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PROCESS FOR THE CONCENTRATION OF ISOTOPES

Filed Jan. 27, 1945

2 Sheets-Sheet 1

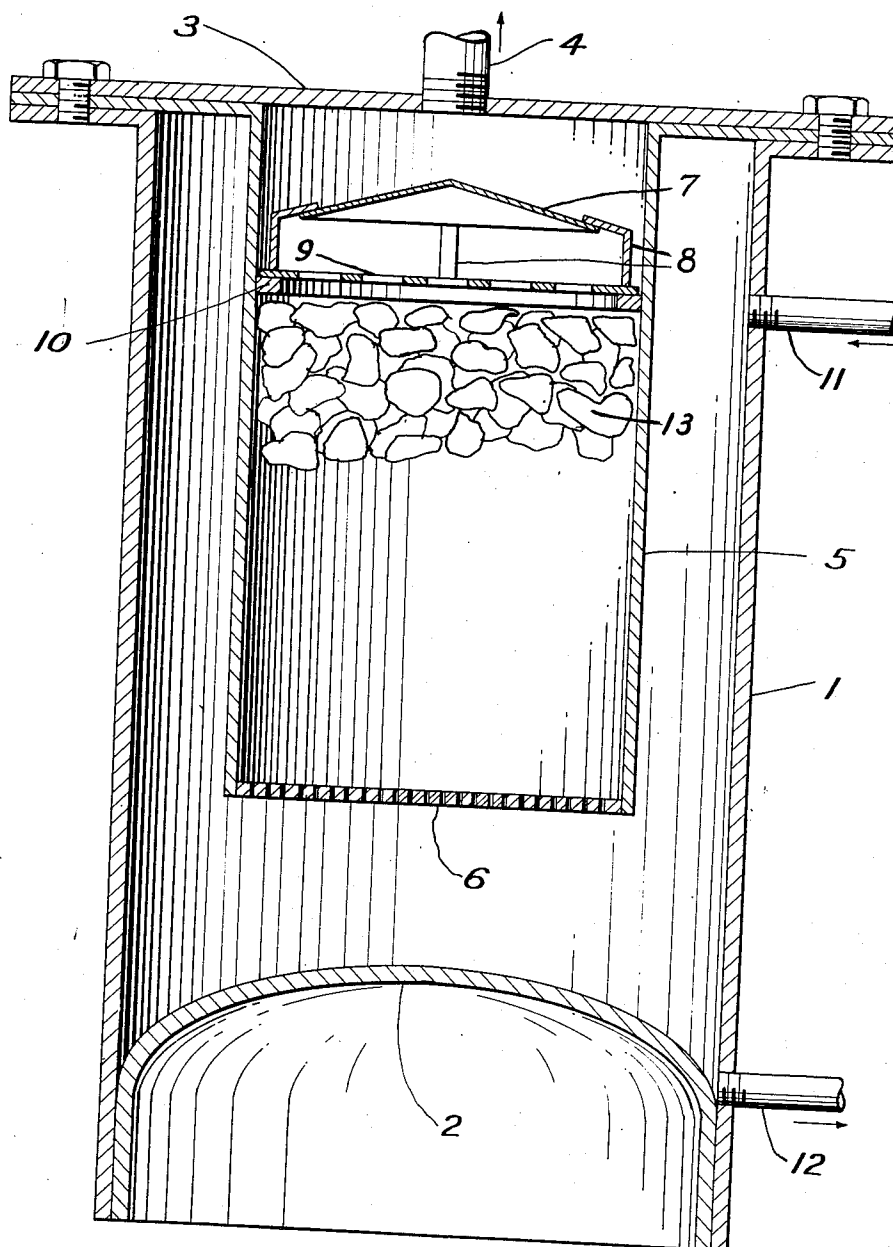


FIG. 1

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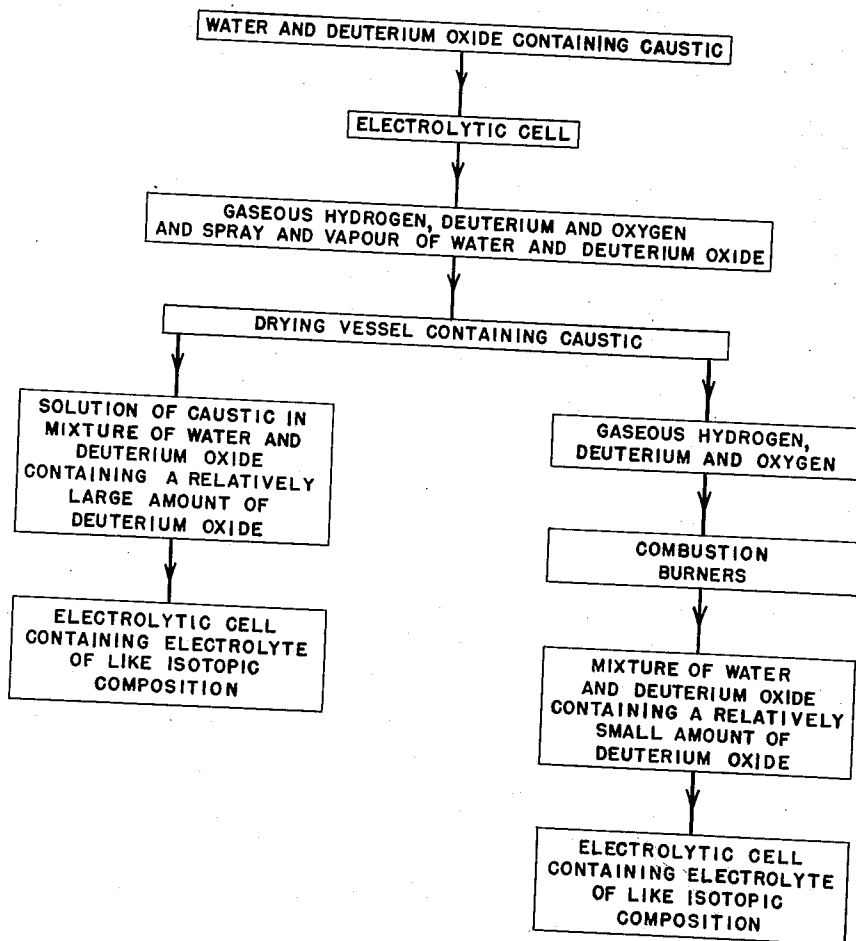


FIG. 2

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PROCESS FOR THE CONCENTRATION OF ISOTOPES

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1 Claim. (Cl. 204—101)

The present invention is an improved method of operating processes for the concentration of the heavier isotopes of hydrogen and oxygen where these gases carry aqueous spray and vapour. This improvement enables a higher recovery to be made of the spray and vapour, and also enables the recovered spray and vapour to be collected and returned to the process in a particularly advantageous and efficient manner, thereby securing increased recovery and economy in the operation of the process.

One of the processes used for concentrating the heavier isotopes of hydrogen and oxygen employs the method of fractionation of the isotopes by electrolysis of an aqueous alkali electrolyte, which relies on the greater tendency of the lighter, rather than the heavier, isotopes to be liberated in the form of gas from the electrolyte by electrolysis. This fractionation process may be carried out as a batch process in a series of successive stages, the electrolyte in each stage containing a higher concentration of the heavier isotopes than the electrolyte in the immediately preceding stage. In this manner, as electrolysis of a batch of electrolyte in any one stage is continued without replenishment with water, the residual electrolyte decreases in volume and there is a progressive concentration of the heavier isotopes in the residual electrolyte. The separation of the isotopic forms of each element in any one stage of fractionation by electrolysis is, however, only partially successful and some of the heavier isotopes in each case is liberated as gas. For this reason, in this type of process, it has been the practice to collect and burn these liberated gases to form water, which, because it contains some of the heavier isotopic material, can with advantage be returned to the fractionation process as feed water to an earlier stage.

In the operation of this type of process, the generated gases carry with them considerable quantities of spray and vapour, particularly the latter, having a concentration of each of the heavier isotopes approximately equivalent currently to their respective concentrations in the residual electrolyte as this becomes progressively enriched in these heavier isotopes. The heavier isotopes are present in lower concentrations in the generated gases than in the electrolyte from which the gases were evolved. Consequently, the water formed on combustion of the gases is fed to an earlier stage of the process in which the electrolyte undergoing treatment contains a lower concentration of each of the heavier isotopes than is contained in the later stage from which the gases were evolved. The spray and vapour carried by the gases accompany the water of recombination, and, as the concentration of the heavier isotopes in the spray and vapour corresponds to that of the electrolyte in the later stage, this concentration in the spray and vapour accordingly becomes diluted by the water of recombination and by the electrolyte of the earlier stage to which it is added. The heavier isotopes in the spray and vapour, on being fed to the earlier stage, and diluted, are again subjected to concentration by electrolytic fractionation, or they may even to some extent be carried off with the gases generated and thus fed to an even earlier stage of the process with further loss in efficiency in the process.

Spray traps of various kinds have been used with a view to recovering part of the spray in this type of process. Sand traps, which have been used to prevent flame from the gas burners from spreading back to the electrolytic cells, have served to some extent to collect spray, but it

has been found difficult to recover the spray collected by the sand.

I have discovered a method which enables a much greater recovery to be made than heretofore of the spray and of the vapour with their content of the heavier isotopes, carried with the gases from the electrolytic cells in this process. My improvement enables recovery to be made of a substantial proportion of the spray and vapour from the gases prior to combustion of the gases, and also enables the recovered spray and vapour with their content of the heavier isotopes to be returned in a suitable form to a stage having an approximately similar concentration of such isotopes, rather than to an earlier stage of the concentration process or to waste, thereby securing increased recovery of these isotopes and greater economy in the process.

I have found that the spray and vapour may be satisfactorily removed from the gases leaving the electrolytic cells by causing the gases containing the spray and vapour to pass through a vessel charged with small lumps of caustic potash or caustic soda, depending on whether potash or soda is used in the cell electrolyte. The caustic, that is to say, either caustic potash or caustic soda, in lump form, serves to remove spray and vapour from the gas stream. To treat the gas collected from a series of cells, one drying vessel, or preferably more drying vessels in series, may be used as desired. It is desirable to have the drying vessels filled with a greater quantity of caustic than is removed by solution, formed with the spray and vapour, during electrolysis of each batch of electrolyte.

The caustic solution collected in this manner in the drying vessels is run off into a storage tank and fed to cells preferably containing electrolyte of approximately similar isotopic concentration to that of the caustic solution, thus automatically serving to supply the alkali needed for conductivity purposes in the electrolyte. When a group of cells in a stage is operated according to the batch system, each batch of electrolyte may conveniently be made up in a storage tank for that stage by charging distilled water into the tank and adding thereto the caustic liquor obtained from the drying vessels of that stage during the electrolysis of the previous batch.

When the fractionation process is carried on as a batch operation and in accordance with my improved method of operating, the caustic solution obtained from the drying vessels may be collected in a series of separate storage vessels, each vessel serving to collect solution containing a different range of isotopic concentration. For example, during the latter part of the batch operation, the deuterium oxide concentration in the solution collected will be greater than that in the earlier part of the operation, and it may be desirable to collect these solutions separately and to feed the solution with the lower concentration to the stage of cells having a similarly low concentration and to feed the solution with the higher concentration to the stage of cells having a similarly high concentration.

The effectiveness of the improvement is well demonstrated by comparative tests which showed that during electrolytic decomposition of a batch of electrolyte when decomposing the electrolyte down to a given residual volume, the use of a drying vessel system, employing caustic potash as the drying agent, enabled 2.04 lbs. of water to be collected per cell compared with only 0.14 lb. of water collected when spray traps employing steel wool were used instead in the gas stream under similar conditions of operation.

It was found that a charge of about 120 lbs. of solid caustic potash in lumps of from 1" to 3" in size in a drying vessel would treat satisfactorily a volume of mixed hydrogen, deuterium and oxygen at 20° C., equivalent to a flow of 8.5 cu. ft. of gas at N. T. P. per minute, to remove about 75 lbs. of moisture and to yield about 150 lbs. of caustic solution containing about 50% caustic potash, in 164 hours. In this case, a quantity of deuterium oxide amounting to 5.8 lbs. was collected in the caustic solution, and if this quantity had been sent back to an earlier stage instead of having been collected and returned to the cells in the particular stage from which it was generated, this quantity would have required re-

3 treatment with resulting losses in product and in economy. After a run of this duration when operating under these conditions, the drying vessel is customarily replenished by adding a charge of about 75 lbs. of solid caustic potash prior to the next run.

A suitable design of enclosed drying vessel is shown in the accompanying drawing. The vessel, which may be of iron, comprises an outer gas-tight shell 1, the side walls of which may conveniently be cylindrical, fitted with a permanent bottom 2 and with a removable cover 10 lid 3 containing a centrally located outlet 4 for gas. There is supported inside the shell a cylindrical basket 5 of which the side wall is gas-tight, but the bottom is a perforated plate 6. The top of the basket 5 is open, but is so located within the shell 1 that its upper edge is flush with the flat cover lid 3 when this is clamped or bolted in place. A baffle 7, which may be of conical shape, is located near the top of the basket 5 and just below the centrally located outlet 4 in the cover lid 3. The baffle 7 is supported by legs 8 fastened to a plate 9 which, in turn, is supported by a ring 10 welded around the inside of the basket 5. Holes are provided in the plate 9 to permit passage of the gas. The side wall of the shell 1 is fitted with an inlet 11 for gas near the top, and with an outlet 12 for drainage solution near the base of the shell. The cover lid 3 is clamped or bolted in place and is readily removable for recharging the vessel with the drying agent. The baffle 7 together with the plate 9 can be easily lifted from the ring 10 to facilitate recharging of the vessel.

In operation, the gas with its burden of aqueous spray and vapour evolved from the electrolytic cells is led into the vessel through the inlet 11 and is passed down around the cylindrical basket 5, and then up through the perforated plate 6 on which is supported the charge of caustic lumps 13.

The gas, with its burden of spray and water vapour, passes up between, around, and in contact with the lumps of caustic, and, before leaving the vessel through the outlet 4 in the cover lid, the gas in a dried condition is deflected around the baffle 7 placed directly beneath the outlet 4.

The caustic solution, formed by dissolution of the lumps of caustic 13 in the spray and water vapour carried by the gas, is allowed to trickle down to the base of the vessel where it is collected to a suitable depth and run off as desired through the drainage outlet 12.

It is preferable to pass the gas up between the lumps of drying agent and countercurrently to the flow of aqueous solution of the drying agent which trickles off the lumps, so that the gas on leaving the drying vessel is in contact with relatively dry lumps of the drying agent. It may be desirable, however, to use two or more drying vessels in series, and in this case the gas may be passed down through the first drying vessel concurrently with the flow of aqueous solution, suitable provision being made in the design of the vessel to accommodate the flow of gas in this case.

Where countercurrent flow of gas and solution is employed in the drying vessel, it is preferable to employ in the drying vessel a basket designed to hold a column of the drying agent such that the dimensions of the column will permit the velocity of the gas in an upward direction therethrough to be slow enough to permit the droplets of solution to fall to the base of the vessel without being carried up and out of the top with the gas stream.

Where the hydrogen gas and the oxygen gas generated in the cells are kept separate in the cells and are evolved separately, each gas may be passed through a separate drying vessel and the resulting aqueous solutions of the drying agent may be returned to the appropriate stage as described herein.

My improved method offers the advantage that the aqueous spray and vapour are collected in a form suitable for feeding directly to the cells and no separation

of the collected water from the drying agent is required, as would be the case and with additional expense were silica gel, or phosphoric anhydride (P_2O_5), or other drying agents employed. In the practice of the present invention, when, as a result of the drying action the lump caustic is exhausted as regards effectiveness as a drying agent, the caustic solution resulting from the drying electrolytic process and serves to supply conducting agent needed for electrolysis of the water. Furthermore, as the caustic dissolves in the moisture removed from the gas mixture, the caustic solution drains off the lumps, leaving these lumps with a freshly exposed surface to dry further quantities of gas, whereas other types of drying agents would become progressively more saturated with water and accordingly less effective as drying agents.

This method is definitely superior to that employing spray traps, as such traps serve merely to remove some of the spray, but not the vapour, and there is a considerably greater weight of moisture carried as vapour than as spray under normal operating conditions in this type of process. When sand traps are used in the system, the tendency of these to become plugged with solid caustic from the spray from the caustic electrolyte when spray traps are used, is largely avoided by placing caustic drying vessels, instead of spray traps, ahead of the sand traps.

This improvement is of particular value where the electrolysis is conducted in cells operated by the batch method and where these cells are used in the final stages of the concentrating process, in which high concentrations of the heavier isotopes, for example, deuterium, are being treated.

I claim:

In a process for concentrating deuterium oxide in a caustic electrolyte consisting of caustic dissolved in feed water by electrolytic fractionation in successive stages of progressively higher deuterium content in which the major portion is of the electrolyte in each stage electrolytically decomposed to a gaseous mixture of hydrogen, deuterium and oxygen carrying water in liquid and vapor phase form containing deuterium oxide, until the deuterium content of the gaseous mixture from each stage approximates the deuterium content of the feed water to that stage, the improvement which comprises drying the gaseous mixture generated during electrolysis in each stage by contacting said gaseous mixture with solid caustic drying agent to form an aqueous solution of the drying agent and adding said solution to a stage wherein the electrolyte contains a concentration of deuterium oxide approximately equivalent to that of the solution.

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