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BONDING—III

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to June 30, 1958

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ABSTRACT

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 UAl_3 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 UAl_3 diffusion zone.

I. INTRODUCTION

The study of diffusional bonding is of considerable practical importance to metallurgists working in the field of reactor fuel element technology, since most fuel element designs call for a corrosion-resistant cladding metallurgically bonded to the fuel-bearing core. The formation and growth of intermetallic phase layers at the interface of many fuel element core-cladding combinations have been investigated extensively in a qualitative fashion. However, relatively little has been done to investigate the fundamental aspects of the phenomenon. Consequently, the present program was begun in December 1954 to determine the mechanisms and the kinetics of intermetallic layer growth. As a start, the aluminum-nickel alloy system was chosen for examination, since previous investigators had observed that layer growth in this system could be influenced by alterations in the magnitude of the applied pressure⁽¹⁾. Therefore, the kinetics of growth were explored as a function of pressure in aluminum vs. nickel diffusion couples and in incremental diffusion couples consisting of aluminum vs. γ phase (Ni_2Al_3) and of nickel vs. β phase (NiAl_3)⁽²⁾. Concurrently, a study was made of the diffusional theory pertaining to multi-phase, two-component systems⁽³⁾, which could be used to predict quantitatively the kinetics of growth of intermetallic layers, provided that the following conditions were met: (1) Reactions at the phase interfaces were not so slow as to be rate controlling; (2) The values of the concentrations at the phase interfaces were known and remained constant; and (3) The values of the diffusion coefficients in each phase were known. Relatively little work had been done to verify this theory experimentally. On the basis of the results obtained from the investigation of the aluminum-nickel system^(2,4,5) it was concluded that most of the observations could be rationalized by the assumption that volume diffusion was the rate-controlling factor. Pressure, therefore, exerted its inhibiting effect on the growth kinetics by reducing volume diffusion, most probably by reducing diffusion coefficients, but perhaps also by changing interface concentrations. Since the phase equilibria in the aluminum-nickel system are quite complicated, it was not practicable to attempt calculation on a theoretical basis of the kinetics of layer growth so as to provide experimental verification of the theory. Consequently, investigations were started to check experimentally the validity of the theoretical solutions in two- and three-phase systems such as antimony-tin, antimony-aluminum and silver-copper. The phase equilibria in the latter systems are much simpler than those in the aluminum-nickel system, where four intermetallic and two terminal phases exist.

While the investigation of the aluminum-nickel system was under way, LeClaire and Bear⁽⁶⁾ published the results of their investigation into the inter-diffusion of uranium and aluminum, which complicated considerably the task of explaining the general role played by applied pressure. Contrary to the observations made in the aluminum-nickel system, it was shown that pressure accelerates growth of intermetallic layers in the aluminum-uranium system. It was concluded by the investigators that the effect of pressure in the aluminum-uranium system cannot be attributed to reductions in the magnitude

of the diffusion coefficients, as appeared to be probable in the aluminum-nickel system. It was suggested that pressure could accelerate layer growth by one of the following means: (1) producing changes in the interface concentrations; (2) producing allotropic changes in existing phases; or (3) causing the nucleation of new phases which would be stable under the pressure conditions employed. In view of the interesting results found by LeClaire and Bear, it was decided to explore further the reported phenomena in the aluminum-uranium system, in an attempt to reconcile the apparently inconsistent behavior of pressure in the two systems and thus to obtain a better fundamental understanding of the pressure effect.

During the present reporting period, the major effort has been directed towards investigation of the aluminum-uranium system, including the effect of pressure on layer growth and the nature and distribution of structural defects found in the layers. In addition, as has been mentioned, work is under way to check the experimentally determined layer growth data in suitable systems against calculated growth rates obtained from solutions of the theoretical equations. Also, in view of the importance of knowing precisely the values of interface concentrations, the attempt is continuing to perfect an X-ray absorption device for determining composition as a function of position in diffusion couples. The progress made to date is reviewed in the succeeding sections of this report.

II. EXPERIMENTAL PROCEDURE

A. Preparation of Aluminum-Uranium Diffusion Couples

The aluminum-uranium diffusion couples were prepared from high-purity (99.99%) aluminum and reactor-grade uranium rods. Half-inch diameter discs 1/4" thick were machined and given grain-coarsening anneals -- the aluminum being heated to 600°C for 24 hours in air and the uranium to 550°C for 24 hours in evacuated Vycor capsules. The faces of the discs were electropolished in a Disa electropolishing unit (A-2 solution), and the samples were rinsed in water and in acetone and dried. The thicknesses of the discs were then measured with a travelling microscope. Subsequently, diffusion couples were assembled by placing an aluminum disc onto an uranium disc with 0.0005" diameter tungsten wires and 0.001" thick molybdenum strips inserted between the mating surfaces as markers.

All of the diffusion bonding runs were made under dead weight loading conditions in the evacuated hot pressing apparatus described in a previous progress report⁽⁴⁾. After the completion of the diffusion anneals, the specimens were sectioned, mechanically polished (in some cases, samples were etched with 5% ammonium persulfate solution containing a little hydrogen peroxide) and the intermediate phase and remaining uranium and aluminum layer thicknesses determined. Six to

twelve measurements were made of each layer thickness with a travelling microscope, and an average thickness and standard deviation calculated.

B. Preparation of Antimony-Tin, Antimony-Aluminum and Silver-Copper Diffusion Couples

Antimony-tin, antimony-aluminum and silver-copper diffusion couples were prepared as indicated in the previous progress report⁽⁴⁾. The antimony-aluminum and the silver-copper couples were diffusion annealed in the hot pressing apparatus. The antimony-tin diffusion anneals were made in an oil bath in which the temperature was controlled to within 0.5°C by means of a thermostatic device.

C. Determination of Concentration-Penetration Curves

The use of an X-ray absorption device to determine concentration-penetration data in multi-phase diffusion couples has been discussed in the previous progress report⁽⁴⁾. In brief, the relative intensities of the X-ray beams transmitted through an alloy can be converted to compositional data by a knowledge of the absorption coefficients of the alloy components. During the present reporting period, the X-ray absorption apparatus was completed and several aluminum-nickel diffusion couples available from previous work were prepared for analysis by being ground into longitudinal sections 0.005" thick. Subsequently, they were irradiated with CuK_α and MoK_α X-rays, and concentration-penetration curves calculated.

III. RESULTS

A. Aluminum-Uranium System

1. Phases Appearing During Interdiffusion:

The aluminum-uranium phase diagram is reproduced in Fig. 1⁽⁷⁾. There are three intermetallic phases, as follows: δ (UAl_2), ϵ (UAl_3), and ζ (UAl_4) -- all with highly restricted solubility ranges. LeClair and Bear⁽⁶⁾ reported that two phases were metallographically visible in the diffusion couples. The UAl_3 phase predominated and grew very rapidly; also, a relatively thin UAl_2 layer formed between the uranium and the UAl_3 . Other investigators^(8,9,10,11) have also noticed the presence of the UAl_3 phase. DeLuca and Sumsion⁽¹¹⁾ found the presence of traces of both UAl_2 and UAl_4 ; the UAl_4 could not be seen metallographically, but was found in the form of a black powder located on the surfaces of samples parted at the aluminum- UAl_3 interface. In the present investigation, similar results have been obtained. Two phases are metallographically observable (Figs. 2(a) and 2(b)), the thick phase being UAl_3 and the thin phase UAl_2 . In addition, a two-phase region (Figs. 3(a) and 3(b)) is found to exist in some of the diffusion

couples, in which finger-like growths presumably of UAl_2 co-exist with UAl_3 . The nature of this phenomenon is not yet understood.

2. Physical Appearance of Diffusion Layers:

It was observed by previous investigators(6, 8-11) that the diffusion zone in aluminum-uranium couples is brittle, porous, and cracks easily. In the present investigation, similar behavior has likewise been noticed. In addition, a series of interesting correlations exist between the applied pressure, the diffusion temperature, the diffusion time, and the appearance of the defect structure in the diffusion zone. In Fig. 4, the effect of diffusion temperature is shown on diffusion couples annealed under an applied pressure of 5 tsi for times sufficient to produce diffusion zones 4-5 mm thick. In the 390°C specimen, the diffusion zone is cracked in several directions and a relatively fine porosity is uniformly distributed throughout the zone. In the 520°C case, several large cracks are visible which run roughly parallel to the interfaces. Large voids exist in parallel layers and occupy about 1/4 of the specimen surface. The 570°C specimen has a much sounder appearance, and the voids are fewer in number and are located close to the Al- UAl_3 interface. The effect of increasing diffusion time in specimens diffused at a given temperature and under a given applied pressure is shown in Fig. 5. It appears as though a porous band of constant thickness (about 1.4 mm thick) forms and persists in the vicinity of the Al- UAl_3 interface at all diffusion times; the remainder of the sample is quite brittle and cracked, but is relatively free of porosity. Finally, in Fig. 6, a series of photomicrographs is shown of samples annealed at different pressures, diffusion time and temperature being held constant. It is evident that the diffusion zone becomes relatively more sound as the pressure is increased. The extent of void formation and the amount of cracking is much less in the sample annealed under an applied pressure of 10 tsi than in the 1-1/4 tsi sample.

3. Kinetics of Layer Growth:

Layer growth data for the aluminum-uranium system are listed in Table I. The thicknesses include the contributions of the UAl_3 phase and the UAl_2 phase, but since the latter thickness is usually negligible in comparison with the UAl_3 layer, the data essentially reflect the growth of the UAl_3 phase. It is clear that the UAl_3 phase grows with great rapidity, attaining thicknesses of more than 5 mm after 24 hours at 560°C. The thickness data are plotted on logarithmic scales in Fig. 7 (log thickness vs. log time). In the temperature range for which data are presently available (520° to 570°C), the growth kinetics appear to be conventional at a given pressure after diffusion times of 3 to 8 hours are reached. A parabolic growth law is obeyed, and volume diffusion appears to be the rate controlling factor. Also, increases in applied pressure are seen to increase layer thicknesses -- corroborating the observation reported by

LeClaire and Bear, and others (6,9). At the earlier diffusion times, however, the situation becomes confused. The parabolic growth law is not obeyed by the 10 tsi data, and at a diffusion time of 1/2 hour, the layer thicknesses of samples diffused at 520°C are greater than for samples diffused at 570°C -- again confirming LeClaire and Bear's observations. It appears, therefore, that layer growth behavior in the aluminum-uranium system is similar to that in the aluminum-nickel system in one respect, but dissimilar in another. In both systems, a "transient" or non-parabolic growth stage occurs which is then followed by the "steady state" period of parabolic growth. The "negative activation energy" phenomenon reported by LeClaire and Bear is clearly transient in nature. The effect of pressure, however, is different. Increasing applied pressure increases layer thickness in the aluminum-uranium system, but produces an opposite effect in the aluminum-nickel system.

4. Other Observations:

- (a) Marker motion: Molybdenum foil and tungsten wire markers placed at the original aluminum-uranium interface are consistently found at the UAl_3 -Al interface after diffusion -- a displacement amounting to at least several millimeters in some cases.
- (b) Hardness of UAl_3 phase: Microhardness traverses (25 gram load) have been made across couples diffused at 520°, 560° and 570°C for times ranging from 8 to 24 hours under applied pressures of 1-1/4 and 5 tsi. The results are roughly similar in each case; the UAl_3 phase has a maximum hardness of about 640 DPH which occurs approximately at the center and then falls off as the distance to the interface decreases.
- (c) Concentration differences in UAl_3 phase: An investigation is under way to determine the concentrations existing in the UAl_3 phase formed during interdiffusion. Samples have been obtained from scrapings removed from the UAl_3 phase near the Al- UAl_3 interface and near the center of the layer (well away from the UAl_2 layer), and the samples have been chemically analyzed by means of a colorimetric technique. At present, only tentative results are available; they indicate that a difference of 5 to 6 w/o may exist in the concentrations of the samples.
- (d) X-ray photographs of UAl_3 phase: Several X-ray photographs have been made of the UAl_3 layer in the following locations: (1) near the aluminum side; (2) at the center of the layer; and (3) near the uranium end. Distinct differences were noted in the appearances of the Debye-Scherrer rings. Near the aluminum end, the rings had a spotty appearance, as though the crystal size were quite large. At the center of the layer, the rings were smooth and sharp, but the rings became quite diffuse and wide towards the uranium end.

A photogram taken quite close to the uranium, but still within the intermetallic layer, also had additional spots visible, presumably from the relatively narrow UAl_2 phase.

B. Antimony-Tin, Antimony-Aluminum and Silver-Copper Systems

1. Diffusion in the Antimony-Tin and Other Systems:

Work has been under way to study the kinetics of interdiffusion in systems in which the phase equilibria are relatively simple compared with those existing in the aluminum-nickel system, for example. As was mentioned in the previous progress report⁽⁴⁾, three systems were originally selected for investigation, as follows: (a) The antimony-tin system, which contains one intermediate phase having a moderately extensive solubility range; (b) The antimony-aluminum system, in which one intermediate phase exists, the range of solubility being highly restricted; and (c) The silver-copper system, in which only one interface boundary exists. The objective of the investigations was to compare experimentally measured interface displacements with those calculated from the available solutions to the multi-phase diffusion problem mentioned earlier.

- (a) Antimony-Tin: Work on interdiffusion in the antimony-tin system has been continued. Additional data have been obtained at 200°C and the layer growth measurements of the β' (antimony-tin) phase are tabulated in Table II. The thickness data are plotted on logarithmic scales in Fig. 8. In the previous progress report, fewer points had been available upon which to base a curve, and it had been assumed that the parabolic growth law was nearly obeyed, the slope of the curve being 0.45 instead of the theoretical 0.50. Now, it appears much more likely that the parabolic law of growth is actually obeyed until a layer thickness of approximately 0.10 mm is reached. Subsequently, the growth curve drops below the ideal curve, and the slope drops to 0.33. Metallographic examination does not show the appearance of voids or major cracks that occur concurrently with the lower growth rate. The only noticeable difference is the presence of larger grains within the antimony-tin phase at the longer diffusion times. At the present time, this phenomenon is being investigated further. Also, experiments are under way to determine the diffusion coefficients in the tin-rich and antimony-rich terminal solid solutions, and in the antimony-tin phase, so that the theoretical calculation of the growth of the antimony-tin layer can be checked against the experimental growth data. A preliminary evaluation indicates that radioactive tracer techniques using available radioactive isotopes of tin and antimony will be suitable. The X-ray absorption technique cannot be used because insufficient contrast exists between tin and antimony. Also, wet chemical analysis of these sections is insufficiently accurate, since the amount of material available per slice is too minute.

- (b) Antimony-Aluminum: In the last progress report⁽⁴⁾, it was pointed out that an extremely irregular antimony-aluminum layer had formed in an antimony-aluminum diffusion couple annealed at 600°C for 96 hours under a pressure of 1 tsi. Two additional runs were made at the same pressure; one at 550°C for 96 hours, and one at 600°C for 405 hours. In both cases, the same irregularity of layer growth occurred as had been noted in the first run. It appeared as though the intermetallic compound was growing predominantly into the antimony and in a highly directional way. In many places along the cross-section of the specimens, it was observed that patches of antimony-aluminum had completely enveloped regions of antimony, producing islands of the latter within the intermetallic layer. The penetration did not seem to occur along grain boundaries; the grain size of the antimony was very large in relation to the spacing of the irregularities. The lack of uniform layer growth made it impossible to obtain meaningful data; therefore work in this system also has been discontinued.
- (c) Silver-Copper: A diffusion run of a silver-copper couple was completed after an anneal of 575 hours at 765°C; 0.0005" tungsten wires and a 0.0005" thick molybdenum ribbon were used as markers. It was found that the markers after diffusion were located within the silver-rich α phase at a distance approximately 0.008 mm from the α - β interface. Unfortunately, it was not possible to locate the displacement of the markers with respect to the original or Matano interface, since the sample had been plastically deformed during the run. Consequently, the results could not be interpreted in terms of intrinsic diffusivities of copper and silver in the α and β phases. In view of the slowness of the observed displacements, work in this system has been abandoned.

2. X-ray Absorption Apparatus:

An example of the type of concentration-penetration curve obtained with the X-ray absorption apparatus as originally built is shown in Fig. 9. It was found in general that the composition varied essentially linearly with distance in the γ (Ni_2Al_3) phase if its thickness were of the order of 0.2 mm. Deviations from linearity occurred in the vicinity of the phase boundaries. Unfortunately, the changes in concentration at the interfaces were not sufficiently abrupt to determine boundary concentrations unequivocally. Also, the location of the linear portion of the concentration-penetration curve was open to some question, since the phase boundary concentrations of the Ni_2Al_3 phase are supposedly 55 to 60 w/o nickel, according to the equilibrium phase diagram. Accordingly, the apparatus and technique are being improved as follows: (a) The length of the slit is to be reduced from 7 mm to 2 mm to minimize the effect of interface curvature and slit

misalignment; (b) The slit edges are to be thickened so as to decrease the transparency of beam-defining edges to the MoK_α radiation used; (c) Experimental checks are being made of the validity of the mass absorption coefficients reported in the literature for nickel and aluminum; and (d) Samples of known composition are being prepared as controls for the X-ray absorption analysis.

IV. DISCUSSION OF RESULTS

The phenomena associated with interdiffusion in the aluminum-uranium system are quite interesting and thought-provoking. A study of the data presently available gives rise to a number of questions which pertain not only to the aluminum-uranium system, but which have general implications as well. Examples of the latter are as follows: (1) What factors determine the relative rates of growth of intermetallic phases in a given system? (2) Why does an increase in applied pressure inhibit layer growth in one system and accelerate it in another? (3) How does interdiffusion take place through an ordered intermetallic phase? and (4) Why do "transient" phenomena appear during the early stages of layer growth? As yet, the data are insufficient to permit a worthwhile discussion of the last point, but sufficient evidence is available to permit some basis for speculation about the others.

There appears to be no major difficulty in explaining the relative rates of growth of the UAl_3 , UAl_2 and UAl_4 phases on the general basis that volume diffusion is the rate-controlling factor and that growth rate values are determined by interface concentrations and by relative values of diffusion coefficients in the phases^(2,3,5). This explanation requires the existence of an appreciable concentration gradient and a relatively large diffusion coefficient value in UAl_3 . The former requirement seems to be incompatible with the aluminum-uranium equilibrium phase diagram (Fig. 1) where no appreciable solubility range is attributed to UAl_3 . However, since the work done to determine the phase diagram was admittedly in the nature of a survey, and since no attempt was made to determine exactly such features as solid solubility limits⁽¹²⁾, the apparent incompatibility may not be serious. The preliminary work done in this investigation indicates that a solubility range of 5 to 6 w/o is probable for the UAl_3 phase.

The effect of pressure on the growth rate of the UAl_3 phase seems to correlate well with the reduction or the elimination of void bands. It may be, therefore, that the observed increase in the UAl_3 layer thickness with increased applied pressure can be explained on the basis of the effective specimen cross-sectional area available for interdiffusion. An increase in pressure (all other conditions being held equal) reduces the number of voids in the specimen. In this manner, the effective cross-sectional area for diffusion could be increased and thicker layers would result. Support for this viewpoint is lent by the results recently reported by Barnes and Mazey⁽¹³⁾. They found that the application of hydrostatic pressure to sandwich diffusion couples of copper and nickel reduced the number of voids which tended to form. If the pressure was equal to or greater than 1300 psi at 1000°C, the formation of voids was prevented altogether. They concluded that pressure suppressed void formation by altering the equilibria in regions where voids would otherwise nucleate

and grow. It remains to be proven that the formation of voids in the UAl_3 phase is likewise the result of the condensation of excess vacancies which are generated in the layer by the action of the Kirkendall effect^(14,15,16). If the voids are caused by excess vacancy condensation, it should be possible to show that the concentration gradient varies sharply in the regions in which the voids are found⁽¹⁵⁾. Anticipating the correctness of the above explanation for the aluminum-uranium system, one could then visualize applied pressure as exerting a threefold effect on layer growth in multiphase systems, as follows: (1) a reduction in the diffusion coefficients, which would result in a decrease in volume diffusion/unit cross-sectional area and a diminution in layer growth; (2) an increase in the cross-sectional area available for diffusion, which would increase layer growth; and (3) an alteration in the equilibria affecting the interface boundary concentrations, which could either increase or decrease volume diffusion/unit cross-sectional area, and thus either increase or decrease layer growth. On the basis of the work done to date, it is felt that the first two phenomena may predominate in the pressure ranges used. However, until better data become available, the magnitude of the pressure effect on interface boundary concentrations and its importance to layer growth remains unresolved.

Concerning the question of interdiffusion in intermetallic phases, there seems to be little, if any, directly applicable data available in the literature, other than the work of Berkowitz, Jaumot and Nix in $NiAl$ ⁽¹⁷⁾. Interdiffusion in layers produced as a result of oxidation and tarnishing reactions have been investigated in detail⁽¹⁸⁾ and the concepts developed should undoubtedly be of help. Indeed, it appears from the results of marker movement studies in both the aluminum-nickel and the aluminum-uranium systems that interdiffusion occurs predominantly by the movement of aluminum ions. It is not yet clear, however, if the aluminum ions migrate interstitially or along vacant sites in the aluminum sub-lattices. The study of diffusion in the UAl_3 phase is appealing, since the structure is simple, being of the copper-gold type. However, the major difficulties involve the preparation of UAl_3 in the bulk, and the acquisition of suitable quantities of Al^{26} isotope.

V. FUTURE WORK

The study of layer formation and growth in the aluminum-uranium system will be continued and concluded. Specifically, the following points remain to be clarified:

1. The extremely rapid growth of the UAl_3 phase, which supposedly has a highly restricted solubility range.
2. The mechanism of diffusion in the UAl_3 phase.
3. The mechanism of macroscopic void formation in the UAl_3 phase.
4. The mechanism responsible for the morphology of the UAl_2 , which

grows in a finger-like manner into the UAl_3 , rather than developing in a more conventional manner as a layer between the aluminum and the UAl_3 phase.

An attempt will be made to prepare incremental couples consisting of bulk UAl_3 and uranium, in order to gain insight into the nature of the UAl_2 phenomenon. As for the first three items, a satisfactory explanation will undoubtedly require the determination of the diffusion coefficients in the UAl_3 phase. Ideally, it would be desirable to determine the self-diffusivity of the aluminum ions by means of a radioactive tracer technique, and consideration will be given to this possibility. However, the limited availability of the Al^{26} isotope and its high cost per unit activity may make its use marginal for this investigation. If the isotopic method cannot be used, an attempt will be made to determine the needed data by the use of incremental UAl_3 phase diffusion couples. In any event, a rough estimate will be made of the diffusion coefficient by calculation from the layer growth data in a manner similar to that already used in the aluminum-nickel system⁽²⁾. Also, the X-ray absorption technique will be refined to permit determination of the concentration-penetration curve and the values of interface boundary concentrations.

The work in the antimony-tin system will be continued. The diffusion coefficients of the three phases will be experimentally determined by radioactive tracer techniques; layer growth rates will then be calculated from the theory and compared with experimentally determined data. If the agreement obtained indicates that reasonable estimates of diffusion coefficients can be gotten from layer growth data, the program will be expanded to include the study of systems having more complicated phase equilibria and more components.

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TABLE I

SUMMARY OF LAYER THICKNESS DATA IN THE ALUMINUM-URANIUM SYSTEM

<u>Diffusion</u>		<u>UAl₃ Layer</u>	
Temp. (°C)	Time (Hr.)	Average Thickness (mm)	Standard Deviation (mm)
<u>1-1/4 TSI Pressure</u>			
520	3	0.62	± 0.02
	24	2.60	± 0.26
570	8	2.79	± 0.05
	16	3.98	± 0.08
	24	4.3	± 0.2
<u>5 TSI Pressure</u>			
390	110	4.30	± 0.06
520	3	1.45	± 0.02
	8	2.33	± 0.05
	16	3.30	± 0.02
	24	4.09	± 0.04
	24	4.21	± 0.03
560	3	1.64	± 0.03
	8	2.78	± 0.03
	16	4.09	± 0.04
	24	5.0	± 0.1
	29	5.44	± 0.09
565	24	5.0	± 0.3
570	3	1.70	± 0.02
	8	2.96	± 0.03
	16	4.23	± 0.08
	24	5.16	± 0.05
<u>10 TSI Pressure</u>			
480	3	1.62	± 0.03
520	1/2	0.85	± 0.01
	3	1.75	± 0.07
	3	1.8	± 0.1
	8	2.55	± 0.04
	24	4.39	± 0.07

TABLE I (Continued)

<u>Diffusion</u>		<u>UAl₃</u>	<u>Layer</u>
Temp. (°C)	Time (Hr.)	Average Thickness (mm)	Standard Deviation (mm)

10 TSI Pressure (Continued)

540	3	1.51	± 0.06
560	3	1.68	± 0.01
570	1/2	0.84	± 0.02
	2-1/2	1.67	± 0.03
	3	1.77	± 0.02
	8	2.95	± 0.02
	8	2.96	± 0.05

TABLE II

SUMMARY OF LAYER THICKNESS DATA IN THE ANTIMONY-TIN SYSTEM

<u>Number</u>		<u>Diffusion</u>		<u>β' Layer</u>	
<u>Sample</u>	<u>Total Measurement</u>	<u>Temp. (°C)</u>	<u>Time (min.)</u>	<u>Average Thickness (mm)</u>	<u>Standard Deviation (mm)</u>
1	68	200	150	0.020	$\pm$ 0.003
1	56		270	0.026	$\pm$ 0.003
1	68		450	0.036	$\pm$ 0.005
1	52		1440	0.066	$\pm$ 0.003
1	67		2880	0.093	$\pm$ 0.004
1	45		8640	0.140	$\pm$ 0.004
1	56		18720	0.179	$\pm$ 0.004
1	55		24600	0.200	$\pm$ 0.006
1	76	220	270	0.032	$\pm$ 0.005
1	15		450	0.042	$\pm$ 0.004
1	30		1560	0.071	$\pm$ 0.007
1	40		2880	0.106	$\pm$ 0.003
1	24		4320	0.109	$\pm$ 0.005
1	31		8640	0.151	$\pm$ 0.005
1	82		18720	0.18	$\pm$ 0.01

FIGURE I. THE SYSTEM URANIUM-ALUMINUM

(a) Etched

100X

(b)

Etched

500X

Fig. 2. Phases appearing in U-Al diffusion couple annealed at 520°C for 24 hours under an applied pressure of 5 tsi; etched with 5% ammonium persulfate solution containing a little hydrogen peroxide.

← U

← UAl_2 (?)

+

UAl_3

(a) Not etched 3 hours 100X

← U

← UAl_2 (?)

+

UAl_3

(b) Not etched 8 hours 100X

Fig. 3. Two-phase structure appearing in U-Al diffusion couples annealed at 520°C under an applied pressure of 10 tsi for varying times.

← U →

← UAl_3 →

← Al →

(a) Not etched 110 hrs. at 390°C 13X

(b) Not etched 24 hrs. at 520°C 13X

← U →

← UAl_3 →

← Al →

(c) Not etched 24 hrs. at 560°C 13X

Fig. 4. U-Al diffusion couples annealed under an applied pressure of 5 tsi for varying diffusion temperatures and times.

(a) Not etched 3 hours 13X

← U →

← UAl_3 →

← Al →

(b) Not etched 8 hours 13X

(c) Not etched 16 hours 13X

← U →

← UAl_3 →

← Al →

(d) Not etched 24 hours 13X

Fig. 5. U-Al diffusion couples annealed at 520°C under an applied pressure of 5 tsi for varying times.

(a) Not etched $1\frac{1}{4}$ tsi 13X

(b) Not etched 5 tsi 13X

(c) Not etched 10 tsi 13X

Fig. 6. U-Al diffusion couples annealed at 570°C for 8 hours under varying applied pressures.

Fig. 7. Kinetics of growth of the UAl_3 layer in U-Al diffusion couples at diffusion temperatures of 520°, 560° and 570°C under applied pressures of 5 and 10 tsi.

Fig. 8. Kinetics of growth of the β' (SbSn) layer in the Sb-Sn diffusion couples annealed at temperatures of 200° and 220°C under an applied pressure of 1 tsi.