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[54] COMBINED SPECIMEN REMOVAL AND VACUUM DISTILLATION APPARATUS

[75] Inventors: **Heribert Jentges; Oskar Burz,** both of 8520 Erlangen, Germany

[73] Assignee: **Siemens Aktiengesellschaft,** Berlin, Munich, Germany

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[52] **U.S. Cl.** ..... **266/34 R,** 23/253 R, 356/36

[51] **Int. Cl.** **C21c 7/00**

[58] **Field of Search** ..... 266/34 R; 23/253 R; 356/36, 356/38; 73/61 LM

[56] **References Cited**

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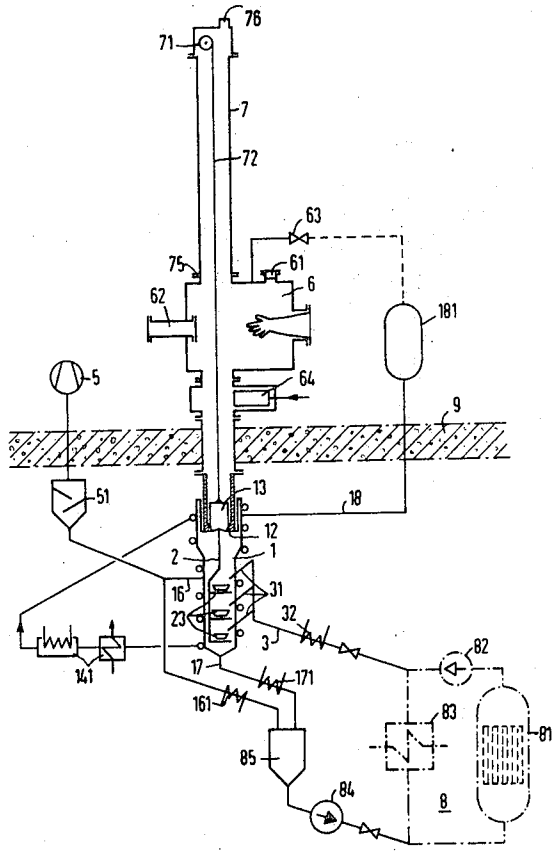
Primary Examiner—Gerald A. Dost

Attorney—Curt M. Avery, Arthur E. Wilfond, Herbert L. Lerner and Daniel J. Tick

[57] **ABSTRACT**

A combined specimen removal and vacuum distillation apparatus for discontinuously checking liquid metal for purity, such as used as a heat carrier in nuclear reactor plants. The apparatus comprises a vessel of stainless steel forming a processing chamber and having wall-temperature control means and a removable top cover, means for positionally securing the cover on the vessel against excess pressure in the chamber, a plurality of removable specimen containers, holder devices mounted on the cover in suspended relation thereto for supporting the respective specimen containers, the holder means having respective heating means and respective temperature sensors, a lifting device having a tube vertically extending above the cover and tightly joined with the vessel chamber, a lifting mechanism disposed in the tube and connected to the cover for lifting the cover and the specimen containers, and a glove box and a specimen sluice interposed between the vessel and the tube of the lifting device.

10 Claims, 2 Drawing Figures



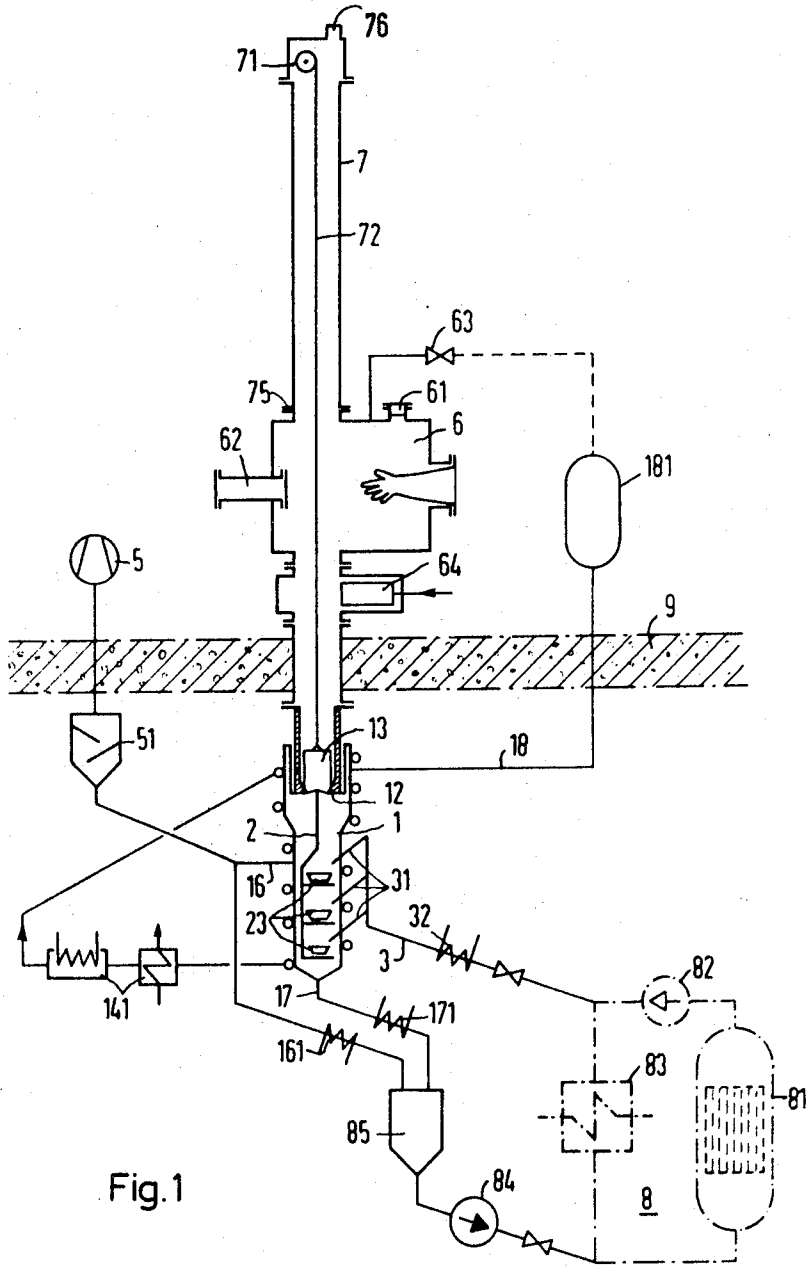

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Fig.1

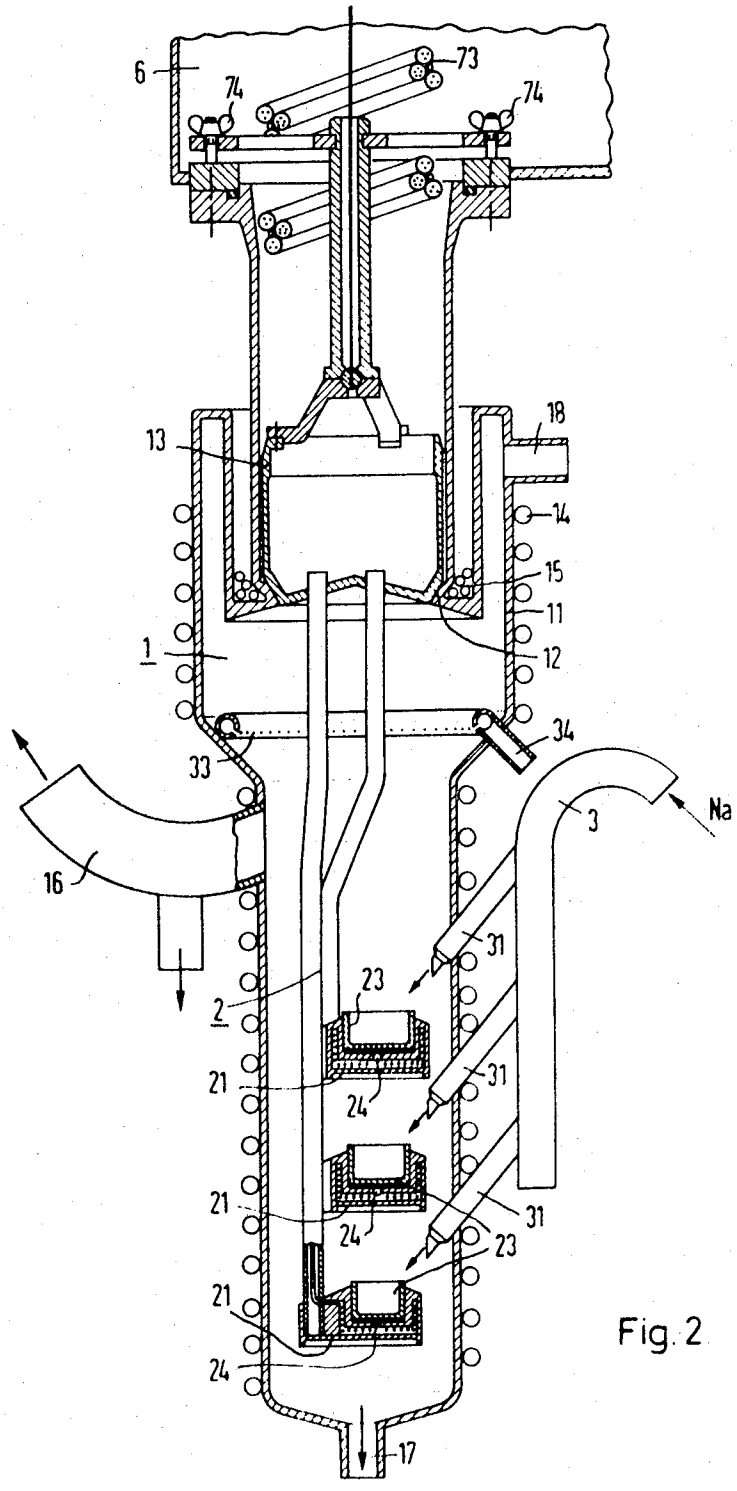
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Fig. 2

COMBINED SPECIMEN REMOVAL AND VACUUM DISTILLATION APPARATUS

Our invention relates to a combined sampling and distilling apparatus applicable to a discontinuous process for checking the purity of liquid metals, such as those used as heat carriers in nuclear reactor plants.

In liquid-metal systems, especially liquid-metal circulation systems for the conveyance of heat, chemical analysis of the liquid-metal melts to detect impurities, especially oxides, is of the highest importance since the rate of corrosion of structural materials depends essentially on the degree of purity of the melt. Since reliably-operating continuous apparatus for determining and recording such impurities do not exist, such determinations can be effected only by discontinuous processes of chemical analysis.

The chemical analysis of liquid-metal samplings, particularly oxygen determination, in any liquid-metal system is difficult since, on account of the extremely minute impurity content, very slight traces of oxygen suffice to critically adulterate the samplings. In nuclear-reactor primary-circulation systems using such liquid metals, radioactive radiation is also present, allowing a chemical analysis of a few cubic centimeters of the melt only after the metal has been removed.

Distillation of the liquid metal is therefore a most promising process as a pre-stage for such chemical analyses, since — with total vaporization of metal — no further metallic oxide can be generated through oxygen, and radioactive radiation due predominantly to the radioactivity of liquid metal is substantially reduced. The residue of such samples can then be investigated for the types and amounts of impurities by means of chemical analysis or flame-spectrometric methods.

Such methods are well known, as illustrated by reports LA-3343 and LA-3879 of the Los Alamos Scientific Laboratory, published respectively in 1965 and 1968 by the University of California. The apparatus set forth therein are very complex and are equipped with vitreous distilling vessels. For safety reasons, they are therefore unsuitable for the continuous operational investigation of liquid-metal samples, particularly if they become radioactive through irradiation in nuclear reactors.

It is an object of the present invention to devise apparatus which allows sampling as well as distillation without interference with the liquid-metal circulation system.

Another object of the present invention is to conserve the distillation residue, if necessary, for subsequent investigation before it comes into contact with oxygen.

Still another object of the present invention is to provide an apparatus which saves time when charging complex industrial apparatus, such as in sodium-cooled nuclear reactor installations.

Another object of the present invention is to eliminate the requirement for transporting the samples or specimens prior to distillation.

Other objects, advantages and features of the present invention will become more apparent from the following description.

According to our invention, the combined sampling and vacuum distillation apparatus consists of a stainless steel chamber with wall temperature control and a removable top cover which prevents excess pressure when in position, and serves as a base for a number of movable sample tanks which include respective holding devices provided with heating means and temperature sensors. Furthermore, a lifting duct is arranged above the cover and tightly joined to the vessel chamber for lifting the cover together with the sample tanks through a glove box and an outwardly discharging sluice.

This apparatus contains no glass components and can be arranged in such a manner that the distillation chamber itself together with the liquid-metal circulation systems — which can also be contaminated with radioactivity — are installed behind a suitable shield, such as cement, while the distillation residue can be taken away in a glove box situated outside the protected area, for further examination outside the chamber. To some extent the further investigation can take place inside the glove box, but the samples can also be taken out through a sluice. Renewed oxygen contamination can be avoided by using a suitably pure protective gas. Since a shield is provided between the glove box and the distillation chamber itself, a further distillation cycle can commence during the examination of the residue. The present invention provides for time saving which is a particular advantage when charging such apparatus on the industrial scale, for instance, in sodium-cooled nuclear reactor installations.

The above-mentioned and further objects, advantages and features of our invention will appear from the following FIGS. 1 and 2 showing the application of the present invention in conjunction with a liquid-metal cooled nuclear reactor installation.

FIG. 1 shows the interaction of the invention with the nuclear reactor plant.

FIG. 2 is an enlargement of FIG. 1 showing only the distillation chamber.

As mentioned, for minimizing danger the present apparatus is constructed of stainless steel and designed to stand an operating temperature of 600°C (1,112°F) and a maximum pressure of 16 atmospheres.

FIG. 1 shows our apparatus in conjunction with a nuclear reactor circulation system **8** for a liquid-metal coolant. System **8** comprises a reactor **81**, a pump **82** and a heat exchanger **83**. The supply conduits to the distillation apparatus further include a a pump **84** and reflux tank **85**. The distillation chamber **1** is connected with the liquid-metal circulation system through first and second outlet lines **16** and **17** and inlet line **3**, with the reactor plant lying behind a shield **9** of concrete.

Beyond the shield, in a vacuum-tight extension of chamber **1** is a glove box **6** and lift tube **7** wherein is mounted a windlass **71** having a rope **72**, whereby a cover **13** of chamber **1** with the distillation residues suspended therefrom can be lifted by means of the rope through the concrete shield into the glove box for further treatment. As most of the radioactivity is in the liquid metal, the distillation residues contain only very little radioactivity and therefore are easily manipulated without fear of contamination. Beyond the concrete shield is a vacuum pump **5** connected to chamber **1** through a liquid-metal condenser **51**, and a storage tank **181** for the protective gas.

As a feature of our invention, there are no plant components subject to repair inside the concrete shield, the sample tanks **23** with their heating means can be driven

for replacement into the lift tube 7, which can be inspected after closing the supply conduit with a vacuum-tight shut-off slide 64 on flanged joint 75, so that repair or replacement of such parts is comparatively easy. The high-vacuum slide 64 can also be closed when charging or discharging the system, thus providing a two-fold protection against an unwanted supply of air, (for instance, the handling gloves 68 becoming ripped).

According to FIG. 2, chamber 1 consists of a longitudinal steel vessel 11, which is provided with an external heating means 14. Since changes of temperature of this vessel are required for accomplishing the distillation process, heating means 14 is capable of cooling, working, for example, through a flow of liquid-metal having a temperature capable of being adjusted, whose circulation system is not shown in FIG. 2. Intensive cooling produces a short distillation time and condensation of the highly radioactive sodium vapor inside the chamber.

Chamber 1 is closed by cover 13 having a sealing seat 12 provided with its own heating or cooling means 15 which reenters into the chamber space 1 to balance the thermal stress. The seat 12 has an upward tubular extension on which the cover 13 is maintained in a tightly sealed position by wing screws 74. The screwing down of the cover is accomplished inside glove box 6 and can be loosened by hand outside the concrete shield.

Cover 13 also serves as a carrier for the sample tanks 23. For this purpose it is provided with a holder frame 2 which is formed by three rigid supporting frames projecting downwardly, onto which the specimen holding devices 21 with heating means 22 for the sample tanks 23 rigidly fastened. The cylindrical shape of sample tanks 23 combined with the rounded transfer from the base to side wall and the cylindrical heating conductor assemblies 22 promotes the heat input by short heat conducting paths and intensive convection to the liquid level such that the short distillation time is achieved without retardation of boiling.

The electrical connections for the heating devices 22 run through the holder frame 2 as do the connecting lines for temperature sensors 24 mounted beneath the sample tanks 23 inside the holding devices 21. These lines are carried upward above cover 13 in the form of a flexible, helical-wound cable 73, and drawn out (in a manner not shown in drawings) through vacuum-tight lead-in means from the lift tube 7 to the height of drum or windlass 71 and connected to respective measuring devices and/or sources of power (not shown).

The comparatively slight bulk of the holder or supporting frame 2 facilitate excellent temperature control, which is of great importance for setting the precise distillation temperature. Chamber 1 is furthermore provided with an inlet 18 for the delivery of protective gas and is connected through supply tubes 31 with a liquid-metal supply 3 merging with the reactor circulation system. The tubes 31 terminate in the interior of chamber 1 in the form of jets and feed the liquid metal to be investigated to respective ones of the sample tanks 23. Chamber 1 is further linked through outlet 16 to vacuum pump 5, the outlet line 16 supplying liquid-metal into a reflux tank 85 when there is an overflow.

Emptying of chamber 1 is accomplished by discharge line or outlet 17 which also leads into reflux tank 85. Closing of these inlet and outlet lines 3, 17 and 16 is effected by freezing of gaskets 32, 171 and 161 which are ganged in a conventional manner with a cooling apparatus (not shown). The freezing of the gaskets ensures a hermetic seal. The interior of chamber 1 is further provided with a rinsing means 33 which is connected to the liquid-metal circulation system through line 34. The sealing of cover 13 against tank 11 is achieved by the freezing-in of the liquid-metal condensate between the chamber and the sealing or closing cone formed between cover 13 and seat 12. The sealing seat is cooled down by the cooling and/or heating means 15° to 30° to 40°C beneath the freezing or solidifying temperature of the liquid metal, so that with heat conduction, the sealing cone resting on the sealing seat is at 20° to 30°C above the freezing point. The liquid metal or the vapor thereof condenses on the bulging bottom of the sealing cone, runs down to the closure gap of the cone and freezes on the sealing seat itself. After the distillation process in chamber 1 terminates, the frozen gasket is melted by warming heating coils 15. The condensate runs off outwardly and is prevented from dripping into the crucibles 23. Normally, this apparatus is not subjected to any special pressure load, but if some unforeseen blockage of overflow 16 were to occur, a maximum excess or overpressure of 16 atmospheres might occur which, as already mentioned, will be absorbed by the cover-fastening screws 74.

The operation of our apparatus as a whole will be described as follows for a better understanding thereof.

First, chamber 1 is evacuated by means of pump 5 and thoroughly baked under the heating and/or cooling means 44. The heating and/or cooling fluid must be fluid and stable in the range from 0° to 650°C and impervious to radioactive radiation which might be emitted from the contents of the chamber. Potassium-sodium melt is suitable, for instance. Subsequently, purified argon is poured into the chamber as the protective gas and the liquid metal, sodium, for example, for examination is squirted through lines or tubes 31 from the reactor circulation system into the sample tanks 23. The overflowing metal through outlet line 17 forms a freezing plug in the gasket 171, so that the liquid-metal level in chamber 1 rises up to the overflow level. The overflowing melt flows back into the reflux tank 85 and from the reflux tank through pump 84 into the reactor circulation system. When the liquid metal initially flows through chamber 1, which is at a temperature of between 500° and 600°C, residual gases especially oxygen are chemically combined and washed away by the liquid-metal or vapor, and therefore no longer interfere with the sample after the metal commences being distilled. The tightsealing of the cover seating is, as mentioned above, secured by the metal condensate. After about 4 hours, the inflow of liquid-metal is shut off and chamber 1 empties through the lower discharge outlet line 17 through the melting of the frozen plug 171. The liquid-metal supply line is shut off by freezing of the metal at point 32.

The chamber cooling circulation flow is now commenced. In particular, the tubes of heating means 14 are supplied with coolant fluid and vacuum pump 5 is energized as soon as the wall temperatures drop to below 100°C and the crucible temperatures to about 350°C, as sensed by means of the temperature sensors 24. As soon as the required vacuum is obtained, the

crucible heater begins to operate, the liquid-metal present is distilled off and again liquefied on the chamber walls and in condenser **51**.

The end of the distillation is indicated by a sudden rise in temperature, so that after a short heating interval, preferable for reliable vaporization of any liquid-metal residues, the heating can be turned off. This completes the distillation process. Chamber **1** is purged with purified argon, and the condensate seal on cover **13** is melted.

Glove compartment **6** and lift tube **7** have been filled during the above processes with the protective gas, enabling cover **13** with the sample tanks **23** dependent thereon to be raised manually or by an electrically operated winch turning drum **71**.

Inside the glove box, the sample tanks are taken off the holder devices **21**, given further treatment there and/or conveyed through sluice **62** to further testing equipment (not shown). A samplings carrier under protective gas is required only if the samples are to be analyzed for elements present in the atmosphere and which react with the residuum. The crucible carrier is subsequently charged with fresh sampling tanks and again set in movement. The next filling and distillation process can immediately start, since a renewed bake-out of wall partitions of chamber **1** is no longer required. The walls of the vessel **11** can be cleansed in rotation through rinsing means **33**, connected through an inlet pipe **34** with a liquid-metal storage tank. Possible residues in the holding devices **21** can be rinsed clean without sample tanks.

With respect to the design of the sampling tanks, they can be made, for example, of superpure nickel, and it is preferable to round off the junction between the lateral walls and the base in order to obviate any boiling of the liquid metal and formation of bubbles. The same end is served also by the direct heating shown in FIG. 2 for the sampling tanks, on the surfaces of the shell, and their indirect heating in the fissure filled with liquid metal between the container base and holder device **21**. This interstice is dimensioned in such manner that on distillation the sampling tank **23** boils first and then the fissure. The vacuum thermocouple **24** senses the temperature of the liquid metal in this fissure.

With a sampling tank capacity of about 15 cm^3 (appr. 1 c. inch) the sodium throughput time will be about 4 hours as previously mentioned and the distillation time about one-half an hour, so that the examination of the distillation residue and likewise of the liquid metal circulating in the circulation system can be effected during this period. It is thus possible, with the plant, to survey the condition of the liquid-metal circulation systems and other continuously-working metering equipment and/or readjust the same.

This constitutes an extraordinary advantage of our apparatus, in that the sampling tanks filled with liquid metal do not have to be transported prior to distillation. Other uses are also obvious, as well as other constructional variations from the example here shown.

It will thus be seen that the objects set forth above, among those made apparent from the preceding description, are efficiently attained and, since certain changes may be made in the above method and apparatus without departing from the scope of the invention, it is intended that all matter contained in the above description and shown in the accompanying drawings shall be interpreted as illustrative and not in a limiting sense.

We claim:

1. A combined specimen removal and vacuum distillation apparatus for discontinuously checking liquid metal for purity, such as used as a heat carrier in nuclear reactor plants, comprising a vessel of stainless steel forming a processing chamber and having wall-temperature control means and a removable top cover, means for positionally securing said cover on said vessel against excess pressure in said chamber; a plurality of removable specimen containers; holder devices mounted on said cover in suspended relation thereto for supporting said respective specimen containers, said holder means having respective heating means and respective temperature sensors; a lifting device having a tube vertically extending above said cover and tightly joined with said vessel chamber, a lifting mechanism disposed in said tube and connected to said cover for lifting said cover and said specimen containers; and a glove box and a specimen sluice interposed between said vessel and said tube of said lifting device.

2. In apparatus according to claim **1**, said specimen containers being arranged above one another.

3. In apparatus according to claim **1**, said vessel having a liquid-metal inlet, a first liquid-metal overflow outlet, a second liquid-metal overflow outlet and a protective gas connection duct; a vacuum pump connected to said first overflow outlet, and a condenser interposed between said vacuum pump and said first overflow outlet.

4. Apparatus according to Claim **1**, comprising a vacuum-tight shut-off slide between said vessel and said glove box for closing and opening said chamber relative to said glove box and sluice.

5. Apparatus according to claim **1**, said specimen containers being crucibles, and said heating means for each of said crucibles being an electric resistance heater arranged on said holder device for heating the crucible content through the crucible wall surface.

6. In apparatus according to claim **5**, said crucibles having a cylindrical cup shape and a rounded edge portion between the bottom and the cylindrical wall of said cup shape.

7. In apparatus according to claim **1**, said lifting mechanism comprising windlass having a rope pull mounted in said tube near the top thereof and having a rope extending from said windlass to said cover.

8. In apparatus according to claim **7**, said rope extending substantially along the center axis of said tube.

9. Apparatus according to claim **1**, comprising flexible helical windings of cables for electrical connection to said heating means and to said temperature sensors, said helical cable windings extending upwardly through said tube in substantially coaxial relation thereto.

10. In apparatus according to claim **9**, said vessel having vacuum tight lead-in means for said cables, said lead-in means being located near said windlass.

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