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STATE-OF-THE-ART SURVEY OF DISSIMILAR METAL JOINING BY SOLID STATE WELDING

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ABSTRACT

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The state-of-the-art of joining dissimilar metals by solid state welding was determined by a literature survey and visits to five selected organizations. Useful data are available on solid state welding of aluminum alloys to stainless steel. Available information on joining of other dissimilar metal systems is limited. General information has been compiled on processing, metallurgical techniques, diffusion phenomena and solid state welding characteristics of similar and dissimilar metals. These data are reported in three parts: a review of pertinent information, a summary of the state-of-the-art visits and an annotated bibliography.

Author

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INTRODUCTION

A comprehensive state-of-the-art survey has been conducted on joining of dissimilar metals by solid state welding. This work was performed as the first phase of Contract NAS 8-20156, "Study of Dissimilar Metal Joining by Solid State Welding". The objectives of this research and development program are: (1) to study the mechanisms by which dissimilar metal joining of various alloy combinations is accomplished by press bonding and roll bonding and means by which such solid state bonding can be facilitated, and (2) to determine the characteristics of completed joints between the dissimilar metal combinations to be studied.

The survey consisted of a literature search for information related to solid state welding of dissimilar metals and a state-of-the-art tour to selected organizations believed to be most able to provide useful information. The prime objective of the survey was to compile data in order to facilitate planning and performance of the experimental phases of the program. The following types of dissimilar metal combinations have been designated for study on the experimental program:

Aluminum alloy to stainless steel Titanium alloy to aluminum alloy Titanium alloy to stainless steel Titanium alloy to nickel-base alloy Stainless steel to nickel-base alloy

The emphasis of the survey was on joining of these material combinations. Consideration was given to joining with and without the use of intermediate materials. Review was made of the various solid state welding methods, but emphasis was placed on press bonding and roll bonding since these techniques will be used in the experimental study.

It became apparent early in the literature search that limited useful data were available on solid state welding of the specified dissimilar metal combinations being considered in the experimental program. The only exception was the joining of aluminum alloys to stainless steel. For this reason, selected information was included on diffusion phenomena and bonding characteristics of related materials or material combinations which would contribute to an improved overall knowledge of the subject.

This report on the state-of-the-art survey is composed of essentially three major parts:

- Review of pertinent information
- Summary of the state-of-the-art tour
- Annotated bibliography

The initial portion of the report reviews pertinent information on solid state welding under the following technical categories:

- Fundamentals of Diffusion Bonding
- Solid State Welding Processes
- Solid State Bonding of Dissimilar Metals

The summary of the state-of-the-art tour is a review of the visits made to five selected organizations and of the technical information obtained which is related to solid state welding of dissimilar metals.

The bibliography contains 71 selected references related to solid state welding. References are listed alphabetically by author. Each reference is accompanied by an abstract or summary, which in some cases was contained in the article but in other cases was prepared or edited to suit this bibliography.

No treatise on this subject would be complete without clarification of what is meant by the term "joining by solid state welding". Evans (31)*, in a review of the subject, defined diffusion bonding... "as a joining technique by which coalescence of clean, closely fitting parts is obtained by applying pressure with or without heating the assembly. No melting and no measurable deformation of the parts result from diffusion bonding". This definition appears to be somewhat too restrictive since in his summary of diffusion bonding techniques, he discusses forge welding and eutectic diffusion brazing. Forge welding involves extensive deformation and eutectic diffusion brazing involves melting in the form of a transient liquid phase.

A somewhat better definition has been provided by Albom (1); "Solid state bonding is a joining technique in which time, temperature and pressure produces, through diffusion, coalescence of the base materials being bonded". For the purposes of this report the latter definition adequately describes solid state welding. To minimize confusion in this report, the terms - solid state welding, solid state bonding, solid state diffusion bonding and diffusion bonding-will be considered synonymous. Processes which involve a transient liquid phase variously called "diffusion brazing", "diffusion sink brazing" or "eutectic diffusion bonding" will be considered as related processes and will be covered also in this survey.

^{*}Numbers denote references in the Bibliography

FUNDAMENTALS OF DIFFUSION BONDING

The technical literature contains numerous reports on fundamental studies of diffusion phenomena. Although many of these were concerned only with learning more about fundamental reactions, a number of reports were reviewed in which the studies were directed towards diffusion phenomena as applicable to development of dissimilar metal diffusion bonding processes.

Castleman (10-17) and his co-workers conducted extensive fundamental investigations from 1954 to 1962 on the diffusion bonding problem related to fabrication of components for nuclear reactors. Detailed studies (10) were conducted on formation of intermediate layers in the diffusion zone of Ni-Al diffusion couples. Only the Ni₂Al₂ and the NiAl₃ phases were observed to form layers in specimens press-bonded at temperatures of 750° to 1160°F. The other two possible intermetallic phases NiAl and Ni₃Al did not appear to form during diffusion bonding. A significant part of this work was devoted to determining the effect of pressure on phase formation.

Subsequent work (13,18,19) conducted with very long diffusion times, disclosed that the two missing stable intermetallic phases, NiAl and NizAl were indeed present but in diffusion bonding the layers were too thin to be detectable.

Investigations (11,12) were conducted on interdiffusion and pressure effects on the somewhat less complicated U-Al system. Increased pressure was observed to increase the formation of the UAl₃ phase. Similar pressure effects were not obtained for the Ni-Al system.

Investigations (15,16) were conducted on the Cu-Zn system with particular emphasis on growth kinetics of beta-brass layers in the alpha-brass vs gamma-brass diffusion couples. Studies were also initiated on diffusion phenomena of the more complex ternary Cu-Zn-Ni system.

The interdiffusional aspects of the diffusion bonding problem were viewed from an analytical approach by Castleman (9). Three idealized types of dissimilar metal systems were discussed.

One Phase - Complete mutual solubility
Two Phase - Partial solid solubility
Three Phase - Growth of an intermediate phase

For the two-phase and three-phase systems, a detailed analytical evaluation was presented of the important roles played by the boundary and interface concentrations and the diffusion coefficients in controlling interface movements and interdiffusion.

McEwan and Milner (46) studied diffusion bonding phenomena of dissimilar metals using roll bonding techniques. The metal pairs studied were divided into four groups:

Immiscible Pairs - Cd-Fe, Fe-Pb, Cu-Pb, Cu-Mo
Partially Miscible Pairs - Cu-Fe, Cu-Ag, Al-Zn
Miscible Pairs - Cu-Ni, Fe-Ni, Mg-Cd
Intermetallic Forming Pairs - Cu-Al, Fe-Al, Ca-Pb, Mg-Al, Ag-Al, Fe-Mo

On roll bonding at high temperatures or on post-bonding heat treatment, the properties of the joints were found to be determined by the extent and effect of interdiffusion. Solid state bonded joints of immiscible and partially miscible systems were generally stable. Miscible systems could be readily bonded but sometimes the bond weakened upon heat treatment due to formation of diffusional porosity. Two types of behavior were observed for intermetallic forming pairs depending upon whether or not the interface layer possessed any ductility. In most cases a brittle intermetallic layer was formed and the bond was strong if the layer was maintained below a critical thickness, but weak if it grew above this thickness. In some cases a ductile intermediate phase was formed. These joints were usually strong, with failure generally occurring in the weaker metal.

LeClaire and Bear (44) investigated the interdiffusion of uranium and aluminum and related the diffusion zone width as a function of temperature, pressure and time. Their experiments showed an appreciable accelerating effect of pressure on the rate of interdiffusion. It was suggested that this phenomenon might result from one of the following pressure induced changes: (1) changes in the interface concentrations, (2) allotropic transformations in existing phases, or (3) nucleation of new stable phases. Castleman (12) confirmed the pressure induced acceleration of diffusion and attributed it to increased formation of UAl3 phase possibly resulting from a decrease of voids in the diffusion zone.

Baird⁽³⁾ studied formation of intermediate alloy layers in the interdiffusion of metals and observed that parabolic growth of phase layers takes place at relatively high temperatures where volume diffusion is the controlling factor. At low temperatures the growth takes place predominantly by grain boundary diffusion in accordance with the following growth equation:

 $x = Kt^{1/n}$; where n may be between 2 and 4

In this equation x is distance, t is time and K is a constant.

Eckel (29) studied diffusion across dissimilar metal joints using diffusion couples of ferritic steels to austenitic stainless steels or nickel-base alloys. Carbon was observed to migrate in the direction of higher alloy content and also in the direction of lower carbon content. Evidence indicated that Fe diffuses into an austenitic steel from a ferritic steel, that Ni diffuses into the ferritic steel and that Cr diffuses away from the interface in both directions.

SOLID STATE WELDING PROCESSES

In simple terms, the only requirement for solid state welding is to bring the mating clean surfaces close enough together to permit their interatomic attractive forces to become effective. The essential ingredients in solid state welding are time, temperature and pressure. These controlling factors are interdependent.

Solid state welding per se is not a new joining technique; in fact, forge welding is one of the oldest joining methods known. However, in recent years, solid state welding has received increased attention. Review of the literature reveals a variety of solid state welding processes and in addition a number of different terms are used to describe essentially the same process. For purposes of this survey, the various solid state welding processes will be placed in one of the following classifications:

Press Bonding
Roll Bonding
Gas-Pressure Bonding
Explosive Bonding
Resistance Diffusion Welding

Press bonding and roll bonding are of primary concern in this survey, and the other techniques will be reviewed briefly as related processes.

Press Bonding (Pressure Bonding)

Press bonding may be accomplished at room temperature, in which case it is sometimes referred to as cold pressure welding (4), or it may be performed at elevated temperatures, in which case it has been termed by various investigators as hot pressure bonding, (59) recrystallization welding (55) or diffusion bonding (48,63)

Tylecote (61) conducted an investigation to establish certain of the principles involved in the process of pressure welding. The degree of deformation required to produce good welds at room temperature was determined for various metals and alloys in both similar and dissimilar pairs. Minimum deformation ranged from 10% to 90% for the various materials. Particular attention was given to determining the connection between weldability and properties of the surface oxide film.

Practical applications of pressure welding from 1946 to 1956 were summarized by Tylecote. (62) Techniques and advantages of the process were discussed. The main applications were given as: (1) lap joints in copper and aluminum alloy sheet and (2) butt joints in steel bar or sections.

Parks (55), as a result of investigating pressure welding at elevated temperatures, reached the conclusion that recrystallization is the basic mechanism of solid phase welding and stated that the process should be rightly called "recrystallization welding". Test data are presented to show relationship between recrystallization temperature, degree of deformation, weld temperature and weld strength of various common sheet metals. It is stated that welding below the recrystallization temperature can only be achieved if the surface layers are severely deformed during welding. Such welding is described as "deformation recrystallization welding".

Pressure diffusion bonding has been adapted to the fabrication of expandable honeycomb core in a process called Astroweld. (67) This process has been used to produce titanium, stainless steel, superalloy and refractory alloy honeycomb core with and without the use of intermediate alloys. (63) Titanium alloy honeycomb sandwich panels have been successfully fabricated by low pressure (creep controlled) bonding to avoid crushing of the core. (47)

Although the more pertinent references have been reviewed, a number of other investigations of solid state welding by press bonding techniques involving both similar and dissimilar metals have been included in the bibliography.

Roll Bonding

Roll bonding has been extensively studied for joining of both similar and dissimilar metals and has led to the successful development of processes for cladding of sheet and plate and more recently for fabrication of structural shapes. Appreciable deformation, in the order of 25 to 90 per cent in thickness, generally is required to achieve good metallurgical bonds during roll bonding.

Cladding of aluminum with stainless steel by roll bonding has been developed by Dulin. (28, 71) Using this process, a stainless steel clad aluminum product has been marketed under the trade name "Duranel." (64)

Roll bonding has also been used to clad steel with titanium for applications in the chemical industry. (40,51) In the cladding process, the temperature during roll bonding is relatively low in order to minimize the interdiffusion of the bimetals and to avoid formation of brittle interface layers.

Recently roll bonding has been applied to fabrication of structural shapes for aerospace applications. (70) The process has been developed for fabrication of titanium alloy roll-bonded sandwich structures. Extensive deformation, in the order of 60% in thickness, is involved and steel inserts forming the internal voids are removed by acid leaching.

Gas-Pressure Bonding

The gas-pressure bonding process makes use of high isostatic pressures at high temperatures to produce solid state diffusion bonding. (54) No deformation results during gas-pressure bonding except that necessary to bring the mating surfaces into intimate contact. The components are assembled in an expendable container or are edge welded to provide a pressure tight evacuated envelope. The assembly is heated to an elevated temperature in an autoclave containing an inert gas at high pressure.

This process has been used to produce metallurgical bonds between a variety of similar and dissimilar metals with and without the use of intermediate materials. Gas pressures generally range up to 15,000 psi and bonding temperatures up to 3000°F are used. (8)

Explosive Bonding

Explosive bonding is a relatively new concept in metal bonding and extensive development is still required before this method will be used for general application. Studies on explosive bonding of similar and dissimilar metals have been conducted by Pearson and Hayes. (56) Developments by several companies have been reviewed by Fairlie. (32) Potential applications include bimetal cladding of sheet and plate and bimetal lining of tubes. A fundamental study of explosive bonding by Cowan and Holtzman (23) indicated that three types of bonds can be achieved by high velocity impact of plates: (1) a direct bond, (2) a uniform layer of solidified melt and (3) a wavy interface of regions of direct bond and solidified melt. Bonding is related to jetting of the metal surfaces in the space ahead of the region of impact.

Resistance Diffusion Welding

The resistance diffusion welding process has been used to form solid-state metallurgical bonds in both similar and dissimilar metals using conventional resistance welding equipment. Applications for this process have included bonding of dissimilar metal combinations which form brittle compounds upon melting and to similar metal bonding where melting causes degradation of joint properties. Hess and Nippes (37) investigated resistance diffusion bonding of 3003 aluminum alloy to AISI 4140 steel. The best joint properties were developed using electroplated silver as an intermediate material.

SOLID STATE BONDING OF DISSIMILAR METALS

Review of the state-of-the-art of solid state bonding of dissimilar metals can be presented most effectively by dividing the subject on the basis of two joining systems: joining of bare metals and joining using intermediate materials.

It is apparent from a survey of the literature that joining of dissimilar metals with intermediate materials has achieved far greater success than using bare metals in both the application of diffusion bonding to production assemblies and to the development of optimized bonding parameters in research investigations. The following reasons have been indicated for the greater attention given to use of intermediate materials: (31) (1) the proper choice of intermediate materials permits bonding at temperatures below where any important metallurgical changes in the base metals take place, (2) another metal in the joint area may permit bonding to occur without the formation of detrimental intermetallics between the base metals, (3) the use of plated intermediate layers overcomes the problem resulting from the formation of tenacious oxides on the surfaces, and (4) it is possible to "diffusion braze", a process whereby a transient liquid phase forms at the interface and is then completely diffused away into the base metal.

In the following review of solid state welding of dissimilar metals, primary emphasis is placed on press bonding and roll bonding. Particular consideration is given to the following factors:

- Dissimilar metal combinations
- Time, temperature, pressure parameters
- Intermediate materials
- Atmosphere
- Surface condition and preparation

Joining of Bare Metals

Storcheim (59) reported on a pressure bonding investigation with Al-Ni, Al-Cu, Al-Fe and Al-Zr diffusion couples. Variables of time, temperature and bonding pressure were studied with regard to joint strength and metallographic structure. In general, as the temperature increased, joint strength increased, reached a maximum and then decreased when brittle intermetallics were formed. Time at bonding temperature had the same effect. At properly chosen temperatures and pressures, depending upon the diffusion couple, the strength increased with increasing bond pressure. Metallographic studies indicated that detrimental diffusion-zone thicknesses can be controlled by proper application of pressure. Increasing pressure decreases the intermetallic zone thickness for the Al-Ni and Al-Cu systems, while the reverse is true for the Al-Fe system.

Specimens were bonded in a vacuum of less than 5 microns. Mechanical abrading of specimens immediately prior to bonding gave the most expedient and reproducible results. Data presented for Al-Ni couples showed bonding pressures to 40,000 psi, bonding temperatures from 750°F to 1110°F and

times from 1 to 10 minutes. Maximum ultimate tensile strength (20,000 psi) was obtained using 40,000 psi pressure, 930°F temperature and 4 minutes bonding time.

Cooke and Levy⁽²²⁾ investigated the diffusion bonding of various aluminum alloys to 18-8 stainless steel. Diffusion bonding was accomplished by the following three methods:

- 1. By pressing a bar of aluminum to a bar of stainless steel (butt joint). At the desired pressure and temperature the bars were twisted in relation to each other to produce the bonded joint.
- 2. By inserting a tapered stainless steel rod into a tapered hole of an aluminum block and heating to the desired temperature followed by pressure to force the steel rod into the tapered hole to produce the bonded joint.
- 3. By pressing an aluminum alloy block against a stainless steel block (butt joint) at the desired temperature. Pressures approximating four times the yield strength were used. Both parts were enclosed in a die to prevent the aluminum from flowing.

Pressure bonding was conducted in air at temperatures from 400° to 850°F. Parts were cleaned by mechanical methods prior to bonding. Tensile strengths of the bonded joints varied from 10,000 to 30,000 psi.

Strengths of bonded assemblies were found to be increased by as much as 100 per cent by subsequent annealing. It was suggested that this increased strength was due to formation by diffusion of a very thin (submicroscopic) film of an intermediate phase, probably an intermetallic compound. When this phase is thick enough to be observed microscopically, then the bond strength is greatly reduced.

Both 32S and B18S aluminum alloys bonded to 18-8 stainless steel (AISI 303, 310, 321) frequently could be strengthened by solution heat treatment and aging, without lowering the bond strength.

II. S. Patent 2,908,073⁽⁷¹⁾ issued to Dulin and assigned to Alcoa, relates to methods for bonding aluminum alloys to steel and copper alloys. The methods outlined are adaptable for joining flat plates of aluminum to steel for use in the manufacture of electric flat irons and cooking utensils. Prior to joining, both the aluminum and the dissimilar alloy are cleaned by mechanical or chemical means. The parts being joined are pressed together to exclude air from the joint and are heated to a temperature range of 700 to 950°F. Pressure is increased to cause the aluminum to flow laterally. The pressure required varies from 15,000 to 50,000 psi depending on the temperature used. The aluminum thickness is reduced from 10 to 50% during bonding. The patent claims bonded joints can be obtained which have high peel strength and without the formation of intermediate phases.

- Dulin (28) discusses various applications for the pressure bonding of aluminum to steel and copper alloys using the process described in the above patent. A number of examples are described of aluminum alloys bonded to stainless steel for fabrication of flat irons, cooking utensils and transition tubes.
- Wood (64) discusses a stainless steel clad aluminum sheet product fabricated by the above process which is being marketed under the trade name "Duranel". The laminated sheet is composed of AISI 304 stainless steel roll bonded to 3004 aluminum alloy. The metallurgical bond can withstand forming into various shapes without detrimental effects.
- Tylecote (61) pressure bonded Al to Fe and Al to Ni at room temperature with 50% and 60% deformation, respectively, required to achieve good bonds. Heat treatments of joints at 750°F caused no change in joint strength while heat treatment at 930°F caused severe loss in joint strength. Attempts to bond Ni to Fe at room temperature were unsuccessful even with deformations as high as 72%.
- Albom⁽¹⁾ briefly described an example of diffusion bonding of titanium to stainless steel. Formation of intermetallic layers is cited and the use of intermediate material is suggested.
- Hughson (40) described a process developed by Lukens Steel Company for producing titanium clad steel plate. An enclosed sandwich of titanium sheet and A-204 low-carbon manganese steel plate was flushed continuously with argon while being heated before rolling, to eliminate impurities that interfere with the bond. Window frame type spacers were used to separate the titanium sheet and steel plate during heating in order to permit argon flushing and to prevent diffusion. Two clad plates, with the titanium sheets back to back, were roll bonded in one sandwich. Preheat temperature limit before rolling was 1700° to 1750°F. A reduction of at least three to one was required for good bonding. No intermediate material was used. Formation of brittle compounds was prevented by limiting the time at elevated temperature in which diffusion could take place.
- Moore (51) described the use of titanium-clad steel for process equipment. The methods of producing titanium clad steel are briefly described. The first method is the Lukens Steel process whereby the composite is produced by simple roll bonding at a temperature below that at which brittle compounds can form. Explosive cladding and brazing are described as additional methods for producing titanium-clad steel plate. Limited information is presented on fabrication details.

Diffusion phenomena were reviewed extensively in a previous section, "Fundamentals of Diffusional Bonding" and these references will not be repeated. However, it should be emphasized that formation of brittle phases at the interface of Al-Fe, Al-Ti, Ti-Fe and Ti-Ni couples is one of the key factors which is responsible for the lack of progress in development of processes for bare metal joining of these metals and their alloys.

Joining With Intermediate Materials

The literature contains only limited information on solid state bonding of dissimilar metals using intermediate materials. Most of these references involve joining of aluminum alloys to steel. However, the very use of an intermediate metal provides a dissimilar metal interface. For this reason pertinent information can also be obtained by review of similar metal joining using intermediate material and several of these references are included in this section.

Crane, et al, (24) developed a diffusion bonding technique for joining of 2219 aluminum alloy to 321 stainless steel using electroplated silver as an intermediate material. Successful diffusion bonding was accomplished at temperatures of 500° to 600°F, pressures of 20 to 25 KSI and times of 2 to 4 hours. The joint faying surfaces were electroplated with silver prior to bonding. The silver prevented the formation of oxide-film barriers and controlled formation of embrittling phases.

Processing techniques for diffusion bonding of large diameter assemblies were successfully developed and demonstrated by fabrication and testing of four 20-inch diameter joints. An unique diffusion bonding method was developed utilizing simple differential thermal expansion tooling. The method can be economically adapted to production requirements.

It was concluded that diffusion bonding is the most satisfactory method for joining large diameter stainless steel to aluminum alloy cylinders and is superior in strength and reliability to welding or brazing.

Orysh, Betz and Hussey (53) investigated procedures for diffusion bonding 3003 aluminum alloy to 304 stainless steel and 2024 aluminum alloy to 1020 steel. In all cases the steel was precoated with a silver brazing alloy (BAg-10) using an oxyacetylene torch. The silver layer was then ground flat to a thickness of .005 inches.

Prior to bonding, the silver layer was cleaned with alcohol and the aluminum was chemically cleaned using an alkaline etch followed by dilute nitric acid and water rinse. Bonding was attempted at temperatures varying from 200°F to 800°F, varying pressure and time. From this screening test the authors selected a bonding temperature of 800°F and a pressure of 2400 psi (2.3% joint deformation) as being optimum.

The following data show the shear test results as a function of bonding time for silver coated 1020 steel bonded to 2024 aluminum alloy using a bonding pressure of 2400 psi and a bonding temperature of 800°F.

Time, min.	Shear Strength, psi
10	2070
3 0	1900
60	2730
120	3330
180	3110
240	2300

^{*}Average of three specimens

Gatsek⁽³⁶⁾ reported that preliminary studies have shown feasibility of a process for diffusion bonding 2219 aluminum alloy to 321 stainless steel. The process involves applying and bonding an intermediate alloy to the surface of the stainless steel and plating the aluminum alloy with the same intermediate material. The interface material serves two purposes; it is used as a bonding agent and as a barrier material between the aluminum and the steel. Tin and silver are mentioned as interface materials but comparative results are not indicated.

Hess and Nippes (37) investigated joining of 3003 aluminum alloy to SAE 4140 steel using resistance welding techniques. It was found necessary to use an intermediate material to avoid overheating the steel. The intermediate material was applied by electroplating the steel. The metals used for plating were tin, zinc, silver, copper, nickel, chromium and cadmium. Aluminum was resistance bonded to electroplated steel using an 800 pound electrode force and 10 cycles welding time. Current was varied to produce the desired penetration into the aluminum side of the joint. Silver was selected as the most desirable intermediate metal for bonding the 3003 aluminum alloy to 4140 steel. Copper was considered satisfactory but required more rigid welding controls to prevent brittle joints. It was observed that plating procedures must be rigidly controlled because much of the bond strength of the joint depends upon the adherence of the plating to the steel.

Barta⁽⁵⁾ investigated low temperature (300°-450°F) similar metal diffusion bonding of 7075, 6061 and 2219 aluminum alloys using diffusion aids in various forms including electroplated, vacuum deposited, plasma sprayed, loose shim material and clad aluminum. The best intermediate material was clad aluminum. Shear strengths of 9,000 to 10,000 psi were obtained on 7075 aluminum alloy clad with 7072 aluminum alloy which was bonded at 325°F for 1 hour at 24,000 psi pressure.

Electroplated Cu, Sn and Zn did not prove satisfactory as diffusion aids when bonded at low temperatures. Electroplated Ag gave good shear strengths (6,700 psi) when bonded at 450°F for 4 hours at 16,000 psi pressure. Silver was not considered as a good intermediate material because it would create a galvanic reaction with aluminum under certain atmospheric conditions.

Search of the literature disclosed no references to diffusion bonding of titanium alloy to aluminum alloy, titanium alloy to stainless steel or titanium alloy to nickel-base alloy.

Several references were found on diffusion bonding of titanium to titanium using intermediate materials, but in most cases these involved a transient liquid phase. For example, DeCecco and Parks (26) investigated bonding titanium using silver as an intermediate material. The silver melted and was squeezed out of the joint and served primarily as a cleaning and flushing agent, while the clean titanium surfaces were forced into intimate contact and bonded by recrystallization welding.

Clark (20) investigated silver brazing of titanium to titanium in which the silver diffused away from the joint area by holding for 1 hour at 1850°F in a vacuum. Tensile strengths up to 67,000 psi at room temperature were

obtained in Ti-5Al-2.5Sn alloy. Diffusing of the silver away apparently resulted in a ductile joint by dispersing of the detrimental silver-titanium intermediate phase.

No references were found on diffusion bonding of stainless steel to nickel-base alloy using intermediate materials. Davies and Stephenson (25) developed similar metal joining procedures to diffusion bond a nickel-base alloy utilizing nickel foil or electroplate. The process included final diffusion and aging treatments. Several other interlayer materials were investigated and found to be unsatisfactory. Titanium and aluminum interlayers resulted in formation of brittle intermetallic compounds, and cracks were observed after the final diffusion treatment. Successful joints were obtained with copper interlayer, but shear strengths were lower than for nickel, and higher bonding temperatures were needed.

Eckel⁽²⁹⁾ used nickel foil as a diffusion barrier in studying diffusion across dissimilar metal joints of ferritic steel to austenitic steel. The nickel barrier reduces but does not completely prevent carbon migration. The formation of voids in the barrier layer was attributed to nickel diffusion. The diffusion joints were bonded at 1300°F and then given diffusion treatments at 1300°F for times up to 8000 hours in vacuum.

Niemann, et al, (52) investigated techniques to diffusion bond beryllium copper to Monel at the aging temperature (650°F) of the beryllium copper. In all tests, two electroplated material combinations were used, one on the beryllium copper and a different metal on the Monel. A bonding pressure of 6,700 psi and 3 hours bonding time (aging time of Be-2Cu) were found to be adequate. Diffusion bonding was conducted in an inert gas atmosphere. Ductile, high strength joints were obtained with Au-Cu and Au-Ag plating combinations. Joints bonded with Au-Pb and Au-Al plating combinations were found to be quite brittle. The average shear strengths obtained for Au-Cu (10,150 psi) and Au-Ag (10,550 psi) systems were assumed to provide a tensile strength of at least 20,000 psi for these joints. The tensile strengths of gold and silver are about 20,000 psi and for copper about 32,000 psi. It was indicated that joints made with these metals could be as strong or stronger than the metals themselves because of alloying that takes place and the state of stress that exists because of joint dimensions.

STATE-OF-THE-ART TOUR

State-of-the-art visits were conducted to five selected organizations believed to be most able to provide information directly related to solid state welding of dissimilar metals. The tour was taken to supplement the literature survey in determining the work on solid state bonding which has been completed or is in progress by industry, government agencies or research institutions.

Organizations to be visited were selected on the basis of information gathered by the literature search followed by direct confirmation with the organization. Visits were made to:

Alcoa Research Laboratories
Battelle Memorial Institute
North American Aviation
Northrup-Norair
Westinghouse Astronuclear Laboratories

The two current Boeing-NASA contracts on joining dissimilar metals were reviewed with each organization in order to promote a better discussion of diffusion bonding techniques.

The general conclusions drawn from these visits are as follows:

- 1. Extensive R&D work is being conducted on diffusion bonding, but only a very limited amount of this effort is devoted to joining of dissimilar metals.
- 2. Organizations engaged in diffusion bonding development consider their work very proprietary and extremely competitive, and for these reasons are reluctant to discuss details of bonding procedures or to show their laboratory facilities.
- 3. The primary benefit resulting from this state-of-the-art tour was the personal cognizance of the work being conducted under active and completed R&D contract effort on diffusion bonding and the sources for obtaining the reports prepared under these programs.

Following is a summary of pertinent technical information obtained:

ALCOA RESEARCH LABORATORIES, ALUMINUM COMPANY OF AMERICA - New Kensington, Pa.

Principal Contact: Dr. P. T. Stroup

Assistant Director of Research

Alcoa has been active in development of solid-state diffusion bonding and transient liquid phase bonding methods for joining of aluminum alloys and joining of aluminum alloys to dissimilar metals, particularly stainless steel and copper. Solid-state diffusion bonding processes have been developed which include a process for roll bonding of stainless steel clad aluminum and a process for pressure bonding of aluminum to stainless, carbon and alloy steels, and to copper.

Roll-bonded stainless steel clad aluminum sheet is marketed under the trade name "Duranel" and consists of a thin sheet of 304 stainless steel bonded to 3004 aluminum alloy. This material is currently in production for cooking utensils. Alcoa is promoting stainless steel clad aluminum for other applications and The Boeing Company, Commercial Airplane Division, has shown interest in this material for engine fire walls and for water storage tanks.

Dr. Stroup indicated that they have no currently active R&D programs for solid-state diffusion bonding of either aluminum alloys or dissimilar metals. Although Alcoa has conducted diffusion bonding work with electroplated silver, they felt that the problem of galvanic corrosion between silver and aluminum limited the potential applications for this process.

Recent R&D efforts have been directed at liquid-phase diffusion-controlled eutectic bonding techniques. The R-260 bonding process has been developed for joining aluminum and the R-318 bonding process for joining copper to aluminum. Both of these techniques utilize a carbon block brazing press with electrical contact resistance heating. Copies of reports on the eutectic bonding techniques and several other useful reports were obtained.

BATTELLE MEMORIAL INSTITUTE - Columbus, Ohio

Principal Contact: Donald C. Carmichael

Assistant Chief, Materials Development Division

Battelle has been very active in development of solid state diffusion bonding techniques with primary emphasis on similar metals although some effort has been devoted to dissimilar metal joining. Major developments have been accomplished in gas-pressure bonding, diffusion bonding of refractory metals and roll bonding of sandwich structure.

R&D programs on solid-state bonding have been conducted by the Materials Joining, Materials Development and Non-ferrous Metallurgy Divisions. The Materials Development Division which is responsible for gas-pressure bonding has performed the major part of past R&D contracts on diffusion bonding. The Non-ferrous Metallurgy Division is responsible for development of roll bonding of sandwich structure. The Joining Division is responsible in general for solid-state bonding development with the exception of gas-pressure bonding and roll bonded sandwich structure.

A list was obtained of fifteen diffusion bonding programs completed or in progress at Battelle. Four of these may provide useful information for the current Boeing-NASA contract.

	Title	Agency	Contract No.
1.	Preliminary Study of Diffusion Bonding Techniques for Use in Making Tubing Connections in Stainless Steel	NASA-GE	Sub-contract from General Electric
2.	Fundamentals of Solid-State Welding and Their Application to Beryllium, Aluminum and Stainless Steel	Army Missile Command, Red- stone Arsenal	DA-01-021-AMC- 11706 (Z)
3.	Effects of Surface Preparation on Solid State Bonding	AEC	W-7405-eng-92
4.	Development of Techniques for Making Throat Blocks for Gas Dynamics Facility by Diffusion Bonding	USAF	af40(600)-706

The following reports in connection with these programs were recommended:

"Preliminary Study of Diffusion Bonding Techniques for Use in Making Tubing Connections in Stainless Steel" by P. A. Kammer, R. E. Monroe, and D. C. Martin, dated July 29, 1964.

Obtain from:

Mr. Forrest O. Rathbun Advanced Technology Laboratories General Electric Company Schenectady 5, New York

"Fundamentals of Solid State Welding and Their Application to Beryllium, Aluminum and Stainless Steel".
by D. Hauser, P. A. Kammer, and D. C. Martin, dated July 15, 1965

Obtain from:

Army Missile Command Redstone Arsenal Huntsville, Alabama

"Effects of Surface Preparation on Solid State Bonding" by D. J. Diersing and D. C. Carmichael

Obtain from:

AEC (Expected to be released by October 1965)

A number of abstracts of pertinent literature were obtained during a visit to the DMIC library.

NORTH AMERICAN AVIATION INC./LOS ANGELES DIVISION, Los Angeles, Calif.

Principal Contact: Julian King, Jr.
Supervisor, Metallurgy, Research and Engineering

North American has been very active in diffusion bonding and has developed processes for electric blanket bonding, resistance spot diffusion welding, roll bonding and press bonding. Mr. King indicated that NAA has been supporting diffusion bonding development on company research funds for the past six years. In view of the proprietary and competitive nature of solid state bonding research, discussion was restricted to government sponsored programs and no tour was made of the laboratories.

The following programs were mentioned as being currently active:

NASA: Development of High-Strength Low-Density Composite Materials for Saturn Applications

Roll Bonded Titanium Alloy "Y" Ring

Roll Bonded Tank Wall Shape

Research and Development for Fabricating a Simulated Titanium Alloy Segment for S-1C

Titanium S-1C Skin Sections (Steel Tank Wall)

USAF: Roll Bonded Conjugate Tank Development

Diffusion Bonded Honeycomb Sandwich Panels

Spot Diffusion Bonding of Beryllium and Titanium

Mr. King was up-to-date on the Boeing-NASA contract for joining of stainless steel to aluminum alloy tubes. He indicated that in view of the success achieved by Boeing on diffusion bonding of dissimilar metal tubing he was considering this technique in connection with some of the NAA programs.

NORTHRUP-NORAIR, Hawthorne, California

Principal Contact: Dr. E. B. Mikus
Chief, Materials Research

Northrup is actively engaged in development of diffusion bonding techniques with particular emphasis on diffusion bonded sandwich structure. Dr. Mikus was most cooperative in discussing their research programs and showing their laboratory facilities. However, he emphasized that information on diffusion bonding of titanium alloy sandwich structure is highly proprietary and competitive and should only be considered as being given on a confidential basis. In any event the Northrup titanium diffusion bonding process is not applicable to the Boeing-NASA dissimilar metals program and specific processing details will not be discussed in this report.

Northrup has not been working on diffusion bonding of dissimilar metals. Current and past diffusion bonding research has emphasized titanium alloys and refractory metals with the greatest advances made in liquid-phase diffusion bonding techniques. They feel this is the most feasible approach for honeycomb sandwich structures because of the light pressures which can be tolerated during bonding. The most significant problems encountered have been core crush and procedures for scale-up evaluation. Solid-phase diffusion bonding is being developed for refractory alloys in addition to the diffusion sink brazing approach. This work is being conducted on several in-house diffusion bonding research programs and the several USAF R&D contracts.

Several unique items of research equipment were observed during the tour of the laboratories. One of these was a welding research instrument, called a "Gleeble" Model 510, manufactured by Duffers Associates Inc. This instrument can duplicate time-temperature or time-temperature-pressure parameters to simulate conditions in welds and heat affected zones on an expanded scale. The instrument contains a dilatometer which is particularly useful for study of phase transformations.

WESTINGHOUSE ASTRONUCLEAR LABORATORIES, Large, Pa.

Principal Contact: David C. Goldberg
Director, Materials Department

Westinghouse Astronuclear Laboratories are currently active in diffusion bonding on classified AEC programs. Mr. Goldberg indicated that this work involves bonding to graphite, and the information would not be applicable to the Boeing-NASA program in any event. In addition to the classified work, the Laboratories recently received two NASA R&D programs on austenitic/refractory bimetallic materials. These are:

Joining of Austenitic/Refractory Bimetal Tubing Evaluation of Austenitic/Refractory Bimetals

The project engineer for these programs is:

Phil Stone NASA Lewis Research Center Cleveland, Ohio

The joining program involves development of gas tungsten-arc and electron beam welding techniques for joining of the dissimilar metal tubing. Tubing will be supplied from another NASA program and will include stainless steel clad and Inconel clad columbium and tantalum alloys.

The bimetal evaluation program involves investigation of thermal exposure and thermal cycling, under simulated space power system environmental conditions, on properties of dissimilar metal sheet. Diffusion and thermal stresses between the dissimilar metals under ultrahigh vacuum and temperatures up to 1550°F for thousands of hours of service are the problem areas. The materials are explosive bonded bimetal sheet consisting of 316 stainless steel and Inconel 600 bonded to Cb-1Zr, FS-85 columbium alloy and T-222 tantalum alloy.

CONCLUSIONS

The state-of-the-art of joining dissimilar metals by solid state welding has been determined. The conclusions reached as a result of this survey are:

- 1. Extensive development work has been conducted on solid state welding, but only a very limited amount of this effort has been devoted to joining of dissimilar metals. Basic research and development work to delineate the problems and their solution is now needed as the first step toward development of practical processes for solid state welding of dissimilar metals.
- 2. A considerable amount of useful information is available on solid state welding of aluminum alloys to steels. Available information on joining of other dissimilar metal systems which can be directly applied to Contract NAS 8-20156 is limited.
- 3. Solid state welding of dissimilar metals has been more successful when intermediate materials are used than when only the bare metals are joined. The principal reasons for this greater success are that intermediate materials have enabled use of lower bonding temperatures, control of detrimental intermetallic formation, control of oxide formation and development of a transient liquid phase bonding process.
- 4. Interdiffusion of Al-Fe, Al-Ti, Ti-Fe and Ti-Ni results in formation of brittle phases at the interface. Control of this interdiffusion is a key factor in the development of solid state welding processes for dissimilar metals. Control of detrimental interdiffusion is greatly simplified by use of intermediate materials.
- 5. Accelerated rate of corrosion is a potential problem in joining of any dissimilar metal combination. The probability of encountering this problem is significantly increased by use of intermediate materials. Careful consideration should be given to corrosion effects in study of solid state welding of dissimilar metals.

ANNOTATED BIBLIOGRAPHY

1. Albom, M. J., "Solid State Bonding", Welding Journal, June 1964, pp 491-504.

A review of the fundamentals of solid state bonding is presented with a limited discussion on actual bonded samples. Beryllium, Ti-75A titanium, Cb-752 columbium, Mo-0.5Ti, and Ta-10W tantalum alloys were pressure bonded. Beryllium was bonded at a temperature range from 1500° to 2800°F for 1 hour in a vacuum of 5x10°5 torr. Molybdenum clamps were used as fixtures with differential thermal expansion between the molybdenum and beryllium providing the pressure needed. The resultant joint showed no evidence of grain coarsening, and pull tests produced 100% joint efficiencies in butt joint round test specimens. For Ti-75A, temperatures less than 1700°F at pressures ranging from 200-600 psi at times up to 15 minutes in a protective atmosphere were used. Satisfactory solid state bonds in Cb-752 were obtained at pressures less than 1,000 psi at 2000° to 2200°F in a protective atmosphere. The Mo-0.5Ti alloy was bonded at 2100°F for 15 minutes at 500 psi in a protective atmosphere.

Examples of dissimilar metal combinations of Mo-0.5Ti to Cb-752, Mo-0.5Ti to Ta-10W, and stainless steel to titanium were also mentioned. Diffusion of titanium and stainless steel was observed to cause formation of brittle interface layers.

2. Anderson, O. L., "Adhesion of Solids: Principles and Applications", Bell Laboratories Record, November 1957.

The principles of adhesion of solids are discussed with emphasis on the effect of localized plastic deformation and piercing of oxide film in facilitating adhesion. Investigation led to development of a technique called thermo-compression bonding for connecting electrical leads to germanium and silicon semiconductors. Features of process are bonding in air without fluxes, low pressures ensure no mechanical or crystal structure changes in semi-conductor, low temperatures ensure no diffusion of metal into semiconductor. Most success is achieved when metallic filament is very soft.

3. Baird, J. D., "The Formation of Intermediate Alloy Layers in the Inter-Diffusion of Metals", Journal of Nuclear Energy, Part A: Reactor Science, Vol.11, 1960, pp 81-88.

An analytical discussion is given of conditions where the equilibrium phases are not formed in a diffusion couple of two metals. The absence of a phase predicted by the equilibrium diagram could be due either to difficulty of nucleation or to a very low growth rate. The latter is probably the reason for many of the reported cases, especially where missing phases have higher melting points (and therefore probably lower diffusivities) than the other intermediate phases in the system. It is postulated that once nucleated, a phase layer should grow parabolically

if growth is controlled by volume diffusion through the layer. However, if grain boundary diffusion predominates, it is stated that non-parabolic growth of a phase layer occurs. In addition, grain boundaries must be considered as a channel which feeds diffusing material into the grains on either side. Grain boundary diffusion is predicted to predominate at low temperatures and volume diffusion at high temperatures. Oxide layers are expected to be important mainly in the earlier stages of inter-diffusion and will limit initial phase growth intermediate between linear and parabolic.

The most striking and unexpected feature of experimental work on layer growth is the large effect of pressure. One explanation of the effect is that the stabilities of the intermediate phases are altered by pressure.

According to the conclusions reached, parabolic growth rates of layers are to be expected at relatively high temperatures where good contact is maintained between the metals by the application of pressure, and where precautions are taken to prevent interference from oxide films.

4. Barnes, W. A., "Cold Pressure Welding", Tool Engineer, Vol. 32(5), May 1954, pp 75-78.

A general summary of cold pressure welding is discussed without getting into any of the technicalities involved. This method is effective only in joining relatively soft materials such as aluminum, copper, lead. tin, nickel, silver, and platinum to themselves or each other.

When joining foils of 0.005 inches or less, a clean oxidized surface is required; materials over 0.006 inches are required to have an abraded or galled surface. Lap or butt welds are feasible. Wires can even be joined by butting them and progressively cold flowing them together. Wire diameters successfully cold welded range from 0.032 in. to 0.375 in.

Impact tools can be used for some applications of cold welding; however, generally uniformly applied pressures are more satisfactory. Tensile and torsion tests on regular butt and lap welds have indicated strengths above 95% of that of the base metal.

5. Barta, I. M., "Low Temperature Diffusion Bonding of Aluminum Alloys", Welding Journal, Vol. 43, June 1964, pp 241s-257s

The diffusion bonding of aluminum alloys can be achieved at 300° to 450°F with clad aluminum sheet providing the best diffusion aid or intermediate material. Plasma spraying showed promise as a technique for applying diffusion aids. Electroplated Cu, Sn and Zn did not prove satisfactory as diffusion aids when bonding at low temperatures. Bare metal tonding proved unsatisfactory at low temperatures for the alloys tested: 7075, 6061, 2219. Surface preparation and cleaning are important for successful diffusion bonding. Machined serrations on interface surfaces give higher strength bonds with low temperature bonding.

6. Buginas, S. J., "Diffusion Bonding: An Annotated Bibliography", Lockheed Missiles and Space Co., Sunnyvale, California, Special Bibliography SB-63-5, March 1963.

This bibliography contains selected references on diffusion bonding and hot pressure welding from the years 1940-1962.

7. Butts, A., and Van Duzee, G. R., "Cold Welding of Silver", Trans. of the Electro Chemical Society, Vol. 74, 1938, pp 327-339.

Time-temperature-pressure relationship in pressure welding of silver was studied at temperatures from 390°F to 750°F and pressures up to 45,000 psi. A curve was obtained showing the minimum time necessary to obtain adherent welds at various temperatures under a pressure of 45,000 psi and under varying pressures at a temperature of 750°F. It was found that 390°F, the recrystallization temperature of the silver, is the lowest temperature at which adherent welds could be obtained, except possibly with greatly prolonged application of pressure. Above 390°F, the time necessary to produce an adherent weld decreases rapidly.

Welds obtained by rolling had greater bond strength than welds obtained by constant pressure in a fixed position. Very thin films of foreign matter between the silver surfaces under pressure caused little interference with welding.

8. Carmichael, D. C., "Diffusion Bonding Sets Sights on the Near Perfect Joint", Iron Age, July 18, 1963, pp 81-83.

Discussion of solid-state diffusion bonding, including the technique of gas-pressure bonding (isostatic bonding), vacuum-diffusion bonding and eutectic-diffusion bonding, of dissimilar metals and ceramics of complex shapes. High joint strengths are obtained, usually equal to that of the base metal. Emphasis on gas-pressure bonding using a high pressure autoclave with gas pressure up to 15,000 psi at up to 3000°F. Atomic diffusion bridges the differences between many dissimilar metals. Advantages are high strength bonds produced with comparatively low temperatures and with minimum of distortion.

9. Castleman, L. S., "An Analytical Approach to the Diffusion Bonding Problem", Nuclear Science and Engineering, Vol. 4., 1958, pp 209-226.

Attention is focused on the interdiffusional aspects of the diffusional bonding problem as it relates to the fabrication of clad fuel elements and their operation at elevated temperatures. Certain idealized cases of core-cladding interdiffusion occurring in single phase, two-phase, and three-phase systems are examined analytically. In the two-phase and three-phase systems, the importance of the roles played by the boundary and interface concentrations and the diffusion coefficients in controlling interface movement and interdiffusion is evaluated in detail.

10. Castleman, L. S., and Seigle, L., "Fundamentals of Diffusional Bonding I", Report No. SEP-227, General Telephone and Electronics Laboratories, Bayside, New York, August 13, 1956.

A detailed study has been initiated of the formation of intermediate layers in the diffusion zone of Al-Ni diffusion couples. This system was chosen because of its significance in fuel element technology. It has been confirmed that only two out of four possible layers form observably during diffusion - the Ni₂Al₃ and NiAl₃ phases. The growth of the intermediate layers occurs initially at a rate different from that predicted by the ideal diffusion equations: however, once a minimum thickness is reached, behavior becomes more normal.

11. Castleman, L. S. and Seigle, L., "Fundamentals of Diffusional Bonding II", Report No. SEP-245, General Telephone and Electronics Laboratories, New York, Feb. 15, 1958.

The study of the kinetics of layer growth in the aluminum-nickel system and the effect thereon of applied pressure has been continued. Additional information has been obtained about the growth of beta phase, and a method was developed whereby the kinetics of growth of the beta phase could be investigated. The implications of Vasileff's equations which provide the solution for interdiffusion in a three-phase two-component system have been examined semi-analytically, and quantitative insight has been gained into the kinetics of interface movement. Finally, an investigation of the kinetics of interface movement and layer growth has been begun on systems which have less complicated phase equilibrium relationships than those existing in the aluminum-nickel system.

12. Castleman, L. S., and Seigle, L., "Fundamentals of Diffusional Bonding III", Report No. SEP-251. General Telephone and Electronics Laboratories, Bayside, N. Y., June 30, 1958.

An investigation has been begun of interdiffusion in the aluminum-uranium system and of the effects thereon of applied pressure. In confirmation of the results of other investigations, it has been found that UAl₃ layer growth is accelerated by increasing applied pressure. The growth kinetics are characterized by a "transient" period, during which growth occurs non-parabolically, which is followed by a "steady state" period of parabolic growth. Interesting correlations are shown to exist between annealing time, annealing temperature, and applied pressure on the one hand, and the nature and distribution of structural defects in the UAl₃ diffusion zone.

13. Castleman, L. S. and Seigle, L., "Fundamentals of Diffusional Bonding IV", Report No. SEP-253, General Telephone and Electronics Laboratories, Bayside, N. Y., June 30, 1959.

The investigation of interdiffusion in the aluminum-uranium system and of the effects of applied pressure thereon have been substantially completed. The kinetics of growth of the UAl₃ phase have been determined in the temperature range 750° to 1110°F and at pressures of 3500, 10.000 and 20,000 psi. The origin of the macroscopic defects

occurring in the UAl₃ layer during interdiffusion has been clarified. The solubility range of UAl₃ has been determined by a nuclear track emulsion technique to be about 2.6 a/o uranium. Exploratory runs have been made with incremental diffusion couples of aluminum vs. UAl₃ and UAl₃ vs. uranium discs to determine the growth rates of the UAl₄ and UAl₂ phases. A theoretical estimate has been made of the extent to which pressure-induced shifts in equilibrium concentrations are important in influencing intermetallic layer growth in the aluminum-uranium and aluminum-nickel systems.

14. Castleman, L. S., Froot, H. A., and Seigle, L., "Fundamentals of Diffusional Bonding V" Report No. SEP-256, General Telephone and Electronics Laboratories, Bayside, N. Y., June 30, 1960.

An exploratory investigation has been completed of the effects of applied pressure on the kinetics of growth of intermetallic layers in the aluminum-thorium system. Contrary to an earlier published report, no appreciable effect of pressure is found, and the growth kinetics appear to be conventional. Also, preliminary data have been obtained on the effects of pressure on the kinetics of growth of the beta brass intermetallic layer in saturated alpha brass vs. saturated gamma brass couples.

15. Castleman, L. S., Froot, H. A., Seigle, L., "Fundamentals of Diffusional Bonding VI", Report No. SEP-258, General Telephone and Electronics Laboratories, Bayside, N. Y. June 30, 1961.

The study of the effects of pressure on the kinetics of growth of intermetallic layers in binary diffusion couples was continued. It was found that pressure diminishes beta-brass layer growth in saturated alpha-brass vs. saturated gamma-brass diffusion couples both above and below the critical transition temperature range for ordering. The effect of pressure appears to be less in the ordered temperature range. In the aluminum-uranium system, the growth of the UAl₃ intermetallic layer contrary to expectations, is not diminished in the pressure range 20,000 to 200,000 psi. An argument is advanced, based on the activation volume concept, that the aluminum ions diffuse in UAl₃ by an interstitial mechanism.

16. Castleman, L. S., Froot, H. A., and Swigle, L., "Fundamentals of Diffusional Bonding VII", Report No. SEP-260, General Telephone and Electronics Laboratories, Bayside, N. Y., June 30, 1962.

An investigation has been completed of the growth kinetics of betabrass layers in alpha-brass vs gamma-brass diffusion couples. It was found that useful estimates of the diffusion coefficients at the interfaces can be made from interface displacement data, and a useful estimate of the mean heat of activation for diffusion can be made from layer growth data. The effects of pressure on layer growth kinetics were explored and found to be consistent with a vacancy model for diffusion in beta-brass in both the disordered and ordered temperature ranges. Preliminary work has been started on investigation of multiphase diffusion in ternary systems. 17. Castleman, L. S., and Seigle, L., "Fundamentals of Diffusional Bonding VIII Final Report: Report No. GTR-21, General Telephone and Electronics Laboratories, Bayside, N. Y., Dec. 15, 1963.

A study was undertaken of interdiffusion in the copper-zinc-nickel system. The identity and sequence of appearance of the intermediate phases found in the diffusion zone were determined by correlation of metallographic observations of microstructure with details of the equilibrium phase diagram and with electron-beam-probe microanalysis. It was found that all except possibly one of the intermediate layers growing in the diffusion zones of the diffusion couples investigated consisted of single phases. In some of the couples the concentration of zinc did not vary monotonically with distance. The sequence of phases in the diffusion couples was independent of time at each temperature. The diffusion paths for couples having the same component as one terminus and a binary alloy of the remaining two components as the other terminus did not appear to cross. The tangent to the diffusion path in the vicinity of the terminal compositions tended to orient itself so that the angle between it and the lines radiating from the apex of the fastest diffusing component was small. In general, the results corroborated many of the observations and conclusions of Clark and Rhines for the aluminum-magnesium-zinc system.

18. Castleman, L. S., and Seigle, L. L., "Formation of Intermetallic Layers in Diffusion Couples", Trans. AIME, Vol.209, 1957, pp 1173-1174; Journal of Metals, October 1957.

Diffusion bonding studies of the aluminum-nickel system were used to demonstrate that the diffusion couple technique must be used with caution when attempting to identify phases present in an equilibrium diagram.

It was shown that the beta (NiAl₃) and gamma (Ni₂Al₃) phases occur at 600°C (1110°F) within minutes. Times in excess of 340 hours at 1110°F were required to produce identifiable delta (NiAl) and epsilon (Ni₃Al) phases. This indicated that high melting intermediate phases grow very slowly in diffusion couples. The delta and epsilon phases were observed to grow very rapidly at temperatures above the eutectic, which occurs at 640°F (1180°F).

19. Castleman, L. S., and Seigle, L. L., "Layer Growth During Interdiffusion in the Aluminum-Nickel Alloy System" Trans. AIME, Vol. 212, 1958, pp 589-596.

The kinetics of growth of intermediate phase layers in the aluminumnickel system has been explored in diffusion couples annealed in the temperature range 750° to 1160°F for times up to 340 hr. and under pressures up to 10,000 psi. It has been found that all phases appear in the interdiffusion zone at 1160°F that are thermodynamically stable at the diffusion temperature; these phases are the NiAl3(beta), Ni2Al3 (gamma), NiAl (delta), and Ni3Al (epsilon) intermetallic compounds. The rate controlling factor affecting the kinetics of growth of the gamma phase in the early stages is not yet clear; after a minimum thickness of 0.025 to 0.030 mm is reached, however, volume diffusion controls layer growth. It is estimated that the chemical diffusion coefficient is 9.1 x 10^{-10} sq cm per sec and that the heat of activation for diffusion is 31,000 cal per mol. An applied pressure of 10,000 psi reduces the diffusion rate by about 27 pct. Volume diffusion controls the growth kinetics of the beta phase after a minimum thickness of 0.040 mm is reached; it is estimated that the 1160° F diffusion coefficient is about 1.8×10^{-11} sq cm per sec and the heat of activation is approximately 27,000 cal per mol. Tentatively, it appears that pressure affects the growth of the beta phase much less than the gamma phase. The growth kinetics of the delta and epsilon phases also appear to be volume diffusion controlled.

20. Clark, E. J., "Vacuum Diffusion Joining of Titanium", Welding Journal, Vol. 38(6), June 1959, pp 251s-259s.

Titanium alloy Ti-5Al-2.5Sn components can be vacuum brazed with fine silver filler and diffused in one operation to give joints with tensile and shear properties equal to that of base metal.

Where a metal-to-metal fit of parts exists, titanium-alloy joints can be vacuum diffusion bonded in the 1800 to 1900°F temperature range, without filler, to give metallographic homogeneity and strengths equal to base metal.

The procedures described in this article offer the basic advantages normally associated with a brazing process together with exceptionally high joint properties which are usually achieved only by welding.

21. Colton, R. M., Fitzpatrick, R., and Rizzitano, F. J., "Solid State Bonding of Prototype Titanium Rocket Motor Casings", U. S. Army Materials Research Agency, Watertown, Mass., ARMA MS64-04, March 1964.

Solid state bonding techniques were successfully used to fabricate large diameter thin wall titanium motor casings. The material used for the investigation was Ti-6Al-6V-2Sn titanium alloy forged rings having a diameter of 22 inches. The rings were joined by bonding using a butt joint. Axial pressure was applied using hydraulic pistons. Heat was applied externally using an oxyacetylene flame ring. The inside surfaces of the ring were protected by argon gas during bonding. An initial pressure of 3.000 psi was applied to the joint. The part was then heated to 1650°F and at the start of plastic flow (upsetting) the pressure was reduced to 4,000 psi. The entire bonding cycle required 2 minutes - 13 seconds.

Prior to bonding, the material had been solution heat treated at 1580°F for 1 hour and water quenched. After bonding, the parts were aged at 1100°F for 1 hour. Tensile specimens taken perpendicular to the bonded interface gave a joint efficiency of 90%. Metallographic examination showed the bonded area was free of embrittled zones and possessed a uniform wrought structure without metallurgical discontinuities.

22. Cooke, V. W. and Levy, A., "Solid Phase Bonding of Aluminum Alloys to Steel", Journal of Metals, Vol. 28(35), November 1949, pp 28-35.

Three procedures for solid-phase bonding of aluminum alloys to various steels have been investigated and found capable of producing high bond strengths. The twist and shear methods are characterized by low bonding temperatures and considerable deformation. The hot-press method employs slightly higher temperatures and no external lateral movement. Variables investigated were bonding temperature, pressure, surface preparation, deformation, composition and subsequent heat treatment.

Regardless of initial bonding technique, strengths of as bonded assemblies were found to be increased as much as 100 pct by subsequent annealing. Both 32S and B18S alloys bonded to 18-8 stainless steel frequently could be strengthened by solution heat treatment and aging, without lowering of bond strengths.

23. Cowan, G. R., and Holtzman, A. H., "Flow Configurations in Colliding Plates: Explosive Bonding", Journal of Applied Physics, Vol. 34(4), April 1963, pp 928-939.

The collision of plates at high velocity sometimes causes them to be metallurgically bonded. A study has been made of the nature of the bonds and of the conditions required to produce bonding. A necessary requirement is the formation of a jet in the space ahead of the region of impact. When the collision region moves along the plates at a velocity which exceeds the bulk sound velocity of both materials, the relative velocity of the plates must exceed a critical value for jetting. Clarification of these critical conditions in symmetric collisions, and a correct extension to asymmetric collisions have been obtained by consideration of the effect of downstream boundary conditions on the configuration of the shock waves attached to the collision line in the jetless case. When the velocity of the collision is subsonic, bonding is obtained when the elastic strength of the material is exceeded. Metallographic examination shows that three types of bond may be formed: (1) a direct bond, (2) a uniform layer of solidified melt. and (3) a wavy interface with discrete regions of solidified melt alternating with regions of direct bond. Consideration of the flow indicates the following: (1) the direct bond results from the removal of the surface layers as part of the jet, (2) the alloy results from the melting caused by entrapment of the high velocity jet between the plates, and (3) the wavy interface is caused by an oscillation in the jet flow which produces the mixed type of bonding. The thickness of the continuous alloy layer, which greatly exceeds the calculated jet thickness, indicates that entrapment of the jet dissipates most of the kinetic energy lost in the collision. Analysis of the layer indicates a composition and structure which is expected from the rapid quenching of a well-mixed melt.

24. Crane, C. H., Lovell, D. T. and Baginski, W. A., "Research Study for Development of Techniques for Joining of Dissimilar Metals", The Boeing Company, Seattle, Washington, for NASA, Huntsville. Alabama, Contract NAS8-11307

Quarterly Progress Report 1, October 15, 1964 Quarterly Progress Report 2. January 15, 1965 Quarterly Progress Report 3, April 15, 1965 Quarterly Progress Report 4, July 15, 1965 Literature Survey - "Joining of Dissimilar Metals", October, 1964

A research program was conducted to: (1) study and investigate potential methods for joining 2219 aluminum alloy to 321 stainless steel and (2) demonstrate the feasibility of adapting the most promising methods to production requirements, i.e., 20 to 50-inch cylindrical assemblies capability of cryogenic service.

State-of-the-art cognizance was established by a comprehensive literature survey and analysis of present technology. The three most promising methods - diffusion bonding, brazing and fusion welding - were selected for further study and development.

This evaluation proved diffusion bonding to be the best method for joining 2219 aluminum alloy to 321 stainless steel. The unique bonding method produced high strength, ductile joints and utilized diffusion aids as well as low processing temperatures and short times to effectively control formation of embrittling phases at the joint interface.

Successful diffusion bonding was accomplished at (1) temperatures of 500°F to 600°F, (2) pressures of 20 to 25 ksi and (3) times of 2 to 4 hours. The joint faying surfaces were electroplated with solder prior to bonding. The silver prevented the formation of oxide-film barriers and controlled formation of embrittling phases.

Processing techniques for diffusion bonding of large diameter assemblies were successfully developed and demonstrated by fabrication and testing of four 20-inch diameter joints. A unique diffusion bonding method was developed utilizing simple differential thermal expansion tooling. The method can be economically adapted to production requirements.

It was concluded that diffusion bonding is the most satisfactory method for joining large diameter stainless steel to aluminum alloy cylinders and is superior in strength and reliability to welding or brazing.

25. Davies, R. J. and Stephenson, N., "Diffusion Bonding of Nimonic 90 Nickel-Chromium-Cobalt Alloy", British Welding Journal, Vol. 9(3), March 1962, pp 139-148.

Techniques were sought for joining components of Nimonic 90 Ni-Cr-Co alloy for applications in which the strength of the joint is required to match, or at least approach, that of the parent material, but where marked deformation of the components is unacceptable, and conventional fusion-and resistance-welding processes are unsuitable. The procedures examined were (i) hot- or cold-pressing, using soft metal or alloy interlayers, followed by heat treatment causing diffusion in the solid state, and (ii) pressure brazing, also followed by diffusion treatment. The interlayer materials and brazing fillers used were types likely to alloy with the parent material during diffusion treatment.

Butt joints, having good stress-rupture strength at 1500°F were made by pressure brazing in a protective atmosphere with a Ni-Mn-Pd filler alloy. followed by diffusion and ageing treatments without pressure. Joints similarly produced by hot-pressing, with electrodeposited nickel or nickel foil interlayers, 0.001 in. thick, were strong at room temperature but developed relatively poor stress-rupture strength. However the joints made with the electrodeposited or foil interlayers were at least as strong in stress-rupture as those brazed without hot-pressing, using joint gaps 0.001 in. wide. Good shear and tensile properties were developed in joints made by pressing a tapered plug into a socket, with the surfaces separated by nickel electroplate or foil, afterwards subject to diffusion treatment.

26. DeCecco, N. A., and Parks J. M., "The Brazing of Titanium", Welding Journal, Vol. 32(11), November 1953, pp 1071-1081.

A review of titanium brazing is presented. Equilibrium diagrams for Ti-Ag, Ti-Cu and Ti-Al are considered as well as surface films and brazing fluxes. Brazing techniques include oxyacetylene, furnace and resistance methods. Recrystallization welding is also discussed.

Brazing studies were limited to commercially pure titanium and the recrystallization welding investigation was limited to Ti-7% Mn and CP titanium. Fine silver braze alloy produced ductile joints forming a TiAg compound by solution of titanium in molten silver. Copper braze alloy produced brittle joints forming TiCu₃ and Ti₂Cu₃ compounds. Pure aluminum was less satisfactory than silver forming TiAl₃ and TiAl the tormer possessing brittle properties. Surface films on titanium were studied by electron diffraction showing that TiO₂ (Rutile) was formed at elevated temperatures and unidentified films less than 100 Å formed at room temperature.

Metal depositing fluxes in the form of metal chloride mixtures were shown to produce adequate fluxing action for silver brazing. Oxyacety-lene brazing was evaluated for 1/8 inch sheet material, using 1/8 inch lap joint. Ultimate shear strengths were obtained by dividing shear load by actual sheared area of the fractured overlap surface. Furnace brazing was accomplished in inert atmosphere using metal chloride containing fluxes.

Recrystallization welding was accomplished by surface machining and then pressing the machined surfaces between heated dies to make lap-type specimens. Interface temperatures above 1000°F were required for both CP titanium and Ti-7% Mn alloy when times of two minutes were used.

27. Dowson, A. G., "A Note on the Forge Welding of Silver", Journal of the Institute of Metals. Vol. 71, 1945, pp 205-212.

Pure silver is used by the chemical industry for the fabrication of special chemical equipment. High quality fusion welds are difficult to achieve in silver. This investigation developed techniques for pressure welding silver fittings and tubing to silver storage vessels. The diffusion bonding was conducted at temperatures between 380° and 720°F. Prior to bonding, material having an as-rolled surface was cleaned with methanol. Specimens (lap joint) were prepared by placing the material between two dies while being heated in a muffle furance. After reaching the desired temperature, the dies were removed and placed in a hydraulic press. Pressure was applied to reduce the joint thickness by 50%. Shear testing showed that the 720°F temperature gave the best bonded joints.

28. Dulin, C. H., "Alcoa Pressure Bonding Process", Alcoa Green Letter, August 1960.

A pressure bonding process (U.S. Patent 2,908,073) is described for bonding of aluminum to stainless steel, carbon and alloy steels and copper. Various applications for this process are illustrated.

29. Eckel, J. F., "Diffusion Across Dissimilar Metal Joints", Welding Journal, April 1964 pp 170s-178s.

The review and study of element migration across a ferritic and austentic diffusion couple are discussed. The experience on dissimilar welded joints in power generating equipment has shown that the migration of carbon from the ferritic material to the austenitic material is a major problem.

Studies of diffusion bonded joints between the dissimilar alloys, AISI 304 (.06C) and SA 212 (.25C), have shown that C migrates from ferritic steels across the interface in the direction of the alloy (304) having the higher chromium content. This migration of interstitial carbon results in extensive precipitation of carbides. Accompanying the migration of interstitial C is a migration of the substitutional atoms Fe, Cr and Ni. Evidence was obtained indicating that Fe diffuses into an austenitic steel from a ferritic steel, that Ni diffuses into the ferritic steel and that Cr diffuses away from the interface in both directions. Interposition of a Ni barrier between a ferritic and austenitic stainless steel reduces but does not prevent C migration.

As the carbon content increased in the AISI 304 steel, the following carbides were formed in order: (Cr, Fe)₄ C (Cr, Fe)₇ C₃, (Cr Fe)₃ C. The effect of more rapid grain boundary diffusion was apparent in that martensite (caused by nickel migration in the SA 212 steel) extended beyond the volume diffusion front in the SA 212 steel and isolation of ferrite grains occurred. Diffusion studies were conducted in helium and vacuum atmospheres.

30. Elliott. R. P., "Diffusion in Titanium and Titanium Alloys", USAF Report ASD-TDR-62-561, October 1962.

The self-diffusion of titanium and the interdiffusion of aluminum, zirconium, molybdenum, vanadium, and oxygen in titanium have been investigated in the temperature range 600°-1300°C. Diffusion couples were prepared by roll-bonding or press-bonding techniques. Electron microprobe methods were used to determine the penetration of the substitutionally dissolved solutes; vacuum fusion analysis was used to determine the penetration of interstitially dissolved oxygen. The electron microprobe analysis could not be used to determine the penetration curves of aluminum in titanium because of the very high absorption of characteristic aluminum X-radiation by titanium.

The self-diffusion of titanium was investigated by studying penetration of Ti⁴⁴, formed by bombarding scandium with protons. Diffusion couples were formed by dissolving Ti⁴⁴-enriched TiO₂ into the titanium. The self-diffusion coefficients of alpha Ti and beta Ti and the interdiffusion coefficients of Zr, Mo. V and O in alpha Ti and beta Ti were determined and the data presented.

31. Evans, R. M., "Notes on the Diffusion Bodning of Metals", Defense Metals Information Center, Battelle Memorial Institute, Columbus, Ohio. DMIC Memo 53, April 1960.

Notes from a literature survey on diffusion bonding are presented. Most of the practical development work has been limited to the application of diffusion bonding to particular products such as nuclear reactor components. A definition is given for diffusion bonding and applicable diffusion bonding processes are reviewed. Various aspects of diffusion bonding are discussed with the subject divided on the basis of two joint systems: metal-to-metal with intermediate materials in the joint area and metal-to-metal without intermediate materials. It is pointed out that the metals joining industry is in need of basic research programs in this area.

32. Fairlie, J., "Explosive Welding and Forming Open Another Door to Industry". Welding Engineer April 1959, pp 61-64.

Possible applications of explosive forming and welding are presented. Preliminary welds have been made on materials such as: aluminum to aluminum, aluminum to Inconel, aluminum to stainless steel, stainless steel to itself and aluminum sandwiched between stainless steel.

33. Feduska, W., "The Nature of the Diffusion of Brazing Alloy Elements in Heat Resisting Alloys", Welding Journal, February 1961, pp 81s-89s.

A microstructural and microhardness study was made of the diffusion of single and binary combinations of elements, present in commercial high temperature brazing alloys, into heat-resisting alloys. Base metals used were AISI 410, AISI 347, Alloy 3, Alloy 4 and Alloy 5. Data obtained on 105 diffusion samples isolated the effects of individual diffusion elements during the interface reactions which take place between the brazing alloy and base metal.

34. Feinstein, L., "Diffusion Bonding", Stanford Research Institute, Menlo Park, Calif., Presented at the ASM Sponsored 1964 Golden Gate Metals Conference, San Francisco, Calif., Feb. 1964.

Diffusion bonding is defined and the features which distinguish diffusion bonding from brazing are described. Diffusion mechanisms are discussed and examples given of diffusion in bonding of similar and dissimilar metals. Since diffusivity is temperature dependent, the bond will be formed faster with higher temperatures. When a liquid phase is present at the interface, diffusion rates are greatly increased. With a well designed system it is possible to form at relatively low temperatures a bond which has a melting point and bond strength approaching those of the base metal.

35. Gaiennie, B., "Diffusion Bonding: The State of the Art", Machinery, March 1965, pp 139-143.

The general aspects of diffusion bonding are presented. The state-of-the-art is fairly advanced as indicated by prototype production in several areas. General research is directed toward optimizing temperature, pressure and time combinations, interface preparation, joint design, interface catalyst and development of bonding techniques and equipment. It was found that maximum diffusion of the interfaces could be obtained at approximately one-half the melting temperature of the metal involved. It was also determined that pressure and time elements used, can vary inversely through a wide range without affecting bonding results.

36. Gatsek, L. F., "Bonding and Welding of Dissimilar Metals", Light Metal Age, April 1965, pp 10-13.

A summary of dissimilar metal joining techniques is presented with emphasis placed on aluminum to stainless steel joining. The processes discussed include brazing diffusion-bonding, resistance-welding, soldering, percussion stud-welding, ultrasonic welding and roll bending. Brazing and diffusion bonding are the two processes of practical application to large diameter tube joining. Brazing of 6061 aluminum to 304L stainless steel is accomplished by hot-tinning the steel and dip brazing using Alcoa 718 alloy filler. The tin on the 304L alloy serves as a barrier material even though it is in a molten state at the brazing temperature. An interface material is applied to both aluminum and stainless steel when diffusion bonding is used. Silver and copper have been used as interface material for diffusion bonding aluminum to aluminum in an argon-gas atmosphere.

37. Hess, W. F., and Nippes, E. G., "A Method for Welding Sheet Aluminum to SAE 4140 Steel", Welding Journal, March 1946, pp 129s-148s.

The joining of aluminum to steel by resistance welding was investigated. The application was for joining aluminum cooling fins to SAE 4140 steel aircraft engine cylinders. The authors determined that the resistance welding had to be accomplished without melting of the steel because of the formation of brittle iron-aluminide phases at the interface. It was also

necessary to create a weld (or bond) without heating the steel above its austenizing temperature to prevent a martensitic structure from forming during cooling of the weld (or bond).

An interface metal was required to obtain a satisfactory weld. Tin, zinc, chromium, cadmium, copper and silver were electroplated to the steel. Of these metals, silver was the most satisfactory providing high-strength, ductile welds over a wide range of welding current and plating thickness.

38. Hirano, K., Agarwala, R. P., and Cohen, M., "Diffusion of Iron, Nickel, and Cobalt in Aluminum", Acta Metallurgia, Vol. 10, Sept. 1962, pp 857-863.

Interdiffusion coefficients of iron, nickel and cobalt in aluminum have been determined over a temperature range of 660° to 1170°F, using radio-active tracers and the residual-activity technique. The experimentally determined frequency factors and activation energies are quite small compared to those for zinc diffusion in aluminum and for self-diffusion in aluminum. This contrast in behavior is explained on the basis of the extremely low solid solubilities of iron, nickel and cobalt in aluminum, which reduce the flux through the lattice to the point where diffusion along short-circuiting paths becomes controlling. The appropriate solution of the diffusion equation and the quantitative results obtained suggest that normal concentrations of dislocations can provide the necessary short-circuiting paths.

39. Hollander, M. B., "Welding Metals by Friction", Materials in Design Engineering, February, 1962, pp 79-81.

A summary of friction welding is given. This process involves rotating one work piece against the other, developing frictional heat at the interface. When interface softening occurs, rotation is stopped and the pieces are upset forged together. The basic process variables are axial pressure. surface sliding speed and time. Materials joined by this method include the various steel alloys and selected light metals. Primary application has been joining oil well drill pipe.

40. Hughson, R. V.. "New Process for Ti-Clad Steel Plate", Chemical Engineering, Vol. 68(10), May 15, 1961. pp 194-198.

A process developed by Lukens Steel Company for producing titanium clad steel plate is described. An enclosed sandwich of titanium sheet and A-204 low carbon manganese steel plate is flushed continuously with argon while being heated before rolling, to eliminate impurities that interfere with the bond. Two clad plates with the titanium back to back are roll bonded in one sandwich. Preheat temperature limit before rolling is 1700° to 1750°F. A reduction of at least three to one is required for good bonding. No intermediate material is used. Method of forming, cutting and welding of Ti-clad steel are discussed.

41. Kinzel, A. B., "Adams Lecture - Solid Phase Welding", Welding Journal, Vol. 23(12), December 1944, pp 1124-1144.

A comprehensive review of solid-phase welding is presented. This process is broadly defined as joining without the formation of a liquid phase during the process. The best criteria for evaluation of a solid-phase weld is elimination of the original interface, this occurs by crystal growth and diffusion. Phase transformation was shown to have an important effect on the weld. Temperature plays a major and even dominant role in solid-state welding. For ferritic steels the temperature must be above the critical temperature for trans-interface crystallization in order to provide a satisfactory bond. Theoretical consideration of diffusion and related variables is presented for application to solid-state welding.

42. Kuper, A. B., Lazarus, D., Manning, J. R., and Tomizuka, C. T., "Diffusion in Ordered and Disordered Copper-Zinc", Physical Review, Vol. 104(6), December 1956, pp 1536-1541.

The diffusivities of ${\rm Cu}^{64}$, ${\rm Zu}^{65}$, and ${\rm Sb}^{124}$ in single crystals of 47-48 atomic percent zinc copper-zinc (beta brass) have been measured over the temperature range 510°-1500°F, by using sectioning techniques. The diffusion coefficients show a striking dependence on the degree of long-range order at temperatures below the critical temperature 874°F. A slight dependence of the diffusion coefficient on short-range order is noted above the critical temperature. The diffusion coefficients obey an Arrhenius equation only in the fully disordered phase, with temperature dependences given by $D_{Cu} = 0.011 \exp(-22000/RT) \text{ cm}^2/\text{sec}$, $D_{Zn} = 0.0035 \exp(-18,800/RT) \text{ cm}^2/\text{sec}$; $D_{8b} = 0.08 \exp(-23,500/RT) \text{ cm}^2/\text{sec}$. The variation of the diffusion coefficients with temperature in the ordered phase is considered in terms of a simple elastic model. Excellent agreement is obtained by using the measured elastic constants and assuming that the energy for motion of the imperfection is simply related to the smallest (110) shear modulus. In the disordered phase Sb diffuses faster than Zn or Cu, while in the ordered phase Sb diffuses at the same rate as Zn, which is faster than Cu. This result is shown to be inconsistent with an interchange, interstitial, or nearest-neighbor vacancy mechanism for diffusion. The result is consistent with an interstitialcy mechanism.

Layers Formed on Aluminized Steel by Means of the Electron Probe Micro-analyser" Journal of the Institute of Metals, Vol. 92, 1963-64, pp 150-152.

An investigation was conducted of the nature of aluminum coating on aluminized steel containing a double intermetallic layer. The layers were identified by compositional means to be Fe₂Al₅ adjacent to the steel and Fe Al₃ adjacent to the aluminum.

44. LeClaire, A. D. and Bear, I. J., "The Interdiffusion of Uranium and Aluminum", J. Nuclear Energy, Vol. 2, 1956, pp 229-242.

Results are given of measurements of the width as a function of temperature and pressure, of the diffusion zone formed by the inter-diffusion of uranium and aluminum and of the relative penetration of uranium into aluminum and of aluminum into uranium.

The experiments are unusual among diffusion measurements in showing an appreciable accelerating effect of pressure on the rate of inter-diffusion. This is not marked for temperatures below 1000° F. The penetration of the uranium into the aluminum is about 2-1/4 times faster than that of the aluminum into the uranium.

The results of a microscopical and X-ray examination of the diffusion zone are presented, and a discussion is given of the possible origins of the pressure effect.

45. Loenstein, P. and Tuffin, W. B., "Metallurgical Bonding of Dissimilar Metals by Co-Extrusion", Nuclear Metals, West Concord, Mass.

The co-extrusion process lends itself to the production of high quality metallurgical bonded shapes of several dissimilar metal combinations. Mechanical aspects and metallurgical aspects of bonding are described. The importance of bond layer thickness, when brittle compounds form, is discussed. Specific illustrations of applications of the process to rods, shapes, tubes and tubular transition joints, are given. Joining of refractory and reactive metals to ferrous alloys or alloys containing nickel are particularly cited.

46. McEwan, K. J. B. and Miller, D. R., "Pressure Welding of Dissimilar Metals", British Welding Journal, Vol. 9(7), July 1962, pp 406-420.

The roll bending of dissimilar metals has been studied and related to previous experience with autogenous pressure welding. The metal pairs studied were divided into the four groups: Immiscible; partially miscible and intermetallic forming.

The immiscible pairs studied were Cd-Fe, Fe-Pb, Cu-Pb. and Cu-Mo; partially miscible were Cu-Fe, Cu-Ag, and Al-Zn; miscible were Cu-Ni, Fe-Ni and Mg-Cd; intermetallic forming were Cu-Al, Fe-Al, Ca-Pb, Mg-Al, Ag-Al and Fe-Mo.

If the temperature of welding was sufficiently low, so that no appreciable interdiffusion occurred, the only significant difference from autogenous welding was that relative movement occurred at the weld interface, owing to the different deformation characteristics of the two metals, and this enhanced bonding.

On welding at high temperatures, or on post-heat treatment, the properties of the joint were determined by the extent and effect of inter-diffusion. Immiscible and slightly miscible systems gave stable welds.

Some miscible metals exhibited diffusional porosity resulting in serious weakening of the bond. With intermetallic forming systems two types of behavior were observed. In the majority of cases, associated with a brittle intermetallic layer, the bond was strong if the layer was maintained below a critical thickness, but weak if it grew above this thickness. The critical thickness at which the transition occurred varied with the system under consideration, probably being dependent upon the existence of a very small amount of ductility in the intermetallic layer. If the interface layer was ductile the joint was strong and failure occurred in the weaker metal.

47. McGowan. P. R., Williams. N. R., and Allyn. J. I., "Diffusion Bonded Titanium Alloy Honeycomb Panels", Interim Report IR-8-212, Douglas Aircraft Company, Long Beach. Calif., October 1964.

A summary of development of diffusion bonded titanium alloy honeycomb is presented. The alloys used were titanium 8Al-1Mo-1V and commercially pure Ti-75A. Bonding parameters of 1750°F for four hours were selected on the basis of satisfactory bonds in four inch square panels. Panel assemblies are enclosed in a vacuum tight stainless steel envelope (10⁻⁵ to 10⁻⁶ torr). To prevent crushing of the core due to this vacuum, the fixture or outer envelope is evacuated to 29.5 inches Hg. Glassrock insulation, strip heaters, refrasil, internally cooled copper platens and slip sheets are placed on both sides of the honeycomb envelope. Pressure is applied by a vacuum pressure bag between the tooling. Low pressure (creep controlled) bonding was used due to crushing of the core at high pressure.

48. Metealfe, A. G., "Modern Diffusion Bonding Meets Space-Age Needs", SAE Journal, April 1964, pp 53-57.

Describes developments in diffusion bonding processes for joining Ta, W, TD nickel. superalloys, T-111 Ta alloy and Mo and Cb and their alloys for aerospace applications. Comparison of different diffusion bonding processes: yield stress controlled bonding (or pressure welding), diffusion controlled bonding and creep controlled bonding. Diffusion bonding is a method of low temperature bonding that is particularly applicable to dissimilar metals and metals that can be damaged or embrittled by high temperatures.

49. Miller, M. A. and Oyler, G. W., "Pressure Welding Aluminum at Various Temperatures", Welding Journal, Vol. 30, October 1951, pp 486s-498s.

Pressure welding is defined as the formation of a solid-phase weld between metallic materials, by the application of a deforming pressure, at any temperature below the melting point of the components. The three major factors in the pressure welding of aluminum, assuming optimum surface preparation and suitable die design, are temperature, pressure and time. These factors are closely interdependent and capable of graphical representation.

At temperatures below about 500°F, the extent of deformation during welding determines weld efficiency. At elevated temperatures, diffusion is

also an important factor in the establishment of metallurgical bonds. Required unit pressures decrease rapidly as temperatures increase above 600°F, and approach the solidus temperature of the alloy being welded.

The advantages and disadvantages of using low temperatures and high unit pressures, as contrasted to the use of elevated temperatures and low unit pressures, have been discussed.

50. Miller, M. A., "Joining Aluminum to Other Metals", Welding Journal, Vol. 32, August 1953, pp 730-740.

Methods of joining aluminum to other metals by fusion welding, pressure welding brazing, soldering, diffusion welding and resin bonding are discussed. Of the welding processes, are and flash welding are of more general usefulness than are torch or resistance welding. Brazing methods, while of limited usefulness in the joining of aluminum to copper and copper-base alloys, except by the use of transition joints, are of considerable usefulness for joining aluminum to ferrous alloys, particularly if the ferrous parts are given a suitable surface preparation, the best of which appears to be an aluminum coating.

Non-fusion methods methods such as diffusion welding and pressure welding are discussed for joining aluminum to other metals. Either pressing or rolling may be used to join metals by these methods. Silver-plated aluminum may be bonded to pure silver, certain silver alloys and to any metal that can be silver plated, by heating the parts together at 500-600°F for a period of time under pressure; silver readily diffuses across the interface and results in a good joint.

Pressure welding, either at room or elevated temperature, under conditions to give proper deformation, is becoming increasingly important for joining aluminum to metals such as copper and steel.

51. Moore, D. C., "Titanium Clad and Titanium Lined Process Equipment", Welding and Metal Fabrication, July 1963, pp 276-282.

Methods of lining processing equipment with titanium are presented. This can be done by lining a steel vessel with titanium or fabricating the vessel from titanium clad steel. The bimetal plate can be produced by roll bonding, brazing or explosive cladding. The rolling technique does not employ intermediate metals and is accomplished at temperatures below brittle compound formation. A copper/silver alloy of eutectic composition is used in brazing titanium to steel. The explosive method has not been used for production applications and apparently does not employ intermediate metals. Vanadium and silver are mentioned as interlayer metals to provide a suitable metallurgical transition when joining clad material by welding.

52. Niemann, J. T.. Sopher, R. R., and Rieppel, P. J., "Diffusion Bonding Below 1000°F", Welding Journal, Vol. 37, August 1958, pp 337s-342s.

Techniques and metal systems investigated to obtain diffusion bonded joints between beryllium copper and Monel. Results showed that bonded can be accomplished at 650°F or perhaps lower. Good joints were obtained with the following intermediate metal combinations: Au-Cu, Au-Ag. Au-Pb and Au-Al. Superior bonds strengths (13,000 psi shear) were exhibited by Au-Cu and Au-Ag combinations while the Au-Pb and Au-Al were found to be quite brittle. The diffusion bonding technique permits the joining of the Be-Cu alloy cover sheet and the Monel block of a hypersonic wind-tunnel throat at the age-hardening temperature of the Be-Cu alloy.

Orysh, M. S., Betz, I. G., and Hussey, F. W., "Joining of Steel and Other Wear Resistant Metals to Aluminum Alloys" Frankford Arsenal Technical Memorandum M64-8-1, August 1963.

Joining of steel and other wear-resistant metals to aluminum by arc welding and diffusion bonding were studied. The steel members used had a coating of either silver brazing alloy, zinc, or aluminum. Samples of commercial products consisting of aluminum brazed to steel by proprietary processes and aluminum bonded to steel in sheet, plate, and tube form were included in the study. Plates of 2024 aluminum alloy overlaid with two metallized coatings and three experimental hard-surfacing aluminum alloys were also evaluated. It was found that 2024 aluminum alloy can be welded to steel which has been coated with either zinc, aluminum, or a silver alloy, provided the welding arc is directed toward the aluminum member of the joint. The diffusion-bonding process can be used to join 2024 aluminum alloy to silver-alloy-coated steel. The surface hardness of aluminum alloy plates can be increased through the application of an experimental hard-surfacing aluminum alloy or a metallized coating.

54. Paprocki, S. J., Hodge, E. S., and Gripshover, P. J., "Gas Pressure Bonding", Defense Metals Information Center, Battelle Memorial Institute, Columbus, Ohio, DMIC Report 159, September 1961.

A critical review of the literature is presented on the gas-pressure bonding process. This solid-state bonding technique has been successfully utilized for joining, cladding and densifying many material systems.

The gas-pressure bonding process employs a gas at high pressure and elevated temperature in order to join and fabricate metallic or ceramic materials. Temperatures are held well below those required in sintering operations and therefore resulting grain growth is held to a minimum.

Examples are cited of typical nuclear fuel elements and various structural components of interest for aircraft and missile application. Bonding parameters including temperatures, pressures and times are discussed.

55. Parks, J. M., "Recrystallization Welding", Welding Journal, Vol.32, May 1953, pp 209s-222s.

Recrystallization is established as the mechanism of solid phase welding and the author concludes that the pressure welding process should more rightly be known as recrystallization welding. The principal factors controlling recrystallization are discussed and related to welding. Shear strengths of recrystallization welds are discussed. A basic relationship between recrystallization temperature and degree of deformation is derived and analyzed. The importance of clean cold-worked surfaces for recrystallization welding is emphasized.

56. Pearson, J. and Hayes, G. A., "Research in Explosive Welding", ASTME Technical Paper SP6397, 1962-63.

A review of explosive welding is presented. Studies have been mainly devoted to understanding the mechanisms of the process. A few commercial applications have been investigated, these include: cladding of large flat plates, lining of long tubes and rocket nozzles and joining of dissimilar metal billets. A "surface jetting" phenomenon may occur at the joint resulting in an irregular or saw tooth surface. Buffer plates are frequently used to protect the weld plate surface and transmit uniform pressure. Extensive plastic flow is generally rerequired to accomplish a satisfactory explosive weld. Nineteen various similar and dissimilar metal combinations have been joined explosively on an experimental basis. These include representative aluminum, steel, super and refractory alloys.

57. Sawatzky, A. and Jaumont, F. E., "Diffusion of the Elements of the IB and IIB Subgroups in Silver" Transactions of AIME, 1957; Journal of Metals, October 1957, pp 1207-1210.

Data are given for the diffusion of the elements of the IB and IIB subgroups in single crystals of silver. The sectioning technique and high specific activity isotopes were used to determine the diffusion coefficients of Cu and Hg in Ag in the present investigation and of Zn, Cd, Au and Ag in Ag in previous investigations.

With the exception of those for mercury in silver, the data indicate that the activation energies for diffusion of the atoms of a given subgroup in the same solvent are similar, but the frequency factors differ. From this, it is concluded that atomic size, provided solid solutions are formed, probably does not affect the activation energy. The relation of the present results to several of the more popular generalizations concerning diffusion is discussed.

58. Schwartzbart, H., "What is Diffusion Bonding", Welding Engineer, November 1962, pp 46-47.

A general description and definition of diffusion bonding is given. It is described as a group of welding process with appropriate temperature and joint deformation limitations. The characteristic feature of diffusion bonded joints is that the sharply outlined filler metal layer evident in conventional brazed joints is essentially obliterated. Reference is made to diffusion bonding Zircaloy plate-type fuel elements with intermediate layers of copper, iron or nickel.

59. Storchheim, S., "Hot Pressure Bonding", Metal Progress, Vol. 72, July 1957, pp 97-101.

Investigation was conducted on the diffusion bonding, by the press bonding process, of several metallic couples without the benefit of intermediate metals. Emphasis was placed on diffusion systems having aluminum as one-half of the couple. Studies were conducted using Al-Ni, Al-Cu, Al-Fe and Al-Zr couples. The variables of time, temperature and bonding pressure were studied with regard to joint strength and metallographic structure. The effect of time, temperature and pressure on joint strength and intermetallic alloy zone formation are discussed.

60. Stroup, P. T., and Purdy, G. A., "Aluminum Coating of Steel - A Comparison of Various Processes", Metals Progress, Vol. 57, January 1950, pp 59-63, 128-130.

Steel can be coated with aluminum by hot-dipping, electroplating, spraying, chemical reaction and calcrizing. The authors discuss the coatings produced by each of these methods. Particular emphases is given to the importance of controlling the thickness of iron-aluminum compound that forms between pure aluminum surface layer and steel base. The six methods of coating are compared on the basis of structure and cost.

61. Tylecote, R. F., "Investigations on Pressure Welding", British Welding Journal, Vol. 1(3), March 1954, pp 117-138.

The object of this work was to establish certain of the principles involved in the process of pressure or solid-phase welding. It is particularly concerned with the connection between the weldability of various metals at room temperature and the physical properties of the oxide film. The effect of post heat-treatment of welds has been investigated and the results are interpreted in the light of oxide-film solubility and the formation of intermetallic compounds.

Work has also been done on the welding of deoxidized copper in hydrogen to prevent the formation of an oxide film. The results show that the nature of the film is undoubtedly responsible for the poor weldability of copper at room temperature.

The effect of orientation on the weldability of single crystals of aluminum has been determined. The particular crystallographic plane brought into contact has a marked effect on the degree of transinterfacial grain growth upon recrystallization.

62. Tylecote, R. F., "Pressure Welding in Practice", British Welding Journal, Vol. 4(3), March 1957, pp 113-120.

The practical applications of pressure welding during the ten years from 1946 to 1956 are summarized. The main applications lie in two fields: (1) those relating to the butt welding of tubes and sections in steel and (2) the joining of overlapping sheet material in copper and aluminum alloys by pressing or rolling.

The techniques used are discussed, and an attempt is made to assess the advantages of the process.

63. Vicars, E. E. "An Efficient Joining Technique: Diffusion Bonding", Metal Progress, April 1965, pp 125-130.

Methods of diffusion bonding honeycomb panels are discussed. The general advantages over alternative methods are presented as well as the general processing used. Stop-weld materials such as: ${\rm TiO_2}$, MgO and ${\rm ZrO_2}$ are used to prevent bonding during honeycomb joining. The use of intermediate metal at the joint is preferred as removal of stop-weld materials is eliminated. Mention is made of accelerating diffusion by cold working which increases the number of vacancies and the use of high frequency current which increases mobility of surface atoms.

64. Wood, C. L., "A Technical Report on Duranel - Stainless-Clad Aluminum", Aluminum Company of America, January 1963

A new stainless steel clad aluminum sheet product marketed by Alcoa under the tradename "Duranel" is described. Properties and applications for the laminated sheet are discussed. Methods for forming, joining and finishing of the clad sheet are described in detail.

65. Young, J. G. and Smith, A. A., "Joining Dissimilar Metals", Welding and Metal Fabrication, July 1959, pp 275-281.

Methods of joining dissimilar metals are reviewed. Principles and applications of fusion welding, brazing, resistance welding, pressure welding and solid state diffusion bonding are discussed. The metallurgical principles in joining dissimilar metals are discussed and consideration is also given to such factors as cracking and dilution and uniformity of joint composition.

66. Young, W. R., and Jones, E. S., "Joining of Refractory Metals by Brazing and Diffusion Bonding", Air Force Technical Document Report No. ASD-TDR-63-88, January, 1963.

A discussion of joining several alloy combinations by diffusion bonding in vacuum, using high temperature and pressures is presented. Alloy combinations that were successfully bonded and produced ductile joints were Cb-1Zr alloy to Mo-0.5Ti molybdenum alloy, copper to Cb-1Zr alloy, copper to 316 stainless steel and Mo-0.5Ti molybdenum alloy to Rene'41.

67. "Astroweld--Diffusion Bonded Honeycomb", Hexcel Products, Inc., Berkeley, Calif., R&D Technical Report 101, May 1963.

The "Astroweld" process of diffusion bonding honeycomb sandwich is presented. Bonding of core materials is accomplished with or without an intermediate material. Successful solid state bonding has been accomplished with the following materials: steel, copper alloys, stainless steel, super-alloys and refractory alloys. Bonding of the skin members to core material requires application of a high purity intermediate metal on the honeycomb cell edges. These intermediate metals are placed on the honeycomb nodes in micron thicknesses. At bonding temperatures this material tends to yield and absorbs initial surface irregularities.

68. "Diffusion Bonding: 'Cool' Newcomer Makes Joints Disappear", Steel, Vol. 150(14), April 2, 1962, pp 117-120.

A brief summary of diffusion bonding experience within the Aero-space industry is given. Specific applications include: Type 321 stainless honeycomb, Rene'41, titanium alloys, tungsten, beryllium-copper and beryllium. In many instances intermediate layers are used to promote bonding.

69. "Joining Steel to Aluminum", Solar Aircraft Co., San Diego, Calif. A Division of International Harvester Company, RDR 12584, February 1965.

The Solar Bimetal Joining Process is described. Any aluminum alloy with a solidus temperature of 1100°F or nigher may be joined to most stainless steel and superalloys. The essential elements of the process include conditioning the stainless steel by a patented process and brazing it to the aluminum alloy. Typical assemblies produced are stainless steel bellows joined to aluminum rings. Tubular test specimens have been subjected to thermal cycling (160°F to -320°F), helium pressure leak test, pressure surge test at -320°F, Gimbal cyclic test, vibration test and helium leak test without evidence of failure. Corrosion resistance of bimetal joints is similar to brazed aluminum joints. Other dissimilar metal joints which have been joined by brazing are: 6061 aluminum to AMS 6439 steel, beryllium to stainless steel and titanium, titanium to alloy and stainless steel and magnesium to steel.

70. "Roll Bonded Sandwich Structures", Douglas Aircraft Co. and Battelle Memorial Institute, November 1963.

A new technique for producing titanium alloy sandwich structures by roll bonding is described. Titanium sandwich with the voids filled with a dissimilar metal is completely encased in a dissimilar metal pack. The face sheets are diffusion bonded to the core by hot rolling of the metal pack. After forming of the sandwich to the desired shape, the dissimilar metal inserts are removed by chemical means. This method of making sandwich structure is believed to be less costly than welding and the joints have strength equivalent to that of the base metal. Data on properties of typical roll-bonded sandwich structure of Ti-6Al-4V alloy are presented.

71. U. S. Patent 2,908,073, "Method of Bonding Aluminum Metal to Dissimilar Metal", C. H. Dulin, Assignor to the Aluminum Company of America, October 13, 1959.

Methods are described for pressure bonding of aluminum alloys to steel and copper alloys. The methods outlined are adaptable for joining flat plates of aluminum to steel for use in manufacture of electric flat irons and cooking utensils.