QUARTZ TO METAL SEAL

Filed March 16, 1961

Fig. 1.

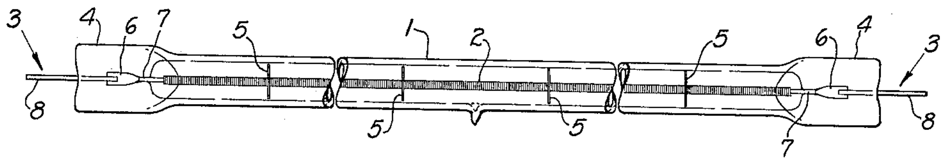


Fig. 2.

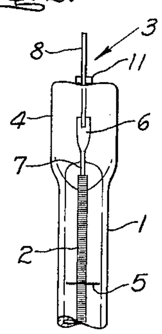


Fig. 3.

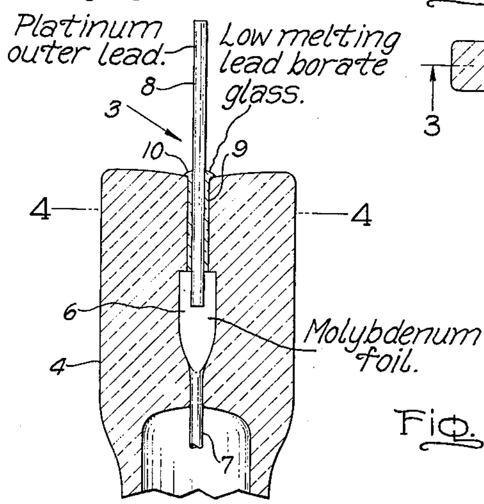


Fig. 4.

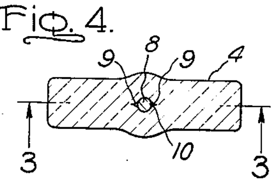
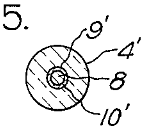


Fig. 5.



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QUARTZ TO METAL SEAL

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This invention relates generally to seals of metallic lead-in conductors in quartz or fused silica for electrical devices comprising a sealed envelope containing electric energy translation means to which electric current is supplied through the lead-in conductor.

As a matter of convenience, and not by way of limitation, the invention will be referred to especially in connection with quartz infrared lamps which comprise a tubular envelope of quartz having an incandescible filament of coiled tungsten wire extending longitudinally therethrough and connected at its ends to lead-in conductors which are hermetically sealed through compressed or pinched seal portions at the ends of the envelope. The lead-in conductors are each generally composed of molybdenum wires having a very thin intermediate foliated or ribbon portion which is wetted to and hermetically sealed in the quartz seal portion. Molybdenum is preferred because of its high melting point whereby it can withstand the high temperatures required to fuse the quartz thereto.

Such seals are entirely satisfactory so long as the seal temperature does not exceed about 350° C. At higher temperatures, the seal fails much more rapidly due to oxidation of the molybdenum. For example, at temperatures not exceeding 350° C. the seal lasts for several thousand hours whereas at a temperature of about 400° C. the average life drops to a few hundred hours. There has been a need for seals which will remain hermetically tight at elevated temperatures.

Many attempts have been made to achieve that objective, including the application of various types of oxidation resistant coatings to the molybdenum leads. Various metals and combinations of metals which have been proposed for this purpose have relatively low melting points so that they melt during formation of the seal. Furthermore, in some cases such coatings prevent the formation of a hermetic seal.

Accordingly, it is an object of the invention to provide quartz to metal seals of increased life at elevated temperatures. More particularly, it is an object to provide a seal structure having a life which may be up to eight hundred times that of the prior art molybdenum-quartz seal in the temperature range of about 500° to 800° C. Such temperatures are encountered in the case of very highly loaded lamps, and even in the case of lamps not so highly loaded but employed in an environment where such temperatures are reached.

The invention will better be understood by reference to the following detailed description taken in conjunction with the drawing wherein:

FIGURE 1 is an elevation of a quartz infrared lamp embodying seals in accordance with the invention;

FIGURE 2 is a fragmentary elevation of one end of a lamp illustrating a step in the manufacture of the seal;

FIGURE 3 is a fragmentary longitudinal cross section, on an enlarged scale, showing the completed seal;

FIGURE 4 is a transverse cross section of the completed seal; and

FIGURE 5 is a transverse cross section of a modified form of seal.

Referring to FIGURE 1 of the drawing, the lamp illustrated therein may be, for example, of the quartz infrared lamp type disclosed and claimed in Patent 2,864,025 Foote et al. which is assigned to the same assignee as the

present invention. The lamp comprises an elongated tubular envelope **1** consisting essentially of fused silica, preferably either clear crystal quartz or translucent sand quartz. A helically coiled tungsten filament **2** extends axially the length of the envelope **1** and is suitably connected at its ends to lead in conductors **3** which are sealed through flattened press or pinch seal portions **4** at the ends of the envelope. The elongated filament **2** is supported from the envelope wall at spaced points along its length by suitable support members **5**, preferably tantalum discs of the type described and claimed in Patent 2,813,993 Fridrich which is assigned to the same assignee as the present invention. The envelope **1** is preferably filled with an inert gas at substantial pressure, for example argon at nearly atmospheric pressure.

The actual hermetic seal between the quartz seal portion **4** and the lead-in conductor **3** is at an extremely thin intermediate ribbon or foliated portion **6**, preferably of molybdenum. The intermediate foliated portion **6** may be composed of a separate piece of molybdenum foil welded at opposite ends to the ends of respective inner and outer lead portions **7** and **8**, or the said foliated portion **6** may be formed as an integral part of a molybdenum inner lead portion **7** by flattening a part of the inner lead portion to a thickness of from .0005 to .001 inch by longitudinal rolling, as disclosed and claimed in Patent 2,667,595 Noel et al. which is assigned to the assignee of the present invention.

When the outer lead portion **8** is made of molybdenum, either as a separate piece welded to the intermediate foil portion **6** or as an integral part of the intermediate foil portion **6** and inner lead portion **7**, a long life in excess of about 5,000 hours is obtained so long as the seal temperature does not rise above about 350° C. At elevated temperatures the seal may fail in three ways which are, in general, characteristic of the temperature range. At temperatures above about 350° C. and below about 450° C., the molybdenum lead is slowly oxidized progressively along the length of the seal until it leaks and air is admitted to the interior of the envelope **1** which causes the filament **2** to vaporize and blacken the bulb and eventually fail by burning through at some point. The lamp may continue to burn for several hundred hours after the leak has started so that the useful life depends upon the degree of blackening that can be tolerated in a particular application.

In the temperature range of about 450° C. to 640° C. failure of the seal is abrupt. It will be understood that because of the difference in coefficient of expansion of the quartz and the relatively heavy outer lead portion **8**, there is a slight space or passage between the quartz and the part of the outer lead portion **8** enclosed therein (as illustrated at **9** in FIGURE 4) through which atmospheric air can reach the outer end of the foil portion **6**. In this temperature range, the oxide builds up at the end of the foil **6** before it can penetrate the seal, and cracks the quartz where it contacts the lead. The crack progresses to the exterior of the quartz whereupon the exposed foil oxidizes rapidly and fuses, usually cracking off a dish-shaped piece of quartz. The hermetic seal remains intact but electrical continuity is broken. At temperatures above about 640° C., molybdenum oxidizes so rapidly that the outer lead portion **8** simply disintegrates, leaving the seal intact.

In accordance with the present invention, seal life is materially increased by a combination of an outer lead **8** of material which resists oxidation at elevated temperatures above about 350° C., and a low melting glass **10** (FIGURE 3) filling the space or spaces **9** between the quartz and the part of the outer lead surrounded thereby. Particularly good results are obtained with the combination of a platinum outer lead **8** and a filling of a lead borate glass composition. For improved results, the lead borate glass is heavily loaded with fluorides, and still further improved results are obtained by the addition of thallium oxide to the glass composition.

A preferred method of introducing the glass composition **10** into the cavities **9** (which may have a width of the order of a thousandth of an inch), is illustrated in FIGURE 2. With the otherwise completed lamp in an upright position, a small ring or bead **11** of the low melting glass is slipped over the end of the outer lead **8** and onto the upper end of the quartz seal portion **4**. Heat is then applied to the surface of the press or pinch seal **4**, preferably by a small oxyhydrogen flame applied locally to the area enclosing the outer lead **8**, to cause the bead **11** to melt and be drawn into the capillary spaces or passages **9** (FIGURE 3). During operation of the lamp, with the seal portions **4** at elevated temperatures, the low melting glass filling **10** serves as a liquid seal to prevent ingress of atmospheric oxygen to the molybdenum foil **6** through the spaces **9**. At lower temperatures the glass **10** begins to harden and then cracks when the seal has become cool.

A composition, in parts by weight, of low melting lead borate glass which has given good results is as follows:

Batch		Analysis (calculated)	
Silica	2.94	SiO_2	3.26
Red Lead	64.20	PbO	75.50
Lead Fluoride	5.72		
Boric Acid	21.76	B_2O_3	13.60
Zinc Fluoride	1.54	ZnO	1.34
Potassium Bifluoride	.93	K_2O	.62
Sodium Bifluoride	.39	Na_2O	.22
Lithium Fluoride	.45	Li_2O	.29
Fluorspar	.40	CaO	.32
Thallic Oxide	1.73	Tl_2O_3	1.92
		F_2*	2.97

* The amount of F_2 is the total equivalent F_2 contributed by all fluorides in batch.

Lamps having seals of the type described, including platinum outer leads and a filling glass **10** of the composition stated above, and operated under conditions such that the seal temperature was above 700° C., had an average life of about 800 hours. This is in contrast to as little as one hour life of lamps with outer leads **8** of molybdenum, and fifty hours with outer leads **8** of platinum but no filling glass **10**. It will be understood that it is not feasible to make the entire lead-in conductor **3** of platinum because the foliated portion, required for formation of a hermetic seal to quartz, would melt at the sealing temperature. The outer lead portion **8** of platinum does not melt because it conducts away the heat sufficiently rapidly on account of its relatively large size, which may be a wire of 30 mil diameter, for example. It is preferred that only sufficient of the low melting glass be used to fill the cavity **9**; in the case of a 30 mil outer lead **8** the glass bead **11** may have a weight of about 3 mg.

While the glass composition listed above has given the best results, some improvement may also be obtained with a simple lead borate, although it is preferred to improve the chemical and physical properties of the lead borate by the addition of the other ingredients in the above composition. The composition may be varied somewhat by employing the ingredients, in part by weight of a calculated analysis, in the following ranges:

	Maximum Range	Preferred Range
SiO_2	0–10	2–5
PbO_2	65–85	70–80
B_2O_3	10–20	12–15
Tl_2O_3	0–7	1–5
ZnO	0–5	1–2
CaO	0–2	.25–1
K_2O	0–2	.25–1
Na_2O	0–2	0–.5
Li_2O	0–2	0–.5
F_2	0–7	1–4

The SiO_2 increases the chemical durability. The addition of CaF_2 (fluorspar) and ZnF_2 provides a means of introducing F_2 to the batch, in addition to that provided by the fluorides of lead, potassium, sodium and lithium, and the CaO and ZnO thus formed increase the chemical durability of the resulting glass. The K_2O, Na_2O, Li_2O and F_2 contribute to a lowering of the softening point and melt viscosity. The F_2 is especially effective in this respect. The thallic-oxide addition is particularly effective and unique in that it reduces the viscosity of the melt and broadens the temperature range of low viscosity.

The most significant physical properties of these glasses are very low softening point (about 320° C. for the preferred composition), melt viscosity and melt surface tension. Contributing largely to those three properties, especially the last two, are the large number of fluoride additions and the thallic-oxide addition.

In FIG. 5 there is illustrated a modified form of seal wherein the tubular quartz seal portion **4'** is evacuated and collapsed upon the lead-in conductor in accordance with a well-known technique, instead of being pressed flat as in FIG. 4. In this case the opening or cavity **9'** around the platinum outer lead **8** completely encircles the lead and is filled with the low melting glass **10'**.

What we claim as new and desire to secure by Letters Patent of the United States is:

1. A seal for electrical devices comprising a member of fused silica and a lead-in conductor sealed in and extending through said member, said conductor including an intermediate foliated lead portion of molybdenum hermetically sealed within said member and subject to oxidation at elevated temperatures above about 350° C., and an outer lead portion of platinum connected to and extending from said intermediate lead portion through said silica member to the exterior thereof with a slight space between the silica member and the part of said outer lead portion enclosed thereby, and a filling in said space of low melting lead borate glass which is molten at elevated temperatures above about 350° C. forming a liquid seal preventing ingress of atmospheric oxygen to said molybdenum intermediate lead portion, said lead borate glass comprising, by weight, 65–85 parts PbO and 10–20 parts B_2O_3.

2. A seal for electrical devices comprising a member of fused silica and a lead-in conductor sealed in and extending through said member, said conductor including an intermediate foliated lead portion of molybdenum hermetically sealed within said member and subject to oxidation at elevated temperatures above about 350° C., and an outer lead portion of platinum connected to and extending from said intermediate lead portion through said silica member to the exterior thereof with a slight space between the silica member and the part of said outer lead portion enclosed thereby, and a filling in said space of low melting lead borate glass which is molten at elevated temperatures above about 350° C. forming a liquid seal preventing ingress of atmospheric oxygen to said molybdenum intermediate lead portion, said lead borate glass containing, by weight, approximately 2–5 parts SiO_2, 70–80 parts PbO_2, 12–15 parts B_2O_3, 1–5 parts Tl_2O_3, 1–2 parts ZnO, .25–1 parts CaO, .25–1 parts K_2O, 0–.5 parts Na_2O, 0–.5 parts Li_2O and 1–4 parts F_2.

3. A seal for electrical devices comprising a member of fused silica and a lead-in conductor sealed in and extending through said member, said conductor including an intermediate foliated lead portion of molybdenum hermetically sealed within said member and subject to oxidation at elevated temperatures above about 350° C., and an outer lead portion of platinum connected to and extending from said intermediate lead portion through said silica member to the exterior thereof with a slight space between the silica member and the part of said outer lead portion enclosed thereby, and a filling in said space of low melting lead borate glass which is molten at elevated temperatures above about 350° C. forming a liquid seal preventing ingress of atmospheric oxygen to said molybdenum intermediate lead portion, said lead borate glass containing, by weight, approximately 3.26 parts SiO_2, 75.5 parts PbO, 13.6 parts B_2O_3, 1.34 parts ZnO, .62 parts K_2O, .22 parts Na_2O, .29 parts Li_2O, .32 parts CaO, 1.92 parts Tl_2O_3 and 2.97 parts F_2.

4. A seal for electrical devices comprising a member of fused silica and a lead-in conductor sealed in and extending through said member, said conductor including an intermediate foliated lead portion of molybdenum hermetically sealed within said member and subject to oxidation at elevated temperatures above about 350° C., and an outer lead portion of platinum connected to and extending from said intermediate lead portion through said silica member to the exterior thereof with a slight space between the silica member and the part of said outer lead portion enclosed thereby, and a filling in said space of low melting lead borate glass which is molten at elevated temperatures above about 350° C. forming a liquid seal preventing ingress of atmospheric oxygen to said molybdenum intermediate lead portion, said lead borate glass comprising by weight, 65–85 parts PbO, 10–20 parts B_2O_3 and about 1–4 parts of equivalent fluorine added as fluoride of at least one of the elements lead, zinc, potassium, sodium, lithium and calcium.

5. A seal for electrical devices comprising a member of fused silica and a lead-in conductor sealed in and extending through said member, said conductor including an intermediate foliated lead portion of molybdenum hermetically sealed within said member and subject to oxidation at elevated temperatures above about 350° C., and an outer lead portion of platinum connected to and extending from said intermediate lead portion through said silica member to the exterior thereof with a slight space between the silica member and the part of said outer lead portion enclosed thereby, and a filling in said space of low melting lead borate glass which is molten at elevated temperatures above about 350° C. forming a liquid seal preventing ingress of atmospheric oxygen to said molybdenum intermediate lead portion, said lead borate glass comprising, by weight, 65–85 parts PbO, 10–20 parts B_2O_3 and about 1–5 parts thallic oxide.

6. A seal for electrical devices comprising a member of fused silica and a lead-in conductor sealed in and extending through said member, said conductor including an intermediate foliated lead portion of molybdenum hermetically sealed within said member and subject to oxidation at elevated tempertaures above about 350° C., and an

outer lead portion of platinum connected to and extending from said intermediate lead portion through said silica member to the exterior thereof with a slight space between the silica member and the part of said outer lead portion enclosed thereby, and a filling in said space of low melting lead borate glass which is molten at elevated temperatures above about 350° C. forming a liquid seal preventing ingress of atmospheric oxygen to said molybdenum intermediate lead portion, said lead borate glass comprising by weight, 65–85 parts PbO, 10–20 parts B_2O_3, about 1–5 parts thallic oxide, and about 1–4 parts of equivalent fluorine added as fluoride of at least one of the elements lead, zinc, potassium, sodium, lithium and calcium.

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