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MICROINDENTATION HARDNESS EVALUATION OF IRIDIUM ALLOY CLAD VENT SET CUPS

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MICROINDENTATION HARDNESS EVALUATION OF IRIDIUM ALLOY CLAD VENT SET CUPS

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Abstract

An iridium alloy, DOP-26, is used as cladding for $^{238}PuO_2$ fuel in radioisotope heat sources for space power systems. Presently, DOP-26 iridium alloy clad vent sets (CVS) are being manufactured at the Oak Ridge Y-12 Plant for potential use in the National Aeronautics and Space Administration's Cassini mission to Saturn. Wrought/ground/stress relieved blanks are warm formed into CVS cups. These cups are then annealed to recrystallize the material for subsequent fabrication/assembly operations as well as for final use. One of the cup manufacturing certification requirements is to test for Vickers microindentation hardness. New microindentation hardness specification limits, 210 to 310 HV, have been established for a test load of 1000 grams-force (gf). The original specification limits, 250 to 350 HV, were for 200 gf testing. The primary reason for switching to a higher test load was to reduce variability in the test data. The DOP-26 alloy exhibits microindentation hardness load dependence, therefore, new limits were needed for 1000 gf testing. The new limits were established by testing material from 15 CVS cups using 200 gf and 1000 gf loads and then statistically analyzing the data. Additional work using a Knoop indenter and a 10 gf load indicated that the DOP-26 alloy grain boundaries have higher hardnesses than the grain interiors.

INTRODUCTION

An iridium alloy is used to encapsulate $^{238}PuO_2$ fuel pellets in the General Purpose Heat Source (GPHS) package used in recent United States radioisotope thermoelectric generators (RTGs). These RTGs are generally used for deep space satellite power systems. The GPHS design operating temperature for the iridium alloy cladding is approximately 1573 K. Iridium is used because of its compatibility with the $^{238}PuO_2$ fuel and the external graphitic components. The high melting point of iridium (2720 K) as well as its excellent resistance to oxidation and corrosion[1] are also very desirable properties in this application. The iridium alloy cladding provides maximum containment of the plutonia fuel during all mission phases.[2]

The iridium alloy currently used as the fuel cladding, DOP-26, consists of 0.3 wt % tungsten, 30 to 90 wt ppm thorium, 20 to 80 wt ppm aluminum, and the balance iridium. This alloy has been used as the fuel cladding material for the National Aeronautics and Space Administration's (NASA) Galileo (mission to Jupiter) and Ulysses (mission to study Sun's polar regions) space probes. Presently, DOP-26 iridium alloy clad vent sets (CVS) are being manufactured at the Oak Ridge Y-12 Plant for potential use in the NASA Cassini mission to Saturn (see Fig. 1).

One of the CVS cup manufacturing certification requirements is to test for Vickers microindentation hardness. The intent of the microindentation hardness specification requirement is to verify further that

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the 1648 K vacuum anneal has achieved essentially full recrystallization (grain size evaluation is primary verification) and to serve as an additional quality indicator that no significant material contamination/ embrittlement has occurred (spark source mass spectroscopic analysis is primary indicator).[3] Sections from 15 annealed CVS cups were tested for Vickers microindentation hardness using 200 grams-force (gf) and 1000 gf loads. The primary reason for switching from the 200 gf test load to the 1000 gf load was to reduce variability in the test data. The mild reduction in variability that was achieved with a higher test load was accomplished by (1) minimizing the indentation diagonal measuring errors with larger indentations, (2) minimizing the effects of superficial sample preparation artifacts with increased indentation depth, and (3) providing an averaging effect of the typical microstructural features - the matrix, grain boundaries, and precipitates - with the increased indentation depth and area.[4] Another reason for switching to the 1000 gf test load was to establish testing parameter consistency between the cups and the wrought/ground/stress relieved blankstock which has always been tested using a 1000 gf load. Use of a 1000 gf test load also permits full compliance with the required specification, American Society for Testing and Materials (ASTM) E 92-82 ("Vickers Hardness of Metallic Materials").[5]

The original microindentation hardness specification limits of 250 to 350 Vickers hardness (HV) were for 200 gf testing. The DOP-26 iridium alloy, like many other materials, exhibits microindentation hardness load dependence[4]; therefore, new limits were needed for 1000 gf testing. Initially, the intent was to establish an upper specification limit only. Further consideration dictated that both lower and upper limits be established for 1000 gf testing, preferably with the same 100 HV range that was used for 200 gf testing. The principal reason for keeping both lower and upper limits as well as the same range was to maintain the same production process quality control check. To establish these new limits testing had to be done at both loads on numerous specimens from different ingots.

Earlier efforts to determine whether the Ir_5Th precipitates present in the DOP-26 alloy are harder or softer than the matrix were inconclusive. Follow-up work for this study addressed grain boundary versus grain interior hardness. This was done using a Knoop indenter and a 10 gf test load.

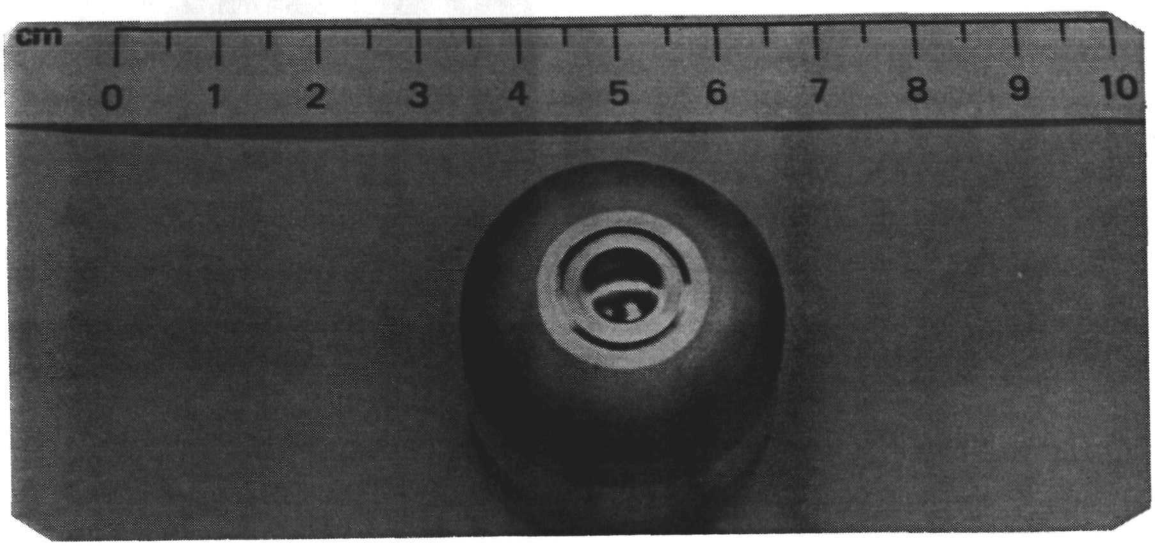

Fig. 1. DOP-26 iridium alloy clad vent set.

METHODS

Specification Limit Change Study. The general manufacturing sequences for both the iridium alloy blanks and the clad vent sets have been described previously.[4] A total of fifteen CVS cups (each from a different heat treat run) were given the standard vacuum (1.3 x 10^{-2} Pa) anneal at 1648 K for 1 hour to achieve full recrystallization. These cups represented material from five different 63 mm diameter ingots. They were then electrical discharge machined (EDM) into sections as shown in Fig. 2. These sections were metallurgically evaluated for grain size, porosity level, and inclusion content to ensure that no anomalous conditions existed before testing for microindentation hardness. The average ASTM grain sizes ranged from 6 to 8 (~20 to 50 μm nominal diameter) while the porosity and inclusion contents were extremely low.

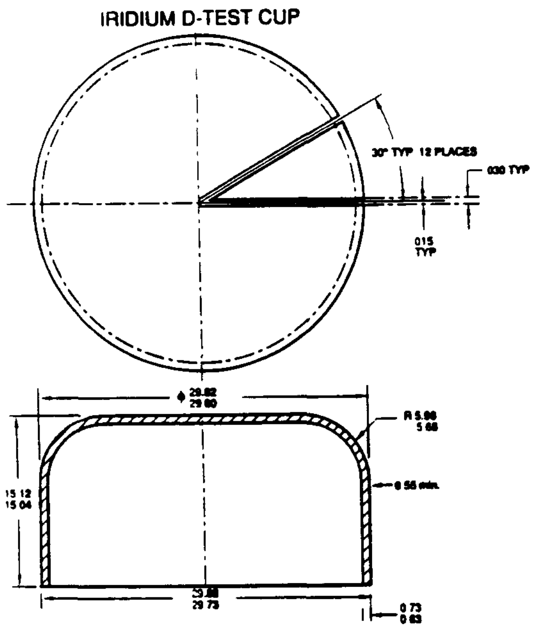

Fig. 2. Clad vent set cup destructive test sections (dimensions in mm).

Microindentation hardness testing was performed with a square-based pyramidal diamond indenter using applied loads of 200 gf and 1000 gf according to ASTM E 92-82.[5] The testing instrument was a Wilson Tukon Model 200 microindentation hardness tester. A 15 s load duration was used for all tests. The indentation diagonals were measured using a video monitor at approximately 1250X magnification. The average of three hardness values equivalently normal to the centerline were taken at the pole, radius, and equator of at least four sample sections (early testing was done with six sections) from each cup. The hardness indentations were made after the specimen surfaces were polished, electrolytically etched, and then lightly re-polished with a vibratory polisher.

Grain Boundary Versus Grain Interior Study. Additional work was done to determine whether the grain boundaries are harder or softer than the grain interiors. A cup section (from cup 3625-00-2360/blank E3-9-4) was tested using the same equipment that was used for the specification limit change study. The testing parameter changes were (1) a Knoop indenter; (2) a test load of 10 gf; (3) the indentation diagonals were oriented parallel to the section centerline (followed cup/grain flow contour) in each pole, radius, and equator position; and (4) the sections were given a final light electrolytic etch to reveal the grain boundaries. Ten indentations were made at the grain boundaries (with the short diagonal parallel to the boundary thickness or width) and ten within the grain interiors in each pole, radius, and equator position.

RESULTS AND DISCUSSION

Specification Limit Change Study. The product non-conformance criteria are based on any of the cup section positional averages (hereinafter referred to as "individual values") being outside the specified limits. There were 192 individual values from the 15 cups tested. These individual values for both test loads were statistically analyzed. A summary of the statistical analysis by test load follows.

Statistical Summary of 200 gf Test Load Data - see Fig. 3

Data passed the test for normality

* Mean - 282.81 HV Standard Deviation - 14.66 HV
* Using 3-sigma limits, projected specification limits would be 238.8 to 326.8 HV
* Observed minimum and maximum individual values - 242.1 and 328.5 HV

Statistical Summary of 1000 gf Test Load Data - see Fig. 4

Data did not pass the test for normality

* Mean - 248.31 HV Standard Deviation - 10.98 HV
* Using percentile method 98% of data found between 229.6 and 287.1 HV
* Observed minimum and maximum individual values - 228.6 and 288.8 HV

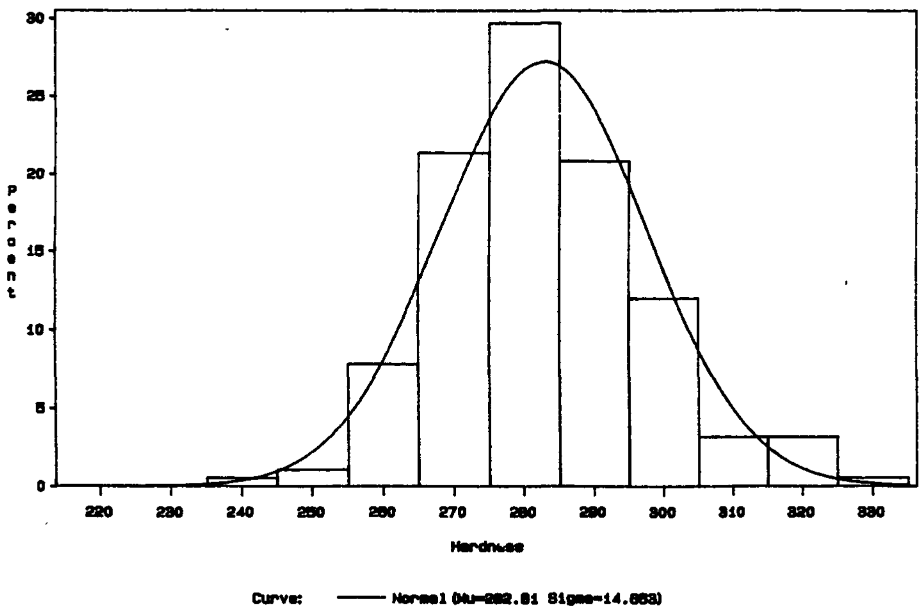

Fig. 3. Distribution of hardness test values for 200 gf test load.

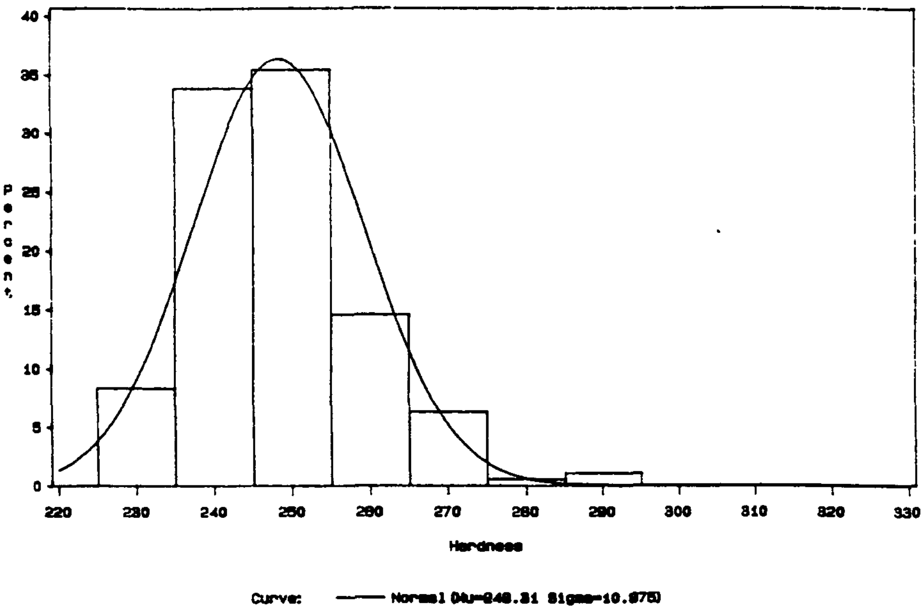

Fig. 4. Distribution of hardness test values for 1000 gf test load.

No statistical differences were found between positions (pole, radius, and equator) at either test load. Also, no consistent relationships were found for parts from the same ingot at either test load. Previous GPHS production data (for Galileo and Ulysses CVS hardware) reported by EG&G Mound Applied Technologies, Inc. (EG&G-MAT) had a mean and standard deviation (for 92 cup sections from 46 cups tested using a 200 gf load) of 291.7 and 17.1 HV, respectively. The low and high EG&G-MAT individual values were 257 and 333 HV (after eliminating a low outlier of 242 HV and two high outliers from the same part of 363 and 383 HV). These data compare favorably with the present 200 gf statistical results. With the present and historical 200 gf results being comparable the present 1000 gf results are expected to be valid for establishing new 1000 gf specification limits.

The upper 3-sigma limit for the Energy Systems 200 gf test load data, ~327 HV, indicates that the upper specification limit of 350 HV has a 23 HV "buffer" in it. Applying a buffer of 23 HV to the 99th percentile data generated from 1000 gf testing yields an upper limit of 310 HV. The lower 3-sigma limits for both the Energy Systems and EG&G-MAT 200 gf test load data (~238 and ~240 HV respectively) indicate that occasionally certification values would fall below the apparently arbitrary lower specification limit of 250 HV. The 99th percentile lower limit for the Energy Systems 1000 gf test load data of ~229 HV indicates that all certification values are expected to be above a 210 HV lower limit. The basis for selecting a rather arbitrary lower limit of 210 HV is to maintain a consistent 100 HV range between the 200 gf and the 1000 gf test load specification limits.

Four iridium alloy samples which were not fully recrystallized after annealing at 1573 K for 1 h (instead of the production annealing temperature of 1648 K) were tested for microindentation hardness at both 200 gf and 1000 gf loads. This was done to verify that 310 HV is an appropriate upper limit for 1000 gf testing. The samples were from cups formed from blanks C1-8-1 and BR6-8-5. The 200 gf hardness values were consistently above 350 HV when testing in the not-fully-recrystallized areas of each sample, whereas the 1000 gf hardness values were consistently above 310 HV. These results verified that both the 200 gf test load upper limit of 350 HV and the new 1000 gf test load upper specification limit of 310 HV will indeed screen out material that is not fully recrystallized.

<u>Grain Boundary Versus Grain Interior Study.</u> The average and standard deviation Knoop hardness (HK) values for the grain boundaries and grain interiors are shown in Table 2. The data show that the grain boundary averages are 45 to 75 HK (~10 to 20%) higher than those for the grain interiors. These results are not unexpected.[6-8] Note, however, that for the Ref.8 data the portion of the microindentation hardness difference between the grain boundaries and interiors that can be ascribed to test load (5 versus 500 gf) dependence is not known. The standard deviations (Table 2) for the grain boundaries are also larger than those for the grain interiors. A Knoop indenter was used with the lowest load available, 10 gf, to limit the indentation area for the grain boundary testing. Generally, the measured Knoop diagonals were 17 to 20 μm long; therefore, assuming ideal indentations, the short diagonals were 2.4 to 2.8 μm long. Other work[6-8] indicates that the short diagonal should have been small enough to test the "grain boundary hardness." The larger standard deviations for the grain boundary data give an indication of the difficulty in getting the long Knoop indenter diagonal fully aligned on a grain boundary.

Interestingly, a number of similarities exist between the iridium DOP-26 alloy doped with thorium and Ni_3Al doped with boron, including the following: (1) the reduction of the intrinsic brittleness of the pure materials with their respective dopant additions,[9] (2) dopant segregation to the grain boundaries,[9,10] and (3) second phase particle formation.[9,11] Boron, however, is reported to reside interstitially[12] in solution (up to 0.35 wt%[9]) with Ni_3Al, whereas thorium probably resides substitutionally if it has any solubility (<30 ppm[11]) in iridium. Doping Ir-0.3 wt% W alloys with thorium tends to increase strength and ductility values slightly when testing at conventional strain rates from room temperature to 1523 K whereas the effect of boron additions in Ni_3Al appear to have a more significant effect at conventional strain rates, but the properties of both alloys are temperature and grain size dependent.[13,14] For the DOP-26 alloy both the grain boundary thorium and the Ir_5Th precipitates are necessary for the desired high temperature impact ductility which is achieved through increased grain boundary cohesive strength and grain size control.[10,11] Unfortunately, unpublished research by Liu (1981) indicates that there is no correlation between room temperature microindentation hardness (200 gf test load) and impact (61 m/s) ductility at 1250 K.

Table 2. Grain boundary versus grain interior 10 gf knoop hardness comparison.

POSITION	GRAIN BOUNDARY	GRAIN INTERIOR
Equator	449 ± 52	404 ± 29
Radius	419 ± 55	355 ± 34
Pole	425 ± 59	350 ± 41
All Combined	431 ± 55	370 ± 42

<u>CONCLUSIONS</u>

Based on this work the following conclusions are made:

1. The previous 200 gf test load specification limits of 250 to 350 HV for the DOP-26 iridium alloy are equivalent to the new 210 to 310 HV limits for 1000 gf testing.

2. The room temperature Knoop microindentation hardnesses (10 gf test load) of the DOP-26 alloy grain boundaries are 10 to 20% higher than the grain interiors.

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