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Liquid Sodium Pump for the Purification of Inert Atmospheres

Russell K. Edwards, Raleigh L. McKisson, and LeRoy A. Bromley

September 28, 1950

Berkeley, California

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Liquid Sodium Pump for the Purification of Inert Atmospheres

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Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
September 28, 1950

Abstract

To maintain inert gas "dry-boxes" free of water, oxygen and other oxidizing gases a liquid sodium pump and contacting tray have been developed. The dry-box gas is circulated over streams of molten sodium.

With this apparatus it is easy to obtain and maintain an atmosphere whose reactivity is less than the best vacuums obtainable in which comparable manipulations may be accomplished.

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Research requiring the use of inert atmospheres of very high purity has necessitated the development of a scavenging device capable of removing traces of oxygen and water vapor from a closed system[1, 2]. In the work cited and in similiar work being conducted in this laboratory, certain handling and sampling operations are required to be carried out on very reactive metals and alloys. The usual technique of sweeping the working chamber - the dry-box[3] - with inert gas proved to be inadequate because of desorption of gases from newly admitted working equipment, and from all interior equipment. Further, if the dry-box has not been in operation for some period of time, desorption takes place slowly, and the sweeping technique required large quantities of high-purity inert gases. Thus, considerable expense is incurred by this procedure.

The use of molten sodium or of sodium-potassium alloy (liquid at room temperature) as "getting" agents within the dry-box was adopted since these rapidly remove both oxygen and water vapor.

The early hopes that a continually clean reactive metallic surface might be achieved due to the sinking of the solid oxides and hydroxides, whose densities are greater than those of the liquid metals, proved to be unfounded. Instead, the oxide material floats, forming a very tenacious surface film which greatly slows down the "getting" process. It was found that continual hand-skimming of the oxide surface did provide a surface which was active enough to achieve a usable atmosphere within about six hours, starting with an atmosphere of high purity attained by

sweeping procedures[1]. Purity was qualitatively judged by the fact that the liquid metallic surface would remain bright for as long as five minutes, but if very little external air is admitted to the dry-box the immediate formation of an oxide film is easily observed.

To eliminate the tedious hand process, an apparatus consisting of a pump and contacting tray was constructed (Fig. 1.). This unit was designed to sit in a container of the liquid metal and to pump the metal up the central tube from which it spilled onto the tray. The intent was that the flow of metal would carry off the oxide and hydroxide being formed, thus leaving a constantly clean active surface in the tray. A fan circulates the atmosphere and directs its flow onto the contacting tray.

This apparatus worked very well after the dry-box atmosphere became reasonably pure but the tenacity of the film formed in the usual starting atmospheres was such that the film remained on the metal surface in the tray and required skimming until the atmosphere improved. This unit was used with both the sodium-potassium alloy at room temperature and with molten sodium at 150°C. The use of the latter was more satisfactory in that the film formed was less tenacious.

A second apparatus is shown in Fig. 2. It consists of a gear pump which circulates the liquid metal and delivers it from a goose-neck delivery tube. A fan blows the atmosphere over the resulting standing stream of metal and due to the high velocity of the stream, the oxide-hydroxide film is carried away nicely even in very impure atmospheres. This device, however, was unsatisfactory due principally to the formation of such quantities of froth that actual spilling over occurred, and due also to binding in the gear pump, caused presumably by carry-through of solid particles.

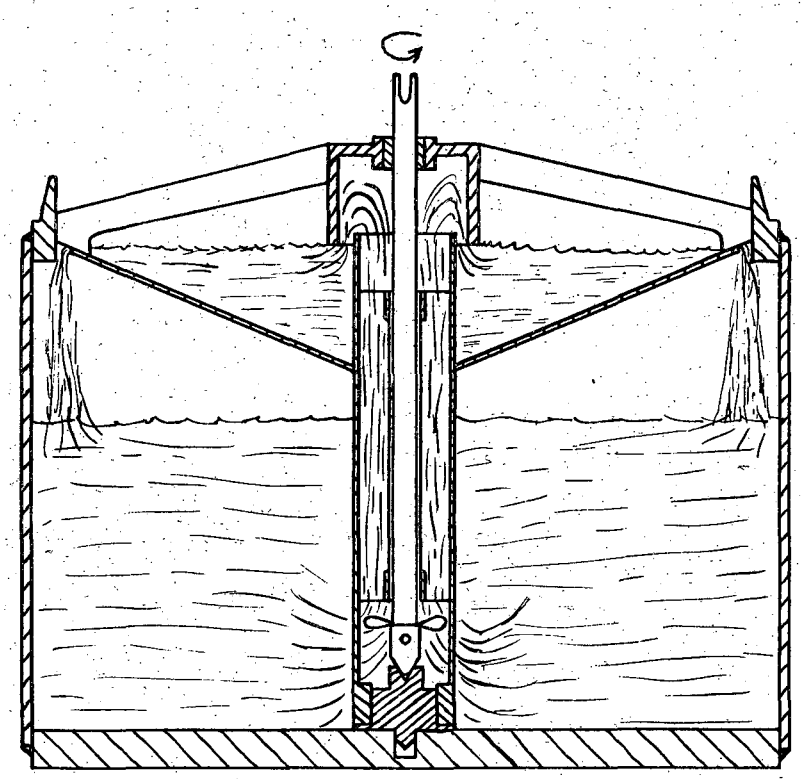

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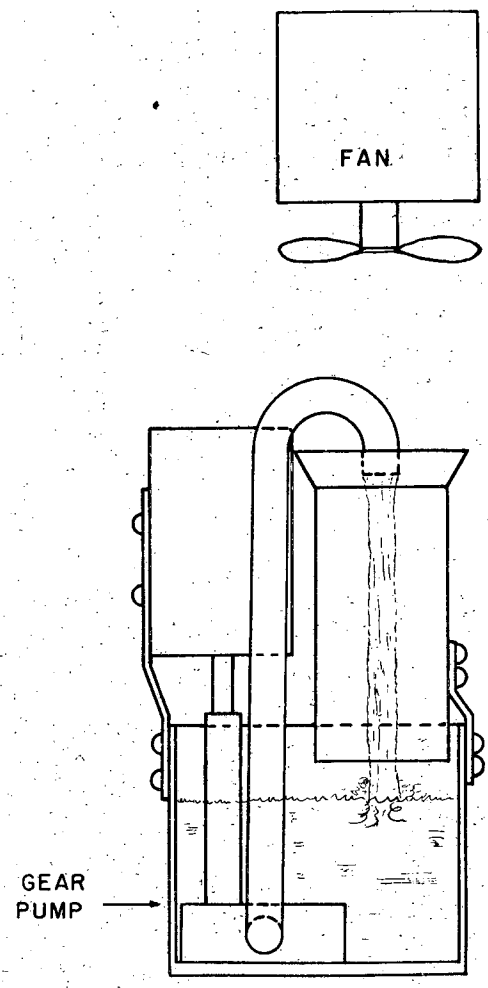
FIG. 1

IMPELLER TYPE SODIUM PUMP



MU 820

FIG. 2

GEAR TYPE SODIUM PUMP

The most satisfactory apparatus (Fig. 3.) consists of a vertical helical pump which discharges streams of molten sodium onto a downward sloping contacting tray. The helical pump element gives the discharged sodium a tangential component which is sufficient to rotate the discharge wier. This gives twelve streams of liquid sodium rotating in a spoke-like fashion and with sufficient velocity to remain free from visible oxide film. This unit operates satisfactorily when starting, and atmospheres have been attained in which drops of molten sodium have remained bright without visible oxide coating for as long as a half hour.

The simple qualitative test for atmosphere purity mentioned above was augmented by the use of the dew-point apparatus shown in Fig. 4. Although analysis was not made, one can assume that the partial pressure of oxygen will certainly be no more than factor of 15 larger than that of water vapor. It will probably be very much less, because of adsorbed water on equipment admitted to the dry-box.

The procedure for starting the scavenging process after a period of inoperation consisted of sweeping the dry-box with a volume of helium followed by two volumes of argon. The helium enters at the top and causes a downward displacement, then the argon enters at the bottom and causes an upward displacement; a total of 21 $ft.^3$ of gases are used in the sweeping operation. A slight pressure above atmospheric is maintained at all times within the dry box so that any leakage will be out rather than into the box. After two hours operation of the scavenger (Fig. 3.), the water vapor pressure is reduced to about 0.5 mm. Hg, and after eight hours it is about 0.05 mm. Hg. The minimum pressure measured corresponded to a dew point of -100°C. or about 10^{-5} mm. Hg,

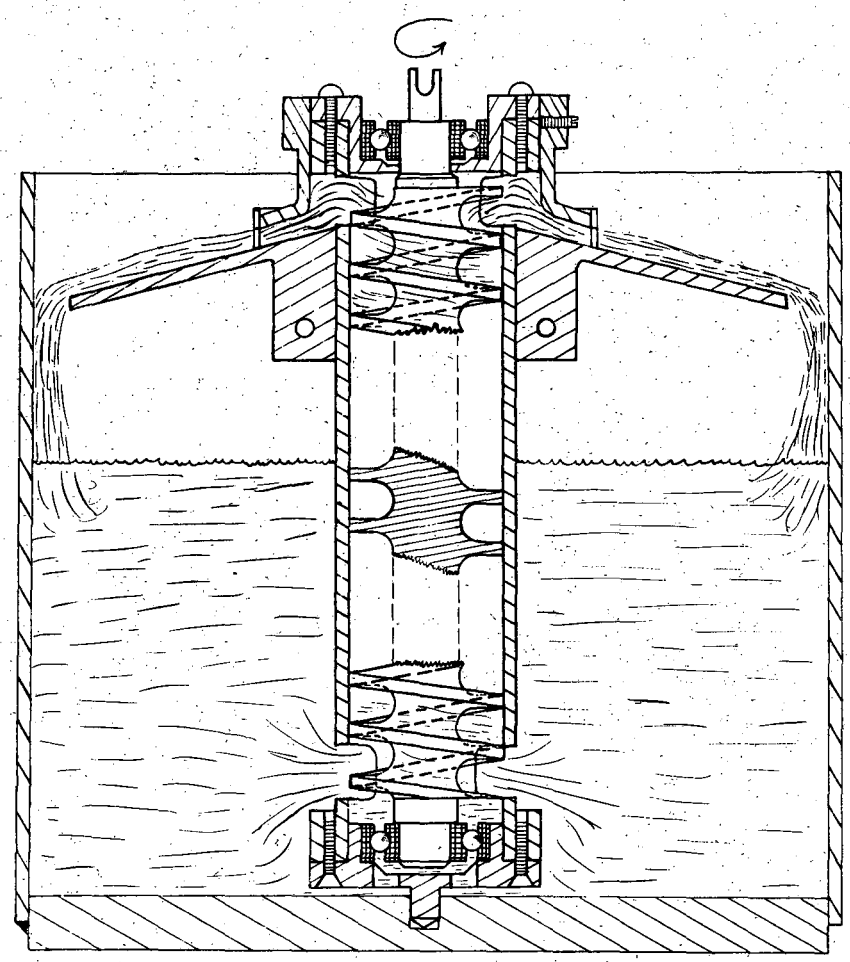


FIG. 3 M-U 821

HELICAL SODIUM PUMP – FINAL DESIGN

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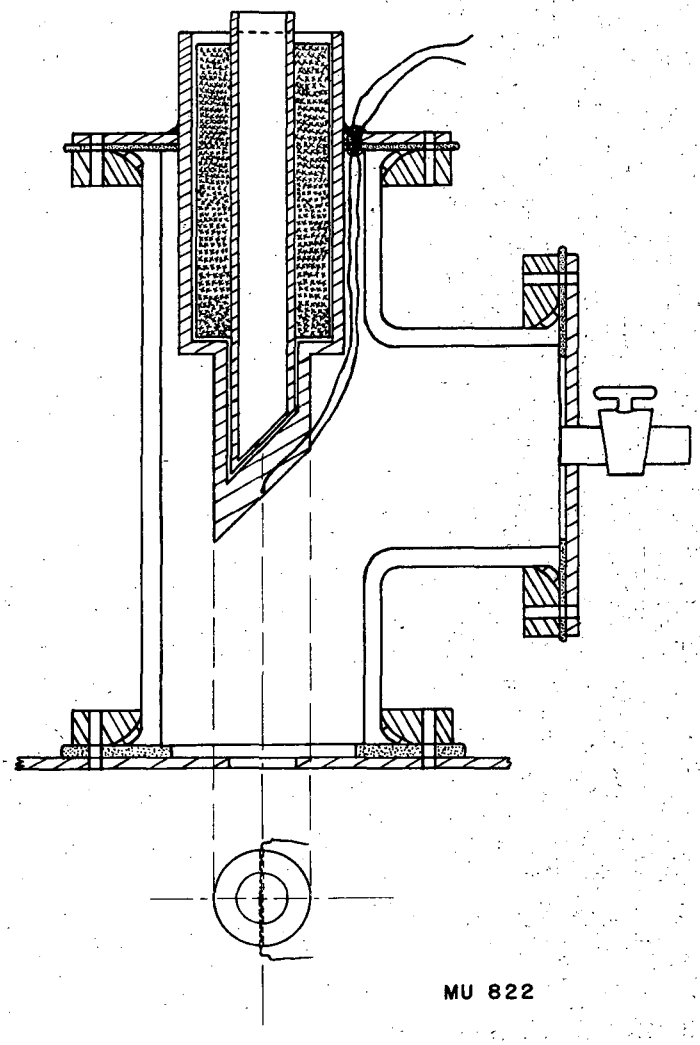

FIG. 4
DEW POINT APPARATUS

which was attained after five days of continuous operation. It should be recognized that the initial rates are very sensitive to the history of the dry-box when not in use. If one starts with a dry-box which has been left open to air, sweeps out as described above, and begins scavenging, the apparant rate of purification is slow. However, if one has obtained a good atmosphere, and has only admitted a piece of equipment through the port system, then purification can be achieved in about two hours.

It is of interest to compare the rates of oxidation of a reactive metal in a vacuum and in a dry-box. It can be assumed that the rate of absorption at the surface of the metal is instantaneous for both cases. Therefore, the rate of reaction is determined by the rate at which reactive material reaches the surface. In a vacuum, this rate is measured by the number of molecules passing through a unit area per unit time; in the dry-box, this rate is determined by the rate of diffusion of the reactive molecules through the inert molecules. The former calculation follows from the kinetic theory of gases, since in a dynamic vacuum the leak rate is equal to the pumping rate, and the mole fraction of deleterious material will usually be near 0.2. The latter calculation depends upon a knowledge of the "film thickness". Heat transfer correlations[4] can be utilized here, and for limits one can take continuous motion of one ft./sec. across a one inch cylinder, and the motion resulting from natural convection for a temperature difference of 1°F. over a horizontal plate. These film thicknesses which should represent a reasonable minimum and maximum are 0.1 inch and 0.25 inch respectively. The effective diffusivity is taken at 0.83 $ft.^2/hr.$ or 0.21 $cm.^2/sec.$ Table I shows the results of the

Table I

Dew Point °C.	v.p. H_2O atm.	Assumed Pres. of Oxygen atm.	Δt = 1°F. over Horiz. Plate: Rate $\frac{mol.}{cm.^2\ sec.}$	Δt = 1°F. over Horiz. Plate: Equiv. Vacuum* atm.	v = 1 ft./sec. over 1" cylinder: Rate $\frac{mol.}{cm.^2\ sec.}$	v = 1 ft./sec. over 1" cylinder: Equiv. Vacuum* atm.
0	6.0×10^{-3}	9.0×10^{-2}	7.8×10^{-7}	6.5×10^{-6}	3.1×10^{-6}	2.6×10^{-5}
-25	6.6×10^{-4}	9.9×10^{-3}	8.7×10^{-8}	7.2×10^{-7}	3.4×10^{-7}	2.9×10^{-6}
-50	4.0×10^{-5}	6.0×10^{-4}	5.1×10^{-9}	4.3×10^{-8}	2.0×10^{-8}	1.7×10^{-7}
-75	1.2×10^{-6}	1.8×10^{-5}	1.6×10^{-10}	1.3×10^{-9}	6.2×10^{-10}	5.1×10^{-9}
-100	1.3×10^{-8}	1.95×10^{-7}	1.7×10^{-12}	1.4×10^{-11}	6.8×10^{-12}	5.7×10^{-11}

*Total pressure (20% oxygen plus water vapor assumed).

calculations outlined above, with an assumed oxygen to water vapor ratio of 15 in the dry-box. A lower ratio would make the dry-box be even better relative to the vacuum chamber.

From Table I, one may conclude that there is a factor of about 10^4 more deleterious material present in a dry-box than total pressure in the vacuum chamber which gives an equal rate of attack. It would be virtually impossible to manipulate objects in a vacuum of 10^{-7} atm. (0.1 micron Hg), and, since a dew point of -50°C. is easily attained, the dry-box offers great advantages to work requiring inert atmospheres and ease of manipulation. Further, since dew points of -100°C. have been attained, the dry-box can surpass even the highest vacuum with respect to preventing oxidation of highly reactive materials.

Acknowledgement

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