[redacted]

ORNL-2065 cy. 122
C-84 – Reactors–Special Features of Aircraft Reactors

AEC RESEARCH AND DEVELOPMENT REPORT

CENTRAL RESEARCH LIBRARY DOCUMENT COLLECTION

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES
3 4456 0350112 8

HIGH-THERMAL-CONDUCTIVITY FIN MATERIAL FOR RADIATORS

H. Inouye

DECLASSIFIED
CLASSIFICATION CHANGED TO
BY AUTHORITY OF AEC 10/25/62
BY R. H. Bridges 7/31/63

CENTRAL RESEARCH LIBRARY
DOCUMENT COLLECTION X-10
LIBRARY LOAN COPY
DO NOT TRANSFER TO ANOTHER PERSON
If you wish someone else to see this document, send in name with document and the library will arrange a loan.

OAK RIDGE NATIONAL LABORATORY
OPERATED BY
UNION CARBIDE NUCLEAR COMPANY
A Division of Union Carbide and Carbon Corporation
UCC
POST OFFICE BOX X · OAK RIDGE, TENNESSEE

[redacted]

LEGAL NOTICE

This report was prepared as an account of Government sponsored work. Neither the United States, nor the Commission, nor any person acting on behalf of the Commission:

A. Makes any warranty or representation, express or implied, with respect to the accuracy, completeness, or usefulness of the information contained in this report, or that the use of any information, apparatus, method, or process disclosed in this report may not infringe privately owned rights; or

B. Assumes any liabilities with respect to the use of, or for damages resulting from the use of any information, apparatus, method, or process disclosed in this report.

As used in the above, "person acting on behalf of the Commission" includes any employee or contractor of the Commission to the extent that such employee or contractor prepares, handles or distributes, or provides access to, any information pursuant to his employment or contract with the Commission.

[redacted]

ORNL-2065

C-84 – Reactors–Special Features of Aircraft Reactors

Contract No. W-7405-eng-26

METALLURGY DIVISION

HIGH-THERMAL-CONDUCTIVITY FIN MATERIAL FOR RADIATORS

H. Inouye

The investigation covered in this report was conducted between November 1952 and August 1954

DATE ISSUED

JAN 2 1957

OAK RIDGE NATIONAL LABORATORY
Operated by
UNION CARBIDE NUCLEAR COMPANY
A Division of Union Carbide and Carbon Corporation
Post Office Box X
Oak Ridge, Tennessee

[redacted]

MARTIN MARIETTA ENERGY SYSTEMS LIBRARIES

3 4456 0350112 8

[redacted]

ORNL-2065
C-84 – Reactors–Special Features of Aircraft Reactors

INTERNAL DISTRIBUTION

1. R. G. Affel
2. C. J. Barton
3. M. Bender
4. D. S. Billington
5. F. F. Blankenship
6. E. P. Blizard
7. C. J. Borkowski
8. W. F. Boudreau
9. G. E. Boyd
10. M. A. Bredig
11. W. E. Browning
12. F. R. Bruce
13. A. D. Callihan
14. D. W. Cardwell
15. C. E. Center (K-25)
16. R. A. Charpie
17. C. E. Clifford
18. J. H. Coobs
19. W. B. Cottrell
20. D. D. Cowen
21. S. Cromer
22. R. S. Crouse
23. F. L. Culler
24. J. H. DeVan
25. L. M. Doney
26. D. A. Douglas
27. E. R. Dytko
28. W. K. Eister
29. L. B. Emlet (K-25)
30. D. E. Ferguson
31. A. P. Fraas
32. J. H. Frye
33. W. T. Furgerson
34. E. J. Breeding
35. W. R. Grimes
36. E. Guth
37. E. E. Hoffman
38. H. W. Hoffman
39. A. Hollaender
40. A. S. Householder
41. J. T. Howe
42. W. H. Jordan
43. G. W. Keilholtz
44. C. P. Keim
45. M. T. Kelley
46. F. [redacted]
47. E. M. King
48. J. A. Lane
49. R. B. Lindauer
50. R. S. Livingston
51. R. N. Lyon
52. F. C. Maienschein
53. W. D. Manly
54. E. R. Mann
55. L. A. Mann
56. W. B. McDonald
57. F. R. McQuilkin
58. R. V. Meghreblian
59. R. P. Milford
60. A. J. Miller
61. R. E. Moore
62. J. G. Morgan
63. K. Z. Morgan
64. J. P. Murray (Y-12)
65. M. L. Nelson
66. G. J. Nessle
67. R. B. Oliver
68. L. G. Overholser
69. P. Patriarca
70. R. W. Peelle
71. A. M. Perry
72. J. C. Pigg
73. P. M. Reyling
74. A. E. Richt
75. M. T. Robinson
76. H. W. Savage
77. A. W. Savolainen
78. R. D. Schultheiss
79. E. D. Shipley
80. A. Simon
81. O. Sisman
82. J. Sites
83. M. J. Skinner
84. G. P. Smith
85. A. H. Snell
86. C. D. Susano
87. J. A. Swartout
88. E. H. Taylor
89. R. E. Thoma
90. D. B. Trauger
91. G. M. Watson
92. A. M. Weinberg
93. J. C. White
94. G. D. Whitman
95. [redacted] (consultant)
96. G. C. Williams

[redacted]

[redacted]

97. J. C. Wilson
98. C. E. Winters
99-100. ORNL – Y-12 Technical Library
Document Reference Section

101-120. Laboratory Records Department
121. Laboratory Records, ORNL R.C.
122-123. Central Research Library

EXTERNAL DISTRIBUTION

124. AF Plant Representative, Baltimore
125. AF Plant Representative, Burbank
126. AF Plant Representative, Marietta
127-129. AF Plant Representative, Santa Monica
130-131. AF Plant Representative, Seattle
132. AF Plant Representative, Wood-Ridge
133. Air Materiel Area
134. Air Research and Development Command (RDGN)
135. Air Technical Intelligence Center
136. Allison Division
137-139. ANP Project Office, Fort Worth
140. Albuquerque Operations Office
141. Argonne National Laboratory
142. Armed Forces Special Weapons Project, Sandia
143. Armed Forces Special Weapons Project, Washington
144. Assistant Secretary of the Air Force, R&D
145-150. Atomic Energy Commission, Washington
151. Battelle Memorial Institute
152-153. Bettis Plant (WAPD)
154. Bureau of Aeronautics
155. Bureau of Aeronautics (Code 24)
156. Bureau of Aeronautics General Representative
157. Chicago Operations Office
158. Chicago Patent Group
159. Chief of Naval Research
160. Convair-General Dynamics Corporation
161. Engineer Research and Development Laboratories
162-165. General Electric Company (ANPD)
166. Hartford Area Office
167. Headquarters, Air Force Special Weapons Center
168. Idaho Operations Office
169. Knolls Atomic Power Laboratory
170. Lockheed Aircraft Corporation (Richard G. Rowe)
171. Lockland Area Office
172. Los Alamos Scientific Laboratory
173. Mound Laboratory
174. National Advisory Committee for Aeronautics, Cleveland
175. National Advisory Committee for Aeronautics, Washington
176. Naval Air Development and Material Center
177. Naval Research Laboratory
178. New York Operations Office
179. North American Aviation, Inc. (Aerophysics Division)
180. North American Aviation, Inc. (Canoga Park)

[redacted]

[redacted]

181. Nuclear Development Corporation of America
182. Office of the Chief of Naval Operations (OP-361)
183. Patent Branch, Washington
184-187. Pratt & Whitney Aircraft Division (Fox Project)
188. Sandia Corporation
189. School of Aviation Medicine
190. Sylvania Electric Products, Inc.
191. USAF Project RAND
192. University of California Radiation Laboratory, Livermore
193-210. Wright Air Development Center (WCOSI-3)
211-235. Technical Information Service Extension, Oak Ridge
236. Division of Research and Development, AEC, ORO
237. Technical Research Group, New York

[redacted]

[redacted]

CONTENTS

Abstract 1

Introduction 1

Survey of Literature 1

Plan of Investigation 1
- Plates 3
- Copper Alloys 3
- Clad Copper 3

Results and Discussion 3
- Plates 3
- Copper Alloys 5
- Clad Copper 7

Conclusions 17

[redacted]

HIGH-THERMAL-CONDUCTIVITY FIN MATERIAL FOR RADIATORS

H. Inouye

ABSTRACT

This report is the result of a study to develop heat-resistant fin materials possessing a high thermol conductivity for air radiators. Since an economical and commercially feasible product was desired, the investigation was restricted primarily to a study of electroplated copper, clad copper, and copper alloys.

Sheet material 0.008 to 0.010 in. thick was evaluated for fabricability and for metallurgical stability and thermal conductivity at 1500°F. From the results of the tests it was concluded that: (1) electroplates were unsatisfactory; (2) clad-copper fins possessing a thermal conductivity of 50% of that of copper are commercially feasible; (3) copper-aluminum alloys possessing a thermal conductivity approaching that of copper at 1500°F are possible. Service tests of clad copper and the copper-aluminum alloys indicate that the choice of materials will be dictated by the requirements of the radiator, since each presents some unique problems.

INTRODUCTION

This investigation was undertaken to develop high-thermal-conductivity fin material for use in a high-velocity air stream between 1300 and 1650°F. Under the conditions of the intended use, the fin material should be heat resistant, metallurgically stable, compatible with Inconel tubing, and capable of being rolled into sheet or strip 0.008 to 0.010 in. thick. Since large quantities of the material are required, an economical and commercially feasible product is desirable.

SURVEY OF LITERATURE

The previous work most applicable to the problem was that of Haythorne,[1] who investigated the thermal stability of numerous heat-resistant alloy sheet materials heated between 1500 and 2000°F followed by a 1-min air cool. Material failures resulted from poor thermal conductivity and high thermal expansion, which are characteristic of heat-resistant alloys. Of the numerous composites and alloys tested, Inconel-clad copper was the most promising; however, the above author failed to report the ratio of copper to Inconel in the composite. The thermal-conductivity data taken from this report are shown in Fig. 1.

[1] P. A. Haythorne, *Iron Age* 162(13), 89–95 (1948).

PLAN OF INVESTIGATION

Of the several metals listed below which might be used for high-thermal-conductivity fin material, the selection was narrowed to the use of copper as the base metal.

Metal	Thermal Conductivity (cgs units at 20°C)
Molybdenum	0.35
Gold	0.71
Copper	0.94
Silver	1.00
Tungsten	0.48

Because of the difficulties expected in protecting molybdenum and tungsten from oxidation, they were not considered further. Gold was not investigated because of its cost, and silver was considered only briefly because of its low melting point and its tendency to dissolve oxygen. Joining silver was also thought to present a problem, since an oxidation-resistant brazing alloy for service up to 1650°F with a melting point lower than that of silver was not known.

Because copper oxidizes in the temperature range of interest, several methods of protection were studied.

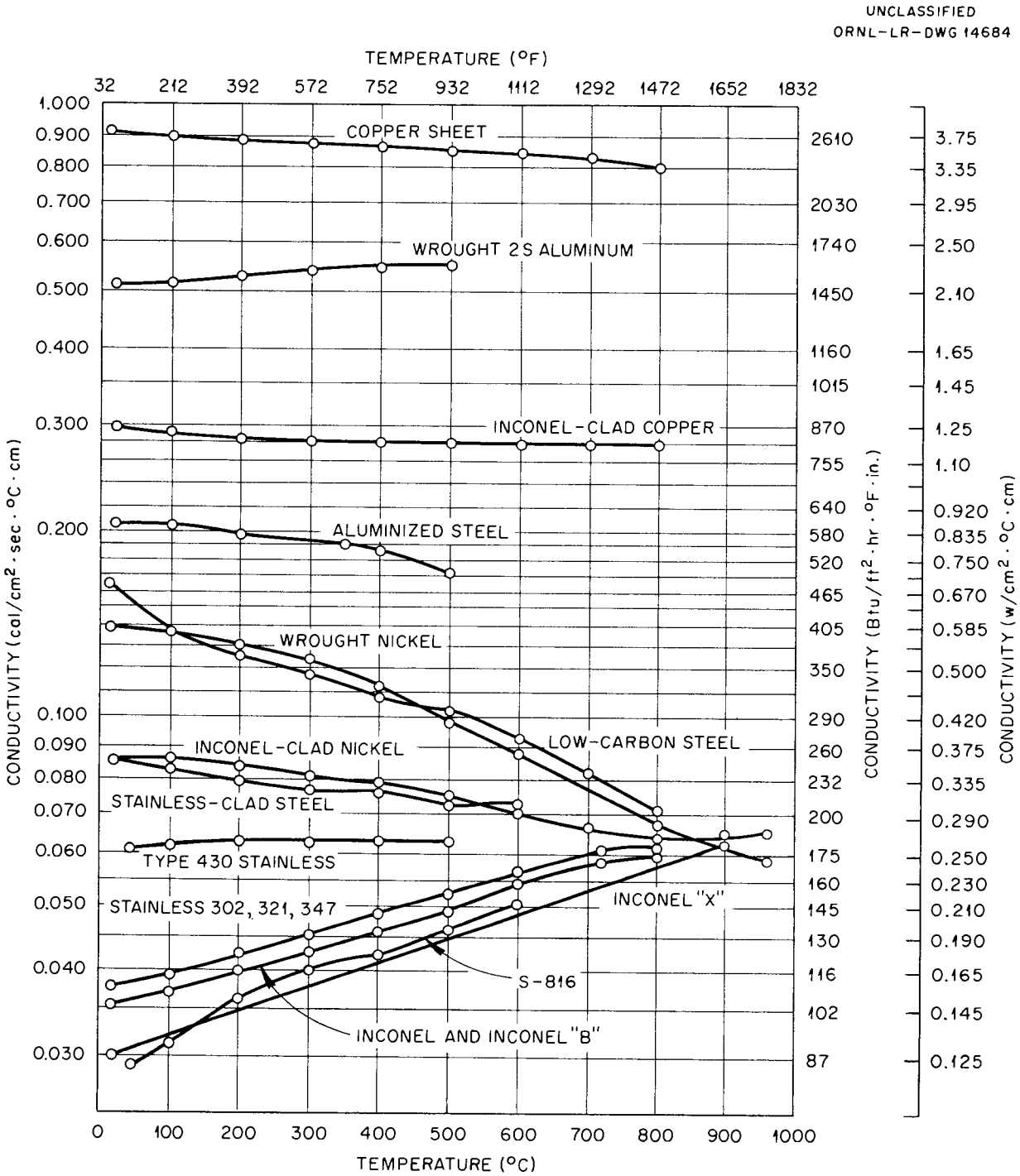

Fig. 1. Thermal Conductivity of Sheet Metal at Various Temperatures.

Plates

Electroplates on copper were tested, since the thin, uniform layers which can be deposited result in a high ratio of copper to the plate. The formation of protective coatings was thought to be possible by interalloying multiple deposits of metal during a diffusion heat treatment. Vapor deposits were also evaluated.

Copper Alloys

From the work of Frohlich[2] on tarnish-free copper alloys and from the studies of Price and Thomas[3] on the formation of protective films on copper alloys by selective oxidation, it was indicated that the copper-aluminum and the copper-beryllium alloys would be oxidation resistant.

Even though an alloy is oxidation resistant, its value under the conditions of the intended use necessarily depends upon its thermal conductivity. The aluminum bronzes show[4] a positive temperature coefficient of thermal conductivity from room temperature to 392°F. Values of the available thermal-conductivity data extrapolated to 1500°F indicated a value of 0.47 cgs units for the 4.5% aluminum–copper alloy and 0.37 cgs units for the 10% aluminum–copper alloy. These values, if correct, are about six times those for the heat-resistant steels and the nickel-base alloys (see Fig. 1). The alloy compositions were therefore selected to determine the minimum amount of aluminum for oxidation resistance and to verify the extrapolated thermal-conductivity values at 1500°F. Although primary emphasis was upon copper alloys, the silver-aluminum and copper-beryllium alloys were briefly studied, since their room-temperature conductivities were high enough to warrant further work.[5]

Clad Copper

Because a high-thermal-conductivity composite was desired, maximum cladding thickness was set at 0.002 in. per side for a total fin thickness of between 0.008 and 0.010 in. The suitability of such a configuration will depend upon the extent of diffusion between the cladding and the copper and the feasibility of applying such thin foils without surface defects. Thermal warpage due to the differential thermal expansion of the composite is also of concern. Studies of heat-resistant alloy-clads were confined to Inconel and types 310 and 446 stainless steels. Type 430 stainless-steel-clad copper was also studied because it was commercially available.

RESULTS AND DISCUSSION

Plates

Electroplates of chromium, chromium-nickel, and chromium-nickel-iron on copper were investigated. Plate thicknesses on either electroformed copper or copper foil were between 0.0005 and 0.002 in. per side. The various composites were tested without edge protection at 1500°F in a slow-moving stream of air in the as-plated and annealed condition. Prior to oxidation testing, the composites with multiple deposits were given a diffusion heat treatment at 1650°F for 18 hr in hydrogen to promote alloy formation – the formation of a Nichrome composition in the chromium-nickel plates and the Inconel composition in the plates of chromium-nickel-iron.

Chromium plates adequately protected copper from oxidation when the plates were adherent and crack-free. The cracking of typical chromium plates under a simulated brazing cycle is illustrated in Fig. 2. The severity of cracking increased with increasing plate thickness. Figure 3

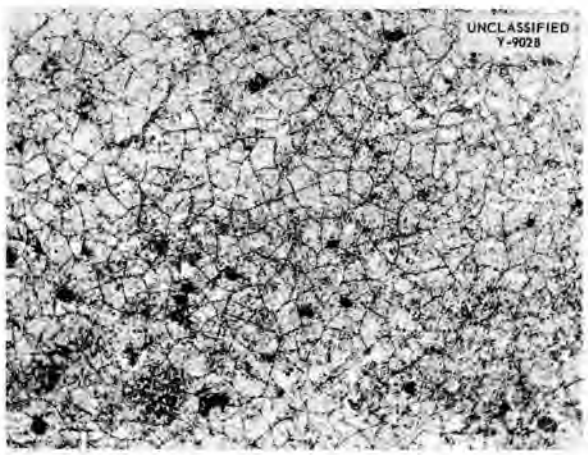

Fig. 2. Surface View of Chromium Plate (0.0005 in. Thick) on Copper After a 1832°F Temperature Cycle. Unetched. 100X.

---

[2]K. W. Frohlich, *Z. Metallkunde* 28, 368–375 (1936).

[3]L. E. Price and G. J. Thomas, *J. Inst. Metals* 63, 21 (1938).

[4]C. S. Smith and E. W. Palmer, *Metals Technology* 2, Technical Paper No. 648 (1935).

[5]P. A. Beck, "Properties of Wrought Beryllium Copper," *Metals Handbook*, p 934, T. Lyman, ed., American Society for Metals, Cleveland, 1948.

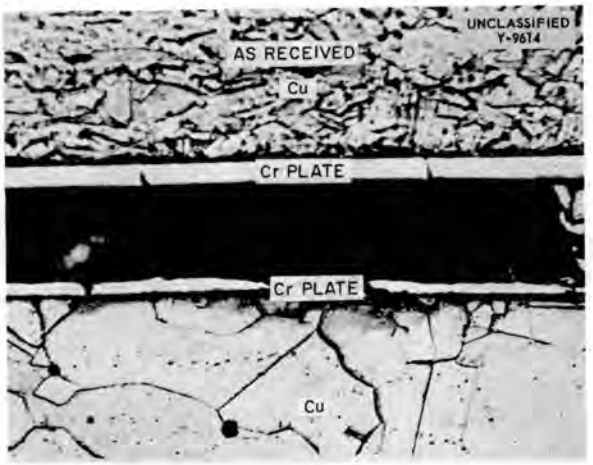

Fig. 3. Cross Section of Chromium Plate by the "Ductile Chromium Process" on OFHC Copper. Above as received. Below after 100 hr at 1500°F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 31%.

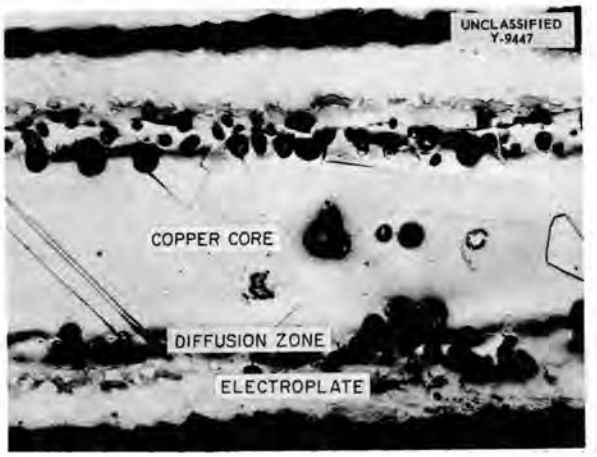

Fig. 4. Photomicrograph of 0.0015-in. Nickel Plus 0.0005-in. Chromium Plate on Copper After 100 hr at 1500°F. Etchant: $NH_4OH + Na_2O_2$. 200X. Reduced 31%.

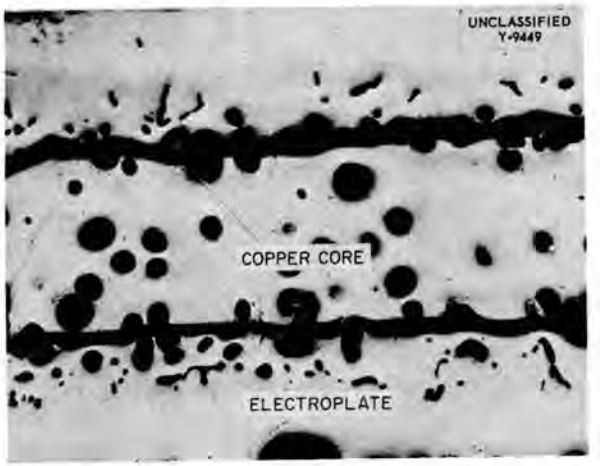

Fig. 5. Photomicrograph of 0.0015-in. Nickel Plus 0.0005-in. Chromium Plate on Copper After 500 hr at 1500°F. Etchant: $NH_4OH + Na_2O_2$. 200X. Reduced 31%.

shows that cracks were present in the "as-plated" condition and that failures occurred in these areas. Another type of failure in the composites was blistering of the plate following the high-temperature anneal in hydrogen. This probably arose from either improper plating conditions or the presence of copper oxide in the copper.

The chromium-nickel combination, after the diffusion heat treatment, showed that the interdiffusion of nickel and copper occurred at a faster rate than between nickel and chromium. Although a heat-resistant coating resulted, the composite was considered unsatisfactory. Figure 4 shows a 100-hr test at 1500°F which resulted in alloying at the copper-nickel interface and the formation of voids due to the unequal diffusion rates of the copper and the nickel. Increasing the service time to 500 hr results in the coalescence of the voids and a complete alteration of the copper core, as shown in Fig. 5.

The combination of chromium-nickel-iron electroplated on copper foil was unsuccessful because of poor adhesion and lack of control of final plate thicknesses. The same combination electroplated on electroformed copper was unsatisfactory because of surface roughness and brittleness.

Deposits of chromium, nickel, and chromium-nickel were vapor-plated onto copper by thermally decomposing their respective metal hexacarbonyls. Brittleness, poor adhesion, and variable plate thicknesses were characteristic of these deposits. The experiments indicated the need for considerably more fundamental work before the process could be attempted on a commercial basis.

The results of a test of vapor-plated chromium on copper are shown in Fig. 6. Even in the rather short exposure time, complete disintegration of the plate and subsequent oxidation of the copper occurred. Because this test was representative of the better deposits on copper, further studies were discontinued.

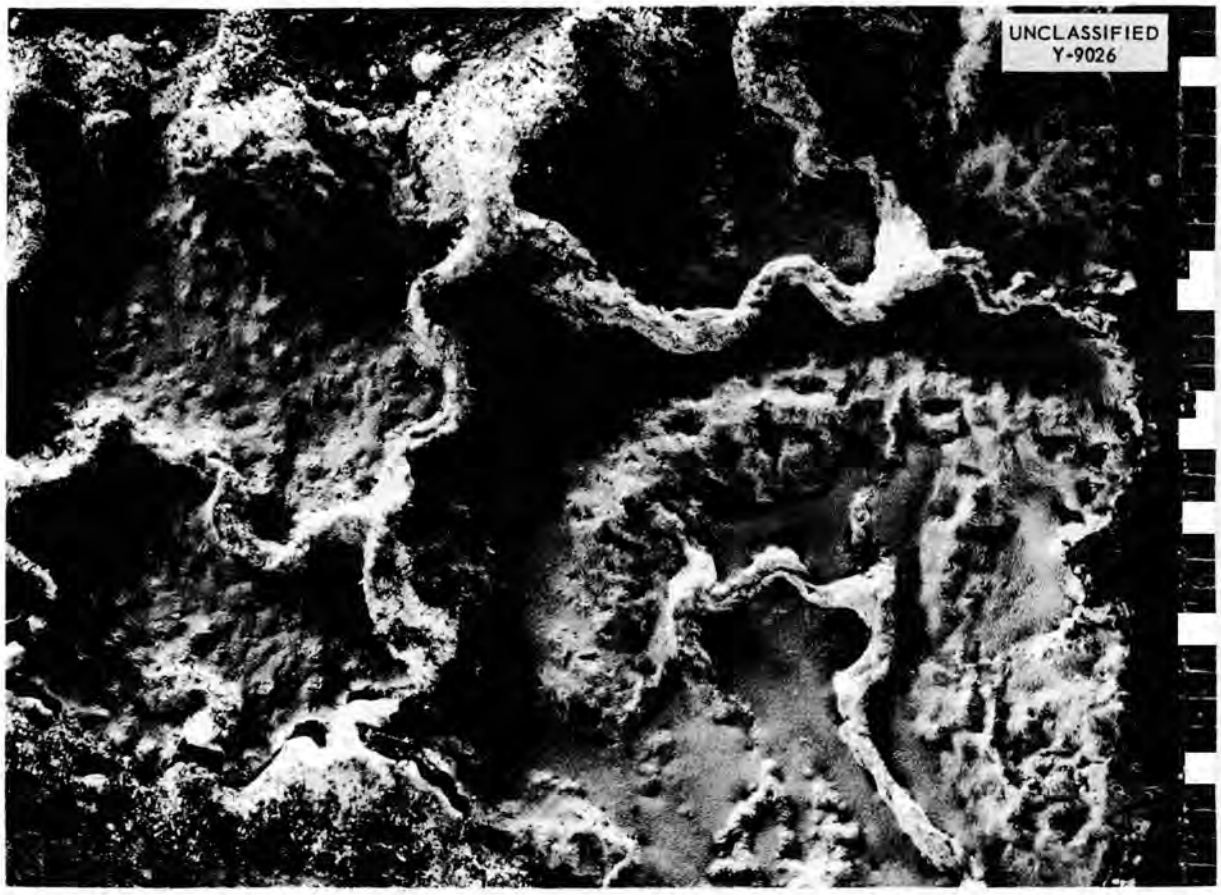

Fig. 6. Vapor-Plated Chromium on Copper Tested 168 hr at 1500°F in Air. Green needles of Cr_2O_3 formed on wrinkled copper oxide. 15X.

Copper Alloys

Several induction melts of copper alloys were made in a graphite crucible and poured to a ½-in.-dia rod. The compositions made were 2, 3, 5, 6, 8, and 10 wt % aluminum–balance copper and a 1 wt % beryllium–balance copper. A composition of 2 wt % aluminum–balance silver was included in this study. All the alloys were cold-swaged to 3/16-in.-dia rods with the exception of the 10% Al–90% Cu, which was brittle.

The oxidation of the mechanically polished specimens was evaluated at 1500°F. The results are presented in Table 1.

The copper-beryllium alloys were not investigated at greater beryllium contents because such increases would result in an alloy with a melting point so low as to prohibit its use at the temperature of interest. The silver-aluminum alloy was investigated only briefly since it was felt that it could not be brazed at a temperature below its melting point.

Because the copper-aluminum alloys with the lower aluminum contents were known to have higher thermal conductivities, attempts were made to preferentially oxidize the aluminum in wet hydrogen. The films of Al_2O_3 which formed were too thin to offer sufficient protection for long-time exposures.

The oxidation rates of the aluminum bronzes were not determined in this investigation because of the previous determination by Dennison and Preece.[6] The results of their work are summarized in Fig. 7. The rapid decrease in the oxidation rate of the several alloys reported by the above authors would indicate that the lower aluminum-content alloys would be satisfactory; however, it

[6] J. P. Dennison and A. Preece, *J. Inst. Metals* 81, 229–234 (1953).

TABLE 1. OXIDATION OF COPPER ALLOYS IN 100 hr AT 1500°F IN AIR

Alloy (wt %)	Weight Loss from Scaling (%)	Remarks
Copper	42.4	Dense black oxide
2 Al–98 Cu	28.5	Uniform scale, dense black oxide
3 Al–97 Cu	1.0	Numerous areas of localized oxidation
5 Al–95 Cu	0.16	Scattered areas of localized oxidation
6 Al–94 Cu	None	No change
8 Al–92 Cu	None	No change
10 Al–90 Cu	0.42	Weight gain, dimensional growth, crystalline Al_2O_3 formed
2 Al–98 Ag	None	No change
1 Be–99 Cu	7.0	Uniform black oxide

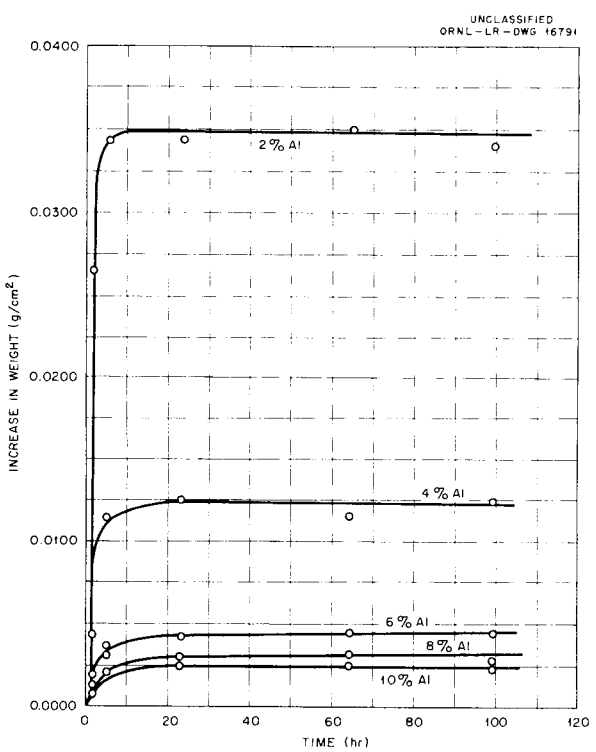

Fig. 7. Oxidation of Copper-Aluminum Alloys After Different Times at 850°C. After Dennison and Preece.

was determined in this investigation that oxidation in alloys containing less than 6% aluminum was localized and would therefore be unsuitable in the form of 0.010-in. sheet.

The areas of localized oxidation characteristic of low aluminum-content alloys are composed of copper as oxide nodules. Their formation is thought to arise from the fact that aluminum is electropositive to copper. The reaction may occur in the following sequence: (1) Both copper oxide and aluminum oxide form initially in air. Coverage of the reaction surface with one or the other oxide is localized since there is an insufficient amount of either to predominate. The copper oxide is then reduced to copper at the expense of the aluminum in the alloy with the resultant formation of porous Al_2O_3. (2) The depletion of the aluminum in the alloy creates areas high in copper. (3) Additional copper oxide is then formed at areas high in copper. The conditions are then favorable for the reaction to continue. The rate of growth of the copper oxide nodules will depend upon the diffusion rate of aluminum atoms to areas high in copper.

Additional tests of the 6 and 8% aluminum–balance copper alloys were extended to longer times and confirmed that these alloys were oxidation resistant. Test durations up to 1000 hr at

1500°F in air showed no adverse effects because of either temperature cycling (warpage) or scale formation.

The thermal conductivities of these two alloys were determined, and it was found that they exceeded the extrapolated values. As was anticipated, the thermal conductivity of both alloys continued to increase with increasing temperatures up to 1562°F (850°C). Figure 8 is a plot of the values obtained in measurements by using iron and aluminum as standards. The assignment of absolute values was uncertain by the amount indicated.

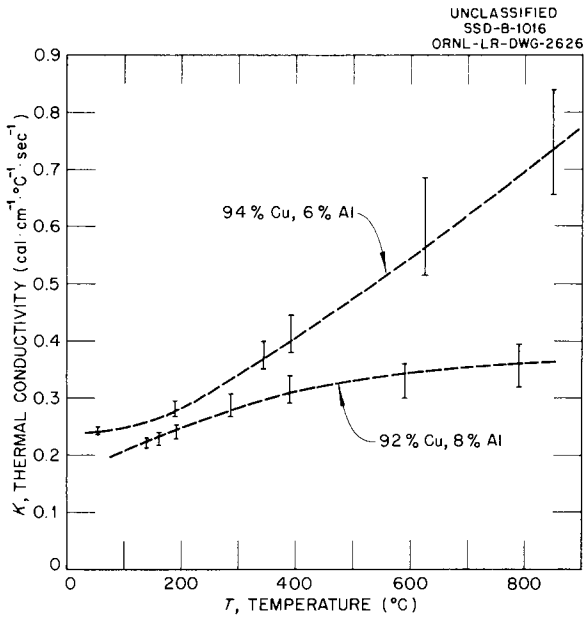

Fig. 8. Thermal Conductivity of Aluminum Bronze.

The comparisons of Fig. 9 show that the objectives of this investigation can be realized in the use of a 6% Al–94% Cu alloy. The joining of the alloy fins to Inconel by brazing is, however, a very great problem. By its very nature, the regenerative type of Al_2O_3 film will render the alloy difficult or unsuitable to braze unless a fluxing type of brazing composition can be developed.

Clad Copper

Fabrication. – Composites for this study were fabricated by roll cladding the copper between oxidation-resistant cover plates at 1832°F. In order to maintain dimensional tolerances, the

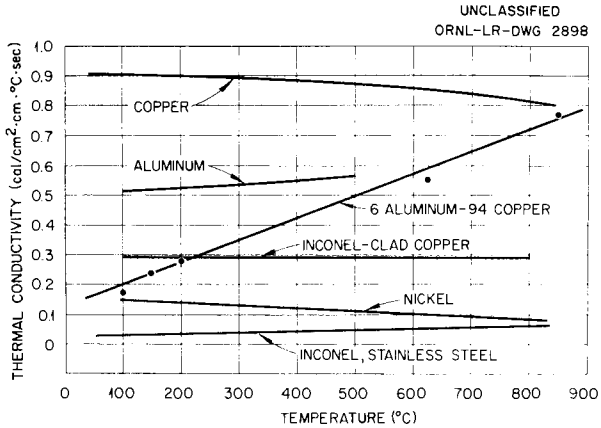

Fig. 9. Variation of the Thermal Conductivity of Several Alloys with Temperature.

copper was closely framed with alloy strips of the same composition as the clad. The cover plates which were welded to the frames formed a capsule, which was evacuated prior to hot rolling. Surface flaws and scaling of the clad were minimized by heating in a hydrogen atmosphere and maintaining "good housekeeping."

Through experience, it was found that as the thickness of the starting capsule was increased, a higher ratio of copper-to-clad became necessary to maintain a precalculated ratio when the capsule was finished to a thickness of 0.008 or 0.010 in. The change in ratio is necessary because the copper is a relatively soft metal compared to the heat-resistant alloys and, therefore, tended to be "squeezed out" during the early hot-rolling passes and especially during the cold-rolling operation. The difficulties were overcome by hot rolling the composite to the maximum total reduction commensurate with the ability of the composite to hold the rolling temperature. This was desirable since the frames prevented satisfactory reduction during cold rolling. By employing these techniques, metal ratios in the composite at the finished size could be held to within ±0.0002-in. variation.

In the initial experiments, reductions in thickness of about 20% per pass with 3-min reheats were made. These reductions frequently caused the rupture of the clad cover plates because of a buildup of a wall of copper within the capsule. The optimum rolling schedule, as determined by the recovery of sound material, was established

to be between 5 and 7% reduction per pass. For clad material low in nickel content, no bonding occurred until the first two passes were increased to 10% reduction. The minimum amount of total hot reductions necessary to achieve metallurgical bonds was about 35% in thickness for Inconel clads, 50% for type 310 stainless steel, and about 60% for type 446 stainless steel. These reductions were indicative of the amount of alloying and diffusion which would be expected between the clad and the copper, as will be shown below.

The alloy frames were sheared from the bonded composite after hot rolling, and the composite was finished to the final thickness by cold rolling. Here again, light (5%) rolling passes followed by anneals after each pass were necessary before a total cold reduction of 40% in thickness could be achieved. Undesirable surface and interface conditions resulted when greater reductions per pass were attempted or annealing omitted.

Evaluation and Inspection. – The clad copper was checked for dimensional tolerance by removing representative rectangles from the compact and dissolving out the copper core with HNO_3. The stripped clad was examined for pinholes. The thickness was measured by a micrometer to find the maximum thickness, and an average thickness, t_{avg}, was determined from a knowledge of the weight, density, and the lateral dimensions according to the formula

$$t_{avg} = \frac{W}{(w)(l)(\rho)(16.4)} = \text{in.},$$

where

W = weight of stripped clad, g,
w = width of stripped clad, in.,
l = length of stripped clad, in.,
ρ = density of clad metal, g/cm^3.

From the two values of thickness obtained, a roughness index was calculated for the copper-clad interface and was adopted as a basis of acceptance or rejection of the composite since areas less than the average must exist.

As an example, in a shipment of Inconel-clad copper specified to be 0.008 in. thick with a clad-to-copper ratio of 1:2:1, the stripped Inconel was found to be 0.003 in. thick by micrometer measurements. A weight measurement showed a calculated average thickness of 0.0018 in. or, according to the above scheme, a roughness index (R.I.) as follows:

$$\text{R.I.} = \frac{\text{maximum clad thickness, in.}}{\text{average clad thickness, in.}} = \frac{0.003}{0.0018} = 1.63.$$

Visual examination of this composite showed surface stretch marks transverse to the rolling direction and pinholes in the clad, as shown in Fig. 10. Tests at elevated temperatures indicate that composites having a roughness index of 1.25 or less were satisfactory up to 500 hr.

A similar unsuitable composite is shown in Fig. 11. The rough interface resulted from hot reductions of about 20% per pass and from being cold rolled in excess of a 50% reduction in thickness between anneals. At reduction schedules of between 5 to 7% per pass and by annealing after a total of 40% cold reduction, the interface roughness was reduced considerably, as shown in Fig. 12.

Clad defects were evidenced by the growth of small nodules of copper oxide on the surface of the test piece after service. Copper oxide nodules which appear after a short-time exposure (100 hr) are usually caused by the presence of pinholes. Nodules which appear after longer exposure

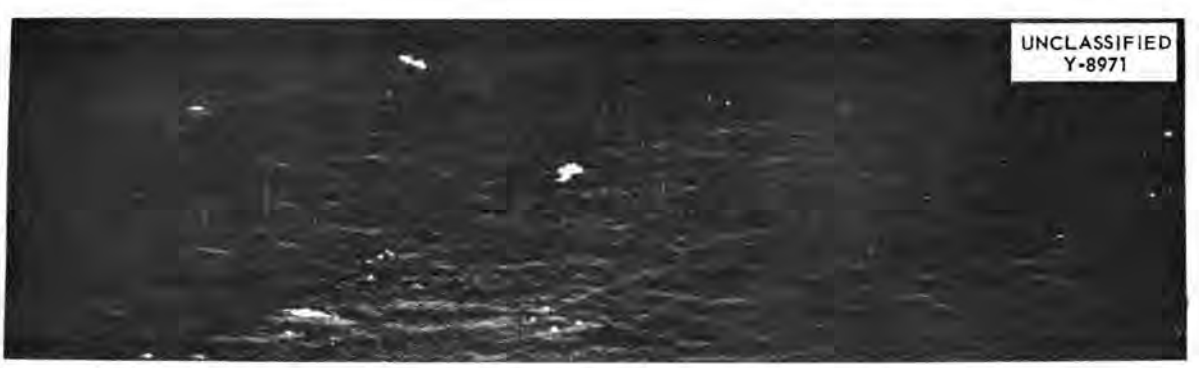

Fig. 10. Silhouette of Inconel Clad Stripped from Copper, Showing Pinholes and Roughness. 4X.

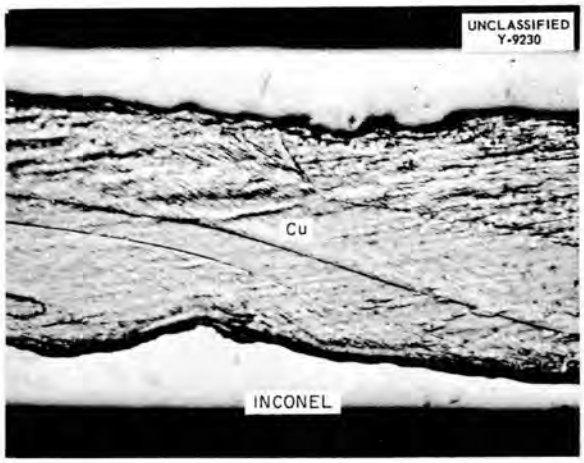

Fig. 11. Inconel-Clad Copper, As Rolled, Clad Variation 1–3 mils. Etchant: $NH_4OH + Na_2O_2$. 200X. Reduced 30.5%.

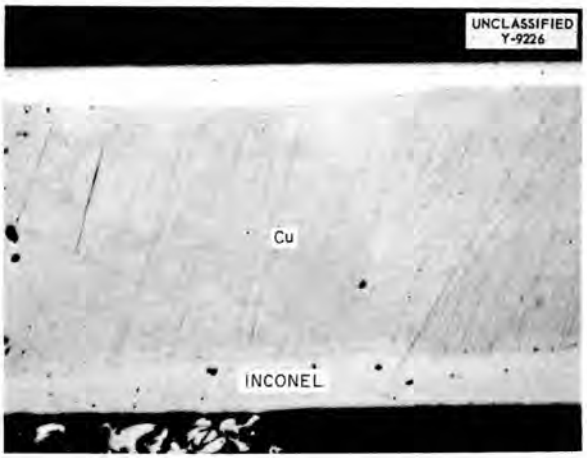

Fig. 12. Inconel-Clad Copper, As Rolled, Clad Variation 1–2 mils. As polished. 200X. Reduced 30.5%.

times are caused by thin areas in the clad which fail from the outward diffusion of copper and from the oxidation of the clad. Provided that the number and size of the defects are small, the performance of the composite is not significantly affected, since the oxidation of the copper within the composite is restricted to a small area under the copper oxide nodule.

Stability of the clad copper was determined by exposure in a slow-moving stream of air at 1500°F for various times up to 1000 hr. After completion of the test, the specimens were examined for warpage, clad failures, and diffusion.

Inconel-Clad Copper. – The initial tests on cladding copper with Inconel were designed to evaluate the "claddability" of oxygen-free high-conductivity copper (OFHC) and the commercial grade of copper (tough pitch). In roll-cladding experiments, it was found that metallurgical bonds were difficult, if not impossible, to obtain when using the tough-pitch copper. These composites were heated in hydrogen during the hot-rolling operation, and it is surmised that the reduction of the copper oxide released sufficient water vapor to cause oxidation of the Inconel.

Composites containing copper cores of both grades were evaluated after a test of 100 hr at 1500°F. As illustrated by Figs. 13 and 14, cross

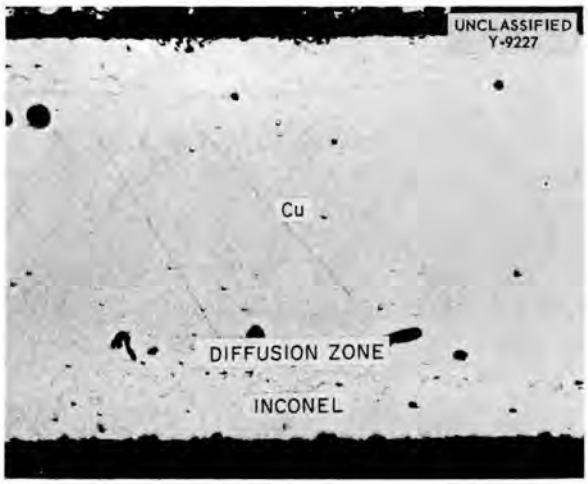

Fig. 13. Inconel Clad with OFHC Copper After 100 hr at 1500°F. As polished. 200X. Reduced 30.5%.

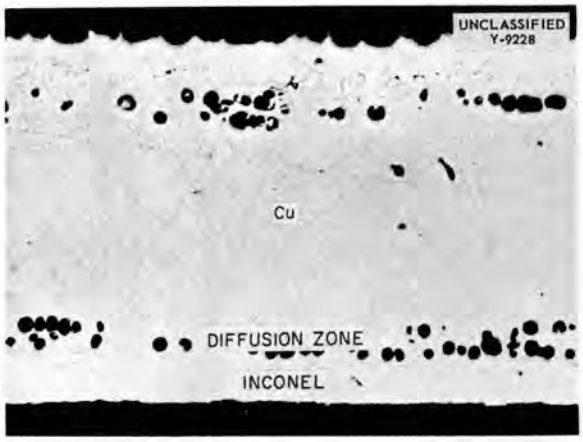

Fig. 14. Inconel-Clad Commercial Copper After 100 hr at 1500°F. As polished. 200X. Reduced 27%.

sections through the composites, diffusion between the Inconel and the copper was pronounced, as evidenced by a color change of the copper near the interface and by the formation of voids. The voids result from the unequal diffusion rates of the copper and the nickel (in Inconel) and were more pronounced in the composite containing the tough-pitch copper. The difference between the two grades of copper is the oxygen content – 0.04% in the tough-pitch copper and nil in the OFHC grade.

A 100-hr test of Inconel-clad copper obtained from the General Plate Division of Metals & Controls Corp. showed similar results (Fig. 15). Increasing the service time to 500 hr causes

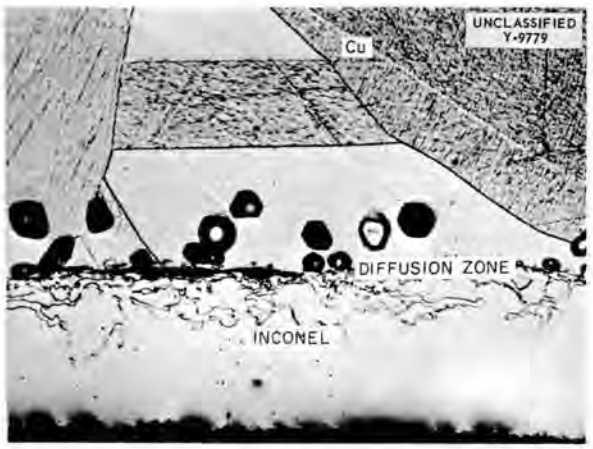

Fig. 15. Inconel-Clad Copper After 100 hr at 1500°F. Etchant: 10% chromic acid. 500X. Reduced 31%.

agglomeration of the voids and the eventual failure of the composite due to the outward diffusion of copper to the air-metal interface as shown in Fig. 16. The metal near the voids in the 100-hr test is white, indicating alteration of the copper (not evident in the photograph). After 500 hr the whole composite loses its characteristic copper color.

Diffusion barriers for the Inconel-copper combination study were selected on the basis of having a sufficiently high melting point and a low solubility in copper. In addition, barrier metals with very high melting points were investigated. Several metals were found which were successful diffusion barriers between Inconel and copper. Thicknesses of the order of 0.0005 to 0.001 in. of types 310

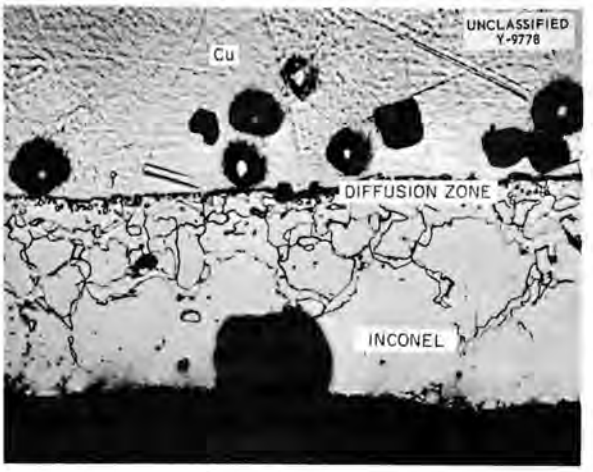

Fig. 16. Inconel-Clad Copper After 500 hr at 1500°F. Etchant: 10% chromic acid. 500X. Reduced 31%.

and 446 stainless steels and of iron were determined to be adequate for 1000 hr at 1500°F (for examples, see Figs. 17 through 19). These combinations of clad copper were fabricated by incorporating a foil of predetermined thickness between the copper and Inconel prior to roll cladding.

The use of high-melting-point metals such as tungsten, molybdenum, and tantalum was unsuccessful, since a continuous barrier layer could not be obtained. The discontinuities resulted from fracturing or the imbedding of the barrier metal in the copper during hot rolling. Both occur because of the large difference in hardness and ductility of the metals at the rolling temperature.

Type 310 Stainless-Steel-Clad Copper. – The diffusion between copper and type 310 stainless steel was slight. A reaction zone, composed of small islands of a new phase, appears in the clad up to about 0.0005 in. in 100 hr at 1500°F, as shown in Fig. 20. Increasing the time to 500 hr does not significantly increase the penetration into the clad but results in a greater number of islands (see Fig. 21). In a 1000-hr test, diffusion is evident but is confined to a small zone at the interface with no apparent alteration of the copper core (Fig. 22).

It was previously shown that the oxygen in copper increased the interface reaction between Inconel and copper. A similar effect is evident between oxygen-containing copper and type 310

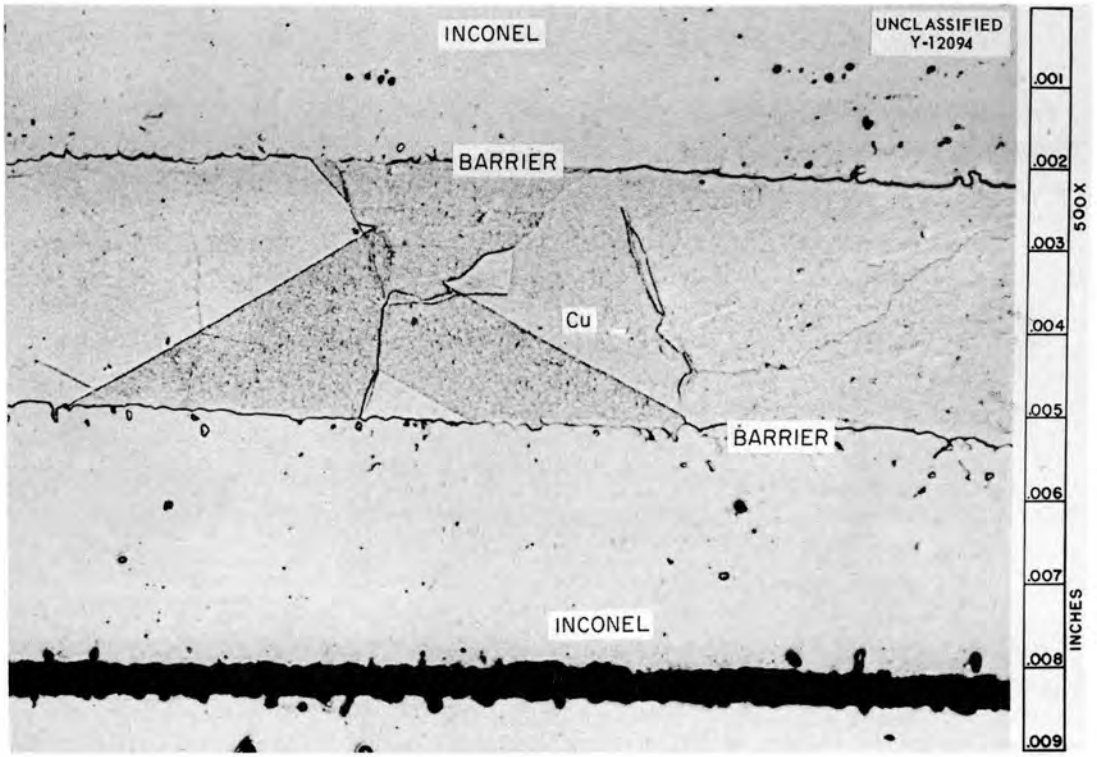

Fig. 17. Inconel-Clad Copper with 0.0005 in. of Type 310 Stainless Steel as a Barrier After 1000 hr at 1500° F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 14%.

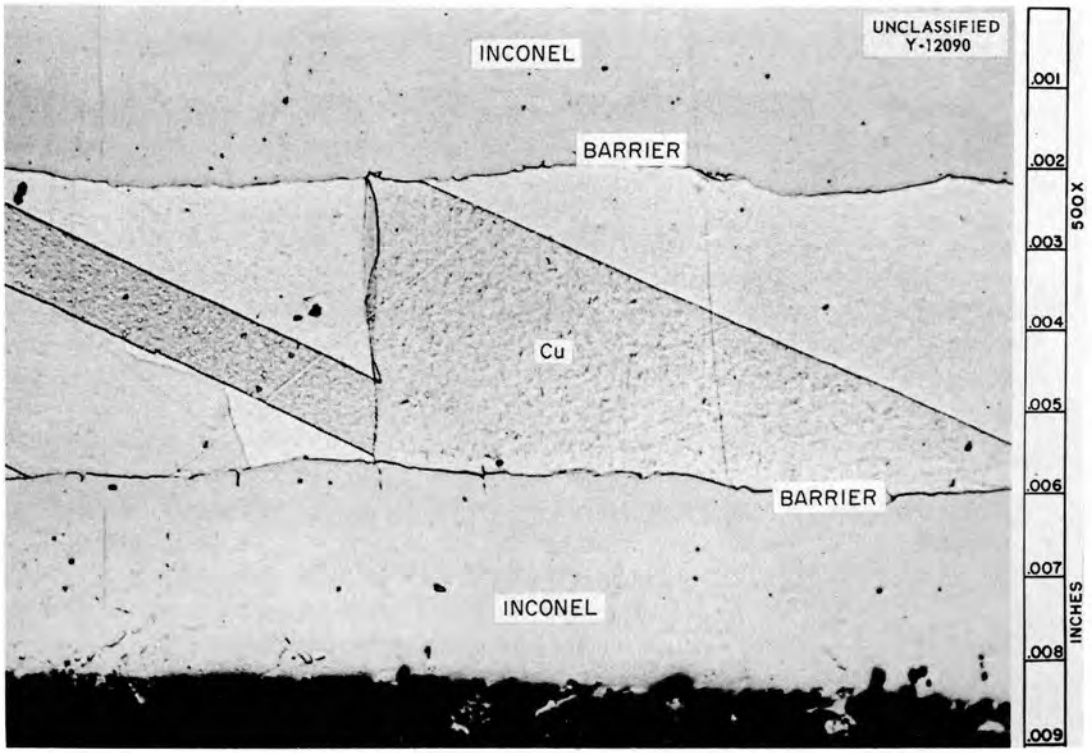

Fig. 18. Inconel-Clad Copper with 0.001 in. of Iron as a Barrier After 1000 hr at 1500° F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 14%.

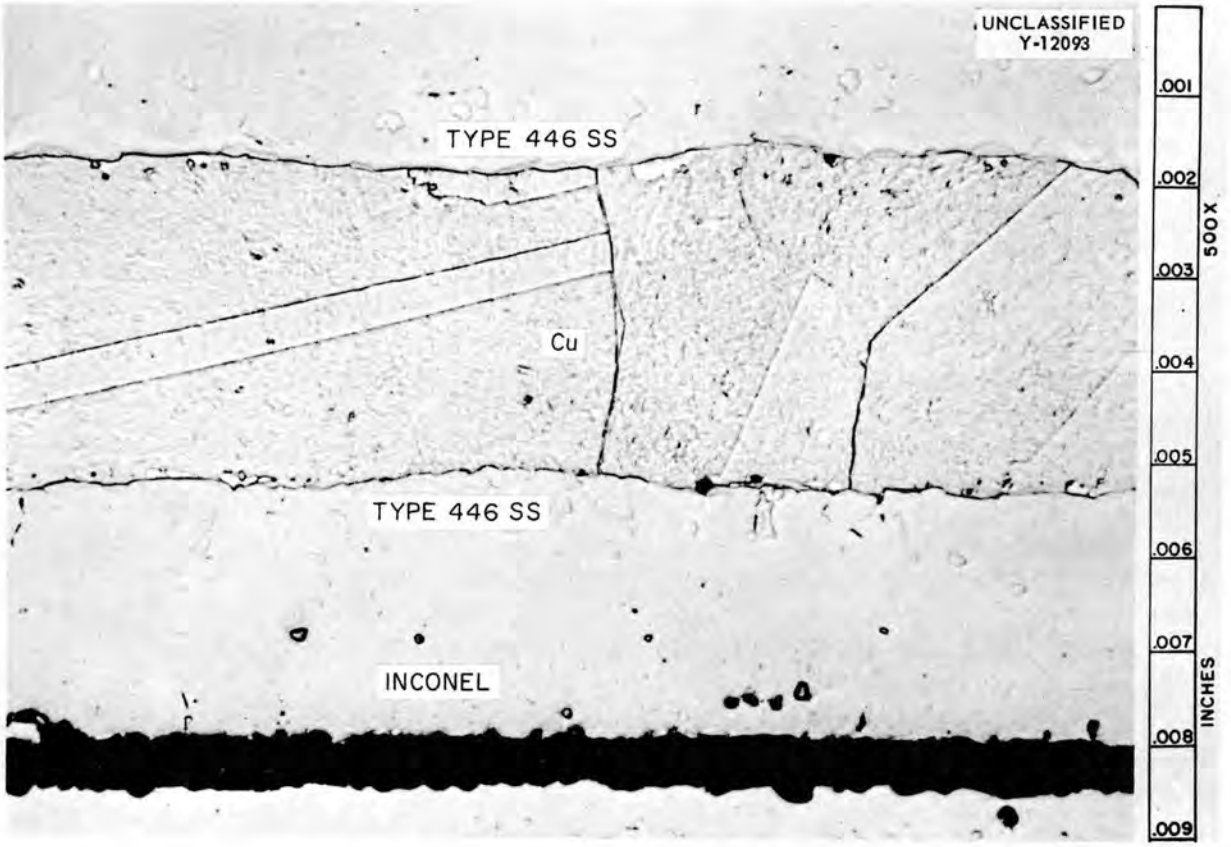

Fig. 19. Inconel-Clad Copper with 0.001 in. of Type 446 Stainless Steel as a Barrier After 1000 hr at 1500° F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 3.5%.

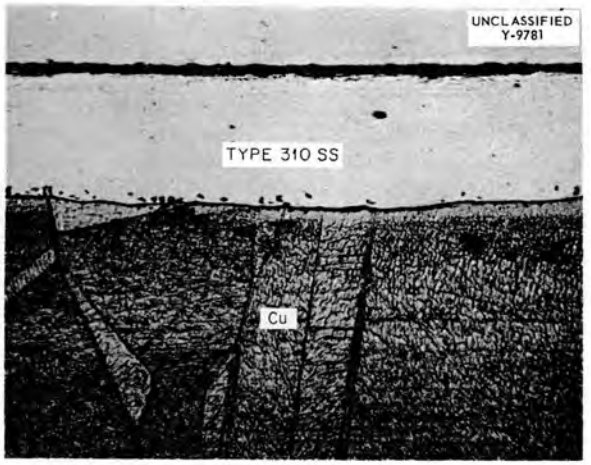

Fig. 20. Type 310 Stainless-Steel-Clad Copper After 100 hr at 1500° F. Etchant: chromic acid. 400X. Reduced 31.5%.

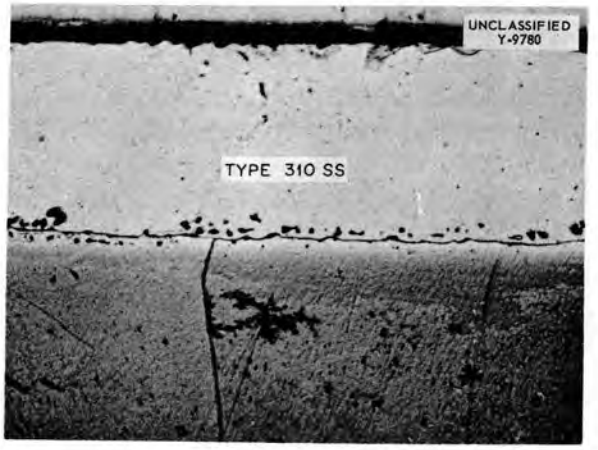

Fig. 21. Type 310 Stainless-Steel-Clad Copper After 500 hr at 1500° F. Etchant: chromic acid. 400X. Reduced 31%.

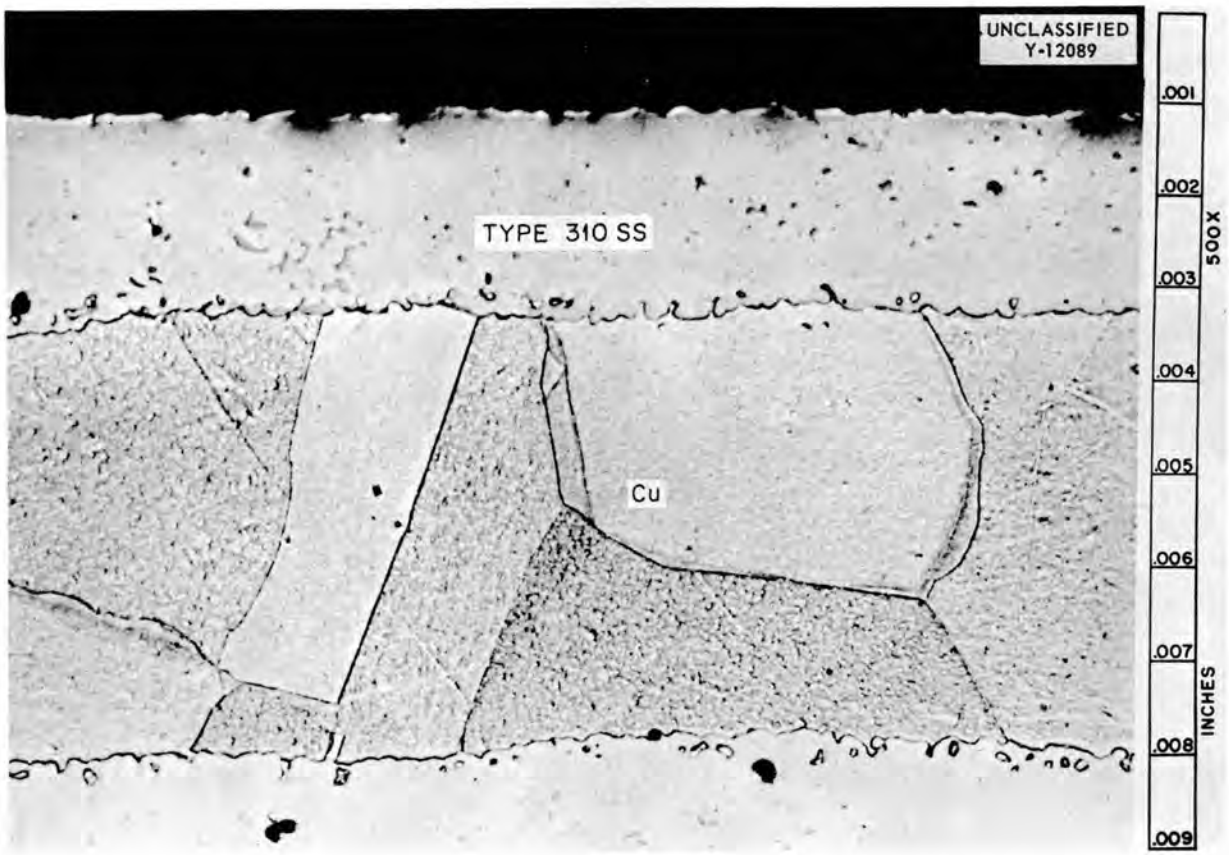

Fig. 22. Type 310 Stainless-Steel-Clad Copper After 1000 hr at 1500°F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 3.5%.

stainless steel. Comparing Figs. 23 and 20, it is apparent that diffusion is more pronounced in the composite containing the commercial copper.

Types 430 and 446 Stainless-Steel-Clad Copper. – In tests up to 1000 hr at 1500°F, combinations of types 430 and 446 stainless steels and copper showed no apparent alteration of the copper core (Figs. 24 and 25). Precipitates appear in the clad which have been identified as carbides and sigma phase. A slight interface reaction occurs, however, and is probably an alloy of copper-chromium-iron (Fig. 26).

Clad-Copper Fabrication by Seam Welding. – During the investigation, it became apparent that "near failures" in the clad would result if attempts were made to reduce its thickness to less than 0.002 in. Moreover, commercial suppliers of these composites were doubtful if thinner clad thicknesses would be amenable to quantity production and still have the desired quality. This

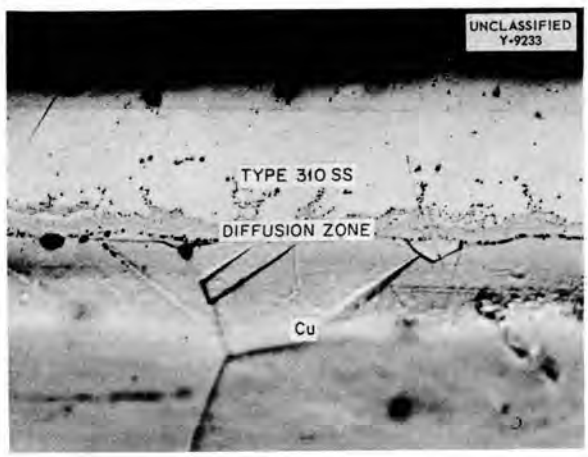

Fig. 23. Type 310 Stainless-Steel-Clad Copper (Commercial) After 100 hr at 1500°F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 31%.

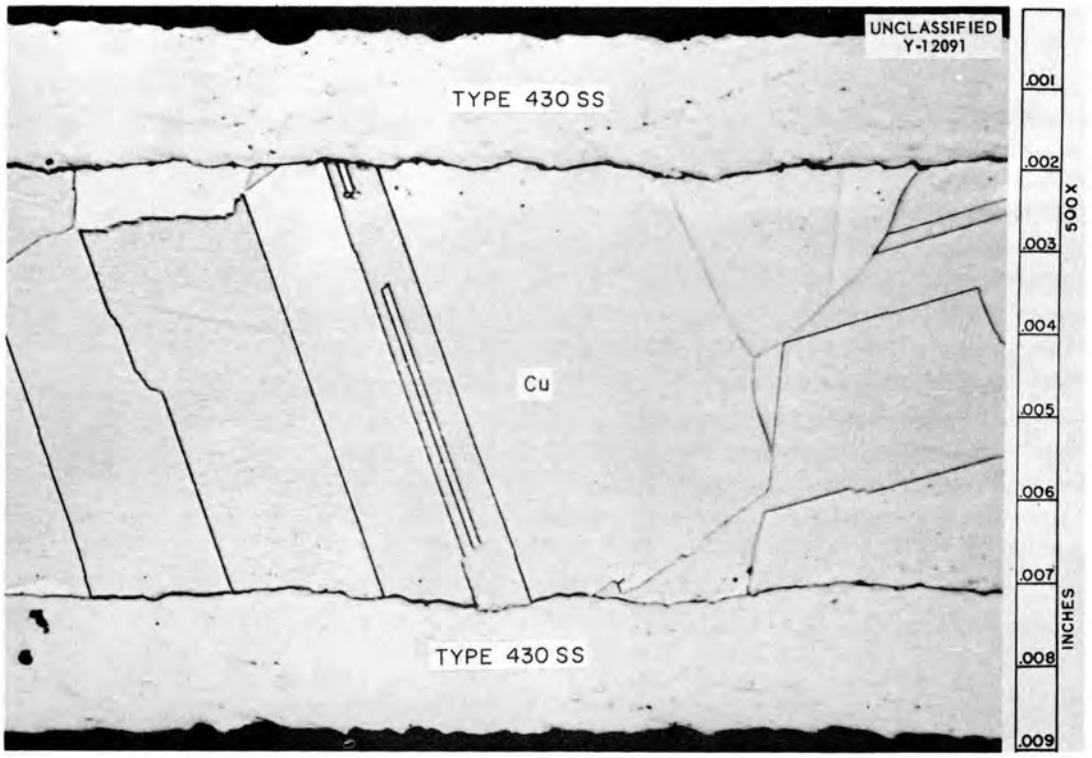

Fig. 24. Type 430 Stainless-Steel-Clad Copper After 1000 hr at 1500° F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 14%.

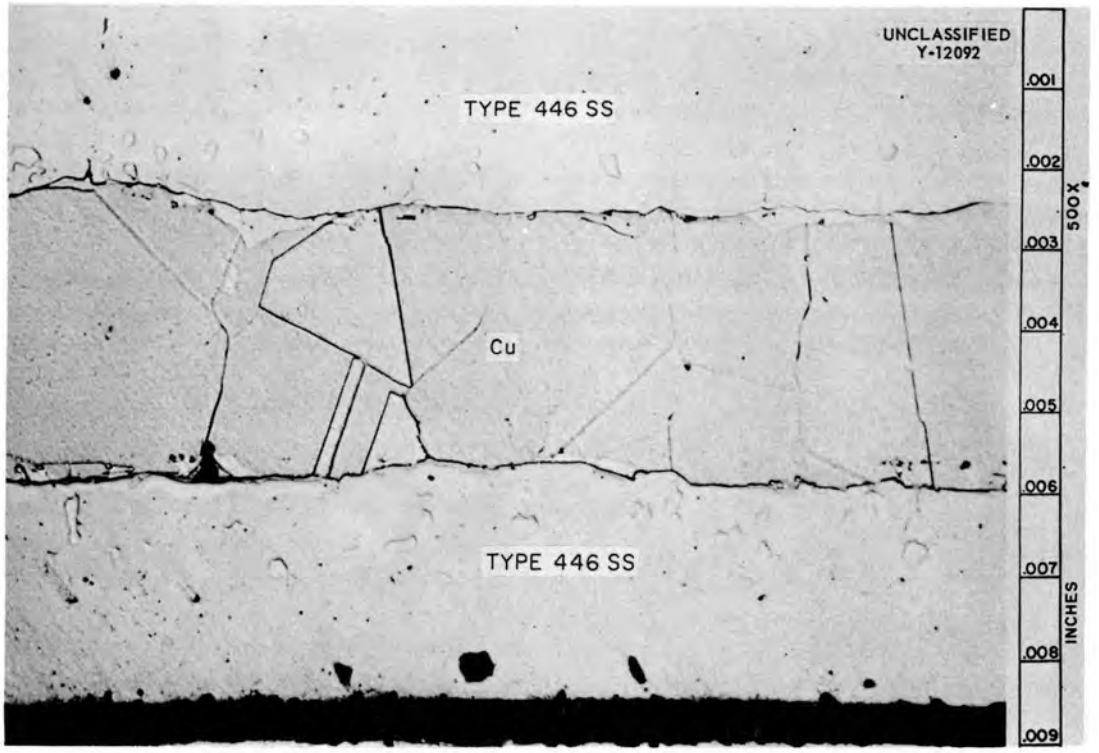

Fig. 25. Type 446 Stainless-Steel-Clad Copper After 1000 hr at 1500° F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 13.5%.

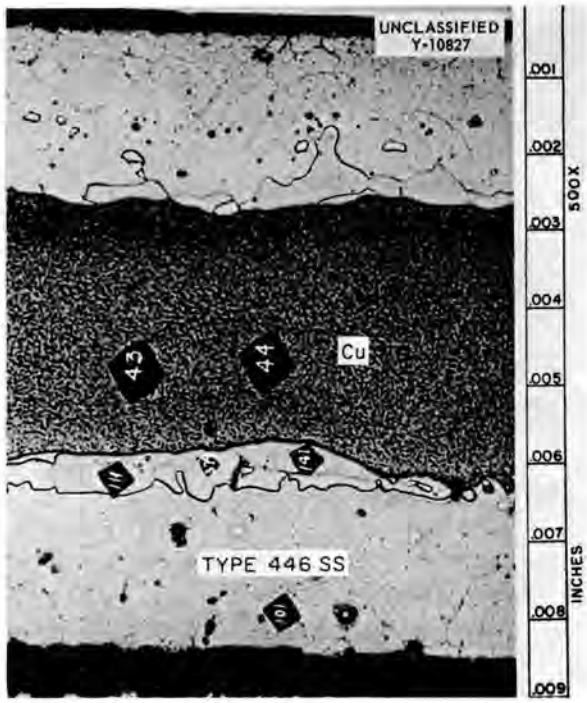

Fig. 26. Type 446 Stainless-Steel-Clad Copper After 1000 hr at 1500°F. Etchant: electrolytic oxalic acid. 500X. Reduced 19.5%.

prompted a search for an alternate method of fabrication aimed at increasing the copper ratio in the composite.

With the cooperation of the Glenn L. Martin Co., test specimens of clad copper were made with the use of a seam-welding machine and evaluated. Metallurgical bonds were obtained by passing a current through the foils of the final desired thickness under pressure. From the few test specimens made, it was apparent that special tooling would be necessary for quantity production of the composite and that the method was best suited for the fabrication of strip.

Such a technique has the advantage of increasing the ratio of copper to clad (hence its thermal conductivity) with no increase in the uncertainty of thin areas since the roughness index remains essentially 1. Furthermore, uniformity of composite layer thicknesses is desirable from the standpoint of strength and calculation of the thermal conductivity. Figure 27 is a composite made by this technique after testing. The uniform layers and the absence of failures should be noted. Composite strip has also been fabricated with the copper edge-protected by seam welding the overlapping clad.

Stress-Rupture Strength of Clad Copper. – It has been demonstrated that the minimum clad thickness on copper is not necessarily restricted by the fabricability of the composite but may be dictated by the strength requirements at the service temperature.

At 1500°F the copper core will not have useful engineering strength and will depend almost entirely on the clad for strength. In order to determine an approximate life of these composites, crude stress-rupture tests of type 310 stainless-steel-clad copper were made at 1500°F. The test specimens were prepared by clamping the composite strips in a steel jig and chiseling them along the jig contour. The rough edges were sanded, and a reinforcing strip of stainless steel was spot-welded on the specimen ends. Gage widths of 0.870 and 0.374 in. were used without edge protection. The specimens were tested in moving air caused by the natural draft in a vertical tube furnace. The stress-rupture tests are summarized in Table 2. The fact that the specimens were not edge-protected may reflect in the rupture life; however, it is expected to be slight in view of the fact that the amount of copper affected in 500 hr at 1500°F was about $\frac{3}{64}$ in.

Figure 28 shows that grain-boundary failure in the clad is beginning to become evident at a stress of 500 psi at 1500°F, but the copper core has not suffered from oxidation. The specimen stressed at 2000 psi (Fig. 29) shows internal fracture of the composite and some oxidation due to cladding failure. Figure 30 is from a specimen stressed at 1000 psi for 500 hr. Here both internal oxidation and core fractures are more severe. Brittle composites resulted in every test which showed elongations of 20% or more, regardless of the stress or exposure time.

Miscellaneous Testing. – *Dimensional Stability.* – The effects of a temperature cycle on these configurations were only noted in tests made for other purposes. In all cases, the composites warped on cooling to room temperature. The severity of warpage depended upon the exposure time. The amount of copper oxide formed on the unprotected edges seemed to be the controlling factor. The warping was most pronounced in those specimens clad with type 400 series stainless steels and was probably caused by the difference in the coefficient of thermal expansion. As a

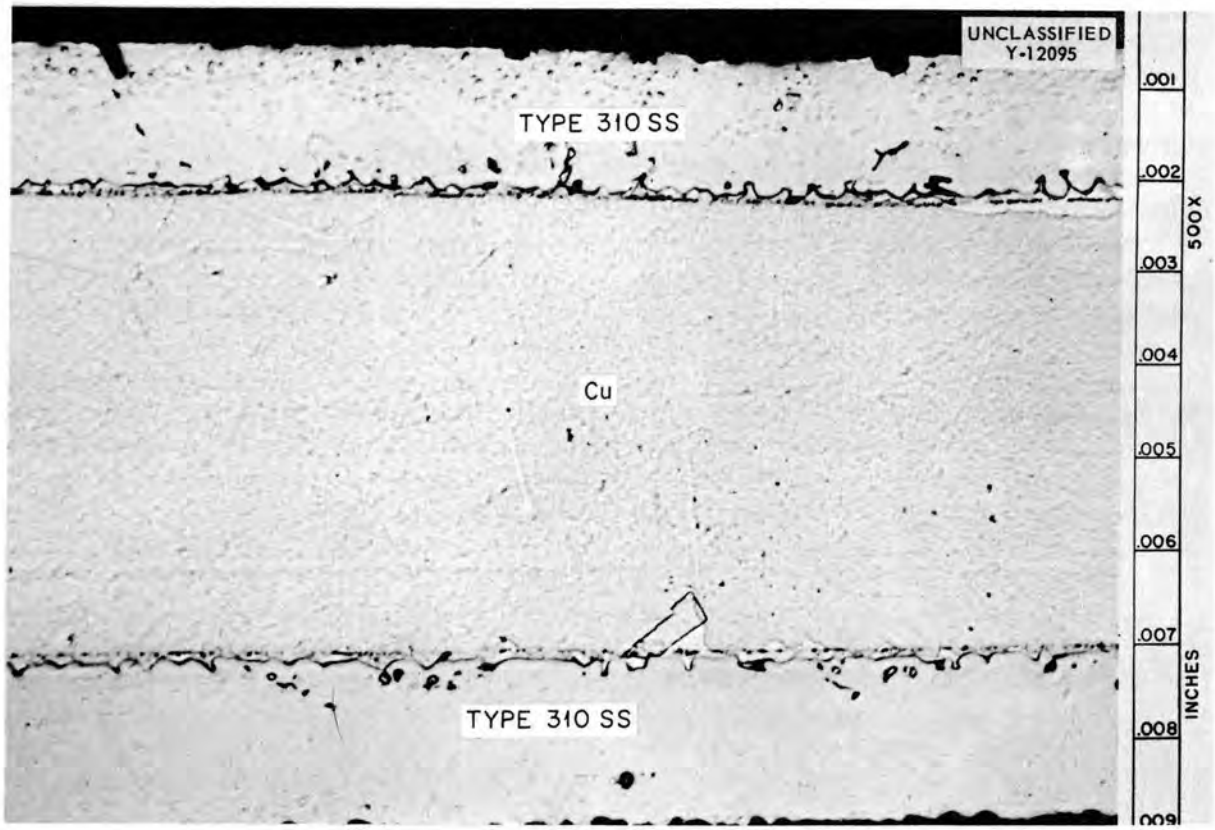

Fig. 27. Type 310 Stainless-Steel-Clad Copper (by Seam-Welding Techniques) After 1000 hr at 1500°F. Etchant: $NH_4OH + Na_2O_2$. 500X. Reduced 4%.

TABLE 2. STRESS-RUPTURE STRENGTH OF TYPE 310 STAINLESS-STEEL-CLAD COPPER 0.008 in. THICK (1:2:1) AT 1500°F IN AIR

Stress (psi)	Test Duration (hr)	Elongation in $2\frac{1}{2}$ in. (%)	Remarks
500	500	5	No failure
500	1000	10	No failure
1000	500	20	Stringers of copper, oxide on gage section
1500	500	$47\frac{1}{2}$	Gage section completely oxidized
1500	887		Rupture
1650	528	40	Rupture
1800	336	50	Rupture
2000	96	39	Rupture

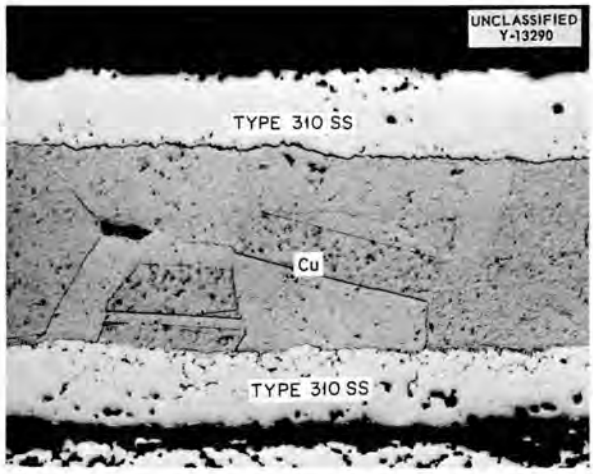

Fig. 28. Type 310 Stainless-Steel-Clad Copper Stressed 500 hr at 500 psi and 1500°F in Air. Etchant: $NH_4OH + Na_2O_2$. 200X. Reduced 27.5%.

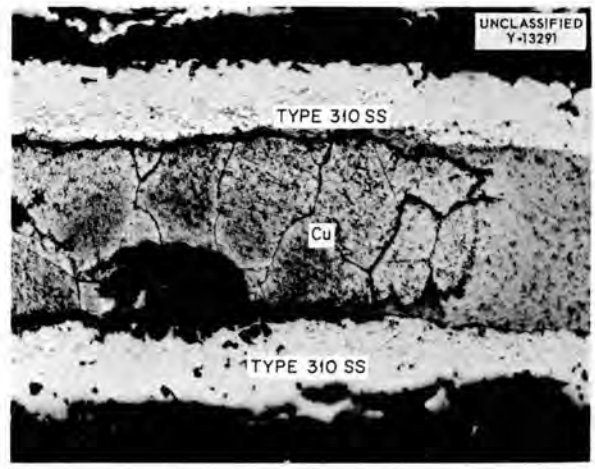

Fig. 30. Type 310 Stainless-Steel-Clad Copper Stressed 500 hr at 1000 psi and 1500°F in Air. Etchant: $NH_4OH + Na_2O_2$. 200X. Reduced 30.5%.

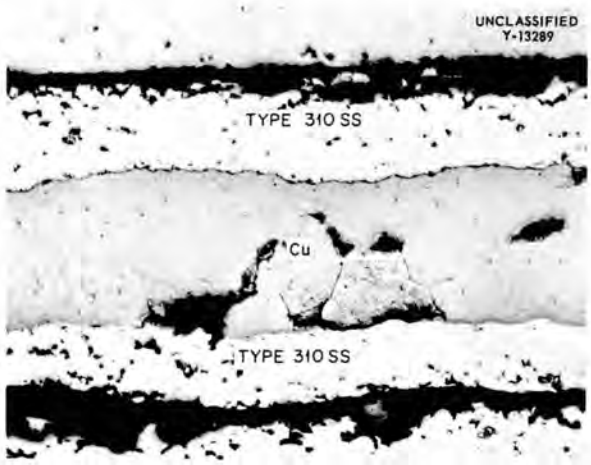

Fig. 29. Type 310 Stainless-Steel-Clad Copper Stressed 96 hr at 2000 psi and 1500°F in Air. Etchant: $NH_4OH + Na_2O_2$. 200X. Reduced 30.5%.

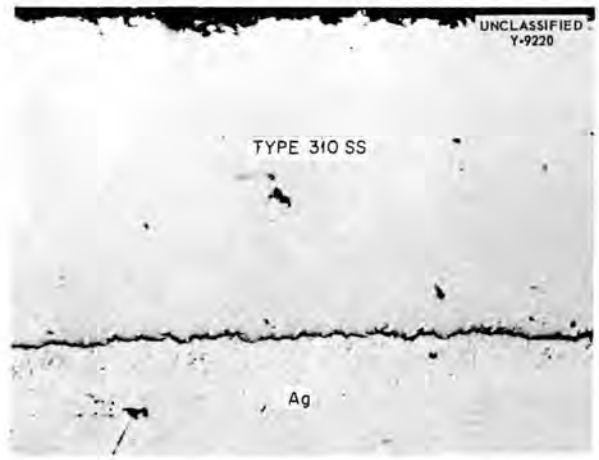

Fig. 31. Type 310 Stainless-Steel-Clad Silver After 500 hr at 1500°F in Air. As polished. 500X. Reduced 31%.

result, the protective oxide scale on these composites flaked off. The 6 and 8% aluminum bronzes showed no tendency to warp.

Type 310 Stainless-Steel-Clad Silver. – One series of tests of clad silver was evaluated. The problem of maintaining the calculated metal ratios was more difficult since the silver had a greater tendency toward being "squeezed out" during rolling. The composites for these tests were made by melting the silver under hydrogen and casting into a flattened stainless steel tube.

The insolubility of silver in type 310 stainless steel is shown in Fig. 31. The dark hairline at the interface resulted from the oxidation of the stainless steel by the oxygen which was dissolved in the silver, since the edge of the composite was not protected.

CONCLUSIONS

Various techniques were evaluated which would permit the use of a metal with a high thermal conductivity in air at 1500°F.

Of three broad schemes to permit the use of copper in this application, cladding the copper and alloying copper with aluminum showed the most promise.

With clad copper, interface reactions are evident; however, the extent of the reactions was not so great that serious effects would be expected under service conditions. Since interalloying was not apparent in the successful composites, the thermal conductivity is expected to be the average values of metals which comprise it. The indications are that the composites studied in this investigation will have a thermal conductivity of about 50% of that of copper and little or no variation as a function of temperature. Larger ratios of copper in these composites are possible but may be limited by the strength requirements in service.

The aluminum bronzes appear to be most satisfactory, since measurements indicate that the thermal conductivity approaches that of pure copper at about 1500°F. Furthermore, dimensional stability, ease of fabrication, and oxidation resistance are good. The major deterrent to the use of these alloys is the lack of a method for joining aluminum bronzes; the layer of Al_2O_3, which adds the oxidation resistance, also inhibits weldability or brazeability.