MAY 4 1965

BNWL-SA-141
~~BNSA-141~~

UNCLASSIFIED

ROUGH DRAFT

CONF-650419-8

MASTER

TUNGSTEN FIBER REINFORCED NICKEL COMPOSITES *

By

R. K. Robinson and P. L. Farnsworth **

Ceramic Materials Development
Ceramics Research and Development
Reactor and Materials Technology Department
Pacific Northwest Laboratories
Battelle Memorial Institute

March 15, 1965

PATENT CLEARANCE OBTAINED. RELEASE TO
THE PUBLIC IS APPROVED. PROCEDURES
ARE ON FILE IN THE RECEIVING SECTION.

- * Paper submitted for presentation at the Tenth Meeting of the Refractory Composites Working Group, April 12, 13, and 14, 1965, Atlanta, Georgia.
- ** Work performed under Contract No. AT(45-1)-1830 between the Atomic Energy Commission and Battelle Memorial Institute.

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, expressed 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, or employee of such contractor, to the extent that such employee or contractor of the Commission, or employee of such contractor prepares, disseminates, or provides access to, any information pursuant to his employment or contract with the Commission, or his employment with such contractor.

UNCLASSIFIED

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

TUNGSTEN FIBER REINFORCED NICKEL COMPOSITES

R. K. Robinson and P. L. Farnsworth

I. INTRODUCTION

Internal stresses which build up in nuclear reactor fuel elements during irradiation sometimes result in rupture of the fuel element cladding material. The development of stronger cladding materials is required if fuel elements capable of operating for longer times, at higher burn-ups, and at higher temperatures are to be produced. One of the most promising approaches to increasing the strength (particularly at high temperatures) of cladding materials is the development of fiber-reinforced composites. Numerous methods exist for consolidating fiber-reinforced composites into dense, useful materials having a variety of applications. One rather unique powder metallurgy technique is a high energy-rate forming method known as pneumatic impaction. While pneumatic impaction is not new, per se, some of its applications in cermet and composite consolidation and forming at Battelle-Northwest are new. This report deals with some preliminary results of reinforcing unalloyed nickel matrices with tungsten fibers by pneumatic impaction.

II. SUMMARY AND CONCLUSIONS

Composite plates containing nominally 10 volume percent continuous, aligned tungsten fibers (0.001 inch diameter) in pure nickel matrices were successfully prepared by pneumatic impaction. Good bonding between fibers and matrix and high densities (~99 percent of theoretical) were achieved during impaction. Little or no breakage of the fibers occurred during fabrication and, for the most part, the fibers remained straight throughout the specimen length. Transverse sections of the composites indicated a random distribution of the fibers although they were loaded in an ordered array.

Significant increases in both the yield strength and the ultimate tensile strength were obtained in the continuous, axially aligned fiber reinforced nickel composites.

TABLE I

PRELIMINARY TENSILE STRENGTH DATA FOR PNEUMATICALLY IMPACTED
TUNGSTEN FIBER REINFORCED NICKEL COMPOSITES

<u>Sample No.</u>	<u>Material</u>	<u>Test Temperature (°C)</u>	<u>0.2% Yield Strength (psi)</u>	<u>Ultimate Tensile Strength (psi)</u>	<u>Elongation (%)</u>	<u>Vol% Tungsten Fibers(.001 dia)</u>
C-N-1	Ni (unalloyed)	25	34,400	53,300	40	0
C-N-2	Ni (unalloyed)	1050	3,100	3,450	6	0
4D-N-1	Ni-Aligned W Wires	25	58,100	61,400	12	9.4
4D-N-2	Ni-Aligned W Wires	1050	4,100	4,100	2	0.7
4D-N-3	Ni-Aligned W Wires	1050	19,000	19,000	8	9.2
1A-NW-2	Ni-Random W Wires	25	34,800	53,600	30	1.
1A-NW-1	Ni-Random W Wires	1050	3,300	3,400	5	1.

(Table I). At room temperature, the yield strength (0.2% offset) of the fiber reinforced nickel was approximately 1.7 times that of an unalloyed impacted nickel standard. At 1050°C (1925°F), both the yield strength (0.2% offset) and the ultimate tensile strength of the reinforced nickel were about 5.5 times that of unalloyed nickel.

At room temperature, fractures in the tungsten fibers do not propagate through the nickel. Many fractures of the tungsten fibers occur before the composite fails. Some localized shear on the nickel-tungsten interface occurs near the tungsten wire fracture area.

At 1050°C (1925°F), fracture in the tungsten fibers does propagate through the nickel. The nickel fails intergranularly. Diffusion between the nickel matrix and tungsten fibers is appreciable during testing at 1050°C.

Small additions (~1 vol% of randomly oriented, chopped tungsten fibers (0.001 inch diameter) did not appreciably affect the strength of pneumatically impacted nickel.

III. PREPARATION OF COMPOSITES

A. Loading Technique

Five plates, each approximately 2-1/2 inches in diameter and 0.1 inches thick, were consolidated simultaneously in the same pneumatic impaction billet. Two plates contained 10 volume percent axially aligned, continuous tungsten fibers in a powdered nickel matrix. The tungsten fiber was General Electric Company's cleaned and straightened drawn wire, 0.001 inch in diameter, with a room temperature ultimate tensile strength of 456,000 psi. The nickel matrix powder was a 50:50 mixture of International Nickel Company's Type 128 (7-9 micron) and Type 340 (<44 micron) nickel powder. Both nickel powders have average typical purities greater than 99.6 % Ni and were produced by the thermal decomposition of nickel tetracarbonyl. A layout of the materials used in preparing the composite specimens is shown in Figure 1.

Alignment of the tungsten wire was achieved by uniformly winding the wire on a mild steel mold using a modified coil winder (Figure 2). One layer having approximately 425 turns per inch was made, then mild steel shim stock (0.005 inches thick)

FIGURE 1. Layout of materials used in preparing fiber-reinforced composites.

FIGURE 2. Modified coil winder used to align and space .001 inch diameter W-wire.

was inserted at the winding studs, and the process was repeated until the thickness of the coil was about 0.2 inches. Assuming no slippage or sagging occurred, the cross section of the wire-wound blank was a rectangular array of 0.001 inch diameter wires spaced 1.35 mils laterally and 5 mils vertically. The extra vertical space was allowance for densification during impaction.

The wire-wound blank was placed in a 310 stainless steel impaction container and unalloyed nickel powder was vibrated into the spaces around the fibers using a 750-pound capacity, 5-5000 cps, electrodynamic Savage vibrational compactor (Figure 3). The as-loaded density was approximately 50 percent of theoretical.

Molybdenum and mild steel spacers were inserted between successive layers. A layer of pure nickel powder (50:50 mixture) was added as a standard. The next layer was composed of 0.005 inch diameter, chopped (1/4-1/2 inch long) tungsten fibers randomly dispersed throughout pure nickel powders. The fiber content of this layer was estimated at 1 vol%. The top layer consisted of numerous 2 inch long pieces of 0.001 inch diameter tungsten wire randomly dispersed throughout the nickel powder matrix. The fiber content of this layer was estimated at 1 vol% fiber.

B. Consolidation Technique

Pneumatic impaction is a high energy-rate forming process initiated by rapid expansion of compressed gases to accelerate a heavy ram into the work piece which is located in a closed die. The die and work piece are located in a bolster plate which simultaneously moves toward the accelerating ram. The rapid deceleration of the ram and bolster plate impact tremendous energy to

FIGURE 3. Electrodynamic vibration compaction unit.

the work piece in the form of high pressures (2,400,000 psi). The work piece is usually preheated to increase the plasticity of the materials to be consolidated and formed. The combination of pressure and temperature together with proper tooling design and materials preparation can produce dense, intricately shaped parts to close tolerances from many metals, refractory materials, and composites. The pneumatic impactation equipment used in this work was a General Dynamics Model 1220B Dynapac machine. A schematic diagram of this equipment is illustrated in Figure 4.

The entire assembly was placed in a 310 stainless container, which was closed by welding. The can was evacuated to 0.5 mm Hg, preheated to 1100°C, and pneumatically impacted at 255,000 psi. The assembly was air cooled to room temperature and the nickel and composite plates were machined out of the can. Density measurements on the pure nickel standard taken at the edge and center of the plate yielded densities of 98.53 and 99.10 percent of theoretical, respectively.

Each plate was surface ground to a final thickness of 0.050 inches and tensile specimens were machined according to the sketch in Figure 5. After machining, the tensile specimens were polished by hand with emery cloth and degreased in ethyl alcohol.

IV. TESTING OF COMPOSITES: RESULTS AND DISCUSSION

Preliminary tests of tensile specimens machined from the pneumatically impacted composites showed promising results. Specimens of (1) pure nickel, (2) nickel containing nominally 10 vol% tungsten fibers aligned parallel to the specimen axes, and (3) nickel containing nominally 1 vol% randomly oriented tungsten fibers were

PNEUMATIC IMPACTION MACHINE

FIGURE 4. Schematic of pneumatic impact machine used to consolidate W-fiber reinforced nickel composites.

G-102-784

STANDARD FIBER REINFORCED
COMPOSITE TENSILE SPECIMENS

FIGURE 5. Sketch of standard tensile specimens used in testing.

tested at room temperature and at 1050°C (1925°F). An Instron universal testing machine equipped with a Kanthal wire-wound resistance tube furnace was used. A means for protecting the specimens with inert gas (argon or argon-8% hydrogen) during testing at 1050°C was provided.

The specimens were pinned (using 0.125 inch diameter Inconel 750 pins) to Inconel 750 pull rods. All tests were made at a machine cross-head speed of 0.02 inch per minute. This corresponds to an effective strain rate in the 0.5 inch gauge length of about 0.04 in./in./min.

The results of the room temperature tests are shown in Figure 6. Although the ultimate strengths of the three specimens are about the same, the yield strength of the specimen containing the aligned tungsten fibers (specimen 4D-N-1) is significantly higher than the yield strength of the other specimens. The jogged behavior of the curve for specimen 4D-N-1 (containing aligned tungsten fibers) is attributed to non-propagating fracture of the individual tungsten fibers. Cracks in the wires do not propagate through the nickel matrix until many fibers have failed. These conclusions are confirmed by metallographic examination of the failed specimen. Figure 7 shows a macroscopic view of a longitudinal section through the fracture area, and Figure 8 shows a higher magnification view of the tungsten fibers in the nickel matrix. Note the fractured tungsten fibers and the absence of fracture propagation through the nickel.

The formation of strong bonds between the tungsten fibers and the nickel matrix during fabrication is indicated by these results. Strong, intimate bonds are required for the efficient transfer of tensile loads from the nickel to the tungsten fibers. If good bonds are not formed, crack nucleation would be expected

<u>Sample No.</u>	<u>Material</u>
4D-N-1	Pneumatically impacted Ni containing 9.4 vol% W fibers aligned parallel to specimen axis.
1A-NW-2	Pneumatically impacted Ni containing 1 vol% W fibers randomly oriented.
C-N-1	Pneumatically impacted pure Ni.

FIGURE 6. Room temperature stress-strain curves for pneumatically impacted nickel and nickel-tungsten fiber composites.

FIG. 7. Fracture area in W-fiber reinforced Ni.
Specimen (4D-N-1) tested at room temperature. 36X

FIG. 8. Fractured W wires in W-fiber reinforced Ni.
Specimen (r-D-N-1) tested at room temperature. 100X

to occur in the nickel rather than in the tungsten, and yield behavior similar to that for pure nickel would be expected.

The wide cracks in the tungsten fibers and the absence of necking in the nickel adjacent to the cracks indicates that interface shear stresses due to end effects when the wires first break are sufficient to permit some interface shear near the crack sites. If no interface shear occurred, one would expect further strain (necessary to open the cracks) to be accompanied by intensive necking of the nickel in the vicinity of the crack. Experimental stress analyses of fiber-reinforced composites have shown high interface shear stress concentrations near fiber ends. Once the tungsten wires fracture, they behave as discontinuous fibers, and the generation of high interface shear stresses near the discontinuity would be expected.

The distribution of tungsten fibers in specimen 4D-N-1 is good. Figure 9 and 10 are photomicrographs of a transverse section near the fracture area.

The results of the high temperature (1050°C, 1925°F) tensile tests are shown in Figure 11. Although specimens 4D-N-2, 4D-N-3, and 4D-N-4 were all originally thought to contain nominally 10 vol% of aligned tungsten fibers, subsequent metallographic examination showed that only specimen 4D-N-3 had an appreciable fiber content (9.2 vol%). A photomicrograph of a transverse section of specimen 4D-N-3 near the fracture area is shown in Figure 12. The other specimens had few tungsten fibers in the gauge section, and the tensile test results for them are not significant, except that they fall between the results for pure nickel and those for specimen 4D-N-3. A photomicrograph of a transverse section of specimen 4D-N-2 (0.7 vol% tungsten fibers) is shown in Figure 13. Further work to obtain better fiber distributions in the composites is required.

FIG. 9. Transverse section of W-fiber reinforced
Ni specimen (4D-N-1). Tested at room temperature
36X

FIG. 10. Transverse section of W-fiber reinforced
Ni specimen (4D-N-1). Tested at room temperature.
500X

Sample No.Material and Test Atmosphere

4D-N-3

Pneumatically impacted Ni containing 10 vol% W fibers aligned parallel to specimen axis. Tested in flowing argon. Good W wire distributions

4D-N-4

Same as 4D-N-3 except containing 4.8 vol% W fibers. Tested in flowing argon -8% hydrogen.

4D-N-2

Same as 4D-N-4 except containing 0.7 vol% W fibers. Tested in air.

C-N-2

Pneumatically impacted pure Ni. Tested in air.

1A-NW-2

Pneumatically impacted Ni containing 1 vol% W wires. Randomly oriented. Tested in flowing argon-8% hydrogen.

FIGURE 11. Elevated temperature (1050°C, 1925°F) stress-strain curves for pneumatically impacted nickel and nickel-tungsten fiber composites.

FIG. 12. Transverse section of 9.2 vol% W-fiber reinforced Ni specimen (4D-N-3).
Tested at 1050°C (1925°F). 36X

FIG. 13. Transverse section of 0.7 vol% W-fiber reinforced Ni specimen (4D-N-2).
Tested at 1050°C (1925°F) 36X

The fracture behavior of the tungsten fiber reinforced nickel composites is dramatically different at 1050°C (1925°F) than at room temperature. Figure 14 shows a macroscopic view of a longitudinal section through the fracture area of specimen 4D-N-3 (good tungsten wire distribution; tested at 1050°C), and Figure 15 shows a higher magnification view of the same area. Compare Figure 15 (tested at 1050°C) with Figure 8 (tested at room temperature). No widespread fiber fracturing is observed in the material tested at 1050°C. (1925°F). The first fracture in the tungsten fibers apparently propagates across the entire specimen. Failure of the nickel matrix material appears to be intergranular. The nickel grain boundaries are apparently not strong enough at 1050°C to support the tensile stresses which are generated when the tungsten fibers break, and they open up (i.e., the crack propagates) at a lower stress than required for shear on the interface. There is some indication that shear on the tungsten-nickel interface may be impeded by interdiffusion of the materials at this temperature. Figure 16 shows a high magnification view of a tungsten-nickel interface in the tested specimen. Note the reaction zone. Further work is required to evaluate the effect of such reactions on the fracture mechanics and strengths of these composites.

The lower ductility of the pure nickel at 1050°C (compared to room temperature) is believed to be due to excessive grain growth at 1050°C. Figure 17 shows rupture sections of pure nickel specimens tested at room temperature and at 1050°C. Extensive grain growth has occurred in the specimen tested at 1050°C. The grains in the pure nickel specimen tested at 1050°C are also much larger than the grains in the nickel matrix material in the tungsten fiber reinforced specimen tested at 1050°C (see Figure 14). The tungsten reinforcing fibers have apparently inhibited grain growth in the nickel. Further work is required to substantiate this observation.

FIG. 14. Fracture area in W-fiber reinforced Ni specimen (4D-N-3). Tested at 1050°C (1925°F). 36X

FIG. 15. W fibers in fractured W-fiber reinforced Ni specimen (4D-N-3). Tested at 1050°C (1925°F). 100X

FIG. 16. W-Ni diffusion zone in W-fiber reinforced Ni specimen (4D-N-3). Tested at 1050°C (1925°F). 500X

(a) Tested at room temperature.

(b) Tested at 1050°C (1925°F).

FIG. 17. Rupture sections of pneumatically impacted Ni specimens (C-N-1) and (C-N-2). 36X

It is necessary to emphasize that the data and conclusions reported here must be considered preliminary. In almost all cases, only one specimen was tested under any given conditions. It is possible that variations in the density and fiber distributions in the fabricated materials and variations in testing procedure or conditions, could have contributed to the results. Further experiments are required to confirm these results. Further work is also required (1) to improve fiber distributions in the pneumatically impacted composites, (2) to determine the effects of tungsten-nickel reactions on the strengths of the materials, (3) to determine the effects of the tungsten reinforcing fibers on the grain growth characteristics of the nickel matrix material, (4) to determine the effects of using larger tungsten fibers, and (5) to evaluate the effects of varying the impaction temperature and the post impaction heat treatments.