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TITLE Microprobe Study of Braze Joint for SNAP 10A Thermoelectric Pump Application

PROGRAM

SUBACCOUNT TITLE

SNAP 10A
Flight Test

T/E Pump &
Emissivity Coatings

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STATEMENT OF PROBLEM Premabrazed grade 409 alloy had shown promise as a substitute braze for the grade 615 alloy used previously for bonding copper block elements to the SNAP 10A pump stainless steel throat sections; but there was a question about excessive diffusion of gold into the stainless steel at operating temperatures. A 1020 hour diffusion test was run at 1200°F to resolve this question.

ABSTRACT

There is a program requirement to produce sound brazed joints between stainless steel and copper metal components that will remain sound after long time exposure to a high vacuum environment at a temperature of approximately 1000°F. The results of a brazing program to achieve this objective will be reported by other personnel in a separate TDR. This report describes the results of a microprobe study of one of the promising brazed joints after long time exposure to high vacuum conditions at a temperature of approximately 1200°F. It was desired to determine the extent of gold diffusion into the stainless steel after long time exposure under accelerated conditions. The brazing test alloy contained 81.5% gold; 15.5% copper; and 3.0% nickel. Microprobe scan photographs are shown for each of the major elements present in the test specimen and a traverse profile sketch of the braze zone is shown for the microprobe equipment set to detect gold and nickel. The results of this microprobe study indicated significant diffusion of gold into the copper metal but no diffusion of gold into the stainless steel. There were indications of minor diffusion of nickel into the copper and perhaps some concentrating of nickel atoms at the surface of the stainless steel.

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Introduction

Grade 615 Premabrazed alloy containing silver, copper and indium appeared to be a satisfactory brazing alloy for joining stainless steel tube throat sections of SNAP 10A thermoelectric pumps to massive copper metal heat transfer blocks. Post test examination of pumps after long time performance runs, however, revealed that this alloy was not satisfactory for this application. Consequently, a development program was initiated to find an alloy that would show higher bond strengths and remain sound after extended periods of operation in high vacuum at a temperature of approximately 1000°F. This development effort is being conducted by other personnel and the results will be reported in a separate TDR. One of the most promising brazing alloys that was selected for test contained a high percentage of gold. There was some concern that there might be serious diffusion of the gold into the stainless steel during extended operation of the pumps at elevated temperature. This alloy, grade 409 Premabrazed, is composed of 81.5 percent gold, 15.5 percent copper and 3.0 percent nickel. A diffusion test was run in high vacuum for 1020 hours at 1200°F (200°F above pump operating temperature) on a specimen of stainless steel and copper metal brazed with this Premabrazed alloy 409. Metallographic inspection of a cross section of this joint afterwards, showed some evidence of intergranular penetration of the braze alloy into the surface of the stainless steel. Penetration did not appear to be severe but it was desired to determine if the gold atoms had penetrated farther into the stainless steel than indicated; so a piece of this brazed joint was examined in the ARL microprobe unit at the Autonetics Plant in Anaheim, California. The following report consists of a description of the microprobe equipment used at Autonetics and a discussion of the microprobe results obtained with this specimen.

Description of Microprobe Equipment

A description of the microprobe equipment and its capabilities is included here in order to clarify the joint traverse sketch and the scan photographs which are shown later in the report.

The microprobe unit at Autonetics was manufactured by Applied Research Laboratories in Pasadena, California. It is equipped with both integrated readout and TV type scan capabilities.

The basic principle of the microprobe is practically the same as X-ray fluorescence except for the size of the beam and the use of electrons instead of X-rays to induce secondary, characteristic X-radiation from a specimen. In the microprobe, an electron beam is focused to cover a one to two micron diameter spot on the surface of the test specimen. The intensity of this electron beam is made sufficiently high to produce characteristic X-ray radiation from a specimen in the area covered by the electron spot. The X-ray radiations from this spot are diffracted from the surface of a curved, lithium fluoride crystal and detected with a proportional counter.

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The crystal and detector may be positioned and locked in such a manner that the radiation characteristic of a particular element is reinforced and the output of the detector then may be compared with standards to give a quantitative measure of the percentage of the element present in the specimen.

Or, on the other hand, all of the elements present in the area of the spot can be determined individually by positioning the crystal and rotating the detector. There are three separate crystals and detectors in the ARL unit. Each detector may be used separately to detect any element in the periodic chart, or all three of the detectors may be read out in succession to detect three different elements at one setting of the electron spot. The specimen may be viewed at a magnification of approximately 400X and cross hairs are provided to indicate where the electron spot is focused.

Two different techniques are available for examining the surface of a specimen, a line traverse across the sample and an electron spot scan of an area approximately 12½ mils square. The specimen is motor driven in a straight line across the section to be studied, in a traverse, and readings are taken at 5 micron intervals across the specimen. The scan technique requires additional equipment which can be purchased as accessories to the standard unit. Two separate oscilloscopes, auxiliary amplifiers, a separate spot sweep mechanism, and a Polaroid camera are required to obtain scan photographs of a selected specimen area. One of the oscilloscopes has a long persistence screen. The other has a short persistence screen and a mounting support for a Polaroid camera. Only one crystal and detector unit output can be fed into the oscilloscopes at one time, but back scattered electrons can be detected separately and viewed on the screens. In the latter case, elements with higher atomic numbers give more back scattered electrons and result in a brighter intensity output on the oscilloscope. The output of a crystal and detector unit, set to pick up a particular element, shows up on the oscilloscopes in a similar manner. Areas in the specimen that contain this particular element show up as a bright field on the screens as the electron spot sweeps across the specimen. This may be visualized better by describing a typical run with a fictitious specimen. Suppose that two pieces of steel were brazed with 2 mils of gold metal foil and examined. A back scattered electron scan across the braze zone would show the gold as a bright line on the long persistence screen, but the iron would show up grey because of its lower atomic number. The gold band would be visible on the short persistence screen as a moving bright spot, however, a time exposure photograph of the short persistence screen would show the gold as a bright line and the iron as a grey area identical in appearance to the pattern seen on the long persistence screen. Now, if one of the crystal and detector units is substituted for the back scattered electron detector and set to pick up gold, the output on the long persistence oscilloscope would show a bright line where there is gold in the specimen. The iron would show up black. A time exposure photograph of the short persistence screen would show a white line for the gold but the iron areas would be black. In a similar manner, if the same probe were

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reset to pick up iron or if another probe were set for iron and substituted, then the photograph of this same area would show a black line for the gold brazing material with a white area on each side where the iron was scanned. In other words, the microprobe scan photographs show up white in the areas where there is some of the element present that is being detected but show up as black areas where there is none of this element present.

Microprobe Examination of Brazed Test Specimen

Microprobe scan photographs were made and a line traverse was run to study the joint of stainless steel and copper metal brazed with grade 409 Premabrazed. Figures 1, 2, 3, 4 and 5 show scan photographs of this joint for the various major elements present in the specimen. Figure 6 is a line traverse through the joint using one probe set to detect gold and another probe set to detect nickel. Figure 7 shows a back scattered electron scan of the stainless steel side of this specimen with a small portion of the braze material visible. Figure 8 is a scan of this same area using a pickup set to detect gold.

Refer to the microprobe scan for gold shown in Figure 1. The lines at the top of the photograph show respectively the width of the specimen scanned by the electron spot and the width of the gold that was detected. The lines at the bottom of the photograph indicate (1) the original thickness of the Premabrazed Grade 409 foil (4 mils), (2) the location of the braze to stainless steel interface, and (3) the relative position of the materials that were joined by brazing. Note that the width of gold detected is wider than the original braze thickness, indicating diffusion of gold atoms out into the copper metal. Gold has diffused approximately 2 mils into the surface of the copper, but there has been only a very slight penetration of gold atoms into the stainless steel.

Figure 2 is a microprobe scan photograph of the brazed joint with the probe set to detect copper. Note that the intensity is less in the braze width than in the balance of the copper but that there is no sharp line of demarcation. Note also, the absence of copper diffusion into the stainless steel.

Figure 3 is a microprobe scan photograph of the joint with the probe set for nickel. Note that the surface of the stainless steel is brighter than the balance of the stainless steel, indicating the possibility that nickel atoms may have accumulated at the braze interface. There also is some indication of a small amount of diffusion of nickel atoms into the copper side of the joint. Note the rather uniform concentration of nickel atoms in the stainless steel. This was type 347 stainless steel which ordinarily contains around 8 percent of nickel.

Figure 4 is a microprobe scan of the joint for iron and Figure 5 is a similar scan for chromium. Both photographs show negligible diffusion of the respective elements into the braze material.

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Figure 6 shows a graph of a semi-quantitative microprobe traverse across the brazed joint using two different detectors. One detector was set on nickel to a reference reading of approximately 8 percent while the spot was out in the stainless steel. The other detector was set on gold to a reference reading of approximately 82 percent at the gold to stainless interface where the reading was the highest. Then a straight line traverse of the brazed joint was made at these read-out reference settings. Referring to the graph, the ordinate for percent nickel is shown at the left and the ordinate for percent gold is shown at the right. The zero abscissa or interface point was taken arbitrarily at the highest reading for gold to establish the worst diffusion condition. The abscissa is marked off in mils of thickness to either side of this interface.

The joint was traversed by starting in the copper metal and moving through the brazed joint into the stainless steel side. Note in the trace for gold that the results show almost an idealized type of diffusion curve for gold diffusing into the copper side of the joint, but that there was an abrupt drop in gold at the stainless steel interface, dropping to background levels in the stainless steel. The trace for nickel started out at background levels in the copper but increased to show approximately 0.6 of a percent of nickel at the original copper to braze interface, indicating some minor degree of diffusion of nickel atoms into the copper. The nickel content in the original braze width increased to a maximum of approximately 2.7 percent at $1\frac{1}{2}$ mils from the interface and then dropped in an irregular manner to a value of approximately 2 percent at the interface. Then there was a slight drop to 1.8 percent followed by a sudden jump to a value of 9.2 percent nickel out in the stainless steel. This correlates with the brighter nickel area visible at the surface of the stainless steel in Figure 3. The nickel content afterwards dropped off in an irregular manner to values of between 7.5 and 7.8 percent in the balance of the stainless steel. Apparently the equipment did not maintain the original nickel setting of 8 percent very well.

Figure 7 shows a microprobe photograph of a back scattered electron scan of the stainless steel side of the brazed joint. In this scan the specimen was moved over to show mostly stainless steel with just a portion of the braze material visible at the left. This trace shows some white dots out in the stainless steel indicating the presence of elements with higher atomic numbers than chromium, nickel or iron. It was thought that these spots might be gold, but a probe scan for gold of the same area, shown in Figure 8, does not show much correlation. The microprobe operator stated that the indications for gold, out in the stainless steel in Figure 8 were just background scatter and did not indicate any gold present, except in the braze material at the left. Referring back to Figure 7, note the hairlike, white network at the surface of the stainless, denoting the presence of a higher Z number element in the grainboundaries. A sufficiently high enough concentration of nickel at these grainboundaries might account for this result. There is some evidence of gold, as hair-like fingers visible in Figure 8 of the scan for gold, but it does not correlate very well with Figure 7. In any event, the distances represented as intergranular penetration, at most, are only a few tenths of a mil in depth and thus do not indicate diffusion of any consequences, even if this were gold.

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It was concluded from these microprobe studies that there was negligible diffusion of gold into the stainless steel after a 1020 hour test at 1200°F. Diffusion of gold into the copper was evident and there was some evidence of nickel diffusion both into the copper and into the surface of the stainless steel; but neither diffusion of nickel in both directions, nor diffusion of gold into the copper is expected to cause much deterioration of the brazed joint. Since diffusion rates are exponential with temperature it was postulated that the Premabraz 409 alloy should give satisfactory performance for thousands of hours at 1000°F without showing excessive diffusion of gold into or through the stainless steel throat sections of the SNAP 10A pump elements.

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Microprobe Scan For Gold

Figure 1

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Microprobe Scan For Copper

Figure 2

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Microprobe Scan For Nickel

Figure 3

Microprobe Scan For Iron

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Microprobe Scan For Chromium

Figure 5

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Width Scanned

Braze

Stainless Steel

Interface

Microprobe Back Scattered Electron Scan

Figure 7

Microprobe Scan For Gold

Figure 8