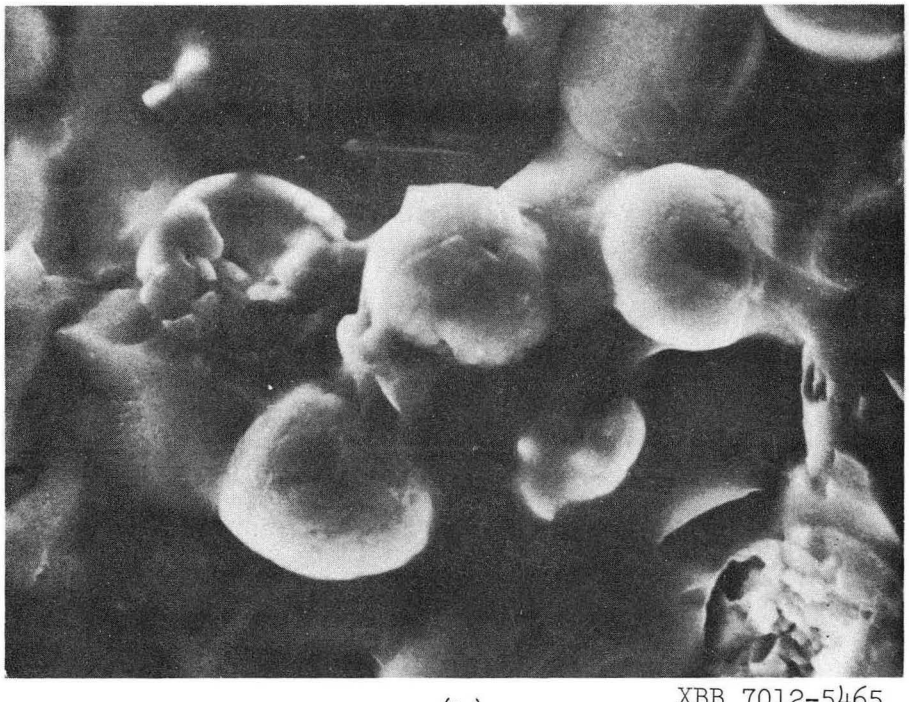


XBL 7010-6723

Fig. 2



(a)



(b)

XBB 7012-5465

Fig. 5.

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