An Analysis of Creep Behavior of Nickel - Copper Laminate Composites

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Abstract

The tensile creep behavior of a series of nickel – copper laminate composites, in which the composition gradients between the $20 - \mu m$ layers were controlled by heat treatment, was evaluated at $500\,\mathrm{C}$. With annealing, the high – temperature creep resistance increased due to the direct effects of solid solution strengthening within the layers. The results correlated to predictions based on a composite model which approximated the continuous composition gradients as a series of discrete compositional zones.

1. Introduction

The high - temperature creep behavior of inhomogeneous materials which possess fine scale microstructural and compositional gradients reflects the composite effects of local variations in creep resistance within the gradients. Inhomogeneous materials include solidified weld metal and castings, welded structures, materials with multiphase microstructures, and composites. In as - solidified microstructures, local composition gradients, with a dimensional scale on the order of the interdendritic arm spacing, are controlled by the partition coefficient for the particular alloy system. In welded structures, transitions in microstructure from the solidified weld metal, through the heat - affected zone, and into the base metal develop as a result of the thermal history associated with welding. In contrast to materials discussed above with compositions and/or microstuctures which vary continuously with position, composites, both artificial or natural(e. g., directionally solidified eutectics), exhibit abrupt changes in properties between constituents.

Creep of inhomogeneous materials depends on the magnitude of the local gradient. Several studies¹⁻³⁾ have used specialized techniques, such as the impression creep test^{2,3)}, to directly measure the creep behavior within a gradient. However, the majority of creep tests still evaluate the effects of gradients on overall properties. For example, in a recent study of the effects of postweld heat treatment on the creep behavior of MONEL alloy 400 wld metal¹⁾, the minimum creep rate decreased and the rupture life increased with heat treatment. The heat treatment,

which tended to homogenize the solidification segregation, increased the creep resistance. It was hypothesized that the creep resistance of weldments and other inhomogeneous materials could be evaluated based on composite modeling techniques, if the position dependent creep resistance within the gradient is known.

Theoretical treatments of high - temperature creep in inhomogeneous systems have primarily involved the application of standard composite modeling techniques, e. g., rule of mixtures for composites subjected to an isostrain loading condition, in which the strengths of the indivedual constituents are expressed as strain rate dependent parameters. Efforts have been made⁵⁻⁷⁾ to model the properites of actual solidified composite structures, such as directionally solidified superalloys and eutectic composites, with composite creep theory. The effects of fiber radius and phase stability on the creep properties have been, to this point, the primary concern. McLean and Quested" pointed out that attempts to rationalize the mechanical behavior of directionally solidified superalloys must take into account the heterogeneities in composition and structure and suggested the possibility of modeling the properties of dendritic structures. However, until now, modeling of the dendritic structure has not been attempted. Therefore, in this study, the creep behavior of a composite material designed to model cast dendritic structures is presented.

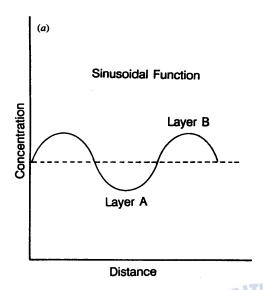
2. Experimental design and theoretical treatment of composite creep within a composition gradient

The effects of variations in local composition gradients on the creep behavior of laminate composites were evaluated on a series of

nickel - copper laminates with 20 - μm - thick layers. The layer dimension was chosen to simulate interdendritic arm spacings in solidified microstructures, as illustrated by MONEL alloy 400 cast weld metal¹⁾. The nickel - copper system was chosen as it exhibits complete solubility and the creep behavior of nickel - copper solid solution alloys is known at the test temperature of interest8). Sheet tensile samples were annealed in a hot isostatic press(HIP) for various times to produce controlled variations in the composition gradients between layers. The HIP treatment also suppressed Kirkendall porosity formation⁹⁻¹³⁾. The laminate composite creep data are intrpreted based on the predictions of composite models modified to incorpoate the effects of gradients.

The composition profiles of heat - treated laminate composites can be approximated by a sinusoidal function, as shown in Figure 1. As described by Flemings¹⁴⁾, during homogenization, the amplitude of a composition gradient decreases, but the wavelength is unchanged with heat treatment. Clearly, the properties will change continuously with position across the gradient. However, to aid in simplifying the composite analysis, the continuous sinusoidal gradient can be approximated as a series of discrete steps, each of which can be treated as an element of a composite and as a specific alloy. The number of steps chosen to approximate the sinusoidal function determines the sensitivity of the following composite analysis. For simplicity, in the range of one half wavelength, the sinusoidal function can be approximate by averaging to produce the step function shown in Figure 1. Each layer of the step function in Figure 1 can be treated as an element in a two-component laminate composite. With these assumptions, the creep rate of laminate composites can be predicted.





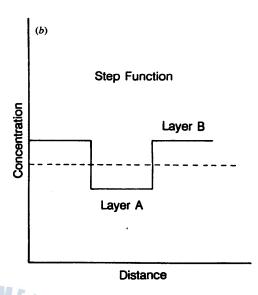


Fig. 1. Schematic illustration of approximation for local composition gradients in an annealed laminate composite: (a) sinusoidal function and(b) step function.

The creep behaviors of laminate and fiber composites subjected to isostrain loading conditions are equivalent and can be predicted with a consideration of the following assumptions:

5,15-17)

(1) In steady state, the creep rates of the composite (identified by c), layer A, and layer B are equal:

$$\varepsilon_c = \varepsilon_A = \varepsilon_R \tag{1}$$

(2) Creep of layers A and B in the composite can each be described by a unified power law creep equation obtained from creep – rate($\dot{\epsilon}$) data on single – phase samples. The appropriate equation is

$$\dot{\varepsilon} = \beta \sigma^n \exp(-Q/RT) \tag{2}$$

where σ is the applied stress, R is the gas constant, T is the absolute temperature, Q is the activation energy for creep, and β and n are constants.

(3) For materials, such as nickel and copper, which exhibit similar creep properties, interfa-

cial strengthening effect5 can be ignored.

If the equilibrium of forces is considered, the stress distribution on each component of the composite can be described¹⁸⁾ by the rule of mixtures given in Eq.[3] at steady state:

$$\sigma_c = \sigma_A V_A + \sigma_B V_B \tag{3}$$

where σ_{c} = stress in the composite;

 $\sigma_A = \text{stress in layer A}$;

 σ_{R} = stress in layer B;

 V_A =volume fraction of layer A; and

 V_B =volume fraction af layer B.

From assumption 2, the stress in each phase at a strain rate, $\dot{\varepsilon}$, can be written as follows:

$$\sigma_A = K_A \dot{\varepsilon}^{1/a}$$
 on layer A
 $\sigma_B = K_B \dot{\varepsilon}^{1/b}$ on layer B (4)
where $K_A = [\beta_A \exp(-Q_A/RT)]^{-1/a}$
 $K_B = [\beta_B \exp(-Q_B/RT)]^{-1/b}$

The terms K_A and K_B in Eq.[4] are temperature – dependent parameters. The values of K_A and K_B may be calculated from the creep properties of the components tested independent



dently. Substituting Eq.[4] into Eq.[3] gives

$$\sigma_c = V_A K_{A \varepsilon}^{\nu_a} + V_B K_B \varepsilon^{\nu_b} \tag{5}$$

If for materials with similar deformation behaviors the strain-rate exponents in Eq.[4] can be assumed to be equal for both layers A and B(i.e., a=b=n), then the creep rate of the composite at a given creep stress can be predicted by Eq.[6], where the constant n replaces the individual strain-rate exponents in Eq.[4].

$$\dot{\varepsilon} = \frac{\sigma_c^n}{\left[K_A V_A + K_B V_B\right]^n} \tag{6}$$

If the sinusoidal function is approximated by multiple discrete steps(*i.e.*, greater than the two steps illustrated in Figure 1(b)), then the analysis above can be modified by altering Eq.[3] as

$$\sigma_c = \sigma_A V_A + \sigma_B V_B + \dots + \sigma_i V_i \tag{7}$$

where A, B, \cdots *i* represent the individual layers in the approximation. If the creep equation for each component *i* is known according to Eq.[2], then direct substitution into Eq.[7] will yield an overall prediction from the composite creep behavior. However, it should be noted that with multiple zones with different *n* values, a simple analytical form similar to Eq.[6] is not available, and appropriate solution techniques must be applied directly to Eq.[7].

3. Experimental procedure

Nickel – copper laminate composites were fabricated according to the procedure outlined in Figure 2 from sheets of nickel 270 and copper $C102^{19}$. The compositions of these materials are given in Table I. Nickel and copper sheets were hot – rolled and cold – rolled to produce thin foils with dimensions of $0.4 \times 100 \times 150$ mm. Prior to cold rolling the surfaces were cleaned

with a solution of 50 pct nitric acid and 50 pct water. Forty foils of each material were stacked alternately to produce a 32 – mm – thick compact. The compact was encased in a welded stainless steel can with a stainless steel tube which was connected to a vacuum system during annealing. Prior to hot rolling, the stainless steel can was preheated at 870°C for 30 minutes. The compact was hot – rolled in five passes to a

Table 1. Chemical compositions of Ni 270 and C102 Cu(Weight Percent)

	Ni 27	70*	C102 C	u**
	>99.97	Ni	>99.98	Cu
	< 0.001	Cu	< 0.005	Fe
	< 0.005	Fe	< 0.005	Pb
	< 0.001	$\mathbf{M}\mathbf{n}$	< 0.003	Ag
F //	< 0.02	C	< 0.004	P
· (/)	< 0.001	Si	<10ppm	O_2
	< 0.001	s		
	< 0.001	Co		
	< 0.001	Cr		
	< 0.001	Mg		
	< 0.001	Ti		

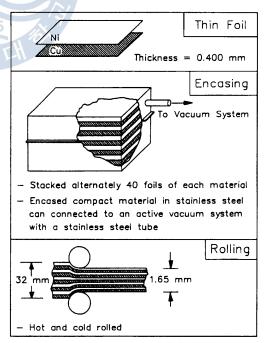


Fig. 2. Manufacturing procedures to produce nickel - copper laminate composites.



thickness of 19 mm; the can was removed; the material was reheated and hot – rolled to a thickness of 3.8 mm. The sheet was cold – rolled to a final thickness of 1.65 mm. The cold – rolled sheet consisted of alternating layers of $20 - \mu m$ – thick nickel and copper.

Tensile specimens with a reduced gage section of $27.0 \times 6.35 \times 1.65$ mm were machined with the tensile axis parallel to the rolling direction. To prepare specimens with different composition gradients, tensile specimens were heat - treated in an HIP at 1000° for times between 30 minutes and 4 hours under a pressure of 172MPa. Tensile creep tests were performed at 500°C under constant load conditions with an initial stress of 17.2 MPa. All tests were performed on a servohydraulic test system with an attached inert atmosphere glove box200 which contained the furnace and associated pull rods. Linear displacement was measured continuously, and the creep tests were carried out until rupture or for a minimum duration of 200 hours.

Specimens for optical microscopy were prepared by standard metallographic techiques and etched for 5 to 20 seconds in a 50 pct nitric acid and 50 pct acetic acid solution. Composition profiles were measured using energy dispersive spectroscopy(EDS) and wavelength dispersive spectroscopy(WDS) techniques with a JEOL JXA 840 scanning electron microscope(SEM) equipped with a TRACOR NORTHERN microanalyzer.

4. Results and discussion

A. Influence of Heat Treatment on Composition Profiles

The microstructure of the as – rolled nickel – copper laminate composite, shown in Figure 3, reveals that each layer has a similar thickness of approximately $20\mu m$ and a distinctive interface. Light micrographs of sample heat treated

in an HIP at 1000° for times between 30 minutes and 4 hours are shown in Figure 4. The heat treatment which promotes interdiffusion of the copper and nickel increased the apparent thickness of the copper layer. Furthermore, as revealed by the etching, the distinct interface was less apparent with an increase in heat treatment time. With annealing, grain growth occurs to produce grains which cross the layers. For all samples shown in Figure 4, the grain size was uniform and approximately 55 µm. It should be noted that in the creep study described below, deformation within the layers is controlled by the composition variations within the 20μm layers and not by the significantly larger grain size.

Figure 5 presents SEM micrographs of etched metallographic samples after heat treatment for 30 minutes and 1 hour at 1000°C Wavelength dispersive spectroscopy compositional line scans for nickel and copper are superimposed on each micrograph and show the variation in composition with position for each element. The profiles are approximately sinusoidal, except that due to diffusivities, the waveshapes differ in the nickel – rich and copper – rich regions. The peak amplitudes decrease with an increase in heat – treatment time.

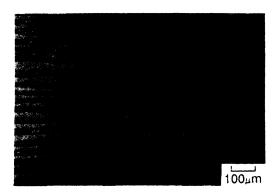


Fig. 3. Light micrograph of the as – rolled laminate composite. The copper appears as the dark layer. Etchant: nitric – acetic.



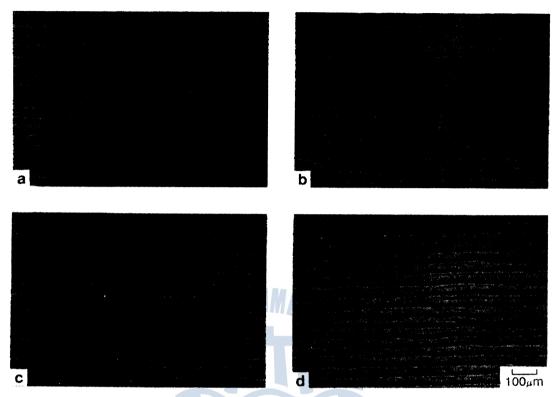


Fig. 4. Light micrographs of laminate composites heat - treated at 1000°C under a pressure of 172 MPa for(a) 30min, (b) 1h, (c) 2h, and (d) 4 h. Etchant: nitric - acetic.



Fig. 5. Wavelength dispersive spectroscopy composition profiles of the laminate composites heat – treated at 1000 $^{\circ}$ for (a) 30 min and (b) 1h.

Results of quantitative EDS analysis of the nickel and copper peak compositions in each layer are shown in Figure 6. With annealing at 1000°C, significant composition changes are observed in the first two hours. The compositions of each layer remain essentially constant with continued annealing. These results show

that the composition profiles of laminate composites were systematically changed by heat treatment under the high pressure.

B. Influence of composition Gradients on Creep Properties

Complete creep curves obtained at 500°C and



17.2 MPa for the as-rolled specimen and the specimen heat-treated for 30 minutes, along with partial curves for the specimens heat-treated longer than 30 minutes, are shown in Figure 7. The lack of a well-defined primary creep region for the sample annealed for 0.5

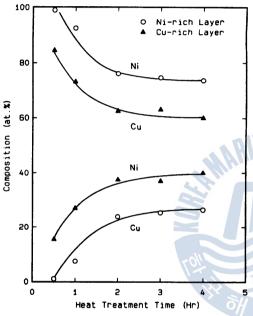


Fig. 6. The effects of heat - treatment time on the peak compositions, obtained with EDS, of each layer of the nickel - copper laminate composite.

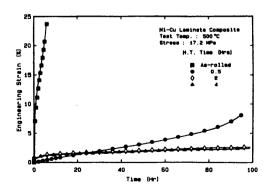


Fig. 7. The effect of heat treatment(1000°C) on the creep curves for laminate composites tested in an argon atmosphere at 500°C and 17.2 MPa.

hour may reflect that with annealing, the apparent effective solid solution alloying classes²¹⁾ change with average layer composition.

All creep curves exhibited a distinctive sec-

Table I. Constant Load Creep Data for Nickel – Copper Laminate Composite Tested at 17.2 MPa and 500℃

Heat – Treatment Time(h)	Grain Size(µm)	<i>ε</i> (s⁻¹)	
0	N/A*	7.22×10 - 6	
0	N/A	$4.83 \times 10 - 6$	
0.5	55	$1.67 \times 10 - 7$	
0.5	· 55	$1.31 \times 10 - 7$	
1	57	$6.10 \times 10 - 8$	
1	57	$4.20 \times 10 - 8$	
2	58	$3.10 \times 10 - 8$	
2	58	$2.50 \times 10 - 8$	
3	58	$2.70 \times 10 - 8$	
3	58	$2.80 \times 10 - 8$	
4	57	$2.90 \times 10 - 8$	
4	57	$2.20 \times 10 - 8$	

^{*}N/A=not applicable.

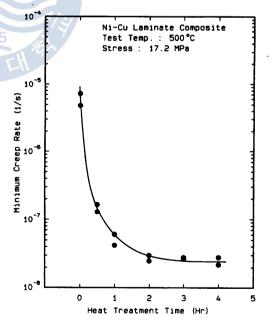


Fig. 8. The relationship between the minimum creep rate and heat – treatment time at 1000 °C for nickel – copper laminate composites tested in an argon atmosphere at 500 °C and 17.2 MPa.



ondary creep region. The minimum creep rates (expressed as engineering strain rates), \dot{e} , are summarized in Table \parallel and plotted in Figure 8 as a function of heat – treatment time. The effect of heat – treatment time on the creep rate is similar to that on the peak nickel atomic percent in the nickel – rich layer, as shown in Figure 6. This comparison shows that the minimum creep rates of laminate composites are significantly affected by the composition change in materials, as there is a greater than two order of magnitude change in strain rate.

The results reveal that the minimum creep rates of laminate composites decreased with increases in heattreatment time, that is, by reducing the composition gradients. A comparison of Figures 6 and 8 shows that the decrease in creep rates with annealing mirrors the composition changes within the layers. Thus, the improvement in the composite creep resistance with annealing reflects the effects of solid solution strengthening within each of the layers.

To illustrate the applicability of the two – component approximation discussed above to the prediction of the composite creep resistance of the nicel – copper laminate composites with continuous composition gradients, consider the following analysis. First, assume that the continuous composition profiles shown in Figure 5 can be approximated by the two – component step function of Figure 1(b). The average nickel

compositions in the nickel – rich layer and copper – rich layer are shown in Table

and were calculated by multiplying by 0.65 and 0.9, respecitively, to the peak nickel composition. The two averaging factors were used to compensate for differences in the shape of the composition profiles in the nickel – rich and copper – rich regions.

Second, assume that the true stress – true stain rate analysis presented in Eqs.[1] through [7], can be applied to the constant load – engineering strain rate results of this study. To evaluate the significance of this assumption, consider the following analysis. The engineering strain rate(e) analogue of Eq.[6] is

$$\dot{e} = \frac{S^{n}(1+e)^{n+1}}{[K_A V_A + K_B V_B]^n}$$
 [8]

where S and e are engineering stress and engineering strain, respectively. As shown in Figure 7, for most samples in this study, minimum creep rates were obtained over strain ranges less than 0.05. Thus, for n=7 and with $S=\sigma$, the ratio of ε to e is less than approximately 1.4, a value which is insignificant with respect to the significant variation in minimum creep rate shown in Figure 8. Therefore, the assumption that the composite analysis in the Introduction applies to the results of this study is valid.

For the compositions shown in Table II, the creep behaviors of homogeneous solid solution

Table ■. The Predicted Creep Rates of Nickel - Copper Laminate Composites Using the Rule of Mixtures

	Ni Composition(At. Pct)		$\mathbf{K}^{\mathbf{a}_{i}}$		
Heat - Treatment	Ni – Rich	Cu – Rich	Ni – Rich	Cu - Rich	- (- 1)
$Time(h)^*$	Layer	Layer	Layer	Layer	$\varepsilon_s(s-1)$
0.5	81.9	18.9	210.7	102.7	1.9× 7
1	77.6	29.3	218.0	148.6	6.4× -8
2	66.9	38.7	144.8	182.8	3.1× 8
3	65.9	38.3	144.8	181.7	3.1× +8
4	65.3	41.0	144.8	189.3	2.8× -8

^{*}All samples were heat - treated at 1000°C



alloys are available from the work of Choi et al^{s_1} . The effects of alloy content on the grain size – independent creep rates for samples tested at 500°C and 17.2 MPa are shown in Figure 9. A minimum in the creep rate is observed with alloy content. Furthermore, the K values in Eq.[4] were determined for each alloy and then also summarized in Table \mathbb{II} . With n=7, a value which approximates the behavior of the nickel – copper solid solution alloys of Choi et al., $\alpha=17.2$ MPa, and $V_A=V_B=0.5$, Eq.[6] can be used to predict the creep rates of the laminate composites as a function of annealing time. The resulting predicted creep rates are also included in Table \mathbb{II} .

To illustrate how the creep data of laminate composites in Table II were predicted, consider the following example for creep in a laminate composite heat – treated for 1 hour. From Figure 9, the creep rates for the nickel – rich(77.6

pct Ni) and copper – rich(29.3pct Ni) regions can be directly read from the graph and are 1.9 $\times 10^{-8}$ and 2.8×10^{-7} s⁻¹, respectively. Then, K values can be obtained by using Eq.[4].

$$K(\text{in Ni-rich layer}) = 17.2/(1.9 \times 10^{-8})^{1/7} = 218.0$$

 $K(\text{in Cu-rich layer}) = 17.2/(2.8 \times 10^{-7})^{1/7} = 148.6$

By substituting these values in Eq.[6], the creep rate predicted with the rule of mixtures in laminate composite is

$$\dot{\varepsilon} = \left[\frac{17.2}{0.5(218.0 + 148.6)}\right]^{7} = 6.4 \times 10^{-8} \text{ s}^{-1}$$

The creep rates of other laminate composites were also calculated with the same method. Because the stress exponent(n=7) and the averaging factors for the composition in each layer for the calculation were representative values for all alloys, the sensitivity of the analysis to variations in n and the composition – averaging

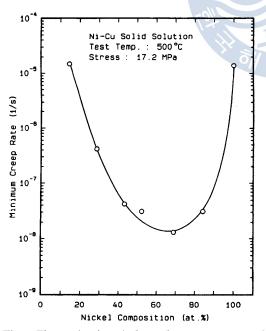


Fig. 9. The grain size - independent creep rates of nickel - copper solid solution alloys".

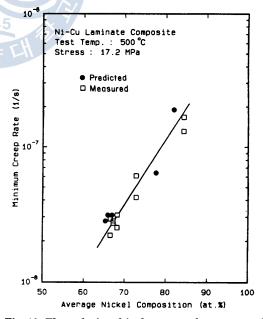


Fig. 10. The relationship between the measured and predicted creep rate of nickel - copper laminate composites tested in an argon atmosphere at 500 °C and 17.2 MPa.



factors was considered. By changing n from 5.5 to 9 and averaging factors 0.65 to 0.7 and 0.9 to 0.95, the creep rates predicted by the rule of mixtures were within \pm 10 pct. Therefore, the creep rates for the laminate composites in Table \mathbb{I} are reasonable.

Both the predicted(Table **I**) and measured (Figure 8) creep rates from the nickel-copper laminate composites are plotted in Figure 10 vs average nickel composition in the nickel-rich layers. The results show that there is good agreement between the predicted and the measured creep rates. Therefore, it is concluded that the creep rate of the lamminate composites with composition gradients can be predicted by the modified rule of mixtures procedure outlined above. The excellent agreement between measured values and those based on the rule of mixtures indicates that interfacial strengthening between layers may be negligible in systems with continuous composition gradients. The two-component approximation to the continuous composition gradient between layers illustrates a procedure which could be extended to multicomponent approximations of materials with continuous composition gradients. Finally, based on this composite study, the improvement with postweld heat treatment of the high temperature creep resistance of MONEL alloy 400 weld metal¹⁾ is interpreted to reflect changes in the solid solution strengthening between the dendrites and interdendritic regions in a manner directly analogous to the composite considered here.

5. Conclusions

This study on nickel – copper laminate composites, in which the composition gradients between layers were systematically controlled by heat treatment, yielded the following primary

conclusions:

- The creep resistance of laminate composites increased with annealing due to the direct contributions of solid solution strengthening within the layers.
- 2. The predicted minimum creep rates of laminate composites, based on the creep rates of solid solutions and the rule of mixtures for the composite creep, showed good agreement with measured creep rates. This result indicates that the creep rate of laminate composites with composition gradients can be predicted by the rule of mixtures for composite creep when the continuous composition or microstructural gradients are approximated by a series of discrete zones.
- 3. Results based on laminate composites provide a basis for interpreting the deformation behavior of solidified weld metal and other materials with composition or microstructural gradients.

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