Hi Nate;

I have put together a brief file of papers, mostly old e-mails, on the history of our attempts at the production of carbon targets at TRIUMF. They are numbered for reference to the notes below. The starting point is to say that graphite targets were in routine use at 1AT1 for many years. They were manufactured in Victoria by Terry Hodges and his group. In 1993 a request was made by Syd Kreitzman to install graphite targets at 1AT2 for use by the musr experimenters.

1. Summarizes the first couple of attempts at making targets for T2. The technique was to put several T1-type targets on a long holder. The targets were quarter pie shaped, 1 or 2 cm thick. The targets lasted only hours before beginning to fail.

2. Measurements made by Syd on half moon shaped targets. The length was still made up by placing several T1-type thicknesses on the holder. These targets were the first to be made at TRIUMF.

3. Minutes of a meeting to discuss the situation.

4. From Syd, who had been checking into getting a heater to test the targets prior to inserting them into the beam.

5. Minutes of another meeting to discuss the general issue of BL1A targets not lasting as long as they had in the past. Terry pointed out that the target cooling had originally been designed with a 100 ua beam in mind, and they were now being run in a 160 ua beam. Up to this time, the beam had generally been more like 140 ua.

6. An idea from John Yandon suggesting screwing as well as soldering the new targets onto the saddle.

7. Terry's response to John's idea (not much in favour).

8. A summary of the situation. This came just after a lot of damage had been done at T2, presumably caused by excessive heating of targets which were in the process of falling off the saddle.

9. More summaries, and a request for help.

10. Some information on brazing carbon.

I believe with an intense effort a solution is not too far from hand. It might well be that simply making the targets in the old vacuum oven would get us over the hump. I do think it would be worthwhile, however, to provide assistance to Tom in these attempts. Someone who can take the time and has the background to learn all about the brazing process and especially about the pitfalls would be ideal. Besides this personnel problem, there is not an easily available power supply on site with enough power to melt the ticusil.

I have reserved the conference room for 3:30pm Tuesday for a discussion.

Rich Helmer

(1)

TRIUMF

TO: J.-M. Poutissou

DATE: March 9, 1994

FROM: R. Helmer

FILE:

RE: use of graphite targets at 1AT2

CC: A. Hurst
J. Beveridge
J. Carey
G. Dutto
S. Kreitzman
G. Stinson

Hi Jean-Michel;

Syd Kreitzman asked me to send you a note about progress on the development of 10cm graphite targets for use at 1AT2. As I'm sure you are aware, we have had two tries at running on graphite. In both instances, it was noted that the surface muon flux in M20 was increased by more than a factor of two, as beam transport calculation had suggested should be the case, while the rate in M9 (M9B the first time and M9A the second) was only marginally affected - perhaps a slight reduction but less than a few percent.

Unfortunately, both targets came to grief after quite a short time. In neither case is it clear what happened to the targets, other than to say the cooling was in some way insufficient. The first time we found the targets had slipped from the saddle which holds them to the ladder, and through which the water cooling flows. Examination of the targets showed that the Ticusil backing on the graphite had partially peeled away (the Ticusil provides a surface to which the solder can adhere). It is not possible to say, however, that this was the cause of the problem. For example, perhaps the solder melted first and then the backing peeled after the contact with the cooling was lost. We haven't yet removed the targets since the second failure, but I expect we will find much the same thing.

Our plan to try to resolve this problem is to make the targets in half moon shapes rather than in quarter sections as is currently the case. Effectively this means that the beam will be spread out over two of the previous sized targets and with twice the previous cooling surface area, and so perhaps the cooling will then be sufficient. This suggestion came from Terry Hodges, but I don't believe he has done any heat transport calculations to check his intuition. In any event, I agree with him that at this stage we might just as well try this simple solution first.

Rich

(2)

From: M20DAC::M20 23-NOV-1994 23:46:10.81
To: REG::SFU
CC:
Subj: The new half-moon carbon target in T2

Summary of tests with the Be and new Carbon target on T2 23:30 Nov23'rd 1994.
Data taken with the Helios spectrometer @1.5T and a 10mm collimator.
The Carbon target in a half-moon geometry.

Started with Be target. After beam stearing, and tuning of DC_Sep and last quads (in that order) rate was 226K. This tune saved as Be10mHel.tic .

Putting the Carbon target in rate went imediately to 408K. After beam steering and tuninf of DC_sep and last quads (in that order) rate was 440K. After tuning the entire beam line rate went to 460K. This tune was saved in C10mHel.tic .

With the same tune (C10mHel) put back in the Be target and rate was 249K.

So in summary, the factor of ~2 in flux is there in the new half-moon Carbon target.

Reported by SRK

(3)

TRIUMF

TO: J. Beveridge
T. Hodges
S. Kreitzman
T. Lyth
C. Mark

DATE: July 12, 1995

FILE:

CC: G. Dutto
J.-M. Poutissou
E. Blackmore
G. Stinson

FROM: R. Helmer

RE: minutes of meeting to discuss status of graphite target at 1AT2
July 11, 1995.

1. Rich and Tom reviewed the difficulties we have had in producing a 10cm long graphite target for use at 1AT2. The target is made up from several smaller lengths approximately 2cm long, and one or two other shorter pieces to make the overall required length. The problem seems to be that the ticusil, which is brazed to each piece of graphite and is used to provide a surface to which solder will stick, breaks away from the graphite. Eventually the graphite piece falls off the saddle; sometimes while still in situ in the beamline, but sometimes not until the target ladder assembly has been removed and placed into the hot cell. In other words, sometimes it seems to take a mechanical vibration to make the graphite piece fall from the ladder. Any pieces that remain on the saddle after the initial jiggling in the hot cell seem to be well attached, and cannot be removed by gentle prodding.

2. Terry reviewed the situation where once before there had been a problem in making a good bond between the graphite and the ticusil. Eventually the problem had been solved by carefully cleaning all equipment used in the manufacturing process, but there was no one item that could be singled out as having been the source of the difficulty. There had been no problem before this instance, and none since until the present time.

3. Rich outlined the approach that had been taken to try to solve the problem in the current situation. The first attempt at a target had been a copy of the 1AT1 graphite targets, i.e. a quarter pie shape, but extended to a 10cm length by adding together several pieces as mentioned above. This target had failed very quickly; most of the pieces had fallen off within a day or two. On Terry's advice, we then made up a new target but in the shape of a half moon, mounted on the saddle with the straight edge vertical. This is

a preferred shape for at least two reasons. First, large stresses develop in the 90 degree corner of the quarter pie targets, and this is where the well-known delamination of these targets occur. The straight edge removes this corner. Second, the surface area over which the heat can be removed is doubled with the half moon. However, this target failed about as quickly as the first one.

We then learned of the previous difficulties mentioned above, so we went through the same procedure of carefully cleaning all of the equipment used in the brazing process, and outgassing the graphite, and so on. The result was perhaps cautiously an improvement; only two pieces fell from the ladder after a week's running, and it is not clear whether or not they fell during actual beam bombardment.

As a result, we thought we had made some progress, but were still not convinced we knew that the problem was in the brazing process itself, rather than in some not understood way with the shape of the target. Our interpretation of the observations have been clouded by the fact that the request for the target came at a time when responsibility for target fabrication was being transferred from Victoria to TRIUMF. Therefore we have had no experience making a successful target with which to compare our failures. As a result, we decided to make from scratch a new target for 1AT1, using all the same techniques we use for the T2 target. How it performs under beam bombardment should help sort out the source of the problem.

4. Two other potential sources of the problem were discussed. One was that perhaps the different expansion coefficients of graphite and copper were introducing some unexpected stresses. Terry didn't think this was likely a problem, but he will check. The other was that perhaps we are not using proper procedures for bonding the ticusil to the graphite, in spite of the success we have had over the years. Clive will contact the manufacturers to see if they have anything to offer in the way of advice.

5. Syd raised the question of whether we couldn't test the targets before they were put into the beamline, since there can be a rather long time delay from a new design or technique to the time it gets tested with beam. It has been Terry's experience that it is very hard to get a high heat deposition (about 8kw in the present case) into a target using a current in a wire. Furthermore it must be done in a vacuum. Jack mentioned he knew of a test facility that used to exist at Berkeley for simulating beams on ISOL-type targets. The device used essentially an electron gun; he will check on its status. He also mentioned that there will need to be a test rig for just this purpose for the ISAC targets, so if the time scale is right we might be able to make use of it. In the meantime, Syd will look for a heating element capable of delivering about 1kw/cm.

6. The conclusion for the moment is that the problem is likely the same as experienced before in Victoria. The new T1 target should help to confirm or deny this. The graphite has already had ticusil bonded to it; the drawings for the saddle will be put into the shop

today, and we will ask for high priority so that the new target can be put in the beam during the upcoming high current running period. On the same ladder there is an old saddle with a new piece of graphite, so we can test this as well. There is also a new 10cm graphite target at T2 which will be tested during the upcoming running.

7. Terry thought most of the equipment needed for bonding the ticusil was still intact at Victoria. If the new target at T1 fails we could try making the bond using this equipment. The bonding was done in a vacuum oven rather than with an RF induction heater as we have been using here, and it may be that the RF heating is too fast. Perhaps slower heating in the oven allows a better bond to form.

8. As an aside from graphite targets, Syd asked whether a beryllium target cooled on three sides rather than four could be used. This would accomplish the same purpose as the graphite target in that the beam could be placed close to the source of surface muons. Terry said that with their current size, there is not enough surface area to get the heat out of the beryllium targets if they are cooled on only three sides. If the target is made bigger to accommodate this problem, then stresses become larger than the yield strength. Jack also pointed out the danger of putting the beam too close to the edge of the target. He reminded us that once before we did put the beam down the edge of the stainless steel jacket, and people are still finding bits of beryllium in the meson channels.

(4)

From: ERICH::MSRORG "SYD KREITZMAN" 14-JUL-1995 15:07:17.40
To: SFU
CC:
Subj: More on "heater" possibilities

Rich: I have further checked into the heater situation. What the company in question (Watlow) has put into their catalouge, are heaters (or heaters with dissipation denisty in power/in^2) that are safe for air operation.

They say that manufacturing a custom made heater, with higher density is eaily done and it will cost less than $200US. Their rep has pointed out however, that for heaters with dissipation of higher than 300W/in^2, one must operate the heater in an environment that removes the heat efficiently (i.e. emersion in a water bath, or perhaps a graphite block) from the surface of the heater.

So, it seams to me that one probably order a heater of circular cross section that has the required heating density, so long it can be effectively installed into the graphite. On the other hand, a better geometry certainly is a rectangular cross section so that the heater can be efficiently clamped to the surface of the graphite. On Monday, I will prepare a quote request for two such heaters, a round model and a rectangular model.

Syd

(5)

TRIUMF

To: G. Battersby, G. Dutto, ~~R. Helmer~~, T. Hodges, T. Lyth, C. Mark, J. Nelson, D. Pearce, M.Stenning, G. Stinson, J. Yandon

Date: June 17, 1996

From: Roman Ruegg

File: T1&T2.doc

cc: J.M. Poutissou
G. Stanford

Re: Minutes from the June 4, 1996 meeting re: Recent Target Failures

My apologies for the delay in the distribution of the minutes. John Yandon presented a summary of the target failures encompassing the last six years, an updated version of this is attached. These show, at first glance, that there appear to have been more target failures over the past year than in previous years. In fact there were three T1 target failures, one T1 target ladder failure, and one T2 target failure, two T2 target ladder failures. Of the three T1 target failures two are directly attributable to low water flow, the third failure was not clearly documented. The T1 target ladder failure was caused by the breakdown of the graphite target saddle, a well known problem for this target material. The T2 failures in the past year were a faulty weld on a target and two 'C' seal failures.

Numerous items were discussed, the following are the highlights:
-Terry pointed out that the targets were designed for 100 μA and that there is not a factor of 2 headroom. He felt that any water flow that was marginal (ie. < 5 l/min.) would cause problems at higher currents (> 100 μA).
-Glen suggested that another water filter be added so that both sides of the resin can are filtered.
-Check records to see if there is a correlation between 'C' seal failure and shutdown periods.

Recommendations and actions:
-be more conservative in the operation of these systems as we are exceeding design parameters.
-reduce the initial beam turn on current to further reduce the thermal shock.
-calibrate the system devices more frequently.
-set warnings and trips to more conservative levels to ensure maximum possible water flow.
-monitor the systems more carefully.
-add a second filter to the resin can when possible (for maximum water flow).
-ensure that there is water flow to the target ladders during a shutdown or ensure that they are drained and dry to prevent 'C' seal corrosion.

T-1 And T-2 Target History

T-1, Mark 1

Nov 1986		"C" Seal
Aug 1987		Bellows
Jan 1994	12 mm Be	Down stream window, 9 years old.
Jan 1996	Graphite	Not in use, saddle failed.
May 1996	12 mm Be	Downstream window.

T-1, Mark 2

Mar 1990	12 mm Be	Beam off center, hit weld
Jul 1994	12 mm Be	Downstream window.
May 1995		"C" seal
Jul 1995		Ferrofluidic Seal
Jan 1996	12 mm Be	
Jan 1996	12 mm Be	Two pinholes, down stream window.

T-2. Mark 1

Feb 1988	10 cm Be	Window
May 1989	10 cm Be	Split case;mis-steered beam
Oct 1990		"C" seal
Feb 1991		"C" seal
Apr 1992		Ferrofluidic seal.
Jan 1994	10 cm Be	
Apr 1995	10 cm Cu	Faulty weld.

T-2, Mark 2

Jan 1986	10 cm Cu	Window
May 1987		"C" seal
Jul 1988	10 cm Be	Weld failure, possible mis-steering
Feb 1989		"C" seal
Nov 1989		"C" seal
Jan 1991		"C" seal
Apr 1992	10 cm Be	Mis-steered beam
Apr 1994		"C" seal corroded in storage.
May 1995		"C" seal
Jun 1996		"C" seal

(6)

From: ERICH::SFU "Rich Helmer" 13-DEC-1996 14:15:38.74
To: HODGES
CC: JYAN,SFU
Subj: carbon targets at T1 and T2

Hi Terry;

I just received the following message from John Yandon. What do you think of the idea? I'm not sure of the status of the targets that failed. I don't know whether they were made with the new backing material, for example, or whether they were made in the rf induction heater or in your old furnace.

I'll be back at TRIUMF next week; where are you these days? Morley O'Neil says he ran into you and Roy at CERN not long ago.

Rich

From: REG::JYAN "John C. Yandon, Vacuum Group Leader" 13-DEC-1996 11:02:19
To: SFU
CC: JYAN
Subj: Carbon Targets

Hi Rich

As you may have heard, the latest carbon targets at T1 and T2 did not last very long. T1 is running on the second C target, while T2 is Be (I think).

I am suggesting that the segments for the T2 be secured with screws that pass through the saddle between the water passages (one per segment) and engage holes tapped in the carbon. The segments would still be soldered for thermal contact,but the strength of the joint would not be important.

What do you think?

John

7

From: TERRY::HODGES 16-DEC-1996 09:07:10.23
To: REG::SFU
CC:
Subj: Graphite targets

Hi Rich
We are still here at UVic but our work on ATLAS makes for a few trips to Cern and occasionally to other collaborator's labs.
I don't think John's idea of putting a retaining screw into the graphite will be much help. It takes very little to hold the graphite in place but if it does fall off, you know the braze/solder joint must have broken up, and there is no effective thermal contact. I think if you hold the graphite in place under these conditions, it will just erode rapidly from the very high temperatures it will experience.
Very puzzling why this continues to plague us.

Cheers,
Terry

8

From: ERICH::SFU "Rich Helmer" 8-JAN-1997 23:54:40.42
To: TLYTH,CMARK,JYAN,RUEGG,MCHIRD,HODGES
CC: JMP,DUTTO,SYD,STINSON,HURST,SFU
Subj: graphite targets in beam line 1

After discussions with several of you, I believe a concensus has emerged on which most people agree.

- We should not run a graphite target again at T2 until we have solved the T1 problem. During the last run, an enormous amount of damage was done to the area surrounding T2, and we can't afford to do so again. Terry has been doing some heat transfer calculations for the graphite target at T2, and indeed we might have an additional problem because of the different design at T2, but we still have a *manufacturing* problem at T1 and until that is solved, we have no hope at T2.

- The T1 problem is that the mode of target failure is different from the way it used to be. Formerly, the corner of the target started to delaminate, there was an increase in the activity detected by various monitors around the site, and that was a signal to change the target. To my knowledge, none of our previous graphite targets simply fell off the ladder as they do now. Therefore, we should not be fooled into thinking the problem at T2 is due to the new target design. I repeat there may be an additional problem there, but until we are sure we are manufacturing the targets correctly we cannot unravel any additional problems brought on by the new design.

- The main remaining difference in the way the targets are manufactured now compared with the way they were manufactured in Victoria is that a RF induction heater has been used recently, whereas an ordinary radiative oven was used in the past. That oven, or rather those ovens - there are two of them - are here at TRIUMF and Tom is cleaning them up. We should recall that there was once a similar problem manufacturing the targets in Victoria, which was ultimately resolved by a thorough cleaning of all apparatus used in their manufacture. It was never proven that this actually solved the problem, but it clearly didn't hurt. Therefore we should start with good, clean ovens this time so we don't repeat mistakes of the past. Terry will be at TRIUMF next week and can advise Tom on any other idiosyncracies about using the ovens.

- There will be some pain in some quarters over temporarily abandoning the effort at T2; we have made steady progress and we were able to run for 10 days with good yield in the last run. However, we don't have enough bodies to clean up the damage every 10 days, so we must solve the problem first in an area where not so much harm can be done.

- Volunteers to help Tom would be appreciated.

Rich

(9)

From: ERICH::SFU "Rich Helmer" 12-JUN-1997 09:26:20.52
To: SYD
CC: TLYTH,CMARK,RUEGG,JYAN,STINSON,REINIGER,HODGES,DUTTO,JMP,SFU
Subj: graphite targets

Hi Syd;

I was wondering if there was anyone in your group who could help with another attempt at making graphite targets that will fill our needs. We don't need anyone with great skills; mostly we need someone with a little time. A student would be perfect.

The situation at present is as follows. We can now make graphite targets for T1 that last for about 2 weeks, but they still fail in the same way as we observed for T2, that is, they slide off the saddle because the bond between the graphite and the ticusil backing breaks. Unfortunately, this means it is still too dangerous to try this type of target at T2. Our best guess for the cause of the problem is that the graphite and ticusil are not heated evenly in the rf induction oven we have been using to bond them together, with the result that stress is built into the graphite and it eventually fractures.

The solution we would like to try is to go back to the old way of heating them in a convection oven, in which everything is heated to the same temperature. We have in hand the oven that was used successfully for many years by the group in Victoria. We have been held up by not having a suitable power supply, but Klaus tells me he now can supply us with one that would do the job. The only hold up now is that with all his other responsibilities, Tom Lyth doesn't have enough time to prepare this equipment - clean the oven, arrange for it to be evacuated, get the power supply in place, and so on. Any help you could provide would be much appreciated.

I believe that as soon as we can make a target at T1 that fails in the old way, that is the target lasts for about a month, and the failure mode is that the graphite itself begins to deteriorate in the region where the beam is striking it, then we can try again at T2 with a reasonable expectation of success.

Thanks,
Rich

RICH HELMER

(10)

Brazing of Carbon and Graphite*

By A.J. Moorhead
Group Leader
Oak Ridge National Laboratory
and
C.R. Kennedy
Research Staff Member
Oak Ridge National Laboratory

CARBON AND GRAPHITE, brazed both to themselves and to metals, are the subject of this article. These materials vary widely in the degree of crystallinity, in the degree of orientation of the crystals, and in the size, quantity, and distribution of porosity in the microstructure. These factors are strongly dependent on the starting materials and on processing and, in turn, govern the physical and mechanical properties of this product. Accordingly, the first section of this article discusses how these materials are produced.

Material Production

Carbons and graphites can be manufactured by several processes that yield materials with a wide range of crystalline perfection and properties. In the most widely used process, polycrystalline graphites are made from cokes produced as a by-product of the residuum from the manufacture of petroleum or from natural pitch sources; the coke is the final product that remains after all volatile materials have been removed after heating to about 930 °F. The coke product is broken up and then calcined at temperatures from 1650 to 2550 °F to reduce the volatile content and create a dimensionally stable filler coke. This stabilization prevents the occurrence of excessive shrinkage during subsequent heat treatments. Calcination is omitted in the production of some special high-strength graphites.

The calcined coke is crushed, milled, and sized through screens into various fractions. The coarse fraction may exceed 0.04 in. in the fabrication of large bodies and may be as small as 0.004 in. in smaller blocks, depending on the desired properties. The shape and properties of the crushed coke particles depend largely on the coke source. Some cokes naturally break up into highly anisotropic particles with platelike or needlelike shapes, while others produce rounder isotropic particles that have a strong influence on the properties of the final body.

The size fractions are mixed according to properties desired in the final material and blended with a hot coal tar pitch. The pitch creates a plastic mix that can be shaped by extrusion or molding. The shape of particles is important in that forming processes tend to preferentially align the anisotropic particles. Particles that are aligned in this manner as the result of extrusion or molding processes produce bodies with highly anisotropic properties. Extruded bodies and molded bodies yield rotational symmetry; however, the axes of the graphite crystals are aligned parallel to the extrusion direction and normal to the molding direction.

The formed body is baked to pyrolize the binder pitch at temperatures from 1470 to 1650 °F, usually in large gas-fired floor furnaces. During baking, the binder experiences approximately 50 to 60% weight loss and an even greater volume loss. The effect is to reduce overall density and subsequently increase porosity. Density can be increased by rebaking following impregnation with low-melting-point, high-viscosity pitches. Special impregnants, such as thermal setting resins or mixtures of resins and pitches, can be used to control porosity.

Final graphitization of carbon artifacts is achieved at temperatures ranging from 4170 to 5430 °F in an Acheson furnace, an electric furnace similar to ones used primarily for the production of silicon carbide. The baked carbon bodies are stacked in a conducting bed that is buried under insulating material. A large electric current (6000 A at 230 V) is passed through the bed using large, water-cooled electrodes at both ends of the furnace until the graphitization temperature is obtained, and then the mass is allowed to cool. The heating cycle generally lasts about 15 days, after which the furnace is disassembled and the graphite blocks are removed.

Two alternate processes may be used to produce carbon bodies. Carbon blacks may be used as the filler material in place of petroleum coke. Carbon blacks, in this case, are mixed with pitch and briquetted. They are then baked, reground, and remixed with pitch. A reforming step completes production. These grades primarily are used in brushes for electrical motors where good wear resistance and conduc-

*Research sponsored by the Gas-Cooled Reactor Programs Division, U.S. Department of Energy, under contract W-7505-eng-26 with the Union Carbide Corp.

tivity are ensured through adjustment of the final heat treatment temperature, which is usually lower than 4530 °F. Carbon fibers also can be made into yarns that are woven into cloth or the desired shape. The product is pitch infiltrated, baked to reduce porosity, and graphitized. These bodies generally have very high tensile strength, as well as low coefficients of thermal expansion, which provides for excellent resistance to thermal shock or stresses. However, carbon bodies fabricated from carbon fibers are very anisotropic, with lower shear and flexure strengths.

Applications

Carbon and graphite find widespread use as electrodes in metallurgical applications and as moderator materials in nuclear applications. Specialized uses include rocket nozzles, guide vanes, nose cones, electric motor brushes and switches, bushings and bearings, high-temperature heat-exchangers, and plumbing, as well as heart valves, synthetic teeth posts, air frame composites, and high-performance brake linings. The physical-property requirements of these products vary considerably, thus illustrating the numerous carbon and graphite structures commercially available. Electrical or thermal conductivity, thermal expansion, and strength requirements may also vary considerably, depending on the application. Also, because of the highly anisotropic crystal structure of carbon and graphite, materials can be produced with physical properties that are capable of being highly anisotropic to structures that are virtually isotropic.

Brazing Characteristics

Wettability. The wetting characteristics of all the carbons and graphites are strongly influenced by impurities, such as oxygen or water, that are either absorbed on the surface or absorbed in the bulk material. Moisture absorption always occurs to some extent, with levels as high as 0.25 wt%. Brazeability also depends on the size and distribution of pores, which can vary significantly from one grade to another. For example, some graphites are so porous that all available filler metal is drawn into them, resulting in alloy-starved joints. Others are so dense and impervious that adherence of filler metal is poor.

Thermal Expansion. A major consideration when brazing carbon and graphite is the effect of the coefficient of thermal expansion of these materials. This can range from about 2×10^{-6}/°C up to 8×10^{-6}/°C between 25 and 1000 °C (75 and 1830 °F), depending on the type and grade of product, as well as within a given piece, depending on the degree of anisotropy. In these materials, expansion coefficients may be less than, equal to, or greater than those of the reactive or refractory metals. However, they are always less than the more common structural materials such as iron and nickel. Before brazing graphite, the type and grade of carbon or graphite must be established to ascertain the expansion characteristics of the particular material. This information is also important when brazing carbon or graphite to itself. Joint failure, particularly during thermal cycling, may occur if too great a difference exists between the coefficients of thermal expansion of the graphite and the brazing filler metal.

Brazing to a Dissimilar Material. If the braze gap increases significantly on heating because of a large mismatch in coefficient, the brazing filler metal may not be drawn into the joint by capillary flow. However, if the materials and joint design cause the gap to become too small, the alloy may not be able to penetrate the joint. In conventional brazing of dissimilar materials, the material having the greater coefficient of expansion is made the outer member of the joint. Joint tolerances are used that do not allow the gap between the surfaces to become too great for capillary flow.

Additional problems occur in brazing dissimilar materials when one part of the joint is a carbon or graphite. Carbons and graphites have little or no ductility and are relatively weak under tensile loading. These adverse conditions are usually compensated for in graphite-to-metal joints by brazing the graphite to a transition piece of a metal, such as molybdenum, tantalum, or zirconium, with a coefficient of expansion near that of the graphite. This transition piece can be subsequently brazed to a structural metal if required. This minimizes shear cracking in the graphite by transposing the stresses resulting from the large difference in thermal expansion to the metallic components. Thin sections of metals, such as copper or nickel, that deform easily when stressed have also been successfully used for brazing dissimilar metals.

A special graded transition piece was developed by Hammond and Slaughter (Ref 1) to accommodate the mismatch between the coefficients of expansion of graphite and of a nickel-based structural alloy (Hastelloy N). Using powder metallurgy techniques, a series of seven W-Ni-Fe rings having different tungsten contents were fabricated. By varying the tungsten content from 97.5% in the first ring down to 40% in the seventh, they were able to fabricate a graded seal with a thermal coefficient of expansion near that of graphite on one end and of Hastelloy N on the other. All of the W-Ni-Fe compacts and the graphite and Hastelloy N terminal pieces were then brazed together in a single operation using pure copper as the filler metal. Brazing of the heavy alloys to the graphite was made possible by a prior metallization of the graphite with chromium, as discussed in the following section of this article.

Filler Metals

Graphite is inherently difficult to wet with the more common brazing filler metals. Most merely ball up at the joint, with little or no wetting action. Two techniques are used to overcome this wetting deficiency: the graphite is coated with a more readily wettable layer, or brazing filler metals containing strong carbide-forming elements are used. Several researchers have developed techniques for coating graphite with either a metallic or intermetallic layer so that brazing can be accomplished with a conventional filler metal.

Example 1. Chemical Vapor Deposition of Graphite With Molybdenum or Tungsten. Graphite was coated with a thin film (0.008 to 0.31 mils thick) of molybdenum or tungsten by a chemical vapor deposition (CVD) process (Ref 2). This was accomplished by passing a mixture of hydrogen and the appropriate hexafluoride gas (molybdenum or tungsten) over the graphite at a temperature of 1470 or 1110 °F, respectively. These metals were selected because of their low coefficients of thermal expansion, which are approximately that of the particular graphite being used, not because of carbide formation at the interface. Carbide formation does not occur at these low temperatures, so the coating-to-graphite bond is essentially mechanical. In this instance, the coated graphite parts were brazed to molybdenum with copper (BCu-1), but other filler metals are equally acceptable. Although the CVD process is not complicated, it is not widely used. There are commercial companies, however, that do utilize this process to apply coatings.

Example 2. Formation of a Chromium Carbide Substrate on Graphite by Chromium Vapor Plating. Hammond and Slaughter (Ref 1) developed a process for treating graphite that produces a metallurgically attached chromium carbide substrate. This treatment is applied by a novel procedure involving vapor plating chromium on the graphite in a partial

vacuum at 2550 °F. The chromium carbide forms by chemical reaction as chromium deposits on the graphite. The chromium vapor is supplied by reacting a mixture of fine carbon and chromium oxide powder, spread over the hearth of a graphite crucible in which the plating is carried out.

Push-pin shear tests were conducted on graphite specimens coated by this technique and then brazed with pure copper filler metal. Reported shear strengths were 20 ksi both at room temperature and at 1290 °F. Metallographic evaluation of failed test pieces showed the specimens coated with chromium carbide to have failed by shear in the graphite pin just inside the bond region.

Filler-Metal Compositions. A number of experimental brazing filler metals have been developed for brazing of graphite either to itself or to refractory metals. These filler metals typically contain one or more of the strong carbide-forming elements such as titanium, zirconium, silicon, or chromium. For example, Donnelly and Slaughter (Ref 3) reported on the successful brazing of graphite using filler metals of composition 48%Ti-48%Zr-4%Be, 35%Au-35%Ni-30%Mo, and 70%Au-20%Ni-10%Ta. In addition, Fox and Slaughter (Ref 4) recommended the use of a filler metal with composition 49%Ti-49%Cu-2%Be for brazing of graphite as well as oxide ceramics. These alloys wet graphite and most metals well in either a vacuum or inert atmosphere (pure argon or helium) and span a fairly wide range in brazing temperatures (from 1830 °F for 49%Ti-49%Cu-2%Be to 2460 °F for 35%Au-35%Ni-30%Mo). However, they have not been evaluated for oxidation resistance or mechanical properties. These materials are not available commercially, and this presents a problem for a potential user who does not have access to arc-melting services.

At least two commercially available brazing filler metals reportedly wet carbon or graphite, as well as a number of metals. One is a modified version of the silver-copper alloy with a small titanium addition to promote wetting of oxide ceramics and graphite. This alloy has the composition of 68.8%Ag-26.7%Cu-4.5%Ti, with a solidus of 1525 °F and a liquidus of 1560 °F. This alloy is suitable for low- to medium-temperature applications but appears to have only moderate oxidation resistance. The second commercially available filler metal for graphite brazing has the composition of 70%Ti-15%Cu-15%Ni. It has a somewhat higher melting range (1670 °F solidus and 1760 °F liquidus) than the first and, by virtue of its greater titanium content, has better oxidation resistance than the silver-bearing alloy.

A considerable amount of work has been done by researchers in Russia on joining of graphite. For example, graphite was brazed to steel at 2100 °F using a filler metal of 80%Cu-10%Ti-10%Sn (Ref 5). In another technique, known as diffusion brazing, a metallic interlayer was placed between the graphite components; the components were pressed together with a specific pressure and heated to the temperature of formation of a carbon-bearing melt or a eutectic (Ref 6 and 7). On heating to higher temperatures, the melt dissociated with the precipitation of finely divided crystalline deposits of carbon that interacted with the graphite base material to form a strong joint. Depending on the physical nature of the metal of the interlayer and on the type of carbon-metal phase diagram, carbon is formed in the joint either during the thermal dissociation of a carbon-bearing melt or a carbide-carbon eutectic. Iron, nickel, and aluminum are typical metals that form carbon-bearing melts when heated at high temperature in contact with graphite. Molybdenum is capable of forming a thermally dissociating carbide-carbon eutectic.

For in situ formation of a liquid film of brazing material that is subsequently dissociated at a graphite interface, a specific compressive force of 0.5 kgf/mm^2 was used, and argon pressure of 0.3 to 0.5 atm was supplied. The optimum temperature range for joining of graphite using an intermediate nickel layer was 3810 to 3990 °F; for iron, 3990 to 4350 °F; for aluminum, 3990 to 4170 °F; and for molybdenum, 4350 to 4710 °F. Metallographic examination of joints made using this technique showed that with increasing temperature the amount of metallic or carbide phase in the joint decreased, but the amount of the graphite phase increased. This increase in graphite content resulted in a marked increase in joint strength as compared to those samples brazed at lower temperatures and having significant metal- or carbide-containing microstructures.

Amato *et al.* (Ref 8) developed a procedure for brazing a special grade of graphite to a ferritic stainless steel for a seal in a rotary heat exchanger. It seems apparent that the selection of type 430 stainless steel was based at least partly on its lower coefficient of thermal expansion (7.3×10^{-6}/°F) as compared to that of a typical austenitic stainless steel (11×10^{-6}/°F). In addition, a joint geometry was developed that minimized the area of the braze joint, thereby reducing thermally induced stresses to acceptable levels. Specimens of graphite brazed in a vacuum furnace to thin type 430 stainless steel sheet with either Ni-20%Cr-10%Si or Ni-18%Cr-8%Si-9%Ti at 2060 to 2150 °F performed well in tests at 1200 °F.

Applicable Heating Methods

As graphite begins to oxidize at about 840 °F (depending on the grade), brazing operations must be conducted in environments that exclude oxygen. This can be accomplished either in a vacuum of $\sim 1 \times 10^{-4}$ torr or less or through the use of high-purity inert gas (argon or helium) protection. Heating typically has been done in a furnace, but induction heating also has been used.

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