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RATE OF GROWTH OF DIFFUSION LAYERS IN U-Al AND U-AlSi COUPLES

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CONTENTS

	<u>Page</u>
ABSTRACT	9
I. INTRODUCTION	11
II. EXPERIMENTAL PROCEDURE	15
A. Preparation of Specimen Materials	15
B. Annealing Procedure	15
C. Diffusion Measurement	16
III. RESULTS AND DISCUSSION	17
A. X-Ray and Metallographic Examination of Diffusion Zones	17
B. Variations of Diffusion with Time and Temperature	20
C. Relative Penetration	25
D. Pressure Effects	31
IV. CONCLUSIONS	32
V. SUMMARY	32
REFERENCES	33

ILLUSTRATIONS

<u>Figure</u>	<u>Title</u>	<u>Page</u>
1	Phase Equilibrium Diagrams for (A) U-Al, (B) U-Si, and (C) Partial Ternary for U-AlSi (KS-18188)	13
2	Typical Diffusion Layers for Various Temperatures In U-Al Couples (KS-18189)	18
3	Typical Diffusion Zones for U-AlSi (KS-18190)	19
4	Time Dependence of Diffusion Layer Thickness in U-Al Couples (KS-18191)	23
5	Time Dependence of Diffusion Layer Thickness in U-AlSi Couples (KS-18192)	24
6	U-AlSi Couples Annealed in Evacuated Pyrex Tubes (KS-18193).	26
7	Results of Present Investigation of Growth Rates of U-Al and U-AlSi Diffusion Layers (KS-18194)	27
8	Comparison of Results from All Investigations of U-Al Reaction Rates (KS-18195)	28
9	Comparison of Results from Investigation of U-AlSi Reaction Rates (KS-18196)	29
10	Relative Penetrations at Various Temperatures (KS-18197).	30
11	Pressure Effects on Growth Rate of U-Al Diffusion Layer (KS-18198)	31

ABSTRACT

The rates of growth of diffusion layers between uranium and aluminum and between uranium and AlSi alloy (eutectic composition) were studied and compared over the temperature range 250 to 550°C. Metallographic observation shows that the U-Al reaction layers form three to five times as fast as the U-AlSi layers over this temperature range. Sixty per cent of the layer growth takes place in the aluminum or AlSi alloy and forty per cent in the uranium.

RATE OF GROWTH OF DIFFUSION LAYERS IN U-AL AND U-ALSi COUPLES

L. S. DeLuca, H. T. Sumsion

I. INTRODUCTION

Interest in the diffusion characteristics of the U-Al system arises from the fact that uranium fuel slugs are sometimes enclosed in aluminum containers. The rate of reaction between these two metals is an important factor in determining the expected service life of the fuel slug. Similarly, since an aluminum-silicon alloy (eutectic composition) is sometimes used as a bonding agent and diffusion barrier between the uranium and aluminum, the U-ALSi reaction rates are also important.

The object of this investigation is to measure and compare the reaction rates of U-Al and U-ALSi under identical experimental conditions.

A review of the literature concerning recent investigations of the U-Al reaction rates shows approximately a factor of five difference between the greatest and smallest reported values. Presumably this is due mainly to the following conditions:

1. Composition differences in the Al and U stock used.
2. Variations in the amount of contaminants, such as oxides, present on the contact surfaces of the two metals.
3. Differences in the amount of pressure applied on the diffusion couples during the annealing period.

One report on U-ALSi reaction rates was found in the literature.

This previous work on the U-Al and U-ALSi systems will now be discussed in more detail. Bierlein¹ and Green² obtained extremely clean surfaces for their investigations by the cathodic etching technique. They report about twice as much diffusion with couples prepared in this manner as with chemically cleaned metals.

LeClair³ evaluated the pressure effect quantitatively and proposes certain relationships between pressure and reaction rate. According to his data there is about a 300% increase in the growth rate of the diffusion layers at 500°C when the pressure is increased from 1-1/4 to 10 tsi.

Murray⁴ reports that the rate of U-Al diffusion is not affected by the presence of grain boundaries or by the orientation of the annealed uranium grains. Vapor transport is not a significant factor for the U-Al diffusion reaction at least up to 450°C according to Bareis.⁵ Williams^{6,7} finds that small additions of Mg to the Al accelerate the reaction while Si, and especially Bi, inhibit it.

The identification and characteristics of the diffusion products are reported in detail by Kiessling⁸ and LeClair³. Both investigators found that all the phases (UAL₂, UAL₃, UAL₄) predicted by the U-Al equilibrium diagram, reproduced in Figure 1, were present in the diffusion zone. However, the relative thickness of each phase layer varied with temperature and pressure. The compounds UAL₂ and UAL₃ form adherent layers while the UAL₄ is porous and easily fractured.

All investigators have assumed that since there is a very small terminal solid solubility of U in Al or Si, and Al or Si in U, that a sufficient quantitative description of the interdiffusion behavior of these two metals can be given by the measurement of the rate of increase with temperature in the width of the visible diffusion zone, as defined by the phase boundaries of the pure metals.

The parabolic growth rate law of diffusion in the form

$$X = Kt^{1/2} \quad (1)$$

was assumed to be applicable by all investigators except LeClair and Murray. LeClair prefers the use of the more general equation

$$X = Kt^{1/n} \quad (2)$$

with n and K both varying with pressure. Murray used the linear law

$$X = Kt. \quad (3)$$

In all cases X represents the width of the diffusion band, t , the time at temperature, and K is the Arrhenius-type temperature-dependent coefficient defined as

$$K = K_0 \exp \left(-\frac{Q}{nRT} \right) \quad (4)$$

where n is the same as in Equation (2).

(A)

(B)

(C)

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FIG. 1 PHASE EQUILIBRIUM DIAGRAMS FOR
(A)U-Al, (B)U-Si AND (C)PARTIAL TERNARY FOR U-Al-Si

A summary of the U-Al and U-AlSi reaction rates determined by the various investigators is given in Table 1 and 2.

TABLE 1. Results of Previous Investigations of
Reaction Rates for the U-Al System

<u>Temperature Range, °C</u>	<u>Q, kcal/gm atom</u>	<u>K₀, inches/hr^{1/2}</u>	<u>Remarks</u>	<u>Reference</u>
200 to 390	14.3	0.93	2S Al, Surfaces cleaned by sputtering, 5 tsi initial pressure.	Bierlein
250 to 450	15.0	0.32	Canned fuel slugs used as diffusion couples.	Williams
540 to 620	16.0	-	High purity Al, annealed in air, max length of time at temperature 3 hr, 1-1/4 tsi continuous pressure	LeClaire
350 to 550	18.2	7.6	High purity Al, 6 tsi initial pressure.	Williams
400 to 550	19.3	94.0	2S Al, anneals of 2 min duration only, pressed powder uranium, 11 tsi continuous pressure	Storchheim
200 to 550	22.0	-	Bonded by dipping uranium in molten Al.	Murray
350 to 550	24.0	27.0	2S Al, 6 tsi initial pressure	Williams
250 to 450	24.0	25.0	2S Al	Bareis
400 to 550	29.4	480.0	2S Al, anneals of 2 min duration only, 11 tsi, continuous pressure	Storchheim

TABLE 2. Results of Previous Investigations of
Reaction Rates for the U-AlSi System

200 to 300	31.5	311.0	Diffusion of uranium into AlSi	Green
	29.7	82.1	Diffusion of AlSi into uranium	Green

II. EXPERIMENTAL PROCEDURE

A. Preparation of Specimen Materials

Diffusion couples were made up as three-layer sandwiches of Al, U, and AlSi. Each layer consisted of a cylindrical disk about 0.970 in. in diameter and 0.140 in. thick. The uranium disks were machined from alpha rolled stock (principal impurity 0.15% carbon) which had been previously given the following heat treatments:

1. Vacuum annealed at 600°C for 6 hr to expel dissolved gases, then furnace cooled.
2. 15-min anneal at 715°C in lead bath followed by water quench to obtain uniform grain size in all samples.

The aluminum disks were punched from 2S stock without further treatment. The AlSi alloy (11.7% Si) was vacuum cast into round bars and machined into disks.

The uranium surfaces were cleaned by abrading under water, first with No. 600 Silicon Carbide paper to ensure surface flatness, then passed twice over coarse No. 120 paper. The second pass was at right angles to the first so that the surface was covered by a cross-hatched series of tiny ridges and grooves. It was thought that when the disks were pressed together many of the ridges would be broken off, and allow the oxide-free uranium surface to come in contact with similarly oxide-free Al or AlSi surface. The Al and AlSi surfaces were wire-brushed immediately before use. All disks were washed in isopropyl alcohol and dried with a purified helium blast before being welded together. An inert marker was placed at the interface of each couple. The marker consisted of a strip of molybdenum foil approximately 0.030 in. wide, 0.0007 in. thick, and 0.900 in. long.

B. Annealing Procedure

The usual procedure in diffusion study is to bond the metals together in an appropriate manner, then in a separate operation carry out the annealing in a vacuum or inert atmosphere. In this investigation the three layer (Al, U, and AlSi) couples, described previously, invariably separated at the U-Al interface during the time at annealing temperature which made it impossible to get reliable penetration data. The U-AlSi part of these couples did not always separate during the annealing and allowed some penetration measurements which are reported.

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The most satisfactory method found for obtaining reproducible diffusion data for both the U-Al and U-AlSi couples was one in which the welding and annealing of the three-layer sandwich was carried out as one operation. The three disks to be bonded, Al, U, and AlSi, were placed in a 1-in.-ID tool-steel die between suitable punches which then was placed between the platens of a bench-type hydraulic press. Approximately 25 tsi were applied and while under this pressure the die was heated to about 200°C by means of a Calrod heating coil. As a result of this process the Al and AlSi disks were deformed sufficiently to encase the uranium virtually in an airtight envelope. The temperature of this pressing operation never exceeded 250°C and the elapsed time was approximately 1 hr. No diffusion was observed metallographically as a result of this operation.

After a couple was bonded and allowed to cool under pressure, the complete assembly consisting of the couple, die, die punches, and Calrod heater was transferred to an arbor press where a dead weight load equivalent to 2 tsi was applied to the couple. Since this pressure is slightly greater than the yield strength of Al at the lowest annealing temperature¹⁰ it was assumed that the metals remained in intimate contact during the diffusion period. The annealing treatments were carried out at temperatures ranging from 250 to 600°C for times ranging to 475 hr. By annealing in the arbor press the pressure applied on the couple was known to remain constant over the entire period at temperature.

Control of the temperature during the annealing was maintained within $\pm 5^\circ\text{C}$ by a potentiometer type controller in conjunction with a calibrated chromel-alumel thermocouple. The thermocouple was positioned in a hole drilled in the wall of the die at such a depth as to be about one-eighth of an inch away from the periphery of the diffusion couple. A second thermocouple was placed in the same hole and connected to a portable potentiometer for periodic checking of the temperature.

Several couples were annealed under a 25 tsi pressure at 250°C in order to determine qualitatively what effect additional pressure above 2 tsi has on the reaction rates of U-Al and U-AlSi.

C. Diffusion Measurements

On completion of the diffusion anneal, each specimen was mounted in a lucite holder and sectional normal to the interface. Measurements were made to the nearest one-eighth of an inch of the magnified image of the diffusion zone projected onto the screen of the metallograph. Magnifications of 100 to 250 diameters were used. The thickness of the diffusion layers, visible microscopically in the as-polished condition, were measured at points of maximum diffusion only. This procedure was adopted on the assumption that any lesser penetration was due to conditions which inhibited diffusion, such as poor bonding or oxide contamination of the interface.

III. RESULTS AND DISCUSSION

A. X-Ray and Metallographic Examination of Diffusion Zones

Uranium-Aluminum

In the U-Al system X-ray diffraction studies revealed that UAl_3 was the main diffusion product at all temperatures. Traces of UAl_4 and UAl_2 also appeared at 475°C and above, while at lower temperatures only traces of UAl_2 remained in addition to the UAl_3 . Figure 2 shows photomicrographs of typical U-Al diffusion layers. Metallographic observation generally revealed only the UAl_3 phase. Some couples which were parted at the interface between the diffusion zone and the aluminum contained traces of a black powder which was identified as UAl_4 .

Uranium-Aluminum-Silicon

In the U-AlSi couples the diffusion zone appeared metallographically as two layers. (See Figure 3.) A hardness survey of the zone of one couple confirmed this observation. However, X-ray diffraction studies indicated that both layers have a simple cubic structure with slightly ($<5\%$) different lattice parameters. A summary of the data is given in Table 3. The two layers which were observed in couples annealed at 400°C correspond to the known (see Figure 1) compounds USi_3 and UAl_3 . At 550°C , however, the observed lattice parameters have values which are intermediate between those of USi_3 (4.03A) and UAl_3 (4.28A). Since it is thought that these compounds are mutually soluble in the solid state,⁹ it is concluded that the two main diffusion products are solutions of these compounds. This conclusion is consistent with the findings of Kaufman and Cullity,⁹ who postulated a limited solubility between USi_3 and UAl_3 . If complete solubility had existed between these two compounds, then only a single layer of diffusion product should have been visible in the couples. The Ternary diagram proposed in the above references to illustrate this division of phases is shown in Figure 1.

TABLE 3. Results of X-Ray Diffraction Studies
of U-AlSi Diffusion Zone

Temperature, $^\circ\text{C}$	Time at Temperature, hr	Lattice Parameter $\overset{\circ}{\text{A}}$	
		Near AlSi Interface	Near U Interface
400	92	4.03	4.27
550	900	4.15	4.23

Fig. 2 Typical Diffusion Layers for Various Temperatures in U-Al Couples, at 50X Magnification

Top - 600°C
Center - 475°C
Bottom - 250°C

Fig. 3 Typical
Diffusion Zones
for U-AlSi

Top - 475°C 100X
Center - 325°C 1000X
Bottom - 250°C 1000X

In addition to the main diffusion products trace quantities of UAl_2 and USi were sometimes detected by X-ray diffraction in some of the diffusion zones. It should be noted that no evidence of the porous phase UAl_4 was found in any of the U-AlSi couples.

Void Formation

Voids were observed in both the U-Al and U-AlSi diffusion zones. In general the voids were largest in size at the highest annealing temperatures and were larger in the U-Al diffusion layers than the U-AlSi layers. In a few cases no voids were found in couples annealed at temperatures as high as $400^\circ C$. No correlation was found between the size and number of voids and the temperature, or time at temperature at which they were formed.

Williams,⁶ investigating the formation of blisters on aluminum jacketed fuel slugs, found that these voids contained various gases including hydrogen and nitrogen. At KAPL¹² it was noticed that if the uranium was vacuum annealed for 24 hrs or more at $600^\circ C$ no blisters formed during the Al jacketing operation. Although voids are known to form as the result of purely diffusion processes, the indications here are that the amount of dissolved gases in uranium could be a strong factor in their production.

B. Variation of Diffusion with Time and Temperature

X vs t The maximum width of the visible diffusion zone, as revealed metallographically, was measured and recorded along with the temperature and time at temperature for both the U-Al and U-AlSi systems. A summary of the data is given in Tables 4 and 5. It was found that the data for both systems fit satisfactorily the parabolic growth rate law, Equation (1), where X is the maximum width of the zone in inches, t is the time at temperature in hours, and K is the penetration coefficient in inches/hr^{1/2}. The plots of the data are shown in Figures 4 and 5. The slopes of the resultant straight lines, which were calculated by the method of least squares, were taken as equal to the penetration coefficients.

An exception was made for the lines which do not pass through the origin (U-Al- $250^\circ C$, U-AlSi- $325^\circ C$). It was assumed that in these cases the diffusion process was initially inhibited by oxide films at the interface. The time at temperature necessary for these films to be dissolved was assumed to be satisfactorily determined by extrapolating to $X = 0$ the straight line obtained on an X^2 vs t plot. After subtracting this indicated incubation period from the total time at temperature of each couple, a corrected curve was plotted from which the penetration coefficient was determined. The corrected curves are shown as dotted lines in Figures 4 and 5.

TABLE 4. Results of U-Al Reaction - 2 Tsi

Temperature, °C	Time at Temperature, hr	Maximum Thickness of Diffusion Layer, in. x 10 ⁻³
600	6	36.0
600	6	36.0
550	1	13.4
550	2	24.0
550	11	33.6
550	25	45.6
475	2	10.1
475	11	24.0
475	20	28.4
475	52	55.2
475	119	64.8
400	2	5.4
400	10	4.3
400	23	12.0
400	96	28.0
400	150	36.1
325	70	7.4
325	120	9.0
325	451	17.6
250	72	3.0
250	96	4.0
250	184	6.7
250	330	10.2
250	475	13.0

Results of U-Al Reaction - 25 Tsi

250	20	3.4
250	92	6.7
250	184	8.9

TABLE 5. Results of U-AlSi Reaction - 2 Tsi

Temperature, °C	Time at Temperature, hr	Maximum Thickness of Diffusion Layer, in. x 10 ⁻³
550	1	4.6
550	2	5.4
550	11	5.8
550	25	9.6
475	11	2.4
475	20	3.8
475	52	6.7
475	119	8.2
400	2	1.5
400	23	4.8
400	96	9.6
400	150	14.1
325	120	3.0
325	139	3.3
325	220	4.4
325	259	5.2
325	281	4.8
325	451	8.0
325	456	7.7
250	475	3.0

Results of U-AlSi Reaction - Vacuum Annealed

550	4	1.5
550	49	4.0
550	100	5.6
550	900	13.0
475	16	1.5
475	100	5.0
475	400	7.7
475	1600	7.9
400	100	2.5
400	400	5.0
400	1080	5.2
400	1600	6.0

FIG. 5 TIME DEPENDENCE OF
DIFFUSION LAYER THICKNESS IN U-ALSi COUPLES

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Figure 6 shows growth rate of those U and AlSi diffusion couples which had been hot pressed into couples and then annealed in evacuated glass tubes without pressure. These rates do not follow the parabolic rate law, probably because of the excessive number of voids which were observed at the interface of all couples. In addition the thickness of each diffusion layer is about one-third or less of that for a similar couple annealed under 2 tsi. No further analysis was made on these couples.

K vs 1/T The temperature dependence of the growth rate for both systems was assumed to be contained within the constant K defined in Equation (2). The log K was plotted against the reciprocal absolute temperature, Figure 7, from which the activation energy, Q, was calculated. The constant K_0 was computed in each case. All slopes were determined by the method of least squares. The data are described by the following relationships

$$\begin{array}{llll} \text{U-Al} & K = 3e^{-\frac{18,500}{2RT}} & \text{Inches/hr}^{1/2} & (1) \end{array}$$

$$\begin{array}{llll} \text{U-AlSi} & K = 0.07e^{-\frac{12,100}{2RT}} & \text{Inches/hr}^{1/2} & (2) \end{array}$$

A comparison of the values of K (thickness of diffusion layer at 1 hr) above shows that the U-Al layers form about three to five times faster than the U-AlSi layers over this temperature range. A graphical comparison between the values above and those of all previous investigations is shown in Figures 8 and 9.

C. Relative Penetration

The growth of a measured diffusion band was assumed to be the result of the penetration of aluminum into uranium and the simultaneous penetration of uranium into aluminum. The ratio of these penetrations was computed by measuring the width of the diffusion zone on each side of the original interface. At the higher temperatures where the diffusion band was continuous, the molybdenum marker was used as the reference position, while at the lower temperatures the original interface was undisturbed at many places and could be used directly.

A number of determinations of the ratio of the penetrations into the aluminum versus the penetrations into the uranium was made and found to have a mean value of 1.5 for all temperatures. That is, 60% of the diffusion band forms in the aluminum and 40% in the uranium; Figure 10 shows typical diffusion bands and molybdenum markers. It should be noticed that the ends of the markers bend into the aluminum indicating that aluminum diffuses faster than the uranium.

FIG. 6 U-ALSi COUPLES ANNEALED IN EVACUATED PYREX TUBES SHOWING SUBSTANTIALLY LOWER REACTION RATES THAN SIMILAR COUPLES ANNEALED AT 2 Tsi IN AIR, FIG. 5

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FIG. 7 RESULTS OF PRESENT INVESTIGATION OF GROWTH RATES OF U-AL AND U-ALSi DIFFUSION LAYERS

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FIG. 8 COMPARISON OF RESULTS, FROM ALL INVESTIGATIONS OF U-AL REACTION RATES

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FIG. 9 COMPARISON OF RESULTS FROM INVESTIGATION OF U-ALSi REACTION RATES

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Fig. 10 Relative Penetrations at Various Temperatures
 Top 550°C, Center 400°C, Bottom 250°C
 50X 50X 250X

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FIG. 11. PRESSURE EFFECT ON GROWTH RATE OF U-AL DIFFUSION LAYER

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In order to form a diffusion zone of roughly the same width on both sides of the interface, the aluminum must diffuse faster since the principal diffusion product is UAl_3 . The ratio of the AlSi-U penetrations was also found to be approximately 1.5 for all temperatures.

D. Pressure Effect

Three triple-layer couples were annealed successfully under a 25-tsi pressure at 250°C. The thickness of each U-Al diffusion layer is plotted versus the square root of time in Figure 11. No diffusion product was visible in the U-AlSi section of these couples. The data for U-Al couples annealed under 2 tsi at 250°C, shown previously, are also plotted on the above graph for purposes of comparison.

As may be seen in the above results the increased pressure on the U-Al couples results in an initial increase of about 0.003 in. in the cross-sectional width of the diffusion product. At longer annealing times the increase becomes progressively smaller. Therefore, it is concluded that for U-Al couples annealed at 250°C pressures in excess of 2 tsi do not affect the reaction rates significantly.

It should be noted, however, that as stated previously, the U-AlSi couples which were annealed under 2 tsi exhibited more than three times the thickness of diffusion product as that found in couples annealed without pressure at the interface. Also the number of voids observed in the diffusion zone of the pressure-annealed couples was always much smaller than that observed in the couples annealed without pressure. Therefore, it is tentatively suggested that pressure serves to increase the rate of reaction only by suppressing void formation and thus maintaining a constant cross-sectional area through which diffusion takes place.

IV. CONCLUSIONS

The results of this investigation show definitely that the U-AlSi reaction layers form at a lower rate than the U-Al reaction layers in the 325 to 550°C temperature range. In addition the formation of the compound UAl_4 is suppressed in the U-AlSi reaction. This is important because this compound does not form an adherent layer in the U-Al reaction but rather cracks to a powder and gives a bad contact zone.

The degree of agreement between the results of the various investigations of the U-Al system is considered good. The differences appear to be due to large variations in experimental conditions (given in Table 1) under which the different programs were carried out. The lack of agreement in the U-AlSi results is particularly significant at temperatures above 350°C. It can be shown that in this temperature range the results obtained at Hanford² indicate a greater layer growth rate in the U-AlSi couples than in similarly prepared U-Al couples. This is in direct opposition to the findings of the present investigation.

V. SUMMARY

The rates of reaction between uranium and aluminum and between uranium and AlSi alloy were studied and compared over the temperature range 250 to 550°C. Measurement by metallographic observation of the progressive growth

of the diffusion layers as a function of time at temperature produced data which for both systems is satisfactorily described by the relation

$$X = Kt^{1/2}$$

where K = penetration coefficient
 X = width of diffusion zone
 t = time at temperature.

The following equations best represent the data;

U-Al

$$K = 3.0 \exp (-18,500/2RT) \text{ inches/hr}^{1/2}$$

U-ALSi

$$K = 0.07 \exp (-12,100/2RT) \text{ inches/hr}^{1/2}.$$

A comparison of the reaction rates shows that a U-Al diffusion layer increases in thickness from three to five times faster than a comparable U-ALSi reaction layer in this temperature range.

Sixty per cent of the layer growth takes place in the aluminum or AlSi alloy and forty per cent in the uranium.

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