EXTENDED LIFETIME AND OTHER BENEFICIAL PROPERTIES OF HAFNIUM CATHODES IN HEAVY ION PIG SOURCES*

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Summary

By replacing tantalum cathodes with hafnium, one obtains longer lifetime in exchange for lower extracted current. Pulsing the arc, when permitted, partially compensates by increasing the beam current. The higher vapor pressure of hafnium best explains this observed difference in performance. A factor of four increase in source lifetime is observed with N_2 feed, and about half that for CO feed. These longer lifetimes would not be expected on the basis of sputtering yield data for pure hafnium, and must instead be due to the formation of compounds of the feed gas and the hafnium. In other cases where N_2 can be used to partially support the arc, some degree of lifetime enhancement usually results.

Introduction

When PIG ion sources are used to produce heavy ions, the source lifetime becomes a major issue if it is small, as the costs of ion source maintenance procedures in materials and manpower can be large, and ion source recycling can become a significant fraction of the accelerator operating time.[1,2] For a PIG source with discharge-heated tantalum or tungsten cathodes, lifetimes fall in the range of 5-20 hours, with the lowest lifetimes generally associated with operation to get the highest charge states of the heavier elements.[3,4,5,6] At NSCL, we have had lifetimes in the range of 5-10 hours with tantalum cathodes in our internal PIG sources.[7,8]

The finite lifetime is due principally to the sputtering of the cathodes by ions from the discharge, as illustrated schematically in Figure 1. Any one of the following consequences of the cathode sputtering can kill a run:

1. Arc short resulting from the build-up of sputtered material in the gap.

2. Low output as a result of sputtered material choking the discharge defining aperture or the source extraction slit.

3. Arc instability resulting from the formation of deep craters in the cathodes and a tendency then for the arc to go out- especially during pulsed operation at low duty factors. In addition, the crater lowers the surface electric field, making it difficult to restrike an arc once extinguished.

Unfortunately the ion bombardment that causes these problems is the essential source of cathode heating for thermionically emitted electrons that sustain the arc.

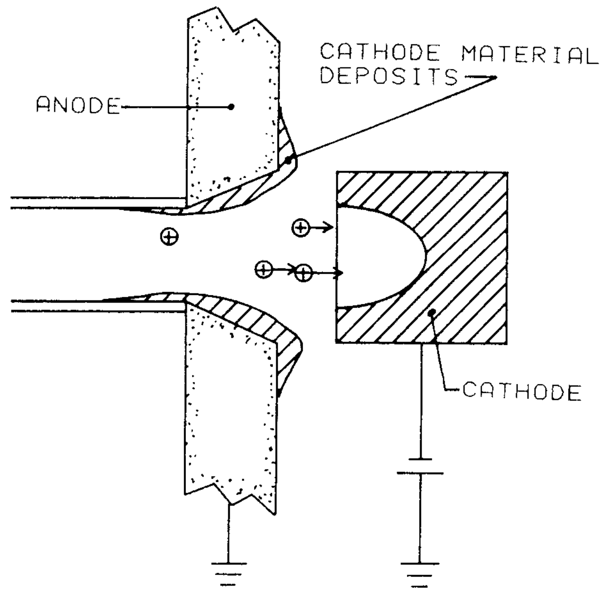

Figure 1. A schematic representation of one end of the PIG arc geometry, showing the anode end, arc defining aperture, and the cathode. The effects of cathode sputtering are illustrated.

Choice of Cathode Material

Good cathode materials for PIG sources must possess low sputtering rates and high thermionic emission current densities. This is why refractory metals are usually the material of choice for PIG sources, as they best meet these two requirements. Other characteristics and practical considerations, such as cost, availability and workability are important, but less so.

Consider the sputtering rate first. In a PIG source large fluxes of positive ions bombard hot cathodes at normal incidence, and we want to know what materials will give the longest lifetimes under these conditions. The most extensive study of the variation in sputtering yield (ejected atoms/ion) was made by Wehner with 30-400 eV mercury ion bombardment first,[9] and then the noble gases over the energy range 50-600 eV.[10,11] Fortunately the data span the energy range of positive ions in a PIG source. The sputtering yields were found to vary nearly linearly with bombarding energy in this range and to increase slightly with bombarding mass. Some of the elements found to have relatively low overall sputtering yields, and have also higher densities, which determines the volumetric sputtering rate, were Ti, Mo, Nb, Hf, Ta and W. These 'pure' atomic physics facts are in good agreement with the experience gained with operating PIG sources with various cathode materials.[1-8,12] In Wehner's studies hafnium, the subject of this paper, had a slightly higher sputtering yield generally than Ta or W, so a slightly higher sputtering rate would be expected.

In addition, cathodes for PIG sources must be good electron emitters at high temperatures. Thermionic emission from pure metals follows Dushman's Equation,[13]

$$J = AT^2 e^{-b/T},$$

where J is the emission current density in Amperes/cm^2, A is a quasi-constant of 120.4 Amperes/cm^{2}deg^2, T is the absolute temperature, and b is the absolute temperature equivalent of the work function. The work functions of the above metals vary only within a factor of two, so that temperature is the dominant parameter, and this favors metals with a high melting point. On that basis Ta and W, from the above list, with melting points of 3269 K and 3683 K respectively, would appear to be the best cathode choices for high power PIG sources. In this regard hafnium has about the same emission current versus temperature as tantalum,[14] but has a much lower melting point (2495 K), so the maximum current would be less.

Hafnium Cathodes

On the basis of the above considerations, as a cathode material hafnium would be expected to perform like the other refractory metals- but not as well as tantalum and tungsten at high arc powers.

Initial Experience- MSU K50 Cyclotron

As a result of an observation that hafnium targets were difficult to produce by sputter deposition, in 1978 tests were made of hafnium cathodes in a heavy ion PIG source in the K50 cyclotron, which was still running at that time. Only a few runs were possible, but some operating characteristics quite different than with tantalum cathodes were observed.[15] With CO feed, a 24 h lifetime was obtained, as compared to 2-3 h for a normal tantalum run, albeit at the expense of lower beam current. One experiment in March 1979 took 1 µA of 70 Mev C^{4+} ions on target for 24 h from a single source equipped with hafnium cathodes.[16] For comparison, with tantalum cathodes the extracted beam current was 15-30 µA. The lower extracted current was thought to be due to a lower maximum cathode operating temperature, as a result of the lower melting point of hafnium. The order of magnitude increase in lifetime was however a surprise, and since the lifetime enhancement was not observed with nitrogen or neon arcs, it was attributed to the formation of a HfC layer on the surface of the cathodes. Hollow tantalum cathodes filled with pressed-in HfC powder were tried, and longer lifetimes were obtained, but the arc operation was erratic and gave unstable beam output.

Since the question of beam intensity required is always a relative issue, it was speculated that hafnium cathodes might find application in the K500 cyclotron when run in the injector mode, where the charge states required for acceleration would be low and intensity would not be a problem.

Experience At Other Laboratories

Other laboratories certainly tried hafnium cathodes after 1979, but generally could not see any advantage over tantalum. The experience at Texas A&M was typical.[17] A longer lifetime was found- but hafnium cathodes were not good for the highest charges states, where large arc powers were required to obtain a useful beam current.

Since higher beam energy, which is limited by the size of the cyclotron ("K") and the ion charge state, is generally more important than source lifetime, hafnium cathodes did not come into wide use.

Hafnium Cathodes in the K500 Cyclotron

In March 1983, we began using hafnium cathodes in the K500 PIG sources, initially for C^{3+} beams, but this has been extended to many other beams as a result of the experience we have gained concerning their properties. (The PIG source for the K500 cyclotron is described in Ref. 8.) Table 1 summarizes the results of using hafnium cathodes under a variety of conditions.

Table 1. K500 Operation with Hf Cathodes. Lifetimes for tantalum cathodes are given were available.

ION	GAS	HAFNIUM		TANTALUM
		t_{mean}(h)	t_{max}(h)	t_{mean}(h)
$^{4}He^{1+}$	1% He+N_2	12.9	20.5	
$^{6}Li^{2+}$	96% ^{6}LiF pellet+N_2	37.3	67.6	
$^{7}Li^{2+}$	LiF pellet +N_2	16.0	42.5	
$^{12}C^{3+}$	CO,CO + N_2	10.6	19.0	
$^{12}C^{4+}$	CO,CO + N_2	14.1	31.6	9.6
$^{14}N^{3+}$	N_2	15.4	27.8	
$^{14}N^{4+}$	N_2	23.4	55.0	5.8
$^{14}N^{5+}$	N_2	24.9	55.1	6.4
$^{16}O^{4+}$	O_2 + N_2	10.8	13.9	
$^{20,22}Ne^{5+}$	Ne	2.5	2.8	2.2
$^{40}Ar^{6+}$	Ar	2.4	3.6	1.8

The largest increase in lifetime is seen for N^{4+} and N^{5+} beams. To get a feel for the magnitude of the lifetime effect, at the end of the first cyclotron operating cycle in June 1983, 340 h of N^{5+} were obtained in only 13 sources! Figure 2 shows long lived hafnium cathodes from a N^{5+} source. The main characteristics of a PIG source operated with Hf cathodes are:

1. Extended lifetime. The best case is with N_2 feed, with CO + N_2 almost as good, and CO the next best. The emergence of N_2 feed as giving the longest source lifetimes was quite a surprise. The lower lifetime with CO results from carbon deposits that form on insulators, making the arc unstable, and not specifically cathode erosion.

2. Multiple source restrikes are possible, even after many hours.

3. Very shallow craters form in the cathodes when operated DC.

4. Less sputtered material builds up in the anode bore, resulting in slower beam attenuation with time.

5. Same behavior as tantalum cathodes for Ne or Ar gas feed- deep craters form in the cathodes and no significant improvement in lifetime is observed.

6. Used cathodes show strong chemical changes after operation with N_2, CO or O_2 gas feed. The cathode color is glossy gold for N_2 feed, burnt black for CO feed, and dark gray for O_2 feed. Often the cathode is found to have fragmented, and for O_2 feed exhibits swelling.

7. There is often evidence of partial cathode melting, especially at the crater base. With more than about four amperes of arc current DC we have melted Hf cathodes in our source.

8. Pulsed operation- roughly the same beam currents can be obtained as with Ta cathodes, but at much lower duty factor. Depending on the duty factor, the lifetime enhancement is still possible however.

9. Normal failure mode for long lived Hf sources- the cathodes crumble and the source dies in a short.

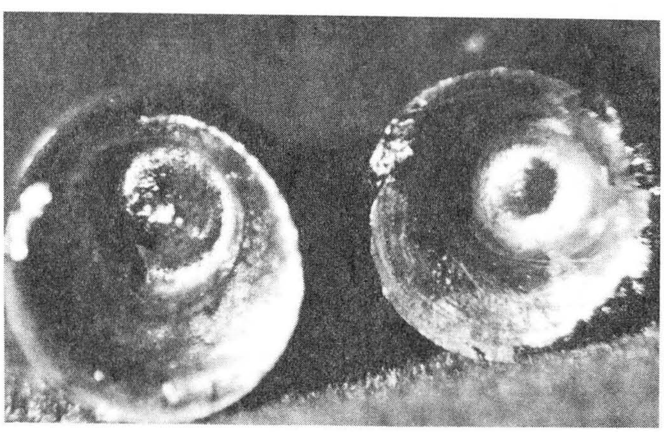

Figure 2. 55 hour lifetime hafnium cathodes for N^{5+} show small molten domes at the base of the craters and crumbled edges- the latter evidence of a structure changing reaction of the hafnium and nitrogen gas. New hafnium cathodes resemble other refractory metals.

Analysis

The dominant cause of these effects is the significantly lowered sputtering rate over tantalum or pure hafnium for N_2 or CO feed and the same operating circumstances.

The key observation is that when a non-reactive gas such as neon or argon supports the arc, hafnium cathodes show the same deep erosion crater formation as tantalum cathodes, with the arc eventually going out in the usual way. It those cases, what we are seeing is the sputtering of pure hafnium, and since it differs from tantalum in a small way, cathode lifetimes are also similar, as seen in Table 1 for Ar or Ne beams.

On the basis of the sputtering yield data for pure hafnium, no enhancement in the lifetime would be expected for nitrogen or carbon dioxide supported arcs. The preponderance of evidence points to the reaction of the hafnium with the arc support gas, and the subsequent formation of at least a surface layer with a much lower sputtering rate at these energies. This kind of lifetime enhancement has been observed before in other elements. In a early study, extended lifetimes were noted in a pulsed low power PIG source, when aluminum oxide or beryllium oxide cathodes were used in place of pure aluminum or beryllium cathodes.[18] In this case it is the nitride and carbide, rather that the oxide that gives the longest lifetime. In that regard, it is no coincidence that hafnium nitride is the highest melting point nitride, and hafnium carbide is the most refractory binary compound of all.[19] As a further corroboration of this effect, HfC cathodes have been used at Karlsruhe and other laboratories since 1972, where it was found that their performance was superior to other materials for production of Li^{3+} in a high power PIG arc.[20] What we have found at NSCL is that one can obtain similar results by exposing pure hafnium cathodes to gases directly in the arc.

Hafnium Cathodes and Beam Current

Up to this point, we have been considering only the lifetime aspect of sources with hafnium cathodes. In addition, we have found that the source output current is affected as well. For low charge state beams (Li^{2+}, C^{3+}, N^{3+}, N^{4+}), we have no difficulty with beam current with hafnium cathodes, and we then choose to run that way because of the large increase in lifetime that it affords. For N^{5+}, under certain conditions, we find that hafnium cathodes can provide adequate current, but for Ne^{5+} and Ar^{6+}, tantalum cathodes must be used. Since N^{5+} is on the edge, we have acquired a lot of data running pulsed or DC, with hafnium or tantalum cathodes. These data are summarized in Table 2.

Table 2. Nitrogen 5+ under a variety of operating conditions. N_2 feed is assumed in all cases.

CATHODE TYPE	DUTY FACTOR (%)	EXT.CURRENT (enA)	MEAN LIFETIME (h)	MAX. LIFETIME (h)
Hf	30	120	24.9	55.1
Ta	100	100	6.4	10.6
Ta	80	150	5.5	7.6

As can be seen, we can distinguish three basic operating modes. In the first, we use Hf cathodes to obtain long life, but find that pulsing at a low duty factor is required to maximize the beam. In the second mode, Ta cathodes are used, the beam intensity is roughly the same, though obtained DC, but the average lifetime 4 times lower. This mode is necessary when the experiment cannot tolerate the pulse structure, such as in coincidence experiments. If a small amount of pulsing can be tolerated, then we go to a third mode with Ta cathodes, and see about a 50% increase in the extracted beam versus DC. (If higher currents are required tnat this, then a lower duty factor with Ta cathodes would be used, but this has not been required and would also result in an even lower lifetime.)

The puzzle here is that if one starts with a hafnium source pulsed at low duty factor, no combination of arc parameters can be found that will allow this same beam current, but at a higher duty factor. On the other hand, if one starts with a hafnium source running DC, and then adjusts arc parameters so as to maximize the beam, one ends up with a low duty factor!

At first one would expect that the lower melting point of hafnium sets a lower thermionic emission current limit, and that this then is the reason for the poor performance. However, we have run tantalum cathodes at low power DC, where the emission currents are similar, and yet the beam current is still higher with tantalum cathodes!

The explanation of this effect requires consideration of the cathode contribution to the source gas pressure, which must be low to see high charge states in the source output current, because then the charge exchange losses are lower.[21] Now the arc voltage is indirectly proportional to the internal pressure, and what we find is that for the same arc current and gas flow, the hafnium source runs at a significantly lower arc voltage than a tantalum source, as shown in Table 3.

Table 3. Comparison of the range of DC arc parameters for sources equipped with hafnium or tantalum cathodes, when used to produce N^{5+} ions in the K500 cyclotron.

CATH. TYPE	ARC VOLTAGE (Volts)	ARC CURRENT (Amperes)	ARC POWER (KW)	EXT. CURRENT (enA)
Hf	200-300	2.0	0.4-0.6	20-40
Ta	500-700	2.0	0.8-1.4	75-150

The difference in arc voltage at the same arc current and gas flow implies that the pressure inside source is higher for a hafnium source. We infer that the higher pressure results from a higher vapor pressure as a function of temperature for hafnium than for tantalum under the same conditions. Since one of the main effects of lowering the duty factor is to lower the average cathode temperature, there would also be a decrease in the source's internal pressure as the duty factor is lowered. A comparison of vapor pressures of hafnium and tantalum in the range of 2000-2500 C, shows that the hafnium vapor pressure is 5 orders of magnitude larger,[22] and that is why hafnium cathodes do not work well DC for N^{5+}.

Impact of Hafnium Cathodes on the Cyclotron Operation

For nitrogen beams, there is a factor of 4 increase in the average lifetime with hafnium cathodes, and on an absolute scale we have had many sources that ran in excess of 50 hours. In addition, hafnium sources can be restruck many times, and after many hours. Sources are often turned off during energy changes, and left in the cyclotron to be restruck, and this further reduces the number of required source changes. Under those conditions, the ion source ceases to be an important operational issue. For instance, manpower dedicated to source recycling can be used for other purposes. This caries over to situations in which nitrogen can be used to partially or completely support an arc used to produce other ions. We have just recently added metal ion capability via back-bombardment sputtering, and support the arc for $^{6,7}Li^{2+}$ ions with nitrogen, and thereby obtain very long source lifetimes as with nitrogen beams. We see the lifetime effect also with CO feed, but CO has a deleterious effect on the HV stand-off insulators in the source, as carbon deposits form on the surface and shorts result. However, by mixing in some N_2, this process can be controlled and the lifetime increased.

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