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DRYING PROCESS AND APPARATUS THEREFOR FOR REMOVING SOLIDS FROM LIQUID MIXTURES

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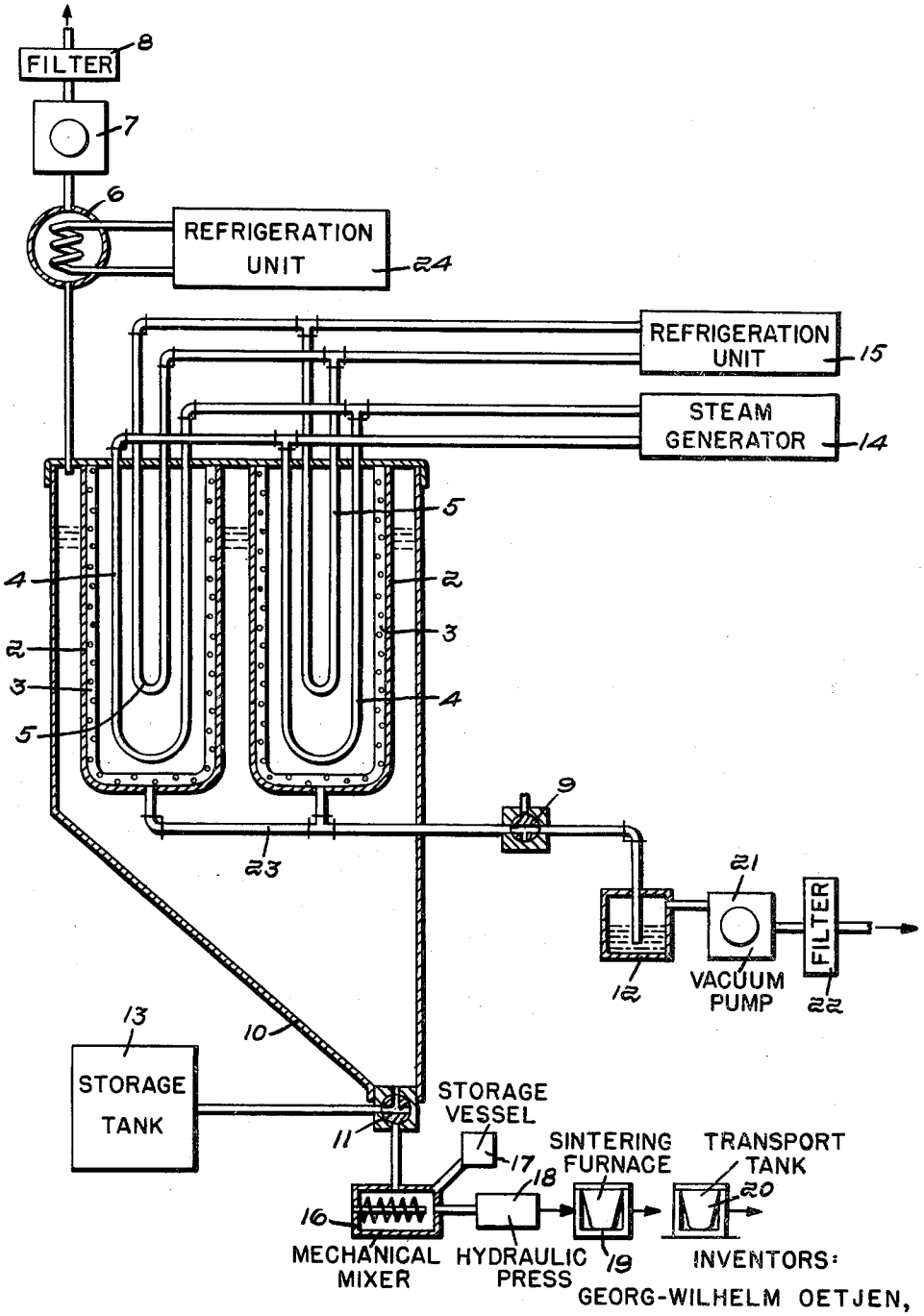

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DRYING PROCESS AND APPARATUS THEREFOR FOR REMOVING SOLIDS FROM LIQUID MIXTURES

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The present invention relates generally to a process and apparatus for converting radioactive waste products into a permanent form for storing and/or transportation purposes, and which may also be used for forming concentrations of radioactive isotopes.

In various nuclear chemistry processes, radioactive waste products in the form of precipitates in liquids are formed. They must be removed from the liquids and changed into a suitable and compact form for transportation or storage purposes. However, in actual practice the concentration and precipitation of radioactive constituents present considerable difficulties.

Accordingly, it is a main object of the present invention to provide a process for separating radioactive substances in liquids into a permanent form so that they may be stored and transported as desired.

A further object of the invention is to provide a process for forming concentrations of radioactive isotopes.

Another object of this invention is to provide a device for accomplishing the concentration and precipitation processes.

These objects and others ancillary thereto are accomplished according to preferred embodiments of the invention, wherein the radioactive substances which are constituents or ingredients of a liquid are separated from the liquid by means of filters. It should be noted that it is unimportant whether or not these ingredients or constituents are insoluble substances produced by a chemical precipitation process, or crystalline deposits which are formed, for example, by lowering the temperature of a solution, or in any other suitable manner in a solution which has a lower amount of radioactivity than the separable radioactive constituents.

The liquid containing the waste products is passed through filter elements so as to separate the waste products from the liquid and deposit them onto the filter elements as a filter residue, which is then subjected to a vacuum freeze drying process, known per se, to thereby dry the filter residue. This residue is then mixed with nonradioactive fillers, if necessary, and may be introduced into a transport or storage tank. Thus, the filter residue is subjected to a freeze drying process which may be carried out in the conventional manner under a high vacuum in a drying chamber which is suitably sealed to be air and gas tight. This vacuum freeze drying process usually yields a powdery dry product as a result thereof so that subsequent comminuation, such as is preformed as a preliminary step for melting or sintering processes, is generally not necesary. The product of the vacuum freeze drying operation may be easily mixed with nonradioactive fillers and may be encased or enclosed in a solid body, e.g. concrete.

If desired, the waste product which has been subjected to the vacuum freeze drying operation may be mixed with a filler and the combined material may be melted or sintered in processes known per se. This may preferably be accomplished by first using a pressing process so that a pressed object is formed and thus a solid body may then

be formed by a melting or sintering process. Although melting and sintering processes of radioactive waste products are already known, particularly desirable and favorable results are obtained when the vacuum freeze dried waste products are used as the starting material.

Upon termination of the vacuum freeze drying process, the vacuum freeze dried waste products have been deposited on the filter elements as a loose coating, and it is desirable to remove this filter residue or at least as much of it as possible, without the need for physically transporting or moving the filter elements and without resorting to operations which are difficult to arrange for remote control. Therefore, a further feature of this invention is the removal of the radioactive substances or the filter residue which coats the filter elements, by gas or steam currents which are suitably directed to accomplish this purpose. Such a current may preferably be generated by ventilating the filter chamber by means of a vacuum. Also, the atmospheric pressure may be utilized to force the liquid, which is to be subjected to a filtering process, into the filtering chamber.

Although in the foregoing description of the process only separation of radioactive products from liquids has been discussed, the process may also advantageously be used to provide concentration of radioactive isotopes. In order to accomplish this, these substances are first presented in the form of a liquid having filterable ingredients or constituents which are filtered out of the liquid and which generally represent a concentration of these isotopes. In this process also the radioactive substances which are deposited on the filter elements as a filter residue are subsequently subjected to a vacuum freeze drying process and thus are in a dry state representing a concentrate.

The filter elements may be used as charging vessels for the radioactive substances during the vacuum freeze drying process. In this event, the filter chamber is designed as a vacuum freeze drying chamber and is connected to pumping devices which are known per se and which are used for the separation of vaporous constituents or ingredients, and especially water vapor. The pumping devices may be constructed of a deep freeze condenser having a single or multiple stage mechanical pump with the use of a Roots pump if necessary. Since there may be radioactive substances in gaseous form and in the form of suspended particles, the pumps must not exhaust the gases or vapors drawn off, directly into the surrounding atmosphere. Instead, a preliminary absolute filter must be arranged to remove these radioactive substances and prevent them from being discharged into the atmosphere.

One device for carrying out the above-described processes may have several filter elements arranged in a filter chamber, with the elements being selectively operatively associated with heating and/or cooling devices. Such filter elements may be filter sacks or bags which are stretched over heating and cooling grates disposed next to each other in pairs. The heating arrangement is important because by this means a rapid runoff or carrying out of the vacuum freeze drying process is assured. It has proven especially advantageous to use a so-called "vacuum steam heating" arrangement in the heating device, which is a system sealed from the atmosphere and in which a vaporous heating medium, such as steam, which is preferable, is used and at an appropriately low vacuum air pressure.

When the present process is used in the separation of radioactive waste products one of the important advantages thereby provided is the great decontamination which is achievable. However, this may be improved even further by adding cooloids or gels to the liquid before the filtering process, on which the substances to be separated will preferably be deposited. The addition of this ma-

terial is also advantageous when used in the concentration of radioactive isotopes.

Additional objects and advantages of the present invention will become apparent upon consideration of the following description when taken in conjunction with the accompanying drawing wherein the sole figure diagrammatically indicates apparatus for carrying out the process.

With more particular reference to the drawing, an inlet filter chamber **1** is provided which may be sealed to be vacuum tight and in which two filters membrane bags or sacks **2** are stretched over perforate pipelines **3**. The interiors of lines **3** form outlet filter chamber which are connected to storage tank **12** for receiving the filtered liquid by means of a connecting line **23**. In this storage tank an appropriate vacuum pressure is produced by means of vacuum pump **21**. This pressure is sufficiently less than the pressure in inlet filter chamber **1** to force liquid in the inlet chamber **1** through the filter sacks into the outlet chambers **3** thereby providing a vacuum filtering process. The gas discharge of vacuum pump **21** is distributed into the surrounding atmosphere through an absolute filter **22**.

Tubular heating and cooling pipes **4** and **5**, respectively, are disposed interiorly of the filter bags **2**. A deep freeze condenser **6**, which is fed by a compression cooling unit **24**, as well as a multiple stage mechanical vacuum pump **7**, are connected to the inlet filter chamber **1**. Thus, the outlet filter chamber **3** and the inlet filter chamber **1** may be used together as a vacuum freeze drying chamber for carrying out the vacuum freeze drying process, which process is known. The gases and vapors drawn off by means of these units have their vaporous constituents separated therefrom in the deep freeze condenser **6**. The condensate from this condenser **6** may be collected in a sealable storage tank (not shown), but appropriate precautionary measures are mandatory. The permanent gas constituents or ingredients drawn off by the multiple stage vacuum pump **7** pass through a further absolute filter **8** before being distributed into the surrounding atmosphere.

A ventilation valve **9** is disposed in the connecting line **23** by means of which the interior of the filter bags or sacks **2** may be ventilated. In order to avoid undesirable conglomerations of the powdery product which is collected in the funnel-shaped collecting chamber **10** of the inlet filter chamber **1**, mechanical or electromagnetic shaking devices, which are known per se, may be provided. These devices at the same time assures satisfactory operation of the closure member of sealable removal opening **11**.

A storage tank **12** receives and stores all the liquid liquid filtered during the filtering process, which liquid is discharged to this storage tank by the vacuum pressure produced by means of mechanical vacuum pump **21**. This liquid has already been filtered. The unfiltered liquid is fed from storage tank **13** to the deepest portion of filter chamber **1** in which a suitable liquid level is produced and may be maintained. The filtrate which is collected in the storage tank **12** may be returned to storage tank **13** by, for example, conventional piping(not shown) if necessary so that a multiple stage filtering process may be carried out.

Preferably, heating pipes **4** are designed as hollow condensation elements which may be supplied with steam from a vacuum steam generator **14** which may have a temperature rating of about 50–80° C. This type of heating provides particularly uniform heat distribution assuring uniform temperature. Cooling pipes **5** are evaporators of a compression cooling unit **15**. For this purpose a particularly advantageous arrangement may be provided under some circumstances whereby the compression cooling unit **15** feeds the deep freeze condenser **6** as well.

The mechanical device **16**, which is connectable with opening **11**, is provided, and to which a filler may be supplied from a further storage tank **17**. During the treating process a hydraulic press **18** treats the material after

mechanical device **16**, and the mixture is compressed to form an appropriately pressed body or object. This object is fed to a melting or sintering furnace **19** in which it is melted or sintered into a homogeneous solid body, preferably having vitreous properties. The product of this melting or sintering process may then be introduced into biologically shielded sealable transport tanks **20** and may be transported to any suitable location in this manner.

In carrying out this process, the initial liquid to be filtered is fed from storage tank **13** into inlet filter chamber **1** until a suitable liquid level is attained, and which is maintained during the filtering process by refilling if necessary. The liquid flows about the outer surfaces of the filter bags **2** and is forced towards the interior of the filter, since a suitable vacuum pressure is produced in collecting line **23** by the vacuum pump **21** which exhausts the permanent gas removed through the filter **22** into the atmosphere. The filtering process may then be carried out in a conventional vacuum filtering manner by maintaining a reduced pressure in the upper portion of storage tank **12**. The filtrate is fed into the lower portion of storage tank **12** while the substance or substances forming the filter residue are deposited as a surface coating on the outer surfaces of the filter bags **2**. The filter bags may advantageously be constructed of a synthetic resin, such as vinyl, for example.

After the filtering process has continued for a period of time for a sufficiently thick coating of the filtrate to be formed on the exterior of filter bags **2**, inlet filter chamber **1** is emptied of the, as yet, unfiltered liquid which is present and this is returned into storage tank **13** and the valve **9** is closed. The filter residue, which is a pasty or paste-like substance, is now subjected to a freeze drying process in the evacuable drying chamber formed by outlet filter chambers **3** and inlet filter chamber **1**. For this purpose the deep freeze condenser **6** is placed into operation together with the multiple stage mechanical vacuum pump **7** which reduces the pressure in the drying chamber by removing permanent gas and exhausting it into the atmosphere through filter **8**. The water vapor which is sublimated from the ice which is thereby formed, is deposited in the deep freeze condenser **6**. This provides a particularly favorable decontamination of the material because no temperature increase is required for driving off the volatile substances, such as is needed, for example, in vaporizing processes. During the vacuum freezing process the temperature of the material to be dried is suitably controlled by the heating and cooling pipes **4** and **5** so that the operation may be carried out under optimum drying conditions, i.e., with the shortest drying time.

Upon termination of the vacuum freezing process, the dry product is in the form of a loose surface coating on the exterior surfaces of the filter bags **2**. This product is discharged from the filter bags **2** by admitting atmospheric pressure to the interior of the filter chamber by means of ventilation valve **9** which causes the product to then drop into the funnel-shaped collecting chamber **10** of the inlet filter chamber **1**. Then, the dry product is removed through the sealable removal opening **11** and is fed to mixer **16** where it may be mixed with a nonactive filler. The pressed object may be formed in the hydraulic press **18**, and may then be melted or sintered in the melting or sintering furnace **19** to thus form a solid body having vitreous properties. In a modification of this method of operation, the melting or sintering process may be omitted and another nonradioactive filler from the further storage tank **17** may be used.

For this purpose the mixture may be placed into appropriate molds or forms.

The plant which is diagrammatically shown in the drawing is, in practice, provided with a number of cutoff valves by means of which individual structural elements may be disconnected from the complete plant for purposes of cleaning or repair. The sealing elements which are to be arranged in addition to the biological sealing are not

shown in the drawing for purposes of clarity. Instead of the two filter bags 2 shown, a larger number of such elements may be used and if necessary a multi-stage filtering process may be provided by removing the already filtered liquid from storage tank 12 to storage tank 13 for additional filtering.

When using this process for the concentration of isotopes, the particular advantage of this process is that the heat effect which could possibly bring about undesired or destructive temperature increases may be eliminated. Even the use of heated grates is not contrary to this, because the heat supplied to the material to be filtered by means of these grates only compensates for the evaporation cooling which occurs during the vacuum freeze drying process thereby preventing an undesired lowering of temperature of the product to be dried, which would substantially increase the drying time. In such concentration processes the starting material may be a concentrated solution wherein, by physical or chemical means, filterable constituents or ingredients having higher radioactivity than the surrounding solution may be obtained. Then, the filtering process already described is carried out. This process is generally interrupted after a certain thickness of the filtered coating has been provided, for example, 1.5–2 cm., and the unfiltered liquid remaining in the chamber 1 may be returned to the storage tank 13. A vacuum freeze drying process of the known type then follows, and the dry product may be further processed as the concentrate of the corresponding istope.

The novel process comprising the present invention is not limited to use with aqueous liquids, but may also find utility with other starting liquids, especially those of the organic type.

Example of process for "Freeze-Drying Method for Radioactive Substances":

The contaminated waste water in the storage container will be treated with 0.4% aluminum-ammonium + ammonia for precipitation. The solution is sucked through the filter cloths 2 by means of the evacuation of the outlet filter chambers 3 and the storage container 12, whereby the precipitating agent deposits with the enclosed radioactive particles on the outer side of the filter cloths. The applied underpressure is, according to the thickness of the filter cake and the vapour pressure of the liquid, about 100 mm. Hg. The filtering process is finished after formation of a thickness of filter cake of 2 cm. in 2.5 hours. At this time the water content of the filtrate is 98.5%. After the residual unfiltered solution in the chamber has refluxed into the storage container 13, the filter cake will be frozen to −30° C. by the cooling pipes 5, which are cooled by means of the refrigerating machine 15. For the drying process itself, which begins now, the inlet chamber 1 will be evacuated to 0.2 mm. Hg by the vacuum pump 7, the cooling condenser 6 is cooled to −40° C. by the refrigerating machine 24, and the necessary heat of evaporation is fed to the frozen product at heater tube temperature up to 60° C. via the heater tube 4 by the steam generator 14. The heater tube temperature of 60° C. is controlled such as to ensure a product temperature of −25° C. to −30° C. The pressure in the chamber is 2×10^{-1} mm. Hg. The drying process lasts 36 hours. By intermittent admission of atmosphere on the inner side of the filter cloths 2 the now dust shaped material is shaken off down the funnel of inlet chamber 1, from where it is led through the valve 11 to the mixer 16 and is thus available for further treatment. The ice frozen on condenser 6 is thawed in half an hour by admission of atmospheric pressure. The contamination of the water thus defrosted from the condenser 6 is by the factor 10^4 smaller than that of the initial solution.

It will be understood that the above description of the present invention is susceptible to various modifications, changes, and adaptations, and the same are intended to be comprehended within the meaning and range of equivalents of the appended claims.

What is claimed is:

1. A process for converting filterable radioactive waste products included in a liquid into a permanent form capable of being stored and transported, comprising the steps of: filtering within an evacuable drying chamber a liquid containing radioactive waste products to separate the waste products from the liquid in the form of a filter residue; and vacuum freeze drying in situ the filter residue thereby forming a product which may be more easily handled.

2. The process according to claim 1 including the steps of removing permanent gas from the evacuable drying chamber during the freeze-drying step, filtering the permanent gas removed from the drying chamber during the freeze drying step, and exhausting the filtered permanent gas into the atmosphere.

3. The process according to claim 1 wherein said filtering step comprises the step of vacuum filtering the waste containing liquid and including the steps of filtering the permanent gas removed during the vacuum filtering step, and exhausting the filtered permanent gas into the atmosphere.

4. The process according to claim 3 including the steps of removing permanent gas from the evacuable drying chamber during the freeze-drying step, filtering the permanent gas removed during the freeze drying step, and exhausting the filtered permanent gas into the atmosphere.

5. An apparatus for rendering radioactive waste material contained in a liquid into a more easily handled form comprising an evacuable drying chamber including evacuable inlet and outlet filter chambers, an inlet for introducing a liquid containing filterable radioactive waste products into said inlet filter chamber liquid, filter means disposed between said inlet and outlet filter chambers and adapted to filter the introduced liquid, an outlet for removing from said outlet filter chamber the filtrate produced by said filter means outlet chamber vacuum pumping means adapted to evacuate permanent gas from said outlet filter chamber, freezing means adapted to freeze the residue remaining on said filter means, heater means for producing sulimation of the frozen moisture contained in the frozen residue, and refrigerated condenser means connected to said inlet filter chamber and adapted to condense the vapor produced by sublimation of the frozen residue.

6. The apparatus according to claim 5 wherein said outlet chamber vacuum pumping means includes a filter device adapted to filter the permanent gas removed from said outlet filter chamber before exhaustion of the permanent gas into the atmosphere.

7. The apparatus according to claim 5 including inlet chamber vacuum pumping means adapted to evacuate permanent gas from said inlet filter chamber, and a filter device adapted to filter the permanent gas removed from said inlet filter chamber before exhaustion of the permanent gas into the atmosphere.

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