FUNDAMENTALS OF DIFFUSIONAL BONDING—IV

Fourth Annual Progress Report [for] July 1, 1958 through
June 30, 1959

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SYLVANIA ELECTRIC PRODUCTS INC.

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ABSTRACT

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 UAl_3 phase have been determined in the temperature range 400° to $600^\circ C$ and at pressures of 1-1/4, 5 and 10 tsi. The origin of the macroscopic defects occurring in the UAl_3 layer during interdiffusion has been clarified. The solubility range of UAl_3 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. UAl_3 and of UAl_3 vs. uranium discs to determine the growth rates of the UAl_4 and UAl_2 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.

I. INTRODUCTION

The nature of the bond which exists between the fuel-bearing core and the surface cladding of a nuclear reactor fuel element is of the utmost importance, since it must be thermally stable, mechanically strong, and capable of good heat transfer. One of the important factors known to affect strength and stability is the formation and growth of intermetallic layers at the original bond interface during elevated temperature service. Accordingly, in the present investigation, a fundamental study is under way of intermetallic layer growth and other interdiffusional aspects of diffusional bonding.

The first phase of the program consisted of an intensive investigation of the aluminum-nickel system, in which the kinetics of growth of the Ni_2Al_3 and other phase layers were determined as a function of interdiffusion temperature and applied pressure.^(1,2,3) As a result of the work, it was concluded that all thermodynamically stable intermetallic phases appear as layers within the diffusion zone, and that the rate-controlling factor is volume diffusion. A detailed analysis was next undertaken of available theoretical solutions to cases involving diffusion in two and three phases in two-component systems, so that some quantitative appreciation could be gained of the factors influencing volume diffusion.⁽⁴⁾ It was concluded that the interface concentrations, as well as the diffusion coefficients of the individual phases, could be important in affecting the course of interdiffusion.

In the aluminum-nickel system, it appeared possible to rationalize the experimental observations, including the pressure-induced diminution of layer growth, on the basis of diffusion coefficient considerations alone. However, the experimental observations of LeClaire and Bear on the aluminum-uranium system,⁽⁵⁾ in which a pressure-induced enhancement of layer growth was reported, pointed out the necessity of including in any phenomenological theory of layer growth a proper evaluation of the role played by shifts in interface concentrations. Therefore, the second phase of the present program has been concerned with a study of layer formation and growth in the aluminum-uranium system, particular emphasis being attached to the effects of pressure on interface concentrations.

In the immediately previous progress report,⁽¹⁾ the initial results of the investigation were reported substantially as follows:

- (a) The acceleration of UAl_3 layer growth by increasing applied pressure was confirmed.
- (b) The kinetics of growth of the UAl_3 phase under applied pressures of 5 and 10 tsi appeared to be conventional, except in diffusion runs at times less than 3 hours.
- (c) Interesting correlations were 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 layer of the diffusion zone.

During the present reporting period, the investigation of the kinetics of growth of the UAl_3 layer in the aluminum-uranium system has been substantially completed, and exploratory runs have been made with incremental aluminum vs. UAl_3 couples and UAl_3 vs. uranium couples to determine the kinetics of growth of the UAl_2 and UAl_4 phases. A nuclear track emulsion technique has been used to determine the solubility range of the UAl_3 layer. The nature of the macroscopic defect structure occurring in the UAl_3 layer has been further clarified by means of metallographic and hot hardness investigations. Finally, 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-nickel and the aluminum-uranium systems.⁽⁶⁾ The details of the progress made to date are reviewed in the succeeding sections of this report.

II. EXPERIMENTAL PROCEDURE

The preparation of aluminum-uranium diffusion couples was described in the previous progress report.⁽¹⁾ For the incremental aluminum vs. UAl_3 and the UAl_3 vs. uranium diffusion couples, it was first necessary to prepare discs made of bulk UAl_3 . This was accomplished by the use of an interdiffusion technique similar to that used in previous work for the preparation of bulk Ni_2Al_3 .⁽¹⁾ A diffusion couple consisting of an 0.075" thick uranium disc and an 0.177" thick aluminum disc was interdiffused at 600°C for 48 hours under an applied pressure of 10 tsi. The UAl_3 intermetallic phase formed was then homogenized at 720°C for 24 hours (to eliminate any traces of UAl_4 which might have been present) and at 900°C for 168 hours within an evacuated quartz capsule.

Several determinations were made of the uranium concentration in the UAl_3 layer of an aluminum-uranium diffusion couple by means of a nuclear track emulsion technique. An Eastman Kodak type NTA nuclear tracking emulsion film was placed in contact with longitudinal sections of diffusion couples for several hours and then developed. The tracks left in selected areas of the emulsion by alpha particles produced in the decay of the U^{238} and U^{235} isotopes were then counted with a counting grid magnified to about 750 diameters on a Zeiss petrographic microscope. The counts were subsequently converted to uranium concentrations with the aid of suitable UAl_3 standards prepared in the manner described above.

All other experimental procedures used were conventional.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Physical Appearance of Diffusion Layers:

It was reported in the previous progress report⁽¹⁾ that interesting correlations exist between the applied pressure, the diffusion temperature, the diffusion time, and the nature of the macroscopic defects occurring within the diffusion zone. Aluminum-uranium couples

annealed at low temperatures (390°C or thereabout) exhibited a "porous" appearance in the diffusion zone, the "porosity" being distributed uniformly through the UAl_3 layer. At higher diffusion temperatures (570°C to 600°C), the UAl_3 layer was relatively sound except for a band of large voids near the UAl_3 - aluminum interface. The effect of an increase in pressure was to diminish the number and size of the macroscopic defects. In addition, a two-phase region was found to exist in some of the diffusion couples near the UAl_3 - uranium interface, especially in those annealed at 520°C, in which finger-like growths, presumably of UAl_2 , co-existed with the UAl_3 phase. It was pointed out that the nature of these phenomena was not understood at the time.

A sufficient number of diffusion couples have now been prepared and examined to permit some clarification of the microstructure. It appears that the growth of the UAl_3 phase into the uranium during interdiffusion is highly irregular, especially in 520°C specimens annealed for 3 hours or less. As a result, the UAl_3 phase isolates regions of uranium which appear as stringers lying roughly parallel to the diffusion direction. Traces of a third phase, identified by X-ray diffraction as UAl_2 , occur as a fine precipitate along the boundaries between the stringers and the UAl_3 matrix. At increasingly greater distances from the UAl_3 - uranium interface, the uranium islands diminish in size and number and eventually disappear; also, the amount of UAl_2 phase precipitate surrounding the uranium islands passes through a maximum and then decreases and disappears.

The "porosity" observed in specimens diffused at low temperature is connected with the formation of microcracks which apparently originate in the vicinity of the UAl_3 - uranium interface (Fig. 1). The microcracks appear in the UAl_3 phase, run transversely to the specimen axis, and are confined to regions lying between the uranium stringers or islands. Their shape becomes more oval as their distance from the UAl_3 - uranium interface increases, and give the specimen a porous appearance at low magnifications. It is strongly suspected that crack initiation is associated with the formation of the UAl_2 precipitate at the periphery of the uranium stringers, and that the role played by the UAl_2 phase in affecting interdiffusion in the aluminum-uranium system is not so insignificant as was hitherto believed. If one considers the volume change accompanying the reaction

it is clear that the formation of the UAl_2 phase must be accompanied by considerable lattice strain. A gram atom in the UAl_2 phase configuration occupies about 6.5 cc more than a gram atom of either of the reactants forming it. (7,8,9) It is well known that the UAl_3 phase is quite brittle at room temperature, as evidenced by the formation of

large cracks in the layer which occur when the diffusion couples are removed from the dies and when they are rough-ground during metallographic preparation. Hot hardness readings taken at elevated temperatures indicate that the UAl_3 phase maintains considerable strength at the lower diffusion temperatures used in this investigation, as the following table shows:

Temperature (°C)	Hardness (DPH)
25	605
450	340
550	233

Thus, at a diffusion temperature of 390°C , where the UAl_3 phase probably has a hardness in excess of 400 DPH, the dilatation strains accompanying the formation of the UAl_2 phase precipitate could initiate fracture in the UAl_3 phase. At higher temperatures, where the "porosity" is not observed, the deleterious effects of UAl_2 phase formation would be reduced as the ductility of the UAl_3 phase increased. Also, an increase in applied pressure would lower the thermodynamic stability of the UAl_2 phase in relation to the neighboring phases and would tend to inhibit its formation.

The origin of the voids appearing at the higher diffusion temperatures is not clear. As was indicated in the previous progress report,⁽¹⁾ a band of macroscopic defects of approximately constant thickness forms in the UAl_3 layer early in the interdiffusion process and persists in the vicinity of the UAl_3 - aluminum interface at all diffusion times (Fig. 5, SEP-251).

One possible explanation is that during the early stages of interdiffusion, cracks are formed by the same mechanism discussed above. However, as interdiffusion proceeds, conditions change, in some as yet undetermined way, and cracks no longer form. Marker motion studies show that diffusion in the UAl_3 interface occurs predominantly by the motion of aluminum ions. If the explanation advanced herein is correct, one would expect to see a band of defects near the UAl_3 - aluminum interface and nowhere else, since the defects would behave as internal markers. They would move relative to the UAl_3 - uranium interface, where the UAl_3 was being formed, and exhibit no displacement relative to the UAl_3 - aluminum interface.

B. Kinetics of Layer Growth:

In the previous progress report, the initial results obtained in the measurement of UAl_3 layer growth were discussed. It was reported that the UAl_3 phase grew with great rapidity, and that increasing applied pressure increased the thickness of the UAl_3 phase, in confirmation

of the observations of LeClaire and Bear and other investigators. The growth kinetics at 5 tsi and in the temperature range 520°C to 570°C appeared to be conventional for diffusion times longer than 3 hours. A parabolic growth law was obeyed and volume diffusion appeared to be the rate-controlling factor. At the earlier diffusion times, however, the situation was unclear.

During the current reporting period, additional growth data have been obtained for the UAl_3 phase, particularly at pressures of 1-1/4 and 5 tsi. The data are listed in Tables I and II; for completeness, the layer growth data obtained previously are also included. The thickness data are plotted on logarithmic scales in Figs. 2, 3 and 4 (log thickness vs. log time). In the temperature range 520°C to 570°C, and for times greater than 3 hours, the data confirm the observations made in the previous progress report. The 400°C data for the 5 tsi runs, however, do not obey the parabolic growth law. The available 10 tsi data are still somewhat fragmentary, and additional data are currently being obtained. Several inconsistencies occur which require resolution. The half-hour and three-hour 520°C points do not lie on the line established by the 8- and 24-hour points (the latter are consistent with the 5 tsi data). Also, the layer thicknesses of the one-half and three-hour points are approximately equal and greater, respectively, than the half-hour 570°C and the three-hour 560°C points. Finally, a number of higher pressure runs have been made at 500°C and 600°C. It appears from the results that pressures above 10 tsi no longer increase the UAl_3 layer thickness at these temperatures.

LeClaire and Bear⁽⁵⁾ have reported the existence of a "negative activation energy phenomenon" in which layer thicknesses increased with decreasing temperature in diffusion couples annealed in the temperature range 520°C to 570°C and at applied pressures of 5 and 10 tsi. The 1-1/4 tsi runs did not show the same anomalous behavior. All of the diffusion runs made by LeClaire and Bear were of 3 hours duration or less. In the present investigation, the "negative activation energy phenomenon" has not been confirmed in the diffusion runs made at 5 tsi for times ranging from 3 hours to 24 hours. In view of the microstructural changes which have been demonstrated to take place as a function of diffusion time in the UAl_3 phase, it is probable that the "negative activation energy phenomenon" is a transient effect occurring at short diffusion times. The data obtained in the present investigation are less susceptible to transient effects, since they cover a much longer time span. Variations in layer thickness due to transient effects occurring initially will obviously be a much smaller fraction of the total layer thickness at long diffusion times than at short diffusion times. In the present instance, it would of course be desirable to obtain short-time diffusion data as well, so that the two sets of observations could be unequivocally reconciled. Unfortunately, the apparatus used for this program is not completely suitable for short-time runs, since it has a high thermal inertia, and an appreciable error develops in the measurement of the duration of short-time runs (runs lasting less than 3 hours).

It is customary to plot kinetics data of the type shown in Figs. 2 and 3 in such a manner as to obtain an activation energy for diffusion in the intermetallic phase. This is done by plotting the logarithm of the reciprocal of the time required for the intermetallic phase to grow to a selected constant thickness vs. the reciprocal of the absolute temperature. The conditions which must be satisfied to make the procedure a valid one have been discussed in an earlier paper.⁽³⁾ In brief, volume diffusion must be the predominant rate-controlling factor for layer growth, and diffusion should effectively be occurring only in one phase -- the phase comprising the bulk of the diffusion zone. In the case of the UAl_3 phase, the second condition is obviously fulfilled, and at any one of the higher temperatures the data indicate that volume diffusion is rate-controlling. A complicating factor arises, however, in that from one temperature to the next the effective cross-sectional area for diffusion must change as the distribution and kind of macroscopic defects change. Thus, the interpretation of the slope of the $\ln(1/t)$ vs. $(1/T)$ plot (where t is the diffusion time to reach constant thickness and T the absolute temperature) becomes complicated. In general, one would not expect to obtain a constant slope under these circumstances. If the data of Fig. 3 are replotted in this manner, it nevertheless turns out that a reasonably straight line is obtained for the slope in the temperature range 520°C to 600°C if the constant thickness value chosen is large. These are the conditions which tend to minimize the influence of the changes in the effective cross-sectional area for diffusion. A value of about 12,000 cal/mol is obtained for the estimate of the heat of activation for diffusion in the UAl_3 phase under an applied pressure of 5 tsi. Since it is the aluminum ions which are diffusing predominantly, the heat of activation value is taken to refer to the energy barrier affecting aluminum ion migration. LeClaire and Bear obtain a value of about 15,000 cal/mol, except in the temperature region where they report a negative heat of activation.

Several exploratory runs with incremental diffusional couples have been completed so that some idea could be obtained of the kinetics of growth of the UAl_4 phase and the UAl_2 phase. There seems to be no special difficulty in obtaining kinetics data for the UAl_4 phase in UAl_3 vs. aluminum couples. One run has been completed in which a UAl_3 vs. aluminum couple was annealed at 600°C for 4 hours under an applied pressure of 2-1/2 tsi. The thickness of the UAl_4 phase (Fig. 5) was 0.087 mm -- less than 1/25th the thickness of the UAl_3 phase obtained in an aluminum vs. uranium couple annealed under similar conditions. The diffusion coefficient calculated from this one run for the UAl_4 phase, using the method outlined in reference (3), is about three orders of magnitude less than that of the UAl_3 phase. The situation is different in obtaining kinetics data for the UAl_2 phase by the use of UAl_3 vs. uranium diffusion couples. The attempts made to date have proven unsuccessful. Two UAl_3 vs. uranium couples have been run at 600°C ; one for 72 hours under a pressure of 2 tsi, and one for 144 hours under a pressure of 5 tsi. In the first case,

examination of the interface after diffusion revealed a very thin layer of a dark phase, presumably uranium oxide, less than one micron thick, and practically no sign of the UAl_2 phase. In the second case (Fig. 6), the presence of the UAl_2 phase was observed in addition to the dark layer, its thickness being about two microns. Since the UAl_2 phase had occasionally been observed to grow to thicknesses of 40 microns in aluminum vs. uranium couples diffused under circumstances theoretically less favorable for its growth than in UAl_3 vs. uranium couples, it was felt that the oxide layer was acting as a diffusion barrier. Consequently, a different scheme was tried in which a restricted thickness of aluminum was used in an aluminum vs. uranium couple. It was expected that the aluminum would be quickly depleted, forming the UAl_3 phase, and that subsequently the UAl_2 phase would grow at the UAl_3 - uranium interface without hindrance. Two couples were diffusion annealed under a pressure of 10 tsi for 144 hours; one at 560°C and the other at 600°C. Surprisingly, even though the UAl_3 - uranium interface was free of the dark oxide layer, no layers of UAl_2 phase were found in appreciable thickness. The conclusion appears inescapable that the formation or growth of the UAl_2 phase is a very slow process under the conditions existing in a diffusion couple.

C. Solubility Range of UAl_3 Phase:

As was indicated in the previous progress report, the very rapid growth of the UAl_3 phase during interdiffusion would be difficult to explain unless an appreciable concentration gradient existed in the UAl_3 phase. As a check on the existence of a solubility range in the UAl_3 phase, several determinations were made of the uranium concentration at two locations in the UAl_3 phase layer of an aluminum vs. uranium diffusion couple by means of a nuclear track emulsion technique. The results obtained are listed in the following table:

<u>Sample</u>	<u>Location</u>	<u>Tracks/Unit Area</u>		<u>Uranium Composition</u>	
		<u>Mean</u>	<u>Std. Dev.</u>	<u>w/o</u>	<u>a/o</u>
U-Al Couple	In U	163.2	± 3.8	100	100
	*In UAl_3 near UAl_3 -U boundary	98.4	± 1.0	76.0**	26.3**
	In UAl_3 near Al- UAl_3 boundary	85.0	± 3.0	73.3**	23.7**
Bulk UAl_3 Standards					
25.5 a/o U	--	95.0	± 3.7	75.2	25.5
23.3 a/o U	--	83.5	± 2.7	72.8	23.3

* In single-phase region of UAl_3 phase.

** Obtained by interpolation or extrapolation.

The solubility range of the UAl_3 phase is about 2.6 a/o or 2.7 w/o uranium. The limits of solubility occur at uranium concentrations somewhat less than 23.7 a/o and somewhat greater than 26.3 a/o. As a matter of incidental interest, one may use the Bragg-Kleeman relationship⁽¹¹⁾ to calculate the ranges of alpha particles in uranium and in the UAl_3 phase, and the alpha counts obtained from the aluminum-uranium couple to determine more approximate values of the uranium concentrations. The values so obtained are 22.4 a/o and 25.9 a/o uranium, as compared to the more accurate values of 23.7 and 26.3 a/o uranium determined by means of bulk UAl_3 standards of known composition.

D. Effect of Pressure on Layer Growth:

A theoretical analysis of the effect of pressure on intermetallic layer growth has been completed and submitted for publication.⁽⁶⁾ The purpose of the analysis was to estimate the magnitude of shifts in phase equilibria as a function of hydrostatic pressure, and to compare the relative importance of such changes with those of pressure-induced changes in diffusion coefficients in controlling the course of multi-phase diffusion. In order that some quantitative insight into the phenomena could be gained, a highly idealized case of interdiffusion was first considered in which the kinetics of layer growth were controlled by equilibrium interface concentrations and by the diffusion coefficient in the intermetallic phase. An equation was derived which enables one to estimate the relative importance to intermetallic layer growth of pressure-induced changes in the diffusion coefficients and pressure-induced shifts in the interface boundary concentrations. The equation shows that the importance of equilibrium concentration changes depends upon the magnitude of the dimensionless parameter $Z = \lambda / 2(Dt)^{1/2}$ where λ is the mean interface displacement, D is the chemical diffusion coefficient of the intermetallic phase, and t is the interdiffusion time. A given change in concentration becomes relatively quite important when Z is very small. The magnitude of changes in concentration was estimated by an evaluation of the extent to which hydrostatic pressure influences the phase equilibria. A simple geometrical argument was used to determine the effects of pressure on the Gibbs free energy curves. It was shown that interesting relationships exist between the equilibrium concentration changes and between (a) the magnitude of the applied pressure, (b) the mean atomic volumes of the phases, (c) the widths of the miscibility gaps, (d) the shapes of the free energy curves, and (e) the inclinations of their common tangents. Next, the pressure-induced increase in the growth rate of the UAl_3 phase of the aluminum-uranium system and the pressure-induced decrease in growth rate of the Ni_2Al_3 phase of the aluminum-nickel system were considered. It was decided that it may be possible to rationalize the growth kinetics of the UAl_3 phase on the basis of the extent to which pressure increases the equilibrium solubility range, but only if maximum theoretical increases are allowed. Since it is doubtful that these would occur, it was felt that the alternative explanation proposed in the previous progress report

was preferable. According to the latter, a purely mechanical effect was envisioned; pressure was assumed to increase layer growth by repressing the tendency for the formation of macroscopic defects and thus effectively increasing the cross-sectional area available for interdiffusion. Finally, it was concluded that the pressure-induced decrease in the Ni_2Al_3 phase diffusion coefficient is more important to the growth kinetics of the Ni_2Al_3 phase than equilibrium solubility range changes.

E. Diffusion in the Antimony-Tin System:

It was pointed out in previous progress reports that it would be desirable to study the kinetics of interdiffusion in systems in which the phase equilibria are relatively simple compared with those existing in the aluminum-nickel and aluminum-uranium systems. One such is the antimony-tin system, which contains one intermediate phase, β' , having a moderately extensive solubility range and two terminal solid solutions. A minor effort has been under way to measure the diffusion coefficients in the three phases, so that layer growth rates could be calculated from theory and compared with experimentally determined data. One diffusion couple consisting of a disc of antimony-tin alloy (3.5 w/o Sb) sandwiched in between two discs of tin has been annealed for 400 hours at 220°C . The diffusion zone has been sectioned into one-mil increments, and a concentration-penetration curve is in the process of being determined by chemical analysis.

IV. FUTURE WORK

The theoretical analysis recently concluded suggests that interface concentrations may be changed by pressure enough to significantly affect the kinetics of layer growth. It would be desirable to verify this prediction experimentally. Ideally, a system is needed for study in which an increase in pressure increases layer growth appreciably -- a condition which insures that the effect of pressure on diffusion coefficients (which results in an inhibition of growth) is of lesser consequence than the pressure-induced shifts in interface concentrations. For this specific purpose, the aluminum-uranium system has proven to be unsatisfactory for three reasons: (a) complications occur at the layer interfaces due to the extremely rapid and irregular growth of the UAl_3 layer; (b) the occurrence of pressure-sensitive macroscopic voids and other defects tends to obscure the effect of pressure on interface concentrations; and (c) the possibility cannot be ignored that the entire pressure effect may be due to the existence of the voids and not to shifts in the equilibrium concentrations. To date, one other system has been reported⁽¹²⁾ in which pressure enhances layer growth, and that is the aluminum-thorium system. Apparently, the difficulties associated with the study of pressure phenomena in the aluminum-uranium system are missing. It is therefore planned to study the kinetics of layer growth

in the aluminum-thorium system so that the pressure effect may be clarified further.

The effect on intermetallic layer growth of specific mechanisms governing interdiffusion has not yet been demonstrated. In this connection, it is felt that valuable information can be gleaned from a study of the effect of ordering on the kinetics of growth of an intermediate phase. The copper-zinc system is well suited for this purpose. In the copper-zinc system, the body centered cubic " β brass" phase is disordered for all compositions at temperatures above 468°C and ordered for all compositions at temperatures below 454°C . Diffusion coefficients have been determined as a function of temperature for the β phase, both in the ordered and disordered states,⁽¹³⁾ and a theoretical picture of the mechanism of diffusion in this and other ordered solutions is emerging.^(14, 15) It is therefore planned to determine the growth kinetics of the β phase in incremental diffusion couples of α -brass vs. γ -brass as a function of temperature and pressure in order to determine the effects produced by the order-disorder phenomenon.

It is expected that the above programs will constitute the major effort during the coming year. Also, as time permits, the work already begun in the antimony-tin system will be continued for the purpose of experimentally validating the theoretical solutions to the three-phase two-component diffusion problem.

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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
	3	1.08	+ 0.03
	16.5	2.61	+ 0.23
	24	2.60*	+ 0.26
		3.05**	+ 0.28
570	3	1.75	+ 0.03
	8	2.79	+ 0.05
	16	3.98	+ 0.08
	24	4.3 *	+ 0.02
		4.42**	+ 0.13
600	3	1.91	+ 0.05
	6.25	2.80	+ 0.05
	24	5.07	+ 0.21
<u>5 TSI Pressure</u>			
390	110	4.30	+ 0.06
400	3	0.27	+ 0.08
	8	0.78	+ 0.05
	16	1.76	+ 0.02
	24.5	2.59	+ 0.06
	72	3.59	+ 0.11
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	5.02	+ 0.28
	29	5.44	+ 0.09
565	24	5.02	+ 0.28
570	3	1.70	+ 0.02
	8	2.96	+ 0.03
	16	4.23	+ 0.08
	24	5.16	+ 0.05

* Inaccurate due to irregular growth

** Maximum values

TABLE I (Continued)

<u>Diffusion</u>		<u>UAl₃</u> <u>Layer</u>	
Temp. (°C)	Time (Hr.)	Average Thickness (mm)	Standard Deviation (mm)
<u>5 TSI Pressure (Continued)</u>			
600	3	1.99	+ 0.01
	4.5	2.42	+ 0.10
	8	3.25	+ 0.11
	16	4.59	+ 0.06
<u>10 TSI Pressure</u>			
480	3	1.62	+ 0.03
520	0.5	0.85	+ 0.01
	3	1.75	+ 0.07
	3	1.81	+ 0.10
	8	2.55	+ 0.04
	24	4.39	+ 0.07
540	3	1.51	+ 0.06
560	3	1.68	+ 0.01
570	0.5	0.84	+ 0.02
	2.5	1.67	+ 0.03
	3	1.77	+ 0.02
	8	2.95	+ 0.02
	8	2.96	+ 0.05
600	3	2.04	+ 0.07
<u>15 TSI Pressure</u>			
600	3	2.03	+ 0.03
<u>20 TSI Pressure</u>			
600	3	2.07	+ 0.03

TABLE II

SUMMARY OF LAYER THICKNESS DATA IN THE ALUMINUM-URANIUM SYSTEM

<u>Diffusion</u>		<u>Average</u>	
Temp. (°C)	Time (Hr.)	Layer Thickness (mm)	Standard Deviation (mm)
<u>UAl₄</u> (<u>UAl₃</u> vs. Aluminum)			
<u>2.5 TSI Pressure</u>			
600	4	0.087	+ 0.006
	24	0.146	+ 0.026
<u>UAl₂</u> (<u>UAl₃</u> vs. Uranium)			
<u>2 TSI Pressure</u>			
600	72	0.0007	nil
<u>5 TSI Pressure</u>			
600	144	0.0019	+ 0.0002

Unetched

500X

(a) Near the UAl_3 interface

Unetched

500X

(b) Interior of the UAl_3 layer

Unetched

500X

(c) Near the $Al-UAl_3$ interface

Fig. 1 $Al-U$ diffusion couple annealed for 8 hours at $400^{\circ}C$ under a pressure of 5 tsi.

Fig. 2. Kinetics of growth of the UAl_3 layer in Al vs. U diffusion couples under an applied pressure of $1\frac{1}{4}$ tsi.

Fig. 3. Kinetics of growth of the UAl_3 layer in Al vs. U diffusion couples under an applied pressure of 5 tsi.

Fig. 4. Kinetics of growth of the UAl_3 layer in Al vs. U diffusion couples under an applied pressure of 10 tsi.

Etched

200X

Fig. 5 Al vs. UAl_3 diffusion couple annealed at 600°C for 4 hours under a pressure of 2-1/2 tsi; etched with hydrofluoric plus nitric acid.

Unetched

750X

Fig. 6 UAl_3 vs. U diffusion couple annealed at 600°C for 144³ hours under a pressure of 5 tsi.