Aug. 25, 1953 J. N. BURDICK ET AL 2,649,993

METHOD OF EVACUATING, FILLING, SEALING, AND RELEASING GAS IN A CONTAINER

Filed Feb. 14, 1947

Fig. 2.

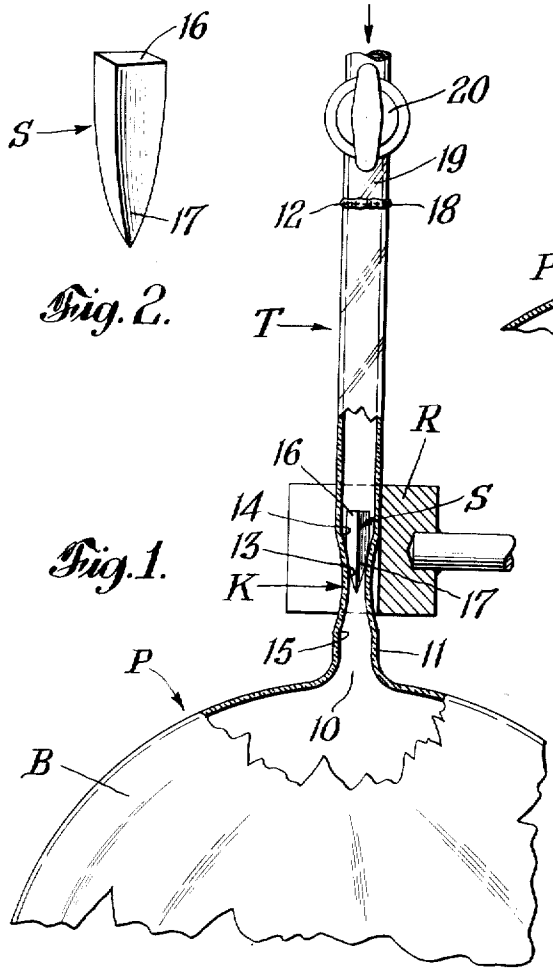

Fig. 1.

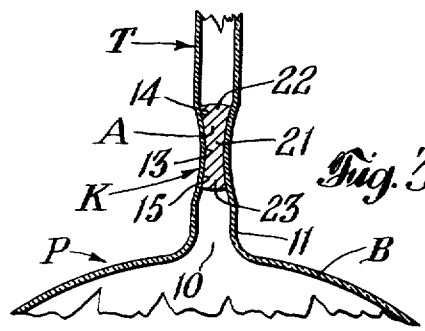

Fig. 3.

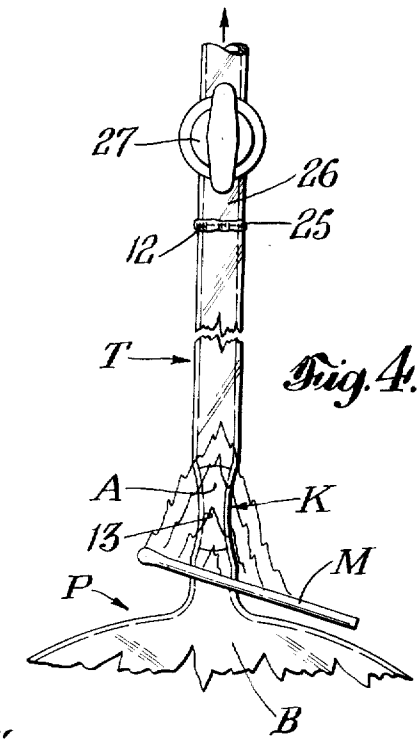

Fig. 4.

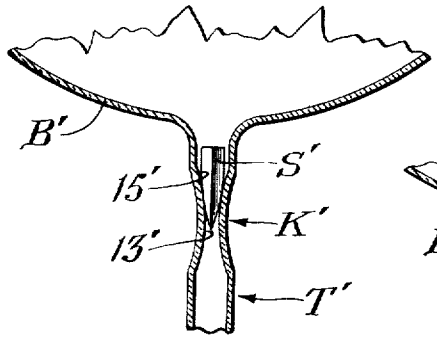

Fig. 5.

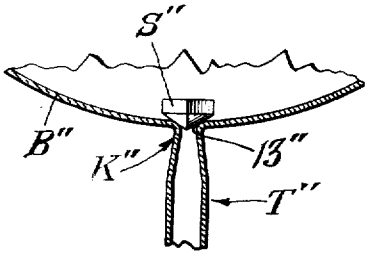

Fig. 6.

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Patented Aug. 25, 1953

2,649,993

UNITED STATES PATENT OFFICE

2,649,993

METHOD OF EVACUATING, FILLING, SEALING, AND RELEASING GAS IN A CONTAINER

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Application February 14, 1947, Serial No. 728,670

1 Claim. (Cl. 222—1)

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This invention relates to a novel container or bulb for storing and transporting rare gas, such as neon, argon, krypton, and xenon, and to improved methods of filling, sealing, and releasing gas in a rare gas container. The invention also concerns the resulting novel package of rare gas as well as a novel fluid-tight seal that is especially advantageous for sealing a rare gas bulb but is also useful for sealing tubular fluid passages connected to other types of containers.

Rare gas such as neon, argon, krypton, and xenon usually is stored and transported in small glass bulbs each containing about one liter of the gas. Heretofore, the empty bulb has been formed with tubular glass connections: one at its bottom through which the bulb is first exhausted, then filled with the rare gas, whereupon this connection is permanently sealed by fusing its outer end; and another connection at the top of the bulb operable to release the rare gas when needed and to lead it into a suitable manifold or receptacle for use. This top gas withdrawal connection comprises a bulb-sealing glass capsule disposed within one end of a glass discharge tube, the adjoining open ends of the capsule and the discharge tube being permanently united to one another and to a top opening in the glass bulb. When the rare gas is to be released from the bulb, a small steel ball or steel-containing weight is placed in the discharge tube, the latter is first evacuated to the atmosphere and then connected to a rare gas receiver, whereupon the weight is lifted by a suitable magnet and then released to drop on and break the sealed end of the capsule, thereby permitting the rare gas to flow from the bulb through the discharge tube into the receiver.

The container just described is expensive to manufacture because the discharge tube and the fragile glass sealing capsule must be welded in place by an experienced glass blower. Also, premature fracture of the capsule and loss or contamination of the rare gas sometimes occurs when the breaking weight is placed in the discharge tube. Furthermore, if the capsule is not broken with sufficient care and at the proper moment, the breaking weight sometimes fractures other parts of the container and the gas escapes. Another possibility of breakage and loss of the rare gas occurs if the rare gas bulb is sparked when the breaking ball or weight is in place, since the spark occasionally jumps from the discharge tube to the ball and then to the capsule, which invariably punctures the latter and eventually results in evacuating all of the rare gas into the atmosphere.

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The principal objects of this invention are: to provide a rare gas bulb or container which is less expensive to construct and simpler and more reliable in use than prior rare gas bulbs; to provide a dependable fluid-tight conduit seal which is formed easily and economically, is unaffected by wide temperature variations, and may be readily broken when desired without fracturing elements associated with the seal; to provide simpler procedure for filling, sealing, and releasing rare gas in containers therefor; and to provide a package of rare gas which is free from objectionable features of the rare gas packages in use heretofore.

The above and other objects and novel features of the invention will become apparent from the following description and the accompanying drawing. In the drawing:

Fig. 1 is a side elevational view of a rare gas bulb, parts being broken away and in section, ready for sealing by the method of the invention;

Fig. 2 is an enlarged perspective view of one type of fusible metal slug useful for forming a fluid-tight seal by the novel method;

Fig. 3 is a vertical sectional view of a part of a rare gas bulb embodying the novel gas-tight seal of the invention;

Fig. 4 is a fragmentary side elevational view of a sealed rare gas bulb, showing how the same is unsealed to permit discharge of its gas content into a receiver; and

Figs. 5 and 6 are fragmentary vertical sectional views of two rare gas bulbs ready for sealing by modified procedures in accordance with the principles of the invention.

The principles of this invention are advantageously utilized in filling, sealing and discharging glass bulbs suitable for storing and shipping rare gas, and this application of the invention will be described in detail hereinafter to illustrate one embodiment of such principles. However, it is to be understood that some features of the invention, such as the novel fluid-tight seal for a tube and the procedures for forming and for unsealing such seal, are of more general utility, and that variations of the exact construction and procedures as disclosed herein are within the scope of this invention.

In accordance with this invention, the novel rare gas container or bulb preferably has only one connection or tube, which provides both the gas-filling and the gas-withdrawal passage for the bulb. One open end of this tube is integrally secured to an opening in the bulb; its other open end is adapted to be connected to evacuating

means and to a rare gas supply when filling the bulb, and to evacuating means and to a receiver when discharging the rare gas from the bulb. When it has been filled with rare gas, the bulb is sealed by a novel gas-tight seal formed within the connecting tube. To assist in forming this seal, the tube desirably is contracted between its ends before joining it to the bulb, which provides a constriction of substantially hourglass shape within the tube and outside the bulb. The seal is completed by disposing a solid body of suitable, easily fusible sealing material in gas-tight sealing engagement with the annular internal surface of the narrowest part of the constriction and also in gas-tight sealing engagement with the annular internal surfaces adjacent both ends of the narrowest part of the constriction. The sealed gas-filled bulb, when connected to a rare gas receiver or customer's manifold, may be readily unsealed by simply externally heating the contracted portion of the tube sufficiently to heat and fuse at least those portions of the fusible body of sealing material which sealingly engage the several surface portions of the internal constriction.

As illustrated in Fig. 1, the rare gas container or package P desirably comprises a spherical glass bulb B provided with a single combined inlet and outlet **10** to which one open end **11** of a single glass tube T is integrally secured so that the tube will extend radially from the outside of the bulb B. The opposite open end **12** of the tube T may be connected to suitable means for evacuating, filling and discharging the bulb B through the tube T. Between its ends **11** and **12**, the tube T is contracted, as by locally heating and stretching the tube before it is secured to the bulb B. This contraction of the tube T provides a constriction K of substantially hourglass shape inside the tube, which constriction has its narrowest section or throat **13** between two adjoining flaring sections **14** and **15** gradually increasing in diameter from the throat **13** to the full inside diameter of the tube. By way of example, the tube T may have an outside diameter of about 7 mm. and a wall thickness of about 1¼ mm.; and the constriction K may have a length of about 20 mm. and the throat **13** may have a minimum inside diameter of about 2 mm.

When the bulb B is to be filled with gas, a solid slug S of suitable fusible sealing material is first inserted through the open end **12** into the tube T, and the bulb B is so supported that the tube will be held with its longitudinal axis inclined to a horizontal plane, preferably in a substantially vertical position, so that the slug S, which is of smaller transverse section than the inside of the tube and too large to pass through the throat **13**, will slide down the tube and be supported directly in contact with the inside of the flaring section **14** above the throat **13**. The shape of the slug S differs from that of the inside of the tube T, so as to leave sufficient space for the passage of gas between the slug and the tube. As shown in Figs. 1 and 2, for example, the slug S may consist of an elongated solid body of fusible metal, such as solder, having a lower fusion temperature and a higher thermal coefficient of expansion and contraction than the glass forming the tube T, and the slug S may have an upper portion **16** of square cross-section and a pyramidal lower end portion **17** which will project into the constriction and center the slug axially within the tube and also leave spaces between the slug and the interior surface of the tube for the passage of gas through the tube into and out of the bulb.

After the slug S is properly positioned in the tube T, the upper open end **12** of the tube is gas-tightly secured, as by a weld **18**, to a glass conduit **19** controlled by a stop-cock **20**, for connection to suitable means for evacuating the bulb and tube and for supplying rare gas to the evacuated bulb. After the bulb B and tube T have been evacuated by a vacuum pump (not shown), rare gas is delivered through the tube T into the bulb B until the desired gas pressure in the latter is reached. Then the slug S is slowly heated and fused so as to gas-tightly seal the filled container P at the constriction K in the tube T.

While it is being heated and melted, the slug S desirably is supported vertically with surface portions of its lower end **17** directly engaging internal surface portions of the constriction K above the throat **13**, so that the fused or molten slug material will flow slowly by gravity down past the throat **13** while the tube T is held substantially vertically. Immediately after some of the fused slug material begins to flow past the throat **13**, heating of the slug is discontinued, whereupon the slug material cools and finally solidifies in gas-tight sealing contact with the internal annular surface of the throat **13** and also in gas-tight sealing contact with both internal annular surface portions of the constriction sections **14** and **15** adjacent the opposite ends of the throat **13**. Under some circumstances it may be desirable to force the molten sealing material into the constriction K by gas pressure, in which event the tube T may be held horizontally if desired.

The slug S may be heated to its fusion temperature by various methods. As shown in Fig. 1, the slug may be indirectly heated by positioning a hot slotted soldering iron R around and touching the outside of tube T adjacent the constriction K, so that its heat is conducted through the wall of the tube to the slug. As the slug melts and flows into the throat **13**, the soldering iron may be gradually lowered until the molten sealing material has reached its proper sealing position, whereupon the soldering iron is removed. Other heating means, such as a low temperature flame, may be used in place of the soldering iron.

The resulting solidified gas-tight seal A, as illustrated in Fig. 3, comprises a neck **21** and bulbous heads **22** and **23** integrally connected to opposite ends of the neck **21**. The neck **21** fills the throat **13** and sealingly engages its internal annular surface, and the heads **22** and **23** occupy larger portions of the constriction adjacent opposite ends of the throat and sealingly engage their internal annular surfaces.

It is essential that the seal A remain gas-tight even when the rare gas bulb B and tube T are subjected to temperatures varying greatly above or below the temperature of the room in which the seal was formed. This is assured by permitting the molten sealing material to flow to sealing positions above and below the throat **13** to form the heads **22** and **23** as the fused sealing material cools and solidifies. Upon discontinuing the application of heat thereto, further flow of the sealing material is controlled and limited by the viscosity of the molten material and by capillary action at the sealing surfaces to form the neck **21** and the heads **22** and **23**. As the seal A cools to room temperature it contracts axially more than

does the tube T, thus pressing the heads 22 and 23 into contact with the surfaces of sections 14 and 15 with sufficient force to cause the seal to conform to any irregularities in these surfaces and to make gas-tight contact with them.

After the seal A has cooled to room temperature and solidified, the tube T is detached from the conduit 19, as by nicking the tube with a file and then breaking it, whereupon the filled and sealed rare gas package P is ready for storage or shipment with the end 12 of the tube T open but with the seal A in position in the constriction K.

When the rare gas package P is subjected to atmospheric temperatures lower than the room temperature at which the seal was formed, the seal A tends to contract axially but remains gas-tight because the heads 22 and 23 are pulled into tighter sealing contact with the internal surfaces of sections 14 and 15 adjacent the ends of the throat 13. Thus, at such lower temperatures a gas-tight seal is assured even though radial contraction of the seal A may have caused its neck 21 to lose sealing contact with the internal surface of the throat 13. When the filled and sealed rare gas package is subjected to atmospheric temperatures higher than the room temperature at which the seal was formed, the seal A expands axially and radially, but its radial expansion is much greater than that of the glass tube T, thereby maintaining the neck 21 in sealing contact with the internal surface of the throat 13. Thus, it appears that tensile and compressive stresses are induced in the seal A at different atmospheric temperatures and cooperate to maintain a gas-tight seal. The seal A also has some degree of elasticity which is important in maintaining its sealing efficiency in service subject to wide temperature changes.

The slug S and resulting seal A may consist of any fusible material having a melting point or fusion temperature lower than the fusion temperature of the material of the tube T and also having a higher thermal coefficient of expansion and contraction than the material of tube T. Preferably the slug S and seal A consist of a fusible metal having a fusion temperature lower than the temperature at which the glass of the tube T softens appreciably, and having a thermal coefficient of expansion and contraction higher than that of such glass to maintain gas-tight sealing engagement of the seal A with internal surfaces of the constriction K over a substantial range of atmospheric temperatures. Low melting bismuth base alloys have been found advantageous as materials for forming the slug S. For example, a bismuth solder slug consisting of a metal alloy having a melting point of 113° C. and containing about 40% bismuth, 40% lead and 20% tin has been used successfully. Another suitable metal is Wood's alloy which contains about 50% bismuth, 12.5% cadmium, 25% lead and 12.5% tin, and has a melting point of 63° C. When the 40% bismuth, 40% lead and 20% tin solder is used the ratio of the length of the constriction K to its minimum diameter may be between 8 to 1 and 12.5 to 1. Both sealing alloys described above have a thermal coefficient of expansion and contraction of about 27×10^{-6}/° C. In contrast, commercial glasses suitable for making the bulb B and the tube T have softening points ranging between 500° C. and 1510° C. and thermal coefficients of expansion and contraction ranging from 3×10^{-6}/° C. to 12×10^{-6}/° C. Tests have demonstrated that gas-tight seals made at 70° F. with bismuth solder in accordance with this invention remain gas-tight at —40° F. and at 212° F.

The rare gas content of the sealed gas package P may be readily released and discharged at will into a suitable receiver or manifold. As illustrated in Fig. 4, the open end 12 of the tube T is first gas-tightly connected, as by a weld 25, to a glass conduit 26 leading through a stop-cock 27 to suitable evacuating means and a suitable rare gas receiver. The cock 27 is then opened and the portion of the connection between the seal A and the gas receiver is evacuated to the atmosphere to eliminate contaminating gas. Thereupon, a suitable source of heat, such as the flame of a lighted match M or any other heating medium, is applied to and moved around the outside of the tube T adjacent the seal A. Such heat melts at least those parts of the seal A which engage the inside surface of the constriction K and loosens the seal sufficiently to unseal the package P. When the seal A has become sufficiently melted to pass through the throat 13, the pressure of the gas in the bulb B blows the seal A from its sealing position up into the upper portion of tube T and the rare gas from the bulb B then passes freely around the seal and through the connecting conduit 26 into the receiver.

It will be evident that a gas-tight seal embodying this invention may be formed by methods which involve variations of the preference seal-forming procedure described above.

For example, as shown in Fig. 5, the sealing slug S' may be inserted into the bulb B' before constricted tube T' is united to the opening in the bulb B'; and for this sealing operation the bulb B' is held uppermost with the tube T' extending substantially vertically downwardly and the sealing slug S' resting directly on the flaring constriction section 15', so that the pointed end of the slug extends into the throat 13'. Then the seal A may be formed in the constriction K' by the heating and cooling steps previously described. As another alternative, a bulb B'' and tube T'' may be constructed so that the throat 13'' of the constriction K'' is located at the joint between the bulb and the tube as in Fig. 6; and in order to form a seal A in this constriction, a sealing slug S'' is placed in the bulb B'' before the tube is joined to the bulb. Then the bulb B'' is held uppermost with the tube T'' extending vertically downward and the pointed end of the slug S'' projecting into the throat of the constriction. Completion of the operation to form the seal may be accomplished by the heating and cooling steps previously described.

Seals in accordance with the principles of the invention are simple, easily formed, and much more economical than the seals formerly used in rare gas packages. For example, the cost of the novel seal is less than half as great as that of the formerly used glass sealing capsule, which constituted an appreciable portion of the cost of the whole package. Moreover, rare gas packages having the novel seals are not subject to premature breakage when the seals are being opened. These important and beneficial advantages are obtained while providing a seal which is gas-tight over a wide temperature range, and which is easily broken when gas is to be discharged from a package such as a rare gas bulb.

It is evident that variations in the above described seal and methods for making and breaking the seal can be made within the scope of the invention. For example, the seal is effective at temperatures at or below the room temperature at which it is formed even though the metal may

not initially have sealingly engaged the narrowest portion of the constriction.

We claim:

A method of filling, sealing and discharging rare gas in a glass bulb having integrally connected thereto one end of a single glass tube constituting both a gas-filling and a gas-withdrawal passage and having a contracted portion outside said bulb providing an internal constriction in said tube, said method comprising inserting, through the other end of said tube and adjacent said constriction, a slug or fusible sealing metal having a lower fusion temperature and a higher thermal coefficient of expansion and contraction than the glass of said tube; evacuating said bulb through said tube; filling rare gas through said tube into such evacuated bulb; sealing said tube and such filled bulb by applying sufficient heat to the outside of the contracted portion of said tube to transfer heat to and fuse said slug while holding said tube substantially vertical, to cause fused portions of said slug to flow by gravity into sealing engagement with the internal annular surface of the narrowest part of said constriction and also into sealing engagement with the internal annular surface portions of said tube adjacent the opposite ends of said narrowest

part; connecting said other end of such sealed tube to a rare gas receiver and evacuating contaminating gas from the passage between such sealed constriction and said receiver, and unsealing said tube and releasing the rare gas in said bulb through said tube into said receiver by applying, to the outside of the contracted portion of said tube, heat that is insufficient to fuse said tube but is sufficient to fuse at least those portions of the sealing metal which engage the several internal surface portions of said tube at and adjacent said constriction.

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