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ONE METHOD FOR MEASURING THE SOLVENT RESISTANCE OF CRYSTAL-TO-CRYSTAL ADHESIVE BONDS

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ABSTRACT

In the course of research on butt-joined ammonium dihydrogen phosphate (ADP) crystal plates, a test having a wide range of application has been developed for the investigation of the solvent resistance of adhesive bonds. Its chief advantage is the elimination of supplementary mechanical tests required in the conventional method of testing. The test showed that, for the cases investigated, the solvent resistance of adhesive bonds is inversely proportional to the bond thickness.

PROBLEM STATUS

This is a final report on a small portion of the larger problem; work on the larger problem, a general study of crystal transducers, still continues.

AUTHORIZATION

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ONE METHOD FOR MEASURING THE SOLVENT RESISTANCE OF CRYSTAL-TO-CRYSTAL ADHESIVE BONDS

INTRODUCTION

Large-sized piezoelectric crystals with resonating characteristics satisfactory for use in sonar transducers have been produced by cementing two crystals together end to end, i.e., butt-joining (1-3). The research that led to this development was based on the necessity of extending the low-frequency limit of crystal transducers of conventional design currently employed by the Navy. In the crystal tranducer used for efficient high-level sound generation, the operating frequency, at the fundamental resonance, is limited by the size (primarily the length dimension) of the crystal; and heretofore, there has been no commerical source of crystals which are large enough to resonate at the low frequencies desired.

A factor of primary importance in the fabrication of a butt-joined crystal transducer is the necessity of matching as closely as possible the frequency characteristics of each of the butt-joined units comprising a group, regardless of the position of the bond in each case. To facilitate assembly of crystal groups, the component crystals should have identical thickness and width dimensions. For these reasons, a bond once effected should be of a permanent type despite any subsequent alteration of design or repair. A primary requisite of an adhesive to be employed in bonding together crystal adherends is its ability to resist the action of solvents customarily employed in the course of sonar transducer repair and maintenance operations.

One method of approach to the problem of determining the solvent resistance of adhesive bonds was provided by the ASTM Standard Method of Test for Resistance of Adhesive Bonds to Chemical Reagents (4). In this test, the bonded specimen is immersed in a solvent for a period of seven days and is then subjected to one of various mechanical tests. It is assumed that the bond will withstand the action of the solvent so that the supplementary mechanical tests may be carried out. In the event of a complete separation before the duration of the stipulated period, the immersion time would have to be shortened accordingly.

For the case of bonds which ultimately separated as a result of solvent action, an alternative procedure which eliminated the necessity of auxiliary mechanical tests was adopted. This resulted in an especially convenient way of investigating butt-joined ammonium dihydrogen phosphate (hereinafter called ADP) crystal plates inasmuch as the brittleness of this material effectively limits the number of mechanical tests which can be performed.

TESTING APPARATUS

The method of operation involved in the test was simple. It was based on the fact that an electrical circuit governed by a microswitch relay remained closed as long as a butt-joined crystal immersed in solvent was intact; a separation of the bond resulted in the circuit's being opened. The time interval which elapsed between the initial immersion and the ultimate separation served as the measure of solvent resistance of the adhesive bond.

The apparatus employed in the test is pictured during actual operation in Fig. 1. The solvent in question was contained in a tank constructed by welding together rectangular sections of quarter-inch, noncorrosive stainless-steel flat stock so as to form a box 14.25 in. long, 6.69 in. wide, and 3.12 in. high. The top edge of the tank walls was covered by a Corprene gasket over which was placed a quarter-inch, wire-mesh glass plate 16 in. long and 7.56 in. wide. The gasket and plate were bolted firmly to the tank in order to minimize the loss of solvent vapor. Right-angled aluminum 24ST brackets were mounted on the top surface of the plate as supports for eight identical testing units. One such unit was made up separately and is shown in Fig. 2. A microswitch was mounted on the bracket so that the pin plunger was in an upright position and supported an actuating lever (aluminum 24ST). The lever in turn was connected to a stainless-steel clamp by a rod (0.186 inch in diameter) of the same material, which was threaded at each end and passed through a circular clearance hole (0.196 inch in diameter) in the glass plate. Two screws set in the clamp held the butt-joined crystal under test in a vertical position.

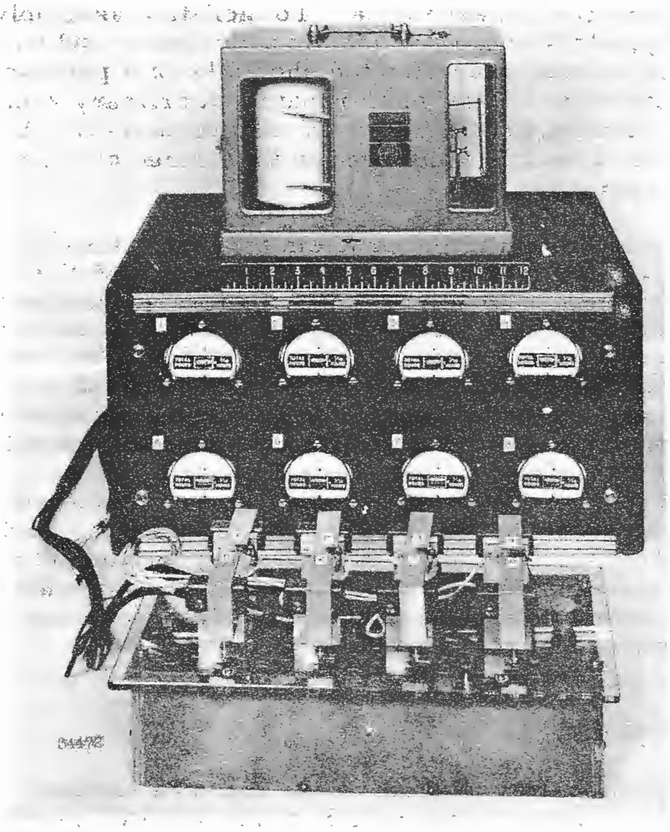

Fig. 1 - Complete apparatus for solvent resistance test. Tank equipped with eight test units in foreground; elapsed-time meter panel and temperature-relative humidity recorder in background.

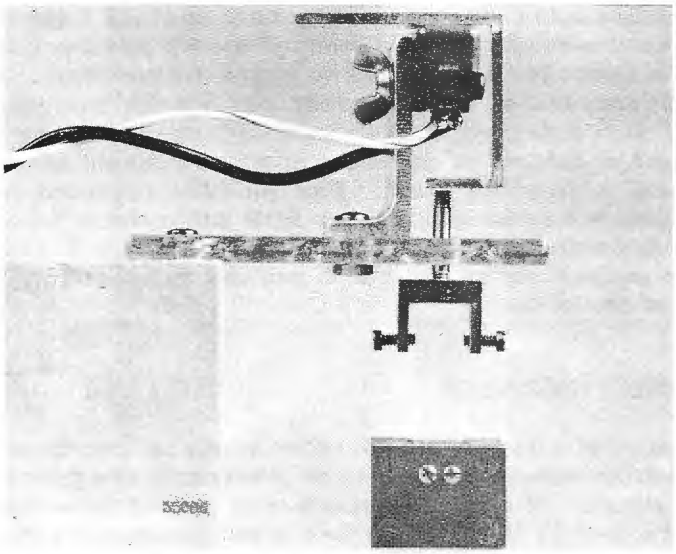

Fig. 2 - Separately constructed test unit with ADP butt-joined crystal in place

The unit was designed so that the axes of the microswitch pin, the steel rod, and the clearance hole, respectively, and the center of gravity of the bonded crystal fell on the same straight line. The clearance of 0.010 in. between the rod and the plate was sufficient to minimize friction which might arise as a result of slight deviations in the balancing of the actuating lever arm with the load. A path was thereby provided for the escape of solvent vapor, although the resulting loss of solvent through evaporation never exceeded 3% of the total volume.

The microswitch, which remained in an open position until the pin supported a load greater than 0.314 lb, was part of a series circuit which also included an elapsed-time meter and a 110-volt 60-cycle source. Once the microswitch relay was closed, weight sufficient to reduce the load on the pin to 0.093 lb had to be removed in order to reopen the circuit. Since the weight in solvent of the lower ADP adherend, which was to drop to the bottom of the tank after separation of the bond, was negligible, an additional weight had to be attached. This took the form of a stainless-steel, hollow, symmetrical block weighing 0.374 lb, which was fastened to the lower adherend by four set screws as shown in Fig. 2. The combined weight of the actuating lever, rod, clamp, set screws, and upper ADP adherend, together with the weight in trichloroethylene of the lower adherend and block, was 0.392 lb, which sufficed to close the microswitch. After the bond separated and the lower adherend and block dropped off, the weight on the microswitch pin was reduced to 0.088 lb, thereby breaking the circuit.

The test procedure was straightforward. After each of the eight crystal units and blocks had been fastened to their respective clamps, the glass plate was bolted down at each corner. Solvent was then slowly poured into the tank through the half-inch fill-hole in the plate to a level of about half an inch above the various gluelines being tested. Since the volume of adhesive material was negligible compared to that of the solvent, no provision for agitating the solvent was required. The initial reading of each elapsed-time meter was recorded, and no further maintenance was necessary until the last bond had separated. The difference between the reading of the stopped meter and the initial reading in each case was the time taken for the solvent to separate the bond. More accurately, the time elapsed represented the time required for the solvent to attack the adhesive bond to such a degree that it could no longer support a tensile stress equal to the weight in solvent of the steel block and lower adherend acting on the cross-sectional area of the bond. For the case reported below, the steel block described above was attached to an ADP adherend 0.50 in. wide and 0.25 in. thick. Accordingly, the stress on the bond was 2.432 lb/sq in. The temperature was maintained at 25° ± 1°C and the relative humidity at 35% ±5% for the duration of the test.

EFFECT OF BOND THICKNESS

Since the action of solvents on adhesive bonds is a surface phenomenon, the less surface which an adhesive exposes to a solvent, the greater should be its ability to resist attack. If one has a number of paired adherends having identical shape, and if each pair is bonded together along geometrically congruent surfaces and differs only in the thickness of the various glue lines, the test described above furnishes a convenient method of measuring the effect of the thickness of the bond on the solvent resistance of the adhesive.

Two adhesives were tested in this manner. Each ADP adherend was 1.12 in. long, 0.50 in. wide and 0.25 in. thick. In the first case an epoxide resin was employed as the bonding agent. The time interval elapsed from the initial immersion in trichloroethylene until the final bond separation, t_E, is plotted against the bond thickness, b, as shown in Fig. 3. A phenolic-elastomer adhesive was used for the second test and the same information is presented in Fig. 4.

Comparison of the two adhesives indicates that the epoxide resin is far superior to the phenolic-elastomer from the point of view of solvent resistance. In fact, if the solvent resistance is proportional to the elapsed time, as is assumed, the former is more resistant to trichloroethylene by a factor of about 12.5 for the range of bond thicknesses investigated.

Each adhesive shows a rapid decrease in solvent resistance as the thickness of the glue line increases from 0.001 to 0.008 in., i.e., from 1 to 8 mils; for greater bond thicknesses, the decrease becomes progressively slower. A reasonably good fit with the experimental data of the two adhesives is obtained by plotting the relation

$$t_E = \frac{K}{b}$$

as is done in Figs. 3 and 4. That is to say, the solvent resistance is inversely proportional to the bond thickness. Here K is a constant which is a property of the adhesive material and of the solvent used, expressed in units of hour-mils if t_E is given in hours and b in mils. For the epoxide, $K \cong 200$; for the phenolic-elastomer $K \cong 16$. The type of behavior exhibited above is similar to that encountered in the experimental study of the mechanical strength of adhesive bonds (5).

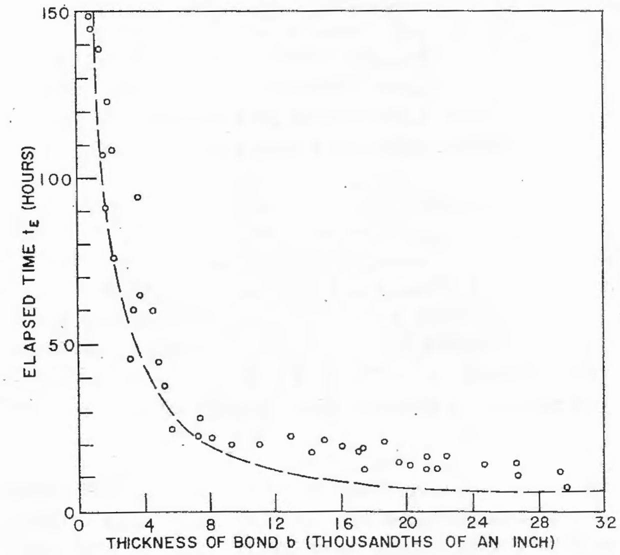

Fig. 3 - Variation of solvent resistance with bond thickness for an epoxide resin adhesive. The curve is a plot of the equation: $t_E = 200/b$

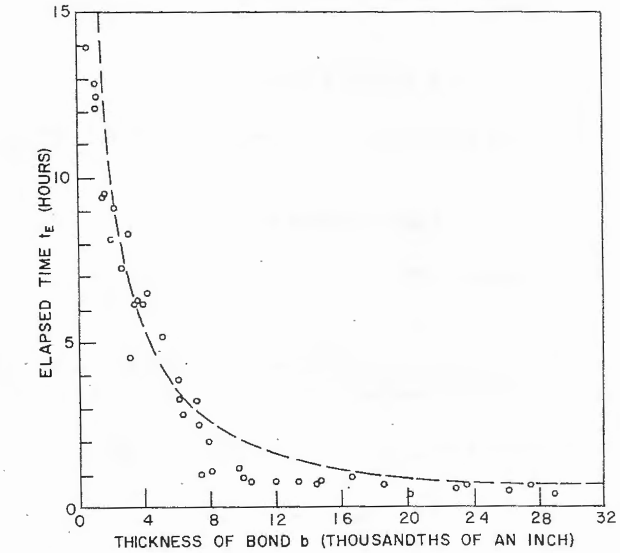

Fig. 4 - Variation of solvent resistance with bond thickness for a phenolic-elastomer adhesive. The curve is a plot of the equation: $t_E = 16/b$

ADDITIONAL APPLICATIONS

There are few limitations as to the nature of the adherends which would make it impossible to use the above test for an investigation of solvent resistance. Extreme brittleness in the adherend material would impede satisfactory clamping. An adherend which lacked sufficient tensile strength to support the required weigth could not be employed. Finally, if the adherend was more susceptible to solvent attack than the adhesive, this adhesive would be eliminated from consideration. On the other hand, the test could readily be utilized to study such bond combinations as metal-to-metal, metal-to-glass, etc.

The test can be of particular value in the determination of the optimum conditions for the preparation and cure of the adhesive. There are four principal variables in this case, viz., the bond thickness, the relative proportion of catalyst to cement, the time duration of the cure, and the curing temperature. By keeping the bond thickness and two of three remaining properties constant, the effect of changes in the fourth variable can readily be examined. It should be pointed out that in such a study a thick bond is far preferable to a thin one since the spread in experimental data is far less in the former case as shown in Figs. 3 and 4.

As a final example of the adaptability of the test, the equipment used in the preceding work could be modified to investigate simultaneously the solvent resistance of an adhesive to different solvents. Each test unit could be mounted on separate glass plates in the manner shown in Fig. 2. Individual solvent containers of identical dimensions would then be fastened to the respective plates and filled with equal volumes of the solvents to be investigated. The weight attached to the lower adherend in each case would have to be adjusted separately so as to equalize the buoyant force exerted by each solvent. If the bond thickness were kept constant at some large value, as discussed above, then the effect of the various solvents could readily be determined.

REFERENCES

1. Faraday, B. J., and Gregan, D. J. G., Report of NRL Progress, October, 12-18, 1954

2. Research Reviews (Office of Naval Research), p. 23, October 1954

3. Faraday, B. J., and Gregan, D. J. G., J. Acoust. Soc. Am. 27:1011, (Abst.) (1955)

4. ASTM Designation: D 896-51, 1952 Book of ASTM Standards, Part 7, 1008-1010. Philadelphia:American Society for Testing Materials (1953)

5. Meissner, H. P., and Baldauf, G. H., Trans. ASME, 73:697-704 (1951)

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