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EB Vs PAM Vs VAR for Uranium Alloys

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Questions were raised during the February '97 PDP Meeting at LLNL as to the advisability of funding three separate melting development programs (Advanced Vacuum Arc Remelt (VAR), Plasma Arc Melting (PAM) and Electron Beam Melting (EB)) through PDP in this era of shrinking research funds. The main issues seemed to be: 1) Have we evaluated the potentials of the three processes sufficiently to eliminate any of them out of hand and 2) Have any of these processes to date produced results which show clear advantages over the other two. The feeling at the meeting toward the latter seemed to be that all three processes required further development before they would be accepted by the complex as the standard. As far as evaluating potential for process improvements, however, we at Livermore did go through somewhat of a trade study in 1993 when we proposed the EB route.

It seemed to us that even with the elimination of the skull caster via a VIM/VAR/VAR route, the problem of recycle limitations in the VIM step due to excessive carbon pick-up limited the potential for a great improvement in material utilization using this method. A single-step, cold hearth route looked to be the preferable choice. Both EB and PAM are used commercially as cold hearth processes, and both have been used for the production of refractory and specialty metals; EB since the mid 1950's and PAM since the mid 1980's. Both seem to have found their individual niches with some applications being suited to EB and others to PAM. Both can be adapted to continuous casting techniques and both are capable of imparting the high heat fluxes necessary to melt refractory metals. The major differences appear to be that for most applications, the EB process is capable of producing a purer product, while PAM results in less loss of volatile components. There exists an abundance of information in the open literature on the results of processing via both routes on various materials[1,2,3]. The attached 1984 paper[4] gives a good explanation of electron beam melting and plasma melting, and, I believe, a fair assessment of the advantages and limitations of both processes.

In the specific case of the U-6Nb alloy, we at Livermore felt that EB melting would be a better match for a variety of reasons. As stated above, EB processing typically produces a purer product[4] and, as both uranium and niobium have low vapor pressures, the only volatile components lost should be high vapor pressure impurities such as Fe, Cu, Ni, Cr, etc. In practice we have found this to be true. Melt losses during EB melting of U-6Nb scrap are typically less than 0.5%, and there is no appreciable change in niobium content from feed material to ingot. In a 1963 study on purification of uranium by electron beam melting, Eikenberry[5] remelted VIMed uranium ingots in a vertical drip melter and reported a 0.2% melt loss. Eikenberry also reports the following data on removal of tramp impurities via EB melting as shown below in Table 1.

Table 1. Uranium Purification via Electron Beam Melting – 1963 data

Element	Feed Analysis - ppm	EB melted ingot analysis - ppm
H_2	1.46	.57
N_2	112	58
O_2	94	57
Al	6	6
C	748	672
Cr	13	9
Mg	8	5
Mn	130	60
Pb	2	2
Zr	67	46

Recent work at Livermore using state of the art melting and analytical equipment confirms this early work as shown in Table 2.

Table 2. Uranium Purification via Electron Beam Melting – 1997 Preliminary data

Element	Feed Analysis - ppm	EB melted ingot analysis - ppm
Cr	23	14
Fe	254	183
Ni	20	17
Mn	14	5
Mo	16	11
Si	104	0
Ti	61	64
Zr	31	20

Where comparisons can be made, the data does appear consistent. The ability to volatize these tramp impurities will of course be more valuable with material more severely contaminated than what has been processed in the current study.

In looking at the Y-12 flow chart, it became apparent that to increase material utilization significantly, a higher percentage recycle of both bulk and low grade scrap was going to be required. One of the big problems with scrap recycle of any metal is dealing with contaminants – the occasional iron nut or piece of copper wire that finds its way into the scrap barrel. During EB processing, which takes place in a high vacuum, these high vapor pressure impurities, as well as dissolved gasses, are removed by evaporation. Plasma melting, which typically takes place at pressures between 1-5 atmospheres, does not appreciably remove these impurities.

We know that U-6Nb feedstock may be required to be fed to a hearth in a variety of forms including bar, briquetted solids, bulk scrap and machine chips. For most forms of scrap, there is little difference in how these are fed to either an EB or PAM furnace. Industrial experience with titanium, however, has shown that machine chips may be fed quite readily to an EB furnace, but tend to be blown about too much by the high pressure plasma gasses to be fed loosely into a plasma furnace[6].

A major challenge to use of the U-6Nb alloy is the banding (alternating regions of high and low niobium) which occurs during solidification of the ingot. As Bob Reiner pointed out in his talk on uranium metallurgy at Y-12, a shallow pool and a flat solidification front is desired in casting U-6Nb to minimize this banding. Here again, EB processing seemed to offer significant potential advantages. Modern beam deflection controllers[7] allow the ability to direct a well focused (two centimeter diameter spot) energy source at high frequency (1,000 Hz) in a narrow ring around the periphery of an ingot. This insures that the edge of the ingot can be maintained liquid, thus insuring a smooth ingot surface, while heat is allowed to radiate from the center producing a relatively flat solidification front[8]. In PAM processing, with the introduction of modern programmable logic control, the hydraulically controlled, mechanical plasma torch has overcome much of its historically unwieldy nature, however being a mechanical system, maximum rotational frequencies around an 8" diameter pool are limited to around .5 Hz and the plasma beam has a less well defined, and definable, beam size (typically around 5 cm diameter).

The potential for cold hearth technology to reduce carbon levels in U-6Nb seems to us to be a significant driver for development of these technologies. Theoretically, and with some confirmatory experiments at LANL, there is the potential for carbon reduction in the U-6Nb alloy via reactions in the liquid state using either process. A 1970's patent[9] claims to have demonstrated carbon reduction in unalloyed uranium via oxygen bled over the hearth during EB processing. Preliminary LASL data reported at the February, 1997 PDP quarterly meeting indicated that carbon could be reduced in U-6Nb at high temperature during PAM via either a C+O=CO reaction in the melt, or a $C+H=CH_4$ reaction using hydrogen as a component of the plasma gas. There is also a 1960's patent[2] showing carbon reduction during EB melting of uranium alloys via the C+O=CO route. We at LLNL feel that recovery of high carbon U-6Nb or enriched uranium would be a significant process improvement, and that investigations into the potential for carbon reduction via both processes should be pursued.

Safety was a significant factor in our decision to pursue the EB route. Water leaks, major and minor, and the resultant steam, and hydrogen generation, are a fact of life in the liquid metal processing industry. Misdirected high power density beams, arcs or plasmas occasionally burn through water cooled devices. When uranium is the liquid metal being processed, safety concerns, of course, become even greater. The consequences of such water leaks vary greatly for the three melting processes. Through the years, vacuum arc remelting, primarily as applied to titanium melting, has seen the largest number of explosions due to water leaks. The Bureau of Mines has compiled an extensive history of explosions in the arc melting field[10,11,12], and as a result, all modern VAR furnaces in the industrial sector are constructed behind blast walls of one sort or another.

Both PAM and EB have excellent safety records. We find no literature reports of explosions during liquid metal processing using either technology. Based upon our experience in establishing safety plans and documentation for a new AVLIS installation at LLNL[13], however, we feel that the EB route offers the most convincing solution to analysis of a base-case accident scenario in the nuclear industry. There are a number of differences between EB and Plasma processing that support this conclusion.

First, water leaks in EB melting systems historically tend to be quite small – typically a few drops per second. These leaks usually result from a misdirected beam hitting a water cooled component, and melting through to a cooling channel. As the first drop of water is liberated, the resultant pressure rise causes the EB gun to arc down and shut off. Thus, the source of melting power is removed, the hole can never become large and the accident is self limiting. In the VAR case, a swinging electrode can strike a high current arc directly to the copper crucible melting a hole through which tens of gallons/min. of water can flow. The arc is sustained through a wide pressure range, and the hole may become relatively large. The resultant intimate mixing of liquid metal and water in a confined volume is often catastrophic[14]. The quantity of water generated in a PAM system is probably somewhere between these two extremes. Secondly, the quantity of liquid metal available to react with water is much less in the case of EB and PAM than with VAR. In a typical VAR process, pool depth in the crucible during steady state melting is at least one ingot diameter. Shallow pools, on the order of .3" for refractory metals, in both a hearth and crucible are the norm for EB and PAM and the potential for intimate mixing of water and liquid metal is not nearly as great as with VAR. Third, the volume into which a sudden gas evolution can expand is quite confined in the case of VAR, and this results in high peak pressures which occasionally rupture the furnace enclosure of blow the lid off the vacuum enclosure. EB and PAM by the nature of their designs have much more open area into which evolved gasses can expand, and this results in lower peak pressures at the vacuum enclosure wall. Finally, EB operates in high vacuum, whereas VAR operates at from 10-500 torr and PAM at 1-5 atmospheres. A shock wave produced during a liquid metal/water explosion in vacuum dissipates to a large extent prior to reaching the chamber walls. In processes operating near atmospheric pressure, the shock wave compresses the furnace atmosphere resulting in much higher pressures at the vessel walls and greatly increases the potential for failure of the furnace walls.

I hope this is of some help in understanding our preference for the EB route. The enclosed paper provides pretty good detail on plasma and EB, but there is, of course, a lot more information in the open literature on specific aspects of uranium processing, safety, etc. for all three processes.

References

1. P.M. Mergeanian, "Electron Beam, Plasma and Other Emerging Melting Processes Practical Comparison and Evaluation", Proceedings of the 1986 Vacuum Metallurgy Conference on Specialty Metals Melting and Processing, June 9-11, 1986, Iron and Steel Society, Warrendale, PA.
2. S. Schiller, G. Foerster, G. Jaesch, "Electron Beams for Metallurgical Processes in Vacuum", Proceedings of the Sixth International Vacuum Metallurgy Conference on Special Melting, April 23-27, 1979, American Vacuum Society, NY.
3. H. Stumpp, et al., "Advanced Production of Superalloy and Titanium Ingots by Cole Hearth Melting", SAMPE Quarterly, April 1989, pages 33-37.
4. D. Apelian, C. Entrekin, "Electron Beam Melting vs Plasma Melting: A Critical Review", Proceedings of the Conference on Electron Beam Melting and Refining, State of the Art, 1984, Bakish Materials, Corp., Englewood, NJ.
5. H.M. Eikenberry, "Electron Beam Melting of Uranium", AEC Research and Development Report NLCO-881, June 14, 1963.

6. B. Camacho, C. Traver, “Melting in Vacuum with the Long-Column Plasma Arc Torch”, Proceedings of the Conference on Electron Beam Melting and Refining State of the Art 1985, Bakish Materials Corp., Englewood, NJ.
7. M. Blum, et al., “Advantages of the New ALD EB-Gun Control System for the Modification of Existing or the Control of New EB-Furnaces”, Proceedings of the Conference on Electron Beam Melting and Refining State of the Art 1996, Bakish Materials Corp., Englewood, NJ.
8. C. Entrekin, “Control of Solidification in Electron Beam Hearth Refining”, Proceedings of the Conference on Electron Beam Melting and Refining State of the Art 1989, Bakish Materials Corp., Englewood, NJ.
9. H.R. Smith, “Improvements in or relating to the Purification of Metals”, US Patent #1,173,014.
10. F. Wood, R. Lowery, “The Contribution of Discharge Irregularities to the Explosion Hazard of Consumable-Electrode Arc-Melting”, USBM-U-571, November 1959.
11. P. Walker, “Explosion of a Titanium Furnace at the Jessop-Saville Works, Sheffield”, R.A.E. Report No. PBW.2, May 1963, Royal Aircraft Establishment, Farnborough, UK.
12. General Recommendations on Design Features for Titanium and Zirconium Production-Melting Furnaces, DMIC Memorandum 116, July 1961, Defense Metals Information Center, Battelle Memorial Institute, Columbus, OH.
13. Safety Analysis of Molten Uranium/Water Interaction in the UDS Separator Vessel, Engineering Safety Note ENE90-049, 1990, LLNL.
14. R. Beall, R. Van Dolah, R. Huber, “Report on Explosion Disaster. Titanium Melting Furnace. Mallory-Sharon Titanium Corporation”, US Bureau of Mines, October 1954.

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