ULTRASONIC SAMPLES USING DIFFUSION BONDING TECHNIQUES*

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The objective of this part of the ARPA/AFML program was to produce a set of standard samples which would be suitable for NDE testing and characterization of different NDE techniques. The idea was to have a standard set of samples that could be used to compare results of various investigators and various techniques. In addition to internal defects it was decided, that in order to couple the previous work, the standard flat bottom hole type of defect should also be produced and so we have done this.

In total, we have produced 54 standard samples to date and more than half of these are being produced by the diffusion bonding method.

Before I get too far into the discussion, I would like to point out that the geometries--both the external sample geometries and the defect geometries--chosen are, to some extent, a compromise. Bruce Thompson and Don Thompson and I sat down and discussed what defect and external geometries we should have in order to most nearly satisfy most of the interested participants in the program. The external sample geometries were constrained to some extent by the equipment variables. Everybody wants to have a bigger sample, and, of course, those who have confidence in their technique want a very small defect, while those who want to characterize the physics of their technique would like a large defect.

I would also like to give some credit here to some of my co-investigators and helpers on this program. Dr. Greg Garmong of the Science Center has been of considerable help both in some of the original work on diffusion bonding and also in actually making some of these samples. Several people, particularly Fred Nevarez and Peter Sauers, have been instrumental in some of the hard work that's gone into making these, and also, I shouldn't omit to give due credit to the people in our machine shop, where they have done an excellent job with some of the very accurate machining that is required to produce these samples to within the specified tolerances.

In this first year of the program, we have chosen four basic geometries as shown in Fig. 1. The flat bottom hole in Fig. 1(a) is being used in order to couple this work to previous work, and this is made by conventional machining techniques and sectioning of samples made early in the program demonstrated that the holes were flat-bottomed within acceptable limits as shown in Fig. 2.

The hole diameters, 400, 800 and 1200 microns correspond to a 1/64, a 1/32 and 3/64 inch diameter which are standard sizes used in the industry.

In Fig. 1(b-d) the defect geometries produced by diffusion bonding are the spherical cavity Fig. 1(b) in the same diameters as were used in the flat

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FLAT BOTTOM HOLE
TYPE 1 DEFECT
400, 800, 1200 μ DIA

SPHERICAL CAVITY
TYPE 2 DEFECT
400, 800, 1200 μ DIA

OBLATE SPHEROID
TYPE 3 DEFECT
800 μ DIA.
200, 400 μ HIGH

PROLATE SPHEROID
TYPE 4 DEFECT
1600 μ HIGH
400, 800 μ DIA

Fig. 1. Defect geometries produced during the program.
Fig. 2. Flat bottom holes 800 µm in diameter in (a) 2024Al and (b) Ti-6Al-4V.
bottom hole; an oblate spheroid 800 microns in diameter and 2 or 400 microns along the minor axis (Fig. 1(c)) and the prolate spheroid 1600 microns high, 400 and 800 microns in diameter (Fig. 1(d)) so that all of these are of comparable size, but geometries that one might expect to find in practice. All of these defects are cavities; we have in the past made defects which contain some inclusions, either nonmetallic inclusions or a metallic inclusion of a composition different from the matrix. We can do this with confidence, but for this initial part of the program these were just hollow cavities.

Now, the diffusion bonded samples were made by first machining two halves. The defect is machined by a number of different techniques in the center of each half, and then, these are diffusion bonded together under conditions that will be discussed subsequently, in order to form a single unit sample with a defect in the center. Then this unit is remachined to give a controlled geometry on the outside of the specimen. One of the major difficulties in making samples by this technique is that one must limit the macroscopic strain in that sample during the bonding process. If you have more than the limit we put on it was 2% strain--the geometry of that defect will change from the desired geometry. So, we have limited the macroscopic strain during the bonding process to 2%. In order to do this, one has to have very careful control of the surface finish and the bonding parameters.

Three different external sample geometries were used in the program. Figures 3 and 4 show samples which are both 2 1/4 inches in diameter and that was constrained by the maximum size of the equipment that we had available early in the program. Figure 3 shows a sample which, when finished, is 2 1/4 inches high. Figure 4 shows a taller sample. The reason for making it in this longer geometry was that subsequently we planned to machine a sphere from this sample which is centered on the defect so that the angular dependence of the scattering of various defect geometries could be investigated. The third sample geometry that is being investigated is a 4 inch diameter sample which is, when finished, 1 inch high and is shown in Fig. 5. And, again, there's a single defect centered in this sample geometry.

The bond in all of these samples which we have succeeded in making with a diffusion bond technique does not seem to influence any test that one wants to do. It is not visible metallographically. In a defect-free-sample, a failure in a tensile test does not take place at the bond line. Of course, if you have an array of defects, then it does take place in the bond line. Figure 6 illustrates a hemispherical defect and, of course, the other defects are made by machining the appropriate defect geometry into the other half of the bonded sample.

Figure 7 shows typical microstructures of the two heats of Ti-6Al-4V that was used to make the bonded samples. Figure 7(a) is the material that was used for the 4 inch diameter specimens, and Fig. 7(b) is the material that was used for the 2 inch diameter specimens. They have both got a fairly uniform grain size and the texture in both cases was determined as less than 2 times random. In other words, a given crystallographic direction does not have more than a 2 times random texture in either of the heats; and that's important because when you diffusion bond two samples together which have different textures, one can be troubled by ultrasonic reflections from a textured interface.
NDE DEFECT SAMPLE ASSEMBLY
FINISHED DIMENSIONS 2.250 ± .002 in DIA
                      2.250" HIGH
MATERIAL Ti-6Al-4V HEAT # D4781 (TIMET)

Fig. 3. Sample assembly for 2-1/4 inch diameter specimen.
Fig. 4. Sample assembly for long 2-1/4 inch diameter specimen intended to be machined to a spherical geometry centered on the defect.
"D" 1/2 DEFECT EACH HALF
MUST BE CENTERED WITHIN
.0005 T.I.R.

4130 ALLOY
STEEL LOCATION
RING
4.081 ± .0005" I.D.

NDE DEFECT SAMPLE ASSEMBLY
FINISHED DIMENSIONS 4.000 ± .002 in DIA.
1.000 in HIGH
MATERIAL Ti-6Al-4V HEAT # D47058 (ORMET)

Fig. 5. Sample assembly for 4 inch diameter specimen.
Fig. 6. Example of internal defect geometry made by diffusion bonding a hemispheroid defect ~ 38 mils in diameter.
Fig. 7. Microstructures of the two heats of Ti-6Al-4V (a) Heat D4705B used for 4 inch diameter samples. (b) Heat D4781 used for 2-1/4 inch diameter samples.
Even though the texture was limited here, in order to further control that, a fiducial mark was placed down the side of the bar from which these samples were machined. When they were reassembled no relative rotation was permitted between the top and bottom half of the samples reducing any effects that might have arisen from any remaining texture. Figure 8 is a view of the 4 inch diameter samples. The top left Fig. 8(a) is a finish machined sample with a defect in it. The two bottom photographs Fig. 8(b, c) are the top and bottom half of a sample in this case, with a prolate spheroid machined in it at the point of the arrow. Fig. 8(d) is a locating ring which is placed around the top and bottom half to make sure the defects in the top and bottom of the sample are properly aligned, which is a critical part of the process.

Now, I mentioned earlier that in doing this, one of the problems was to use a low bonding pressure so that the macroscopic strain was always less than 2%. In some earlier studies on diffusion bonding, Dr. Garmong and myself with Prof. A. Argon, did some theoretical calculations on the time required to achieve perfect contact of the interface of the diffusion bond if you assume that you start out with some sort of a rough surface. The rate controlling process in diffusion bonding is to bring the two surfaces into contact. Once you've done that, in the case of titanium, you have a good bond, because any oxide that is on the surface, which may also influence the bond, is dissolved by the titanium. So, the rate controlling step is just to bring the two surfaces into contact.

Now, we found in these studies that most machined surfaces have a sort of bimodal distribution of surface roughness. There is a long wavelength surface roughness and superimposed on that are fine machining marks as shown schematically in Fig. 9. It's these machining marks that one usually measures in conventional surface roughness measurement techniques. In the calculations that we did, we found that it was this long wavelength surface roughness height, that is H₁ in Fig. 9, that controls bonding time more than the short wavelength surface roughness height, H₂. In other words, what you want in order to get short bonding times and low bonding pressures is to control this long wavelength surface roughness height H₁ to a minimum. Therefore, these samples were lapped so that they were flat within less than 4 optical bands from one side to the other across the 4 inch diameter surface, ensuring a minimum H₁. By doing that and by very carefully controlling the cleanliness of that surface before bonding, we were able to get very good diffusion bonds at 1700°F, 500 pounds per square inch for 30 minutes. Now, you will notice that the surface in Fig. 8(b, c) does not have a mirror finish. This is, by conventional surface roughness measurements, not a terribly smooth surface, but the critical thing is that it's flat. Small scratches don't influence the bonding time to nearly as large an extent as long as long wavelength undulations in that surface.

The other critical thing is to make sure that the surface is absolutely clean. If that surface is washed in tap water, for instance, before bonding, one does not get a good diffusion bond. It has to be very carefully ultrasonically washed in a series of acetone and then water baths. So, the cleaning is also critical.
Fig. 8. Typical diffusion bonded sample (a) finished sample, (b) and (c) lapped top and bottom halves containing a prolate spheroid (arrowed), (d) 4130 steel alignment ring.
Fig. 9. Drawing of the model and terminology used in the bimodal asperity analysis.
The next step in this program was to try and produce diffusion bonded samples of steel and aluminum. Now, this is a considerably more difficult prospect because of the fact that the oxides of steel and aluminum do not dissolve in the metal when heated. In the case of titanium I mentioned, if there is any oxide on the surface, it very quickly dissolves at the bonding temperature giving a clean surface which bonds once it comes into contact with the opposite surface. In the case of steel and aluminum, the oxide does not dissolve, so one has to remove it somehow. This is also true of aluminum.

Figure 10 shows a tensile specimen of A533-B pressure vessel steel, which is a fairly conventional low alloy steel. There is a diffusion bond at the tip of the pencil, and one can see a successful diffusion bond was obtained and failure did not occur at the bond line. The way in which we did this is we carefully lapped the surfaces the way we did for the titanium, installed the two pieces into the high temperature furnace, but before applying pressure, flushed the system with hydrogen at temperature with the sample surfaces separated by about 1/8 inch. What the hydrogen does, is it reduces the oxide on the steel and gives you a clean surface. You get what appears to be a good diffusion bond. Metallographically--as shown in Fig. 11--the diffusion bond, which runs right between the two arrows, looks very good. If you look very carefully, you can see some areas where grain growth is discontinuous across that interface, but by the same token there are many ferrite grains that have grown right across that interface. So that, when you look metallographically at this bond line, it's not surprising that it had parent metal strength and apparently parent metal ductility. The ductility of the bonded sample was the same as that of an unbonded sample given the same heat treatment.

We feel as though we've been successful in bonding conventional low alloy steels. This technique cannot be applied, however, to stainless steels and aluminum alloys because of their oxides cannot be reduced by hydrogen. In order to bond these materials, we are attempting a DC sputtering technique where we put an electrode between the separate surfaces to be bonded, sputter the oxide off those surfaces and then apply the appropriate temperature and pressure to get a diffusion bond. We are far enough along in this now that we feel that it will work although we haven't demonstrated its suitability to the extent that we have in the low carbon, low alloy steels.

In summary, we have produced a series of standard samples which have satisfied most of the investigators in the ARPA/AFML program. Unfortunately, we haven't been able to make enough samples to satisfy everybody in terms of their need for samples as quickly as we would like to have, but we've done pretty much what we set out to do. We've also demonstrated that certain steels can be diffusion bonded so that the next step will be to put defects in these steels, and we think we're on the way to being able to do this in aluminum alloys and stainless steels also.

References

Fig. 10. Failed tensile test specimen with a bond line produced as described in the Figure 7 caption at the location indicated by the pencil.

Fig. 11. Bond line (arrowed) in A5333 steel produced by reducing the surface oxide in H₂ at 1000°C and then bonding at 500 psi for 30 mins.
DISCUSSION

DR. T. WOLFRAM: Questions?

DR. GARY DAU (Electric Power Research Institute): I'm quite interested on the low carbon steel. Do you care to comment on the possibility that you see for making much larger samples, say, 8 to 10 inches thick, a foot or so square or bigger, and the flaws can be bigger also.

DR. PATON: Yes. I see no reason why that shouldn't be possible. As I say, these are some very preliminary experiments and without a great deal of care it worked the first time. In fact, to be a little speculative, I see no reason why this shouldn't be used as a structural joining process. All you have to do is flush with hydrogen and then apply heat and pressure which is something you can do on a large scale, if you are prepared to take the right precautions with dealing with hydrogen.

DR. DAU: We'll contract that out.

DR. PATON: I see no problems really. I think it would work.

DR. SY FRIEDMAN (NSRDC): There is a mixture called "forming gas" which is made up of hydrogen and nitrogen in such proportions as to render it nonexplosive under any and all circumstances. The builders of larger structures, use "forming gas", but it is still reducing atmosphere.

DR. PATON: That's an excellent idea. Yes, I hadn't thought of that. One of the things one has to watch out for here is, of course, you'll decarborize the surface if you apply this treatment for too long, so that careful control is necessary. And if one had a gas which was more easily handled, I think that would facilitate that considerably.

DR. LARRY KESSLER (Sonoscan, Inc.): What is the periodicity of the high frequency ripple compared to the grain size in the titanium sample?

DR. PATON: Well, the grain size is typically 20 \( \mu m \) and this distance in our calculations and measurements is typically of the order of 1/10 of an inch. And this is of the order of 5 to 10 \( \mu m \).

DR. KESSLER: Smaller than a grain?

DR. PATON: Well, it's about the order of the grain size. It could, in some cases, be larger than the grain size.

DR. KESSLER: Thank you.

MR. HARRY BERGER (National Bureau of Standards): Do you use any nondestructive methods to characterize the samples before you send them out?
DR. PATON: I don't, but Bruce Thompson and Bernie Tittmann do and if Bernie is in the audience somewhere, he might like to comment. I don't know, but the answer to that question is, "Yes, we do" and we're not satisfied unless that interface is invisible ultrasonically.

DR. BERNIE TITTMANN (Rockwell International Science Center): Or at least comparable to the noise that the rest of the material gives in the scattering process.

DR. PATON: Thank you very much.

DR. ROBERT GREEN, JR. (Johns Hopkins University): Since I asked you a question this morning, I don't feel so bad because I was thinking about aluminum which I have been having trouble with and it looks like you're getting there but that hasn't been done either, but one thing I wanted to make a comment on--

DR. PATON: I'm sorry. I didn't realize you were talking about aluminum.

DR. GREEN: Yes, I understand that. But we have made bonds with aluminum and I find that I can't detect the interface with ultrasound, but then when I put it in tension, it breaks there. That's what I was saying. So, I don't see anything metallographically either with optical microscope that I can detect. But there must be something there. Maybe an electron microscope or something else might prove useful.

DR. PATON: Well, the oxide that's formed on aluminum during a careful polishing technique might be a 100 A thick and you wouldn't expect to see that optically, but you would expect it to initiate failure at the bond line. So, I agree with you. We bonded a stainless steel sample and we could not see the bond ultrasonically but it failed at the bond line for very similar reasons I assume. So, that's a difficult problem.

DR. TITTMANN: Could you use ESCA to shed light on that thickness of the aluminum oxide?

DR. PATON: Measuring the thickness is not the problem--I don't think. You could use ellipsometry as an obvious technique, but the thing is you have to remove that oxide before you do the bonding.

MR. MIKE STELLABOTTE (NADC): You commented with regard to looking at the bond line to characterize it, but are you looking at the flaws that are being induced in this way? I presume you are making replicate specimens? If there any check being made to see that each replicate provides the same kind of response, ultrasonic response, and secondly, that the material provides the same kind of ultrasonic response? ASTM, in the standards that they have for manufacturing of flat bottom holes, has had to revise their work because of the difference in the technique for producing the aluminum alloys, for example, and it's found that the attenuation changes or has changed over a number of years and, therefore, the technique had to be modified. Would the same sort of problem apply here perhaps?
DR. PATON: Yes, I think it does. That's a very real problem. We have tried to get around that by making all of our samples out of the same bar of material wherever possible. There are two heats of material involved here, for the Ti-6Al-4V and ultrasonically they seem very similar. And we have in the past looked at other materials, of course, and they can be very different for similar-looking microstructures. So, I agree with you. That's a real problem to try to get a standard sample that has about the same background noise level and so forth. I don't know how that problem can be solved except to say that one has to be very careful.