2

Y-12

Y/DV-1321

OAK RIDGE
Y-12
PLANT

MARTIN MARIETTA

THE METALLURGICAL INTEGRITY OF THE FRIT VENT ASSEMBLY DIFFUSION BOND

G. B. Ulrich

June, 1994

MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

Date of Issue: April 26, 1994 Y/DV-1321

THE METALLURGICAL INTEGRITY OF THE FRIT VENT ASSEMBLY DIFFUSION BOND

G. B. Ulrich

Process Metallurgy Department
Development Organization

June 1994

Prepared by:
Oak Ridge Y-12 Plant
P.O. Box 2009, Oak Ridge, Tennessee 37831-8169
managed by
MARTIN MARIETTA ENERGY SYSTEMS, INC.
for the
U.S. DEPARTMENT OF ENERGY
under contract DE-AC05-84OR21400
OS

CONTENTS

LIST OF FIGURES

LIST OF TABLES

SUMMARY

The metallurgical integrity of frit vent assemblies produced by Martin Marietta Energy Systems, Inc. (Energy Systems) were compared with those produced earlier by EG&G-Mound Applied Technology, Inc. (EG&G-MAT). Scanning electron microscope (SEM) photographs were taken (at magnifications of from 126X to 1000X) of the starting frit vent powder and the diffusion-bonded powder in finished frit vent assemblies produced by Energy Systems and EG&G-MAT. Frit vent assemblies also were metallographically prepared and visually examined/photographed at magnifications of from 50X to 1000X. The SEM and metallographic examinations of the particle-to-particle and particle-to-foil component diffusion bonds indicated that the Energy Systems-produced and EG&G-MAT-produced frit vent assemblies have comparable metallurgical integrity. Statistical analysis of the Energy Systems production data shows that the frit vent manufacturing yield is 91%.

INTRODUCTION

Iridium alloy clad vent sets (CVSs) are now being made by Energy Systems at the Oak Ridge Y-12 Plant (see Fig. 1). These CVSs are being made for the U.S. Department of Energy's (NE-53) General Purpose Heat Source-Radioisotope Thermoelectric Generator (GPHS-RTG) program, which is to supply electrical power for the National Aeronautics and Space Administration's Cassini mission to Saturn. A GPHS-RTG has 72 CVSs. Each CVS encapsulates one $^{238}PuO_2$ fuel pellet. The helium gas produced from the alpha decay of the ^{238}Pu is vented through a nominal 0.45-mm-diam hole in the vent cup (see Fig. 2) of each CVS. A frit vent assembly (see Figs. 2 and 3) that is electron beam welded over the vent hole allows helium gas to escape but prevents plutonia fines from exiting.

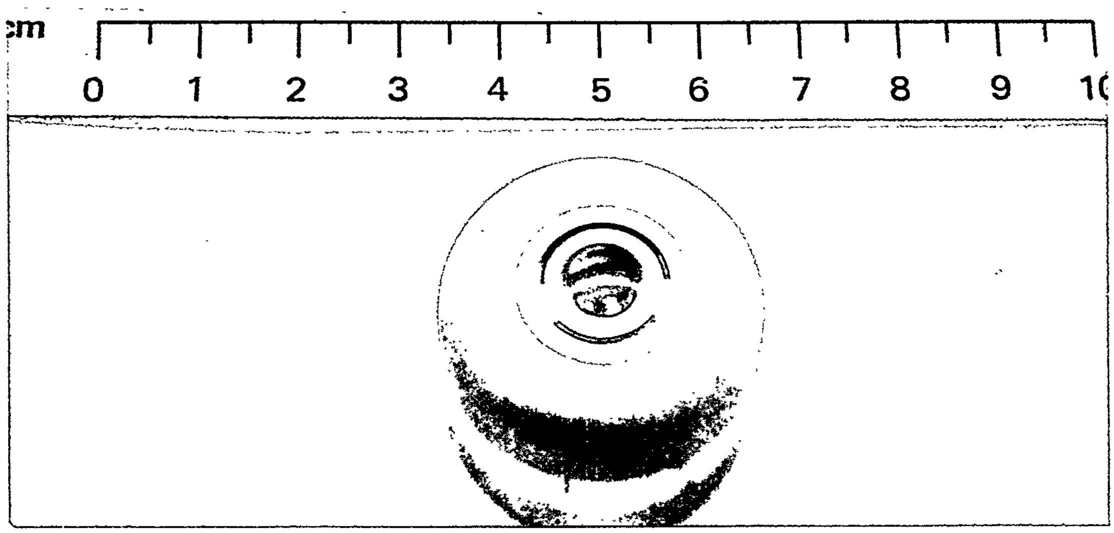


Fig. 1. Iridium alloy clad vent set.

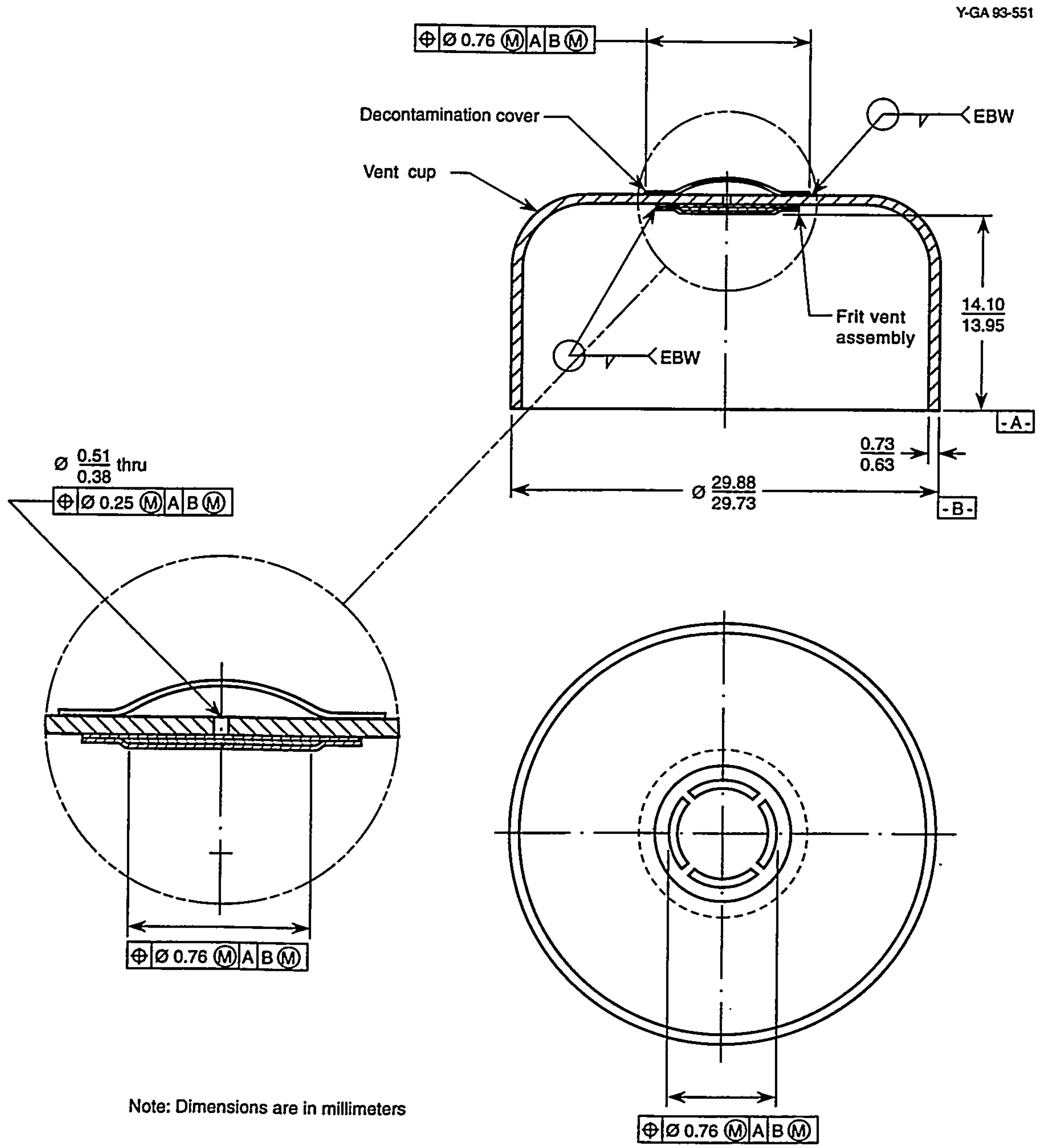


Fig. 2. GPHS-RTG vent cup assembly.

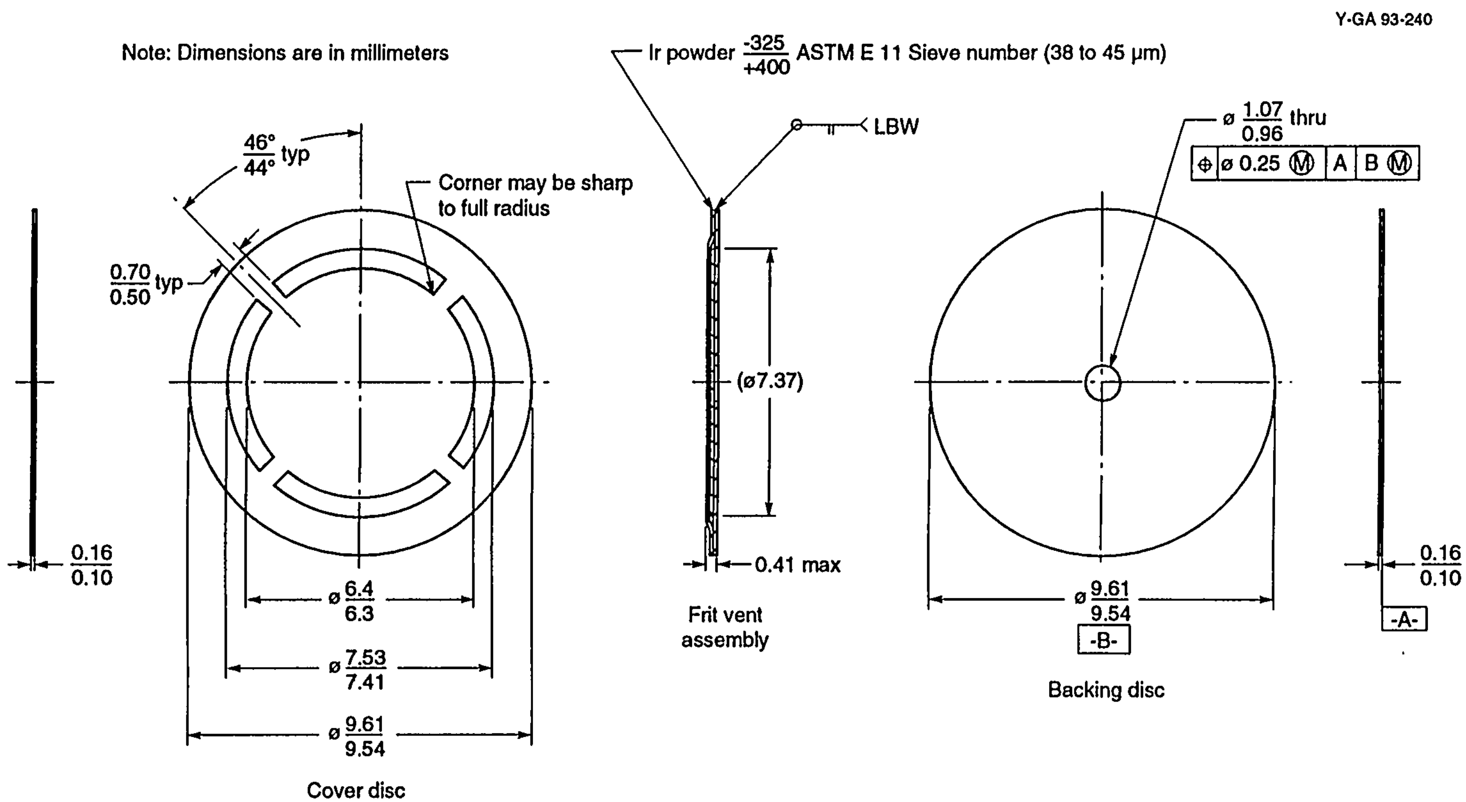


Fig. 3. GPHS-RTG frit vent assembly drawing with cover disc and backing disc.

Originally, the GPHS frit vent assembly was designed, manufactured, and certified by EG&G-MAT.[1] The only certification requirements for the frit vent assembly were a maximum assembly thickness of 0.41 mm and a helium flow rate between 4.5 and 7.5 cc/min at 1 psig input pressure at room temperature. The EG&G-MAT hardware was used for the Galileo and Ulysses missions, which were launched in 1989 and 1990, respectively. EG&G-MAT also produced CVS hardware for the various test programs to support those two missions. No reports occurred of compromised GPHS frit vent integrity from the various test programs. (See Refs. 2, 3, and 4 for results from some of the numerous test programs.)

CVS production at the Y-12 Plant was temporarily shut down from September 1992 to April 1993. During the shutdown, concern was expressed regarding the integrity of the Energy Systems-produced frit vent diffusion bond. This concern was based primarily on earlier reports of questionable powder bonding in frit vents manufactured during Energy Systems preproduction efforts (before the production quality readiness review, which was conducted in May 1990). Concern also stemmed from frit vent manufacturing items that were identified as requiring corrective action before CVS production could be restarted. The corrective actions involved (1) replacement of the aluminum edge pressing tool with a rubber pad to avoid potential aluminum contamination and cracking in cover discs and (2) intense periodic inspection of the laser weld tantalum tooling tips until (3) replacement and redesign of the laser weld tantalum tooling tips with tungsten to avoid potential tantalum contamination. Together with the fact that a significant delay would occur between fabrication of flight-quality hardware and Cassini test program results, these concerns mandated that some kind of assessment be made of the frit vent powder-bonding integrity.

Consideration was given to performing a tensile test to evaluate the powder bond. This idea was rejected, however, because of reported difficulties with a frit vent tensile test done for the Multi-Hundred Watt program,[5] precursor to the GPHS program, and the absence of any GPHS program data with which to compare the results. It was agreed that a metallographic study should be conducted to compare the metallurgical integrity of frit vent assemblies produced by Energy Systems for Cassini with those produced by EG&G-MAT for Galileo/Ulysses.

Ever since CVS production was restarted, Energy Systems has produced hardware for the Cassini mission using essentially the same design, manufacture, and certification schemes that EG&G-MAT used to produce frit vent assemblies for the Galileo and Ulysses missions. Note: the only known manufacturing (see Table 1 for the Energy Systems processing sequence) differences are that Energy Systems

1. performs an edge repress operation before laser welding,

2. omits a vacuum outgas (1500°C/1 h) operation that EG&G-MAT performed before laser welding, and

3. laser welds with a carbon dioxide (CO_2) laser source [backup welder uses neodymium:yttrium-aluminum-garnet (Nd:YAG) source] and tungsten tooling tips instead of welding with a Nd:YAG source and tantalum tips, as EG&G-MAT had done.

Table 1. Frit vent assembly processing sequence

1	Blank cover and backing discs.
2	Flatten and deburr discs.
3	Inspect discs (100% visual at 30X to 45X magnification and sampling plan for dimensions).
4	Clean discs.
5	Sinter iridium powder on backing disc (graphite tooling in vacuum at 1500°C/1 h).
6	Inspect sintered subassembly.
7	Diffusion bond backing disc, powder, and cover disc (graphite tooling in vacuum at 1900°C/2 h with ~2.25-kg load).
8	Air burn residual graphite from tooling (635°C/2 h or more).
9	Press edges (500 psi).
10	Flow test (1 psig helium at room temperature).
11	Compress (if required; typically 1500 psi applied twice)/flow test to establish proper flow.
12	Repress edges, if required, for laser welding.
13	Laser weld (CO_2 or Nd:YAG) circumferential edge.
14	Inspect frit vent assembly (30X magnification).

The Energy Systems final certification inspection is done after laser welding instead of before the air burn-off operation, as EG&G-MAT had done. The Energy Systems visual inspection is more rigorous in that 30X magnification is used to examine for blisters, stains, extraneous matter, missing powder, and cracks whereas the EG&G-MAT frit vents were visually examined with magnification only immediately after the sintering operation. Even the maximum width of the weld rollover is a requirement for Energy Systems frit vents.

For a summary of earlier work on this topic, see Ref. 1.

PROCEDURE

The iridium powder that Energy Systems uses to produce frit vents was examined and photographed by SEM at magnifications of from 126X to 1000X (Fig. 4). Two frit vents were metallographically prepared and examined in each of the following conditions:

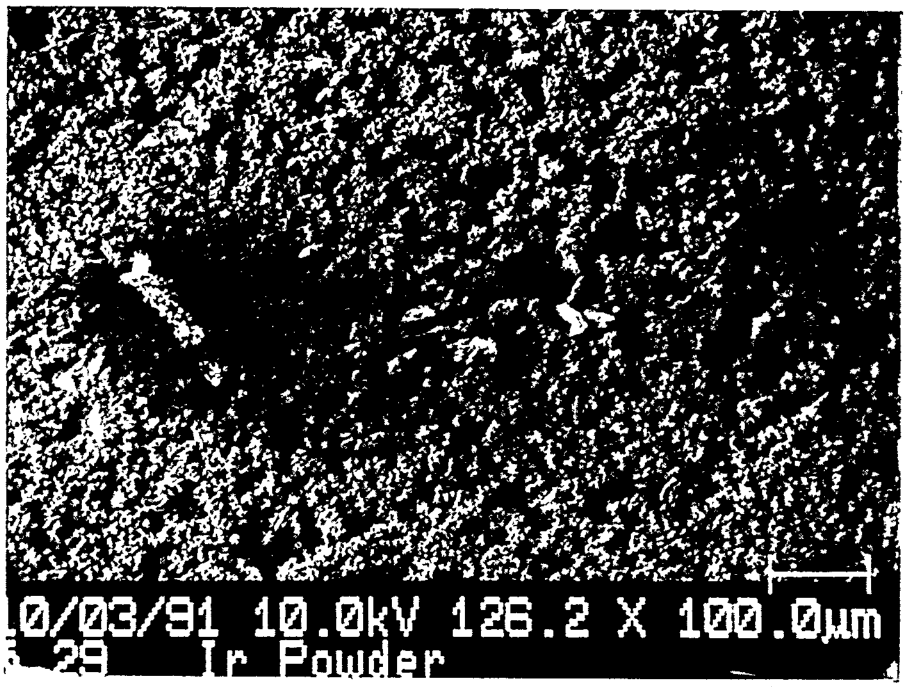
1. diffusion bonded by Energy Systems (Fig. 5),

2. pressed and compressed by Energy Systems (Fig. 6),

3. laser welded by Energy Systems (Fig. 7), and

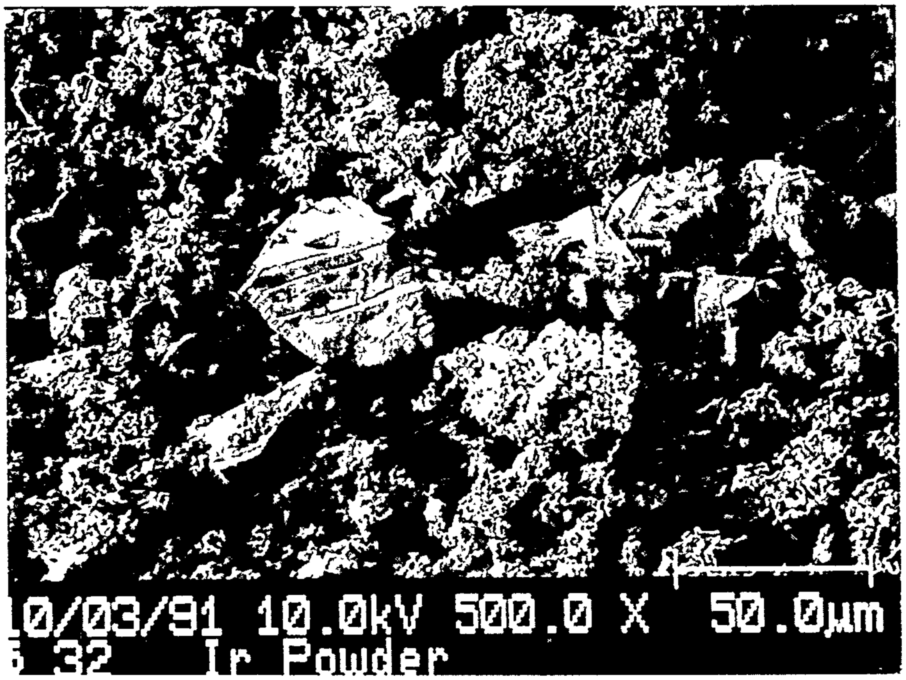
4. laser welded by EG&G-MAT (Fig. 8).

The most informative locations were photographed at magnifications of from 50X to 1000X. SEM photographs were also taken at 126X and 1000X magnifications of both Energy Systems (Fig. 9) and EG&G-MAT (Fig. 10) frit vent assemblies electron beam welded into vent cups. Data collected for frit vent assemblies manufactured since the CVS production restart were summarized statistically.

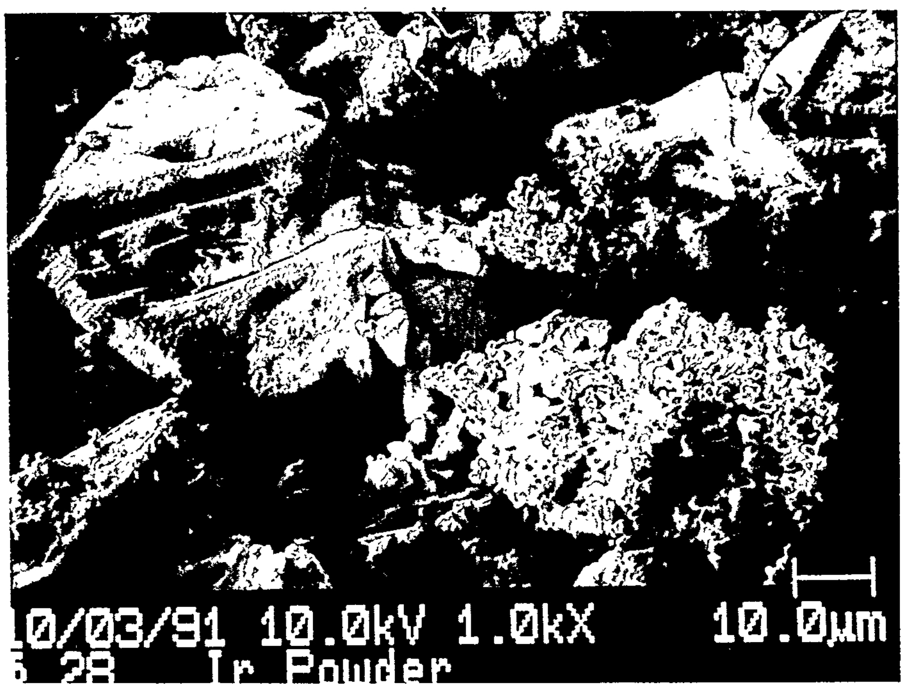


(a)

Fig. 4. Energy Systems starting iridium powder for frit vents. *(a)* 126X, *(b)* 500X, *(c)* 1000X.



(b)

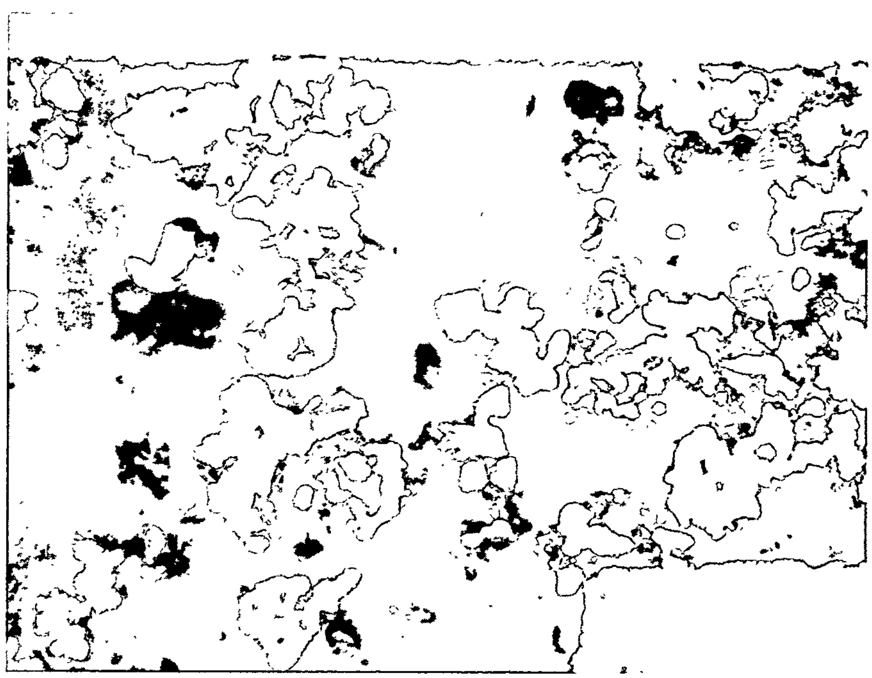


(c)

Fig. 4. Continued.

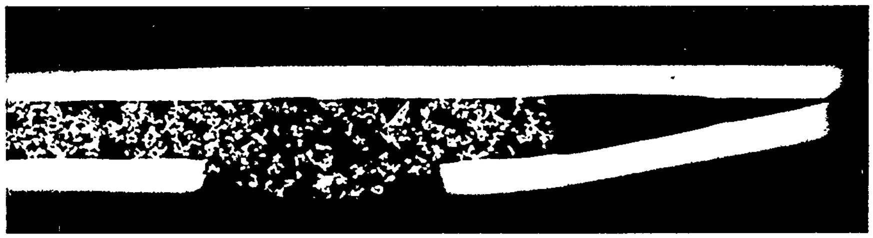
(a)

(b)

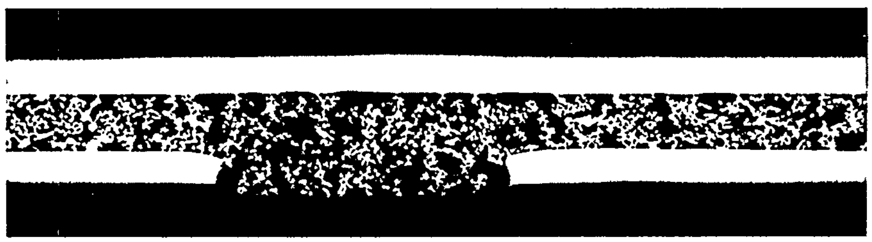


(c)

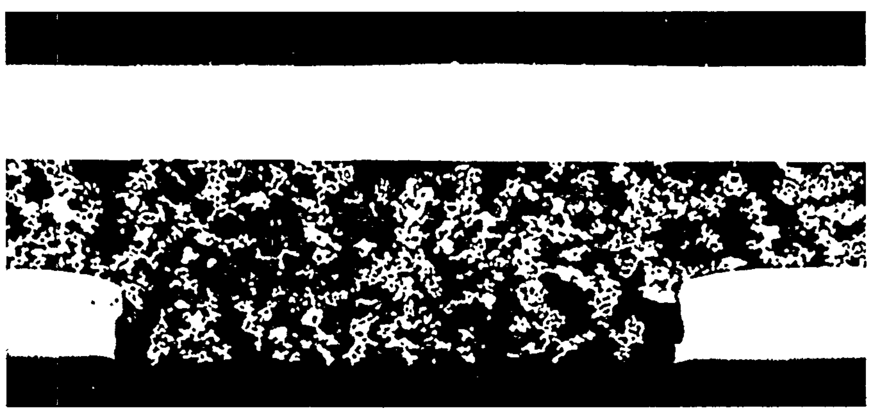
Fig. 5. Energy Systems frit vent 9752-00-3799 in the as-diffusion-bonded condition. *(a)* Backing disc hole region, 50X; *(b)* backing disc hole region, 100X; *(c)* backing disc hole region, 400X.



(a)

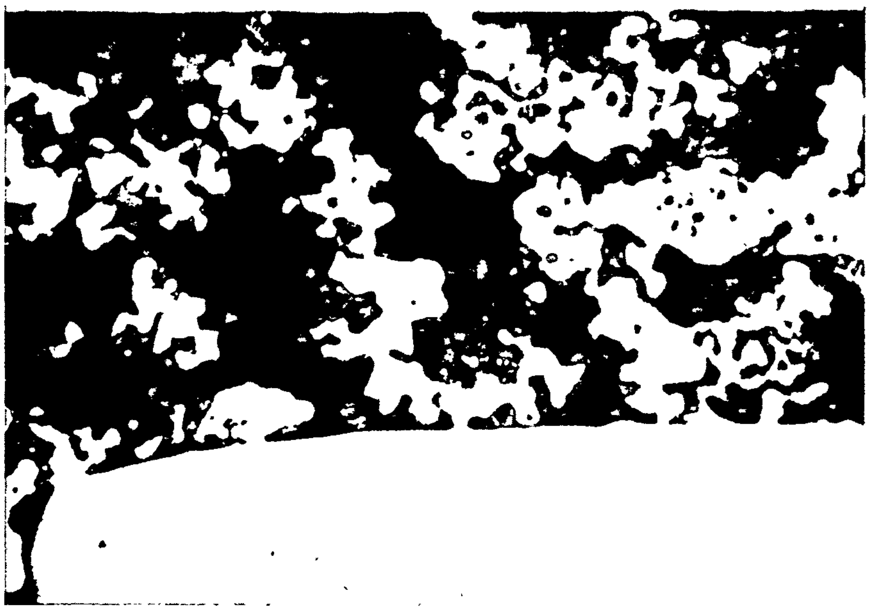


(b)

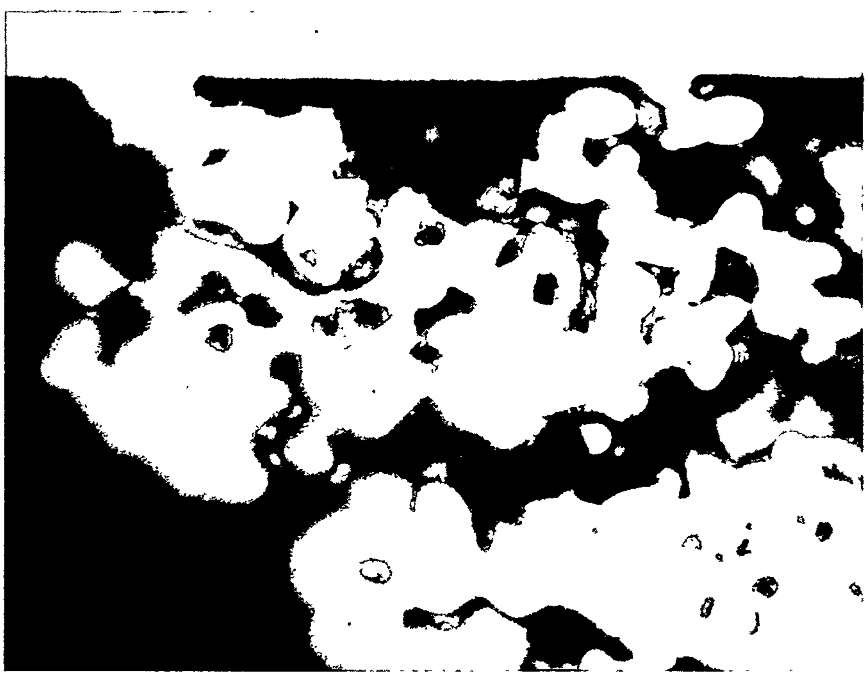


(c)

Fig. 6. Energy Systems frit vent 9752-00-3757 with edges pressed at 450 psi compressed at 1510 psi to establish flow of 5.7 cc/min. *(a)* Pressed edge and slot region with taper in cover disc from rubber pad, 50X; *(b)* backing disc hole region, 50X; *(c)* backing disc hole region, 100X; *(d)* backing disc hole region, 400X; *(e)* backing disc hole region, 1000X.

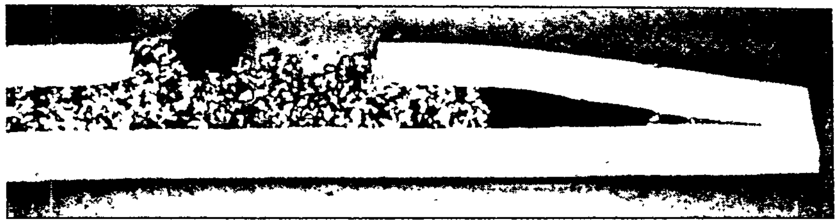


(d)

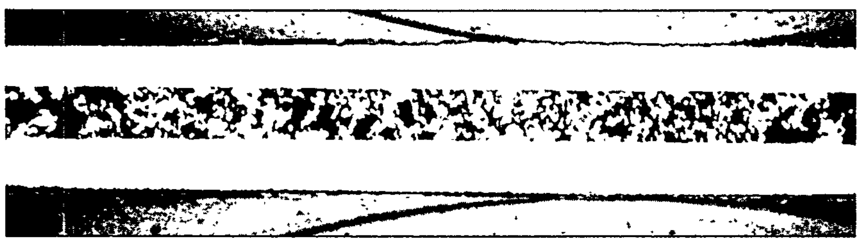


(e)

Fig. 6. Continued.



(a)



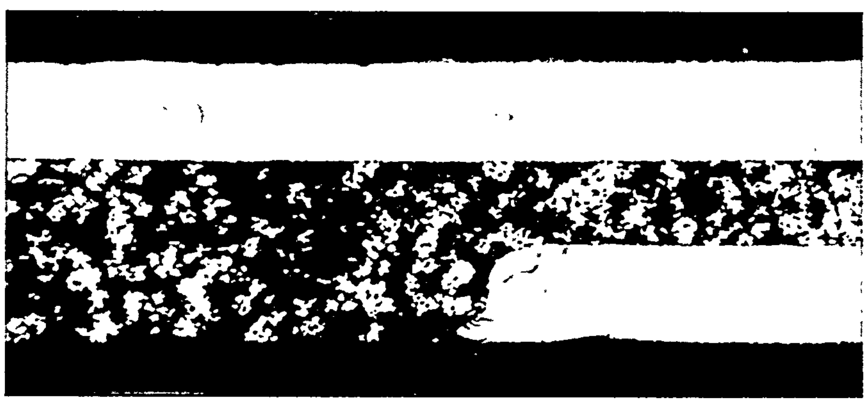
(b)

(c)

Fig. 7. Energy Systems frit vent 9752-00-3760 laser welded. Edges were pressed at 500 psi. No compression was required to establish the flow rate of 6.0 cc/min. *(a)* Laser weld and slot region, 50X; *(b)* central region, 50X; *(c)* central region, 400X.

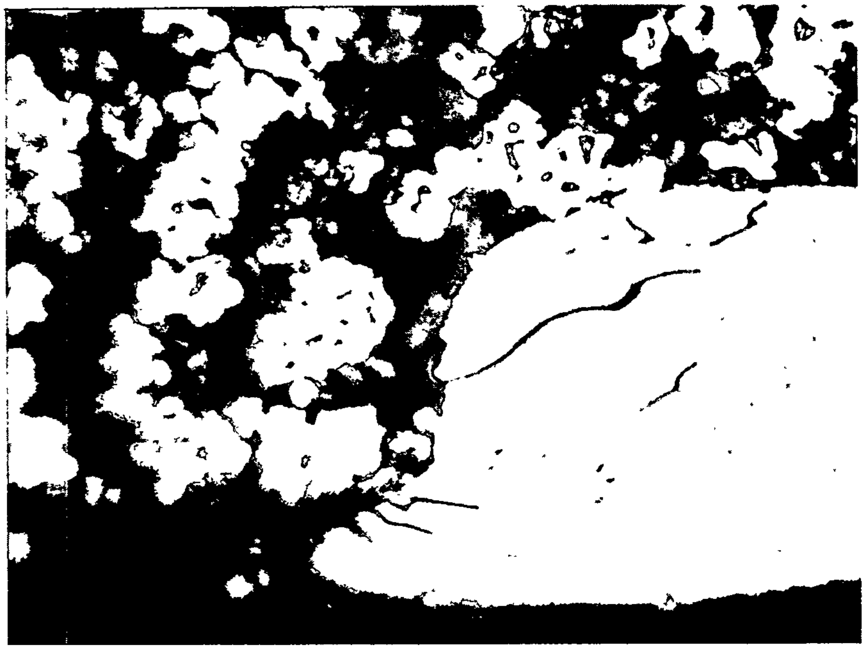
(a)

(b)



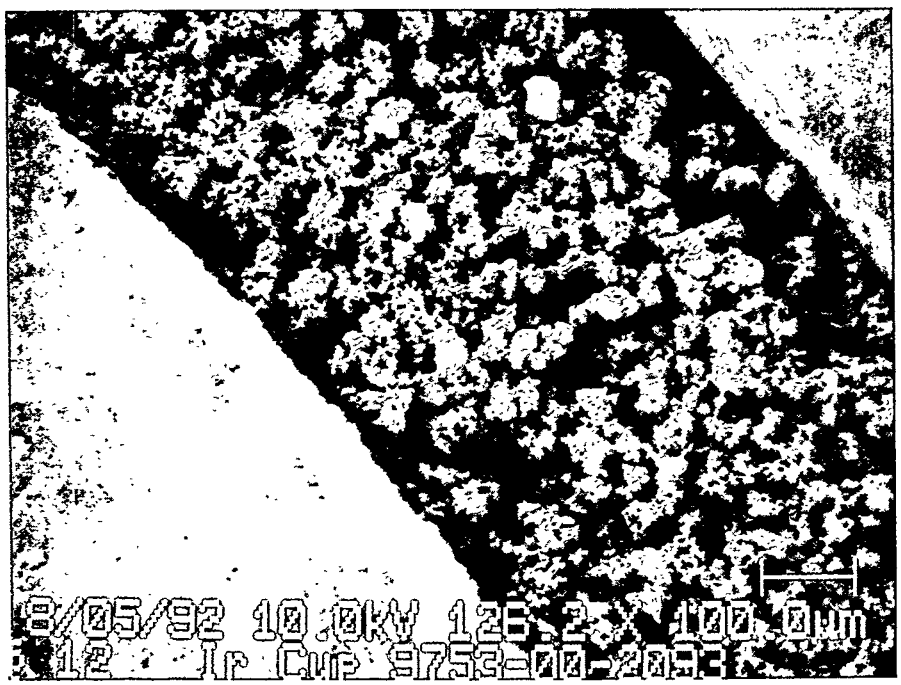
(c)

Fig. 8. EG&G-MAT frit vent P-1082 with flow rate of 4.98 cc/min. *(a)* Laser weld and slot region, 50X; *(b)* backing disc hole region, 50X; *(c)* backing disc hole region, 100X; *(d)* backing disc hole region, 400X.



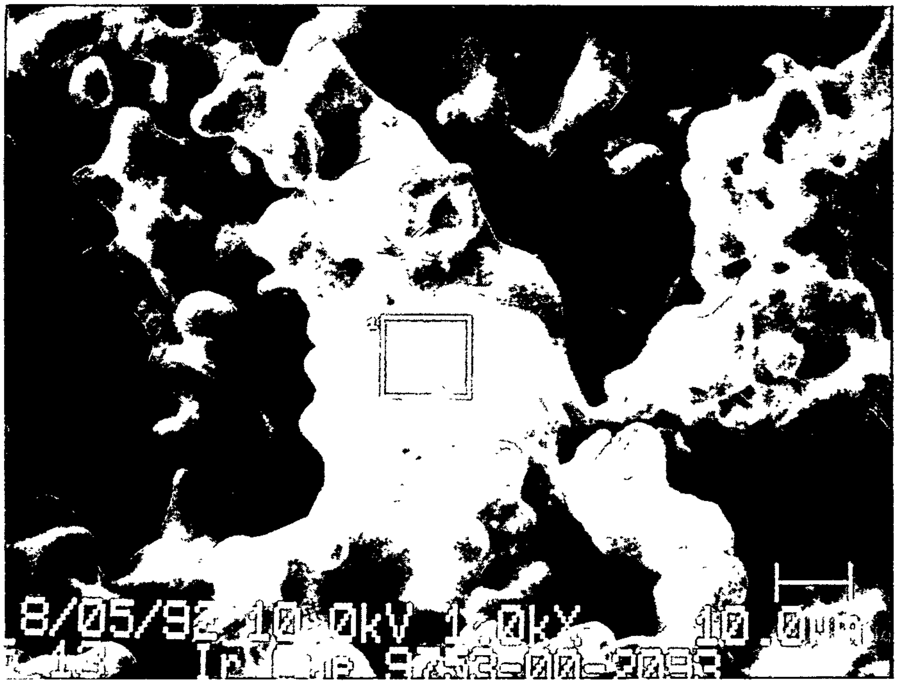
(d)

Fig. 8. Continued.



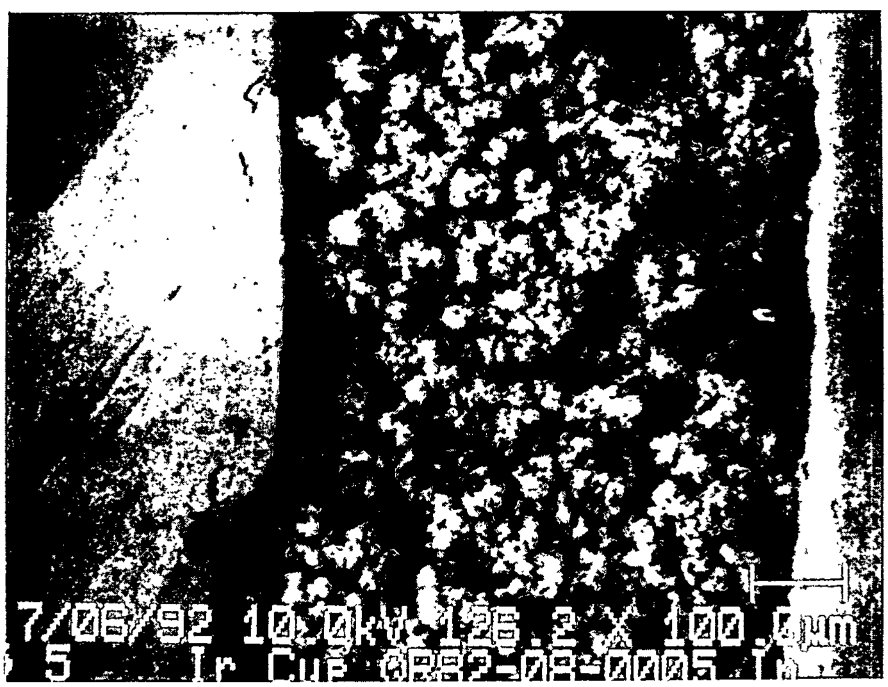
(a)

Fig. 9. Scanning electron microscopy photographs of Energy Systems vent cup assembly 9753-00-2093 showing frit vent assembly and powder after frit vent-to-cup electron-beam weld was made. *(a)* 126 X, *(b)* 1000X.



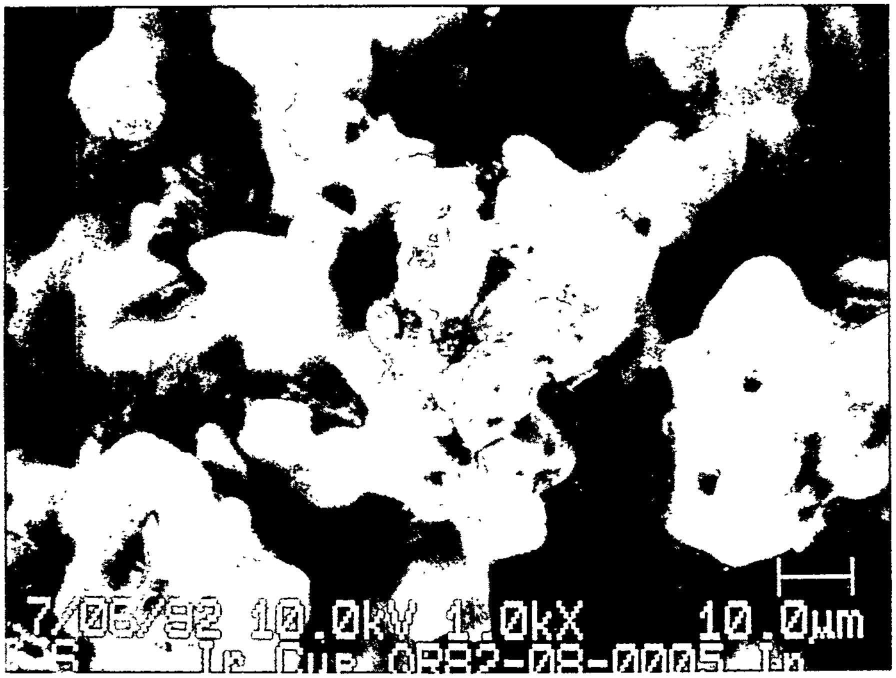
(b)

Fig. 9. Continued.



(a)

Fig. 10. Scanning electron microscopy photographs of EG&G-MAT vent cup assembly QR828-5 showing frit vent assembly and powder after frit vent-to-cup electron-beam weld was made. *(a)* 126X, *(b)* 1000X.



(b)

Fig. 10. Continued.

RESULTS AND DISCUSSION

The chemically precipitated iridium powder used for the Energy Systems frit vents is washed, screened, and certified to specification GPHS-M-187 at the Oak Ridge National Laboratory. The powder is screened to have a particle size of from 38 to 45 μm, but commonly it consists of agglomerates of very fine particles (see Fig. 4).

The photographs of the frit vents in the diffusion bonded (Fig. 5), pressed and compressed (Fig. 6), and laser-welded (Figs. 7 and 8) conditions indicate good particle-to-particle and particle-to-foil metallurgical bonding. Most importantly, the bonding is comparable between the Energy Systems product (Fig. 7) and the EG&G-MAT product (Fig. 8). The metallography work shows, as expected, the reduced density in the four cover discs slots and the backing disc hole regions.

The typical density of the powder in an as-diffusion-bonded frit vent is calculated to be 47% of the theoretical density (22.65 g/cm^3) of pure iridium. This calculation is based on the average as-diffusion-bonded frit vent thickness of 0.039 cm (see Table 2), cover disc and backing disc thicknesses of 0.013 cm each, powder weight of 0.0725 g, and powder area of 0.527 cm^2. The typical density of the powder in a finished frit vent assembly is calculated to be 67% of the theoretical density. The calculation is based on the preceding values except that the average frit vent thickness is 0.035 cm (see Table 2), which is measured after the final flow rate is established. These density differences can be seen qualitatively in Figs. 5, 6, 7, and 8. The frit vent in Fig. 7 is very unusual because no compression was required after diffusion bonding to adjust the flow rate within the specification limits. The qualitatively high powder density in this frit vent shows why no compression was required. The SEM photographs (Figs. 9 and 10) of frit vents electron beam welded into vent cups also indicate that the bonding is comparable for the Energy Systems product and the EG&G-MAT product.

Table 2. Frit vent manufacturing data summary (average ± 1 standard deviation)

Manufacturing stage	Gauge pressure (psi)	Thickness (mm)	Flow rate (cc/min)
Initial (as bonded)	-	0.392 ± 0.021	18.5 ± 7.37
Intermediate	1492 ± 258	-	9.3 ± 1.79
Final	1588 ± 238	0.352 ± 0.014	6.09 ± 0.75

The thickness, compression-gauge pressure, and flow-rate data for 130 frit vent assemblies manufactured since production was restarted in March 1993 have been summarized statistically. Table 2 shows the averages and standard deviations for the frit vent values of initial (as diffusion bonded) thicknesses and flow rates, the gauge pressures for the compression operation to lower the initial flow rates to the intermediate flow rates, and the final gauge pressures that produced the final thicknesses and the final flow rates. The raw data were analyzed by linear regression to determine if any operationally significant correlations or linear relationships existed between any of the seven "variables" in Table 2. The two best correlations were +0.88 for the two sets of gauge pressures and +0.59 for the final gauge pressures and the initial flow rates as well as for the initial flow rates and the intermediate gauge pressures. No useful linear equation could be found that would adequately predict the gauge pressure that should be used to compress a frit vent to a desired flow rate based on the initial flow rate and/or thickness.

The Energy Systems frit vent process yield before the CVS production shutdown was 96% (235/245). All of the rejections (4.1% - 10/245) were for low flow rates. The averages and single standard deviations for the 235 acceptable frit vent flow rates were 6.11 ± 0.59 cm^3/minute while those for the 196 vent cup subassemblies were 6.07 ± 0.75 cm^3/minute. The averages indicate that the flow rates of the Energy Systems vent cup subassemblies tended to be slightly lower than those for the frit vents. Before the shutdown of CVS production, the frit vent flow rate was established after the laser welding operation.

Since the CVS production restart, the process yield has been 91% (348/384). Eleven (2.9%) frit vents were rejected for missing powder, 10 (2.6%) for low flow rate, and 15 (3.9%) for miscellaneous reasons (bad welds, stains, or physical damage). The averages and single standard deviations for the 374 acceptable frit vent flow rates were 5.97 ± 0.51 cm^3/minute while those for the 310 vent cup subassemblies were 5.82 ± 0.61 cm^3/minute. None of the 310 vent cup subassemblies manufactured after the production restart have been rejected for an out-of-tolerance flow rate while more than 30 of the EG&G-MAT vent cup subassemblies had flow rates above the upper limit. The present frit vent process yield of 91% is comparable to EG&G-MAT's yield of 92% (1050 acceptable out of 1140 total)[1] for the Galileo/Ulysses missions, especially considering the more rigorous inspection required for the Cassini mission frit vents. The averages indicate that the flow rates of the current Energy Systems vent cup subassemblies tend to be lower than those for the frit vents whereas the flow rates of the EG&G-MAT vent cup subassemblies tended to be higher[6] than those for their frit vents.

The different directional changes in the flow rates for the Energy Systems and EG&G-MAT vent cup subassemblies versus frit vents may be related to the manufacturing differences described earlier. The current Energy Systems and the previous EG&G-MAT processes involve(d) laser welding the edge of the frit vent and electron beam welding the frit vent to the vent cup between the two flow rate certification operations. In addition, EG&G-MAT vacuum outgassed (1500°C/1 h) the frit vent assembly before laser welding, whereas, Energy Systems does not perform the vacuum outgassing operation. This outgassing operation most likely accounts for the different directional changes in the flow rates for the Energy Systems and EG&G-MAT products.

A technical surveillance is conducted quarterly to assess the adequacy of the frit vent laser weld and the powder bonding. Although laser weld tooling tips have been changed as a result of this surveillance, no deficiencies in powder bonding have been found.

CONCLUSIONS

SEM and metallography work indicate that the frit vents produced by Energy Systems have metallurgical integrity comparable to those produced earlier by EG&G-MAT. They are expected to perform satisfactorily in the various test programs planned to support the Cassini mission. The frit vent manufacturing yield is 91%.

The manufacturing and technical surveillance data indicate that the Energy Systems frit vent manufacturing process is consistently producing hardware that exceeds specification and procedural requirements.

ACKNOWLEGMENT

The author thanks L. Boshears for supplying the frit vent components and the production yield data. He also thanks H. L. Richards for the electron microscopy work, D. S. Johnson and E. W. Wester for the metallographic work, and M. W. Sherrill for the linear regression analyses.

REFERENCES

1. E. W. Johnson, "Current Helium Venting Technology for $^{238}PuO_2$ Heat Sources," in *Proceedings - Seventh Symposium on Space Nuclear Power Systems*, pp. 701-704, CONF-900109, Albuquerque, N.M., January 7-10, 1990.

2. E. W. Johnson, presentation to the Martin Marietta Energy Systems, Inc., In-Process Production Status Review Board, Oak Ridge, Tenn., Autumn 1992.

3. T. A. Cull, T. G. George, and D. Pavone, *General-Purpose Heat Source Development: Safety Verification Test Program (Explosion Overpressure Test Series)*, LA-10697-MS, September 1986.

4. D. Pavone, T. G. George, and C. E. Frantz, *General-Purpose Heat Source Safety Verification Test Series: SVT-1 Through SVT-6*, LA-10353-MS, June 1985.

5. *GE Multi-Hundred Watt Frit Vent Specification, NS0060-02-78*, General Electric Space Division (now Martin Marietta Astro Space Company), Valley Forge, Pa., 1973.

6. L. A. Greene, *Galileo/Ulysses Missions Frit Vent Production Date*, received from EG&G-MAT during CVS technology transfer in 1989.

DISTRIBUTION

Department of Energy - Oak Ridge

L. F. Blankner
R. L. Lahti

Department of Energy - Office of Special Applications

W. J. Barnett

EG&G - Mound Applied Technologies

W. R. Amos/E. W. Johnson (2)

Los Alamos National Laboratory

T. G. George

Oak Ridge National Laboratory

R. H. Cooper, Jr.
J. F. King
M. M. Martin
J. P. Moore
E. K. Ohriner

Oak Ridge Y-12 Plant

H. W. Berry
L.Boshears
G. L. Bowers
K. J. Helle
D. S. Johnson
K. F. Kesterson
A. K. Lee/DOE-OSTI (2)
R. A. Lynch
T. M. Mustaleski, Jr.
W. G. Northcutt, Jr./E. L. Bird
R. B. Richmond
M. W. Sherrill
S. G. Snow
G. B. Ulrich (3)
E. W. Wester
Y-12 Central Files - RC