#### SUPERALLOY MELTING AND CLEANLINESS EVALUATION

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#### Summary

The need for nickel base superalloy cleanliness levels superior to that now attainable in production has been emphasized by the adverse effects of oxide inclusions on low cycle fatigue properties. Developmental activities were conducted to evaluate various melting or remelting processes for improved ingot cleanliness levels in Rene' 95 and IN718 nickel base superalloys. The principles of the current Vacuum Induction Melt plus Vacuum Arc Remelt (VIM + VAR) production practices as well as the Electro-Slag Remelt (ESR) and Electron Beam Cold Hearth Remelt (EBCHR) processes are discussed with respect to potential sources of oxide Advantages and disadvantages for each melt practice are inclusions. Cleanliness rating results for Rene' 95 and IN718 ingot considered. materials produced by these melt practices are presented. Metallographic evaluations like the slag inclusion ratings used for steels were found to be neither practical nor reliable for the low oxide content levels currently found in nickel base superalloys. Cleanliness ratings were established using the Electron Beam (EB) button melt test. Small EB melted ingots (buttons) were produced which concentrated oxide inclusions via flotation from a larger test volume than could practically be metallographically Scanning Electron Microscope/Energy Dispersive characterized. Α Spectrum/Image Analysis (SEM/EDS/IA) system was used to quantify the size distribution and the total oxide content for the concentrated oxide The results of these SEM/EDS/IA evaluations for Rene' 95 and particles. IN718 EB buttons demonstrated that both the ESR and EBCHR practices improved ingot cleanliness over the VIM/VAR process, and that EBCHR showed the greatest potential for improved cleanliness.

#### Introduction

High performance requirements and the need for reduced costs for aircraft turbine components over the past decade have necessitated development efforts to improve the cleanliness of nickel-base superalloys. The need for superalloy cleanliness levels superior to that currently available for either conventional melt plus wrought or powder metallurgy (PM) processes has been emphasized by the documented (1,2) detrimental effects of oxide and other types of inclusions on low cycle fatigue (LCF) properties. Undesirably high life-cycle costs for these PM and conventional melt plus wrought superalloy materials can result from limiting the application life as a result of these inclusions. This paper describes efforts directed at demonstrating a clean melt practice for production of high performance nickel-base superalloys with oxide contents significantly lower than that found in current conventional melt processes.

The evaluations described in this paper were predominantly conducted using the Rene' 95 and IN718 nickel-base superalloys. The nominal chemical compositions of these two alloys are as follows:

# Alloying Element - Nominal Weight Percent

Rene' 95 
$$\frac{A1}{3.5}$$
  $\frac{Ti}{2.5}$   $\frac{Cb}{3.5}$   $\frac{Cr}{13.0}$   $\frac{Co}{8.0}$   $\frac{Mo}{3.5}$   $\frac{W}{3.5}$   $\frac{Fe}{--}$   $\frac{C}{0.06}$   $\frac{Zr}{0.05}$   $\frac{B}{0.01}$  Bal. IN718 0.5 1.0 5.1 19.0 -- 3.0 -- 17.5 0.03 -- 0.006 Bal.

The major strengthening mechanism for these alloys is precipitation hardening; Rene' 95 is strengthened by  $\chi'$  [Ni<sub>3</sub>(A1,Ti,Cb)] and IN718 alloy by  $\chi''$  (Ni<sub>3</sub>Cb).

# Background

Utilization of the full potential of high strength nickel base super alloys in aircraft engines has been found to be limited by early fatigue crack nucleation as a result of small oxides or other types of inclusions. The effect of inclusions on nickel-base superalloy LCF properties and the sources of those inclusions have been documented (1,2). Inclusion size and location have been found to be the two most dominant factors controlling LCF life for these alloys. PM Rene' 95 LCF life degradation was found to be increasingly more severe with increasing inclusion size, and surface—intersecting inclusions were found to cause greater LCF life degradation than equivalent size inclusions at internal test bar locations. Similar results have been found for the cast plus wrought forms of the Rene' 95 and IN718 alloys, although some differences have been noted in the types of inclusions found in the cast plus wrought versus the PM materials. PM materials are subject to introduction of inclusions from the melt process (VIM plus VIM-atomization) and from post atomization handling, whereas the cast plus wrought materials only exhibit melt related inclusions. While the PM process introduces additional processing steps for potential inclusion

introduction, it also reduces problems of macrosegregation found in the cast plus wrought process. However, the predominant type of inclusions for both the PM and cast plus wrought processed material are oxides introduced from the melt-step refractory facilities, and in that respect contamination problems are similar.

## Nickel Base Superalloy Melt Practices

The basic requirements for melting and cooling of superalloys have been described by Schlatter <sup>(3)</sup> as providing: (a) a clean and consistant product (b) chemistry control (c) refining reaction control (d) no contamination from containers or atmosphere (e) applicability to a broad range of alloy compositions (f) solidification control, and (g) high reliability and low cost productivity. Although attempts have been made, no single melting process to date has successfully fulfilled all these requirements. As a result, manufacture of superalloys is accomplished in a multiple melting operation which can be broken down into primary melting, electrode casting, and remelting of electrodes.

The VIM process is currently recognized as the primary melt production technique that provides the best overall chemistry control both in respect to the desired alloying elements and in reduction of undesired impurities. A typical VIM furnace schematic drawing is shown in Figure 1a. Schlatter (4) presents an excellent review of the VIM process. The initial melt charge is typically the base alloy minus the reactive alloying elements. Homogenization and refining are accomplished after the charge is fully molten by a number of outgassing and boiling reaction. Reactive alloying elements are added after the oxygen and nitrogen are reduced to very low levels. Dip chemistry samples are taken and final corrective additions are made prior to casting of electrodes. Electrodes are poured either in vacuum or under inert gas.

The major disadvantages of the VIM process are: refractory erosion and refractory-melt reactions (5,6) that introduce oxide inclusions, lack of control over the solidification rate which results in excessive pipe formation and freezing segregation, and a non-uniform macro-microstructure. The judicious implementation of dams and weirs in the tundish design along with filtering may reduce, but does not eliminate, the ceramic content of the VIM electrodes. VIM electrodes are characteristically remelted to overcome the pipe, segregation and macro-microstructure disadvantages described.

The VAR process is currently the most widely accepted method for remelting VIM superalloy electrodes in the U.S. (7). A typical VAR furnace schematic drawing is shown in Figure 1b. Melting takes place in a water cooled copper crucible at pressures of  $\sim 10^{-3}$  torr. Heat is generated by a low voltage high current arc between the electrode and the molten metal. Melt rate for this process is controlled as a function of input power. Low superheat temperatures are achievable. The VAR solidification rate can be controlled by the melt rate with the large heat sink of the water cooled copper crucible producing a relatively shallow

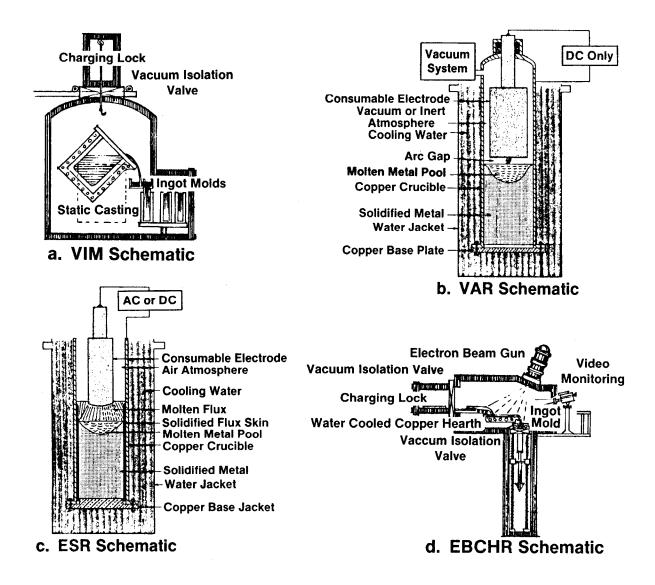


Figure 1. Schematic Drawings of Melt Practices.

molten pool. The controlled VAR solidification rate alleviates those specified disadvantages of the VIM electrodes, but can only remove oxide inclusion introduced by the primary VIM process via flotation.

While VAR does improve the macro-microstructure in comparison to VIM, it still exhibits some segregation disadvantages. The two major types of detrimental segregation that can occur in VAR ingots are "freckles" and "white spots". Freckles are chemical segregation areas with abnormally high solute element concentration. The precise mechanism for freckle formation has not been determined. "White spots" are areas of low solute element concentration. Three mechanisms are generally proposed for their formation. One mechanism involves the condensate on the cold crucible wall known as "crown" and the second involves the solidified metal with entrapped oxides at the edge of the molten pool called "shelf". White spots are generated when either the "crown' or "shelf" are undercut by the arc and fall into the melt. These materials are entrapped in the solidifying pool before they can be remelted or floated (if oxides). White spots formed by these two mechanisms generally exhibit excessive carbides, carbonitrides,

and oxides. They are collectively termed "dirty white spots" and have been observed to be particularly deleterious to LCF properties. The third mechanism proposed for white spot formation is that of melting through a region of secondary pipe in the VIM electrode. Dendrite arms that were formed during VIM solidification can be cut off and trapped via solidification in the VAR ingot. White spots formed by this mechanism do not exhibit excessive carbides, carbonitrides, and oxides, and are generally believed to be less detrimental to LCF properties. While modification of the VAR melt practice may somewhat reduce the frequency of white spots, their total elimination in the VAR ingots has not been demonstrated.

ESR melting has recently received increased interest as a potential means of improving cleanliness of superalloys. A typical ESR furnace is shown schematically in Figure Ic. ESR melting also takes place in a water cooled copper crucible. The electrode is submerged in a refining slag; the slag is resistance heated by electric current passing through it from the electrode to the molten pool. Superheat is dependent on flux chemistry and power input. Solidification rate is again controlled by melt rate. Refining results primarily from reactions occuring during droplet formation on the electrode tip and at the slag-molten pool interface. The major processing advantages of ESR are improved cleanliness and lack of mechanisms for formation of "white spots". The ESR process is not as sensitive to ingot quality (pipe, cleanliness) as is the VAR process. Klein, et al (8) reported improved chemical uniformity, finer dendritic microstructure and more uniform carbide dispersion for ESR IN718 compared to VAR. The major disadvantage of ESR are the potential for slag entrapment in the melt, the potential segregation problems at start up and unstable slags.

The EBCHR process has recently come into prominance as potentially providing the cleanest ingots of the currently available remelt process<sup>(9)</sup>. A schematic of the EBCHR furnace is shown in Figure 1d. The furnace consists of a vacuum chamber containing one or more electron guns, a water cooled copper hearth, a system for introduction of feedstock and an ingot mold. Heat is supplied by high energy electron beams controlled by electromagnetic scan coils. Both beam shape and scan patterns are independently controllable. Control can be either manual or computerized. Refining takes place on the hearth by several mechanisms. Gases and some high vapor pressure tramp elements, e.g., Pb, Bi, Ag, S, Zn, and Sb are Low density inclusions are retained on the hearth by volatilized. mechanical dams or an electron beam barrier. Some oxides may also dissociate and volatilize under the intense electron beam. High density inclusions, e.g., tungsten carbide from tool bits in the case of machining-chip melting, sink to the bottom and are trapped in the solidified skull. The oxide removal refining capability is a major advantage of the EBCHR process. In addition, superheat temperature control is independent of meIt rate. The disadvantage of the EBCHR process is evaporation losses of high vapor pressure alloying elements such as Cr making precise chemistry control more difficult than for the VAR or ESR practices. This disadvantage is currenly controlled via producing VIM ingot input material with higher levels of volatile elements to compensate for the anticipated loss.

## Cleanliness Evaluation Techniques for Nickel Base Superalloys

The initial cleanliness evaluation technique for nickel base superalloys was inadvertently that of LCF testing for the high strength PM materials. Degradation of the LCF properties as a result of oxide or other types of inclusions which were too small for detection by production ultrasonic inspection techniques gave ample evidence that a reduction in the oxide inclusion content was required to achieve the full material capabilities. However, LCF testing was not an economical cleanliness monitoring technique, and therefore, other means for cleanliness control were investigated.

Metallographic cleanliness rating tests of powder compacts similar to the slag inclusion ratings (10) for steels were first considered. The relatively low concentration of oxide inclusions in these superalloys compared to steels required that large surface areas be evaluated to obtain a meaningful cleanliness rating. An Electro-Chemical Machining (ECM) test was subsequently developed to allow observations of the oxide inclusions over large surface areas. Large sections of the test sample, approximately 32.3 cm<sup>2</sup> (5 in<sup>2</sup>), were ECM cut to dissolve the base metal thus leaving the non-conducting oxides standing in relief for observation. Visual inspections were then made to establish the quantity and dimensions of those inclusions. Inclusion ratings were obtained by that technique on the Rene' 95 alloy using 20 ECM cuts to observe about 645 cm $^2$  (100 in $^2$ ) of test While this technique did acceptably quantify the relative inclusion content for the PM and cast plus wrought Rene' 95, it was not Secondary phase surface acceptable for IN718 cleanliness ratings. undulations for ECM cut IN718 yielded a surface topography which masked the oxide inclusions. Although the ECM test or the metallographic inclusions rating technique could both yield reasonable oxide inclusion ratings for the Rene' 95 alloy, the large surface area requirements made those tests impractical both from the material utilization and labor requirement stand points. Further efforts subsequently identified the water elutriation test as a more practical production quality control cleanliness rating for the PM superalloys.

The water elutriation test(11) was the first industry accepted cleanliness rating technique for PM nickel-base superalloy materials. That test uses a water/ powder fluidized bed to separate the lower density inclusions via Stoke's law flotation. The test was developed to evaluate only the larger size range mesh fraction (+230 mesh) to quantify the larger inclusions as a cleanliness rating technique. The powder sample (typically 454 gm, 1 lb) was screened to separate out the +230 mesh fraction, and therefore, particles which could pass through the 230 mesh screen ( 63 um, 2.5 mil) were not evaluated. The major disadvantage with this test was the exclusion of these -230 mesh inclusions in the analysis. Assuming a 2:1 aspect ratio oxide particles, inclusions with a size up to 63 X 126 um (7938  $um^2$ ) or 2.5 X 5.0 mils (12.5 mils<sup>2</sup>) would not be collected and quantified. In spite of that disadvantage, the water elutriation test was used beneficially to quantify inclusions as a relative cleanliness rating technique; it also provided a means to confirm sources for inclusions in the PM superalloys. However, since the water elutriation test was not applicable to cast plus wrought materials and it only provided a partial

relative cleanliness rating for PM materials, further studies were conducted to establish a more universal cleanliness rating technique.

The EB button melt test was initially developed for cleanliness rating of nickel-base superalloys by Brown, et. al. (12) Modifications were made to that test, to yield the practice currently used by the General Electric (GE) Company<sup>(13)</sup>. The basic GE practice now involves electron beam drip melting of 454-680 gms (1.0-1.5 lbs) of material from the test sample into a three inch diameter hemispherical copper water cooled crucible. The purpose of that melt step is to float and collect the oxides from the melted material into a raft on the top central section of the small ingot (button) The EB melt parameters are controlled to minimize melting or volatilization of the oxides and to facilitate their collection into a single raft during solidification. The raft oxides are thereafter characterized for quantity, composition and dimensions using a Scanning Electron Microscope/Energy Dispersive Spectrum/Image Analysis (SEM/EDS/IA) system. The oxides are characterized by the IA system based on their cathodoluminescence image to provide a computer print-out of the number of inclusions, their sizes and major metallic element compositions. Seeding studies (13) have allowed the particle size distribution results to be used to calculate the total concentration (ppm level) of oxides. This technique was used to establish the oxide content for the Rene' 95 and Inco 718 materials evaluated for this paper.

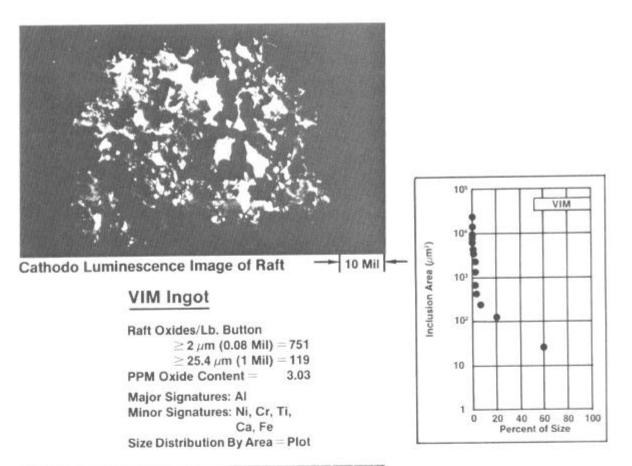
# Rene' 95 and IN718 Cleanliness Evaluations

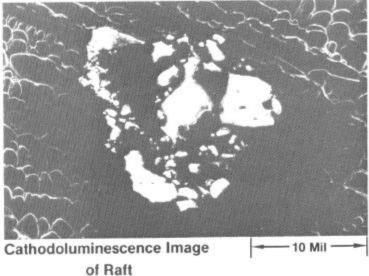
EB button oxide content cleanliness determinations have been conducted on both the Rene' 95 and IN718 alloys, although the majority of the data obtained since implementation of the SEM/EDS/IA evaluation technique have been for the Rene' 95 alloy. Results to date for Rene' 95 and IN718 EB buttons produced from ingot materials made by various melt practices are shown in Table 1. Typical observations for this test technique are shown by VIM and VIM/EBCHR data for Rene' 95 in Figure 2. The results for each alloy/melt practice have been summarized in Table 1 by showing the number of tests and vendors (ingot melt sources) involved followed by the average SEM/EDS/IA data. That data includes the number of surface (raft) oxide particles per 454 gm (1 pound) of button weight, the size of the largest particles found, the total oxide content in parts per million (ppm) by weight, and the EDS signature summary for the majority of those oxide particles. The surface oxide particle count per 454 gm (1 lb) of button is further separated to indicate both the total count and the count for those particles with a single dimension greater than 25.4 um (1 mil). The largest particles are characterized for both the maximum length dimension and the maximum area. The EDS signatures are separated into major elements (major EDS counts) and minor elements. The major element was consistently Al which was generally found in 90+ percent of the particles and generally accounted for 90+% of the EDS counts. The minor elements combined thus generally only accounted for less than 10% of the EDS counts. Where numbers of particles, dimensions and oxide contents are reported, the results are given as the average and one standard deviation.

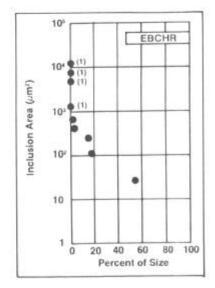
OXIDE INCLUSION CONTENT RESULTS FOR VARIOUS INGOT MELT PRACTICES RENE' 95 AND IN718 EB BUTTON MELT TEST SEM/EDS/IA(1) TECHNIQUE TABLE 1

MELT	NO. TESTS/ NO. VENDORS	SURFACE OXIDE PARTICLES PER LB. BUTTON WT.	RTICLES IT.	OXIDE LARGEST PARTICLES <u>DIMENSION</u>	AREA	TOTAL PPM OXIDE(3)
		>2 µm (0.08 mil)	>25.4 µm (1.0 mil)			
RENE 95						
WIM	8/2	390 ± 187 (2)	125 + 122	257 + 69 µm (10.1 + 2.7 mils)	19770 + 12900 µm <sup>2</sup> (30.6 + 20.0 mils <sup>2</sup> )	1.31 ± 0.8
VIM/VAR	6/1	925 ± 479	267 ± 157	163 + 64 µm (6.4 + 2.5 mils)	11878 + 5806 $\mu m^2$ (18.4 $\pm$ 9.0 mils <sup>2</sup> )	2.84 ± 2.1
VIM/ESR.	2/1	194 ± 50	42 ± 29	94 + 52 µm (3.7 + 2.0 mils)	$4513 \pm 3857  \mu \text{m}^2$ $(7.0 \pm 6.0  \text{mils}^2)$	$0.64 \pm 0.$
VIM/EBCHR	5/2	50 <del>+</del> 13	10 + 5	109 + 48 µm (4.3 + 1.9 mils)	$6409 + 3293  \mu m^2$ (9.9 ± 5.1 mils <sup>2</sup> )	$0.15 \pm 0.0$
• IN718						
VIM/VAR	1/1	1169	420	772 µm (30.4 mils)	70,500 µm <sup>2</sup> (109.3 mils <sup>2</sup> )	2.81
VIM/ESR	1/1	58	10	76 µm (3.0 mils)	2420 µm <sup>2</sup> (3.8 mils <sup>2</sup> )	0.35

SEM/EDS/IA = Scanning Electron Microscope/Energy Dispersive Spectrum/Image Analysis. All  $\pm$  variations shown are for one standard deviation. PPM  $\overline{\rm oxide}$  content calculation based on equation established in Reference 9. 393 3







# EBCHR Ingot

Raft Oxides/Lb. Button  $\geq 2 \, \mu \text{m} \, (0.08 \, \text{Mil}) = 62$   $\geq 25.4 \, \mu \text{m} \, (1 \, \text{Mil}) = 10$  PPM Oxide Content = 0.15 Major Signatures: Al

Minor Signatures: Ni, Cr, Ti, Ca Size Distribution By Area = Plot

Figure 2. Typical EB Button SEM/EDS/IA Results for VIM and EBCHR Ingot Materials.

The EB button SEM/EDS/IA results for Rene' 95 allowed ranking of the previously described melt practices based on the oxide content cleanliness as follows:

VIM/VAR VIM VIM/ESR VIM/EBCHR INCREASING CLEANLINESS

The eight Rene' 95 VIM ingot results were obtained from two melt producer's products, six tests from the first producer's product and two from the second. The six tests indicated an order of magnitude variation in the results, from 0.30 to 3.0 ppm oxide content, to give a result of 1.30 + 1.02 (1  $\sigma$  ). The two tests only indicated a factor of two variation but gave a similar average and  $1 \sigma$  result of 1.34 + 0.98 ppm. These results were considered equivalent and were combined to give the 1.31 + 0.87 ppm result shown in Table 1. The six Rene' 95 VIM/VAR ingot results obtained from a single melt producer's product indicated a 2.84  $\pm$  2.26 ppm oxide content. The results ranged from 1.34 to 7.78 ppm oxides. The single IN718 VIM/VAR ingot result of 2.81 ppm oxides was equivalent to the Rene' 95 VIM/VAR ingot average. The two Rene' 95 VIM/ESR ingot results obtained from a single melt producer's product indicated a  $0.64 \pm 0.16$  ppm oxide content. These results ranged from 0.48 to 0.80 ppm oxides. The single IN718 VIM/ESR ingot result of 0.35 ppm oxides was similar to the low end of th Rene' 95 VIM/ESR results. The five Rene' 95 VIM/EBCHR ingot results obtained from two melt producer's product indicated the cleanest metal at 0.15 + 0.03 ppm oxides with a range from 0.10 to 0.20 ppm.

Even though the cleanliness EB button melt rating results now allow a reasonably substantiated ranking for the stated nickel-base superalloy melt practices, a note of caution must be made on the validity of the results in respect to sampling procedures. The nature of oxide inclusion introduction in ingots does not facilitate uniform contamination; multiple EB button samples are required to obtain meaningful cleanliness results. As described, the actual results for one of the Rene' 95 VIM ingot materials varied by an order of magnitude (0.3 to 3.0 ppm). These variations in cleanliness are believed to be real within the ingot material due to the nature of melt practice introduction of oxides. It is anticipated, however, that powder metallurgy and fine grain drip cast ingot processes may yield a more uniform distribution of the oxide inclusions as a result of the different material handling practices. The data here which show that VIM ingot material is cleaner than the subsequent VAR ingot may be a result of sampling variations; but similar results were presented by Brown (14). The conclusion can at least be reached, in agreement with Brown, that VAR has no cleaning effect on the oxide particle content.

#### Conclusions and Future Direction

Development activities were conducted to evaluate various melting or remelting processes for improved cleanliness levels of nickel base superalloys. In addition, various cleanliness rating techniques were evaluated. An effective cleanliness rating technique for nickel-base

superalloys using EB button melt test has been established. The size distribution and the total oxide content in the button can be quantified by SEM/EDS/IA system. The results of these evaluations for Rene' 95 and IN718 EB buttons demonstrated that both ESR and EBCHR practices improved ingot cleanliness over the VIM/VAR process, and that EBCHR showed the greatest potential for improved cleanliness.

Various programs sponsored by U.S. Government agencies and industry are currently ongoing to develop and refine the ESR and EBCHR processes. Progress to date has been encouraging, although further scale-up melting demonstrations are required to qualify these processes for gas turbine hardware production.

## References

- (I) Chang, W.H., Green, H.M., and Sprague, R.A., "Defect Analysis of PM Superalloys", Rapid Solidification Processing Principles and Technologies III, Ed. R. Mehrabian, Proceedings of the Third Conference on Rapid Solidification Processing, Bureau of Standards, Dec. 8, 1982, pp 500-509.
- (2) Shamblen, C.E. and Chang, D.R., "Effect of Inclusions on LCF Properties of As-HIP Rene' 95", TMS Fall Meeting High Temperature Alloys Committee of AIME, Philadelphia, PA, Oct. 3, 1983.
- (3) Schlatter, R., "Melting and Refining Technology of High Temperature Steels and Superalloys A Review of REcent Process Developments", Superalloy Proceedings of the Second International Conference, Sponsored by Met. Soc. AIME, Battelle, Columbus, Ohio, MCIC-72-10, September 18-20, 1972, pp A1-40.
- (4) Schaltter R, "Vacuum Induction Melting of High Temperature Alloys", Journal of Metals, Vol. 24, No. 5, May, 1972, pp. 17-25.
- (5) Brown, E.E., Stulga, J.E., Jennings, L., and Salkeld, R.W., "The Influence of VIM Crucible Composition, Vacuum Arc Remelting, and Electroslag Remelting on the Non-Metallic Inclusion Content of MERL-76", Superalloys 1980, Eds. J.K. Tien, et. al., American Society for Metals, Metals Park, Ohio, 1980, pp 159-68.
- (6) Snape, E. and Beeley, P.R., "Refractory Melt Reactions in Vacuum Induction Melting of Nickel-Base Alloys", <u>Journal of the American</u> Ceramics Society, Vol. 50, No. 7, pp 349-353.
- (7) Op. Cit. 3
- (8) Klein, H.J. and Pridgeon, J.W., "Effective Electroslag Remelting of Superalloys", Superalloy Proceedings of the Second International Conference Sponsored by Met. Soc. AIME, Battelle, Columbus, Ohio, MCIC-72-10, September 18-20, 1972, pp B1-B26.

## References (continued)

- (9) Shamblen, C.E., Culp, S.L., Lober, R.W., "Superalloy Cleanliness Evaluation Using the EB Button Melt Test", Proceedings of the Conference Electron Beam Melting and Refining State of the Art 1983, Ed. R. Bakish, Library of Congress ISSN 0740-8706, Nov. 6-8, 1983, pp 61-94.
- (10) American Society for Testing and Materials, E45-76 Method D Chart for Determining the Inclusion Content of Steel, ASTM, Philadelphia, PA.
- (11) Op. Cit. 2
- (12) Op. Cit. 5
- (13) Op. Cit. 9
- (14) Op. Cit. 5