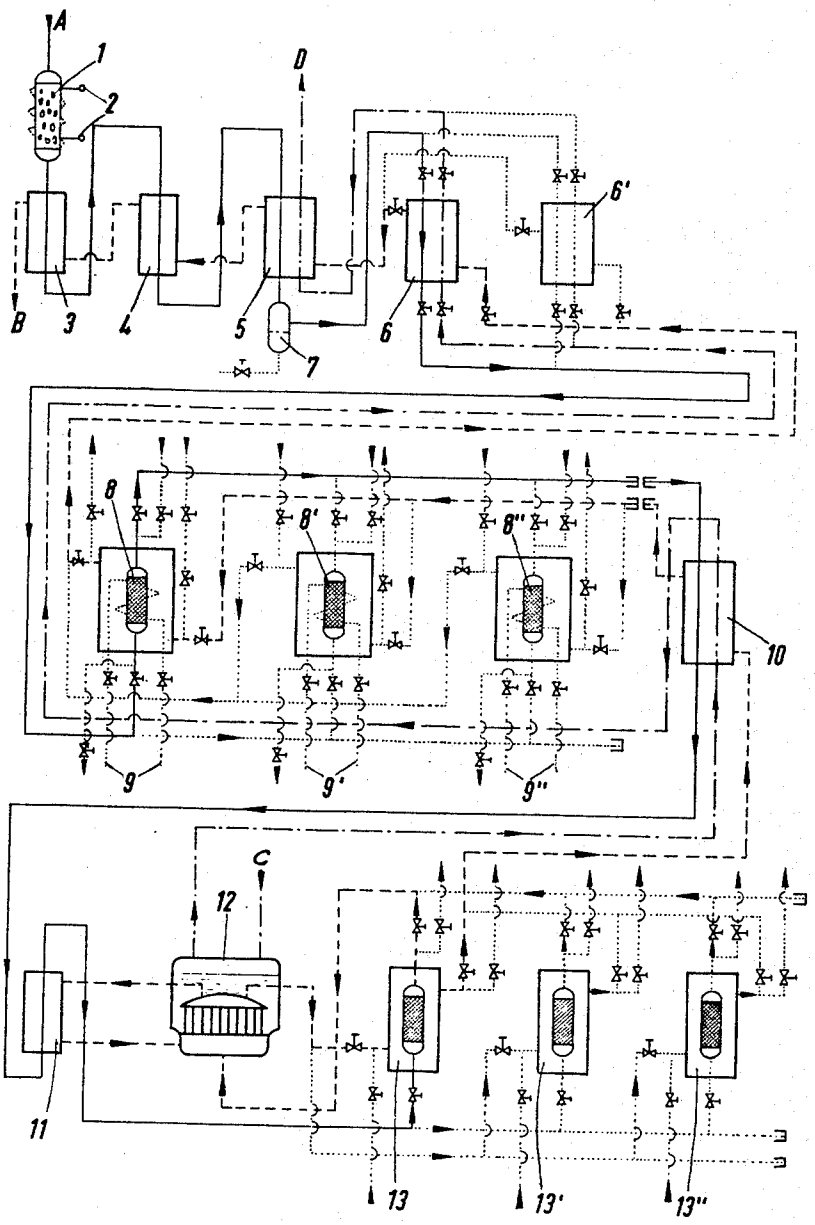
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PROCESS FOR REMOVING RADIOACTIVE IMPURITIES FROM CASES

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PROCESS FOR REMOVING RADIOACTIVE IMPURITIES FROM GASES

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This invention relates to the removal of radioactive components, especially gaseous radioactive components, from gaseous mixtures which may be circulating, such as found for instance in the form of cooling and/or protective gases in atomic piles or other enclosed spaces badly contaminated with radioactivity.

Methods are known by which such impurities can be removed, for instance by the use of adsorbers or other conventional purification processes, especially washing processes and the like. It has also been suggested that such a purification process be carried out in several stages, in the same way that other purifications of gases and liquids with respect to non-radioactive substances are carried out in several stages.

In the case of radioactive impurities, however, which is dealt with in this invention, special conditions prevail. For here a new problem arises, which is that when the purification process is finished the substances used for purification—be it washing substances, adsorption or absorption agents or even purely mechanical or electrical filters—after having taken up the radioactive substances are themselves polluted with radioactivity and must, therefore, in their turn, be rendered harmless to their environment by some means.

The invention relates especially to a type of purification by use of an adsorption process, for instance by means of an adsorptive gel, active charcoal, microfilters and/or the like. On the other hand, the scope of the invention extends beyond this particular embodiment.

The principal object of the invention is to insure that during execution of the purification process the purification agents which have become radioactive can subsequently be rendered harmless as simply and thoroughly as possible.

According to the invention, this object is achieved by carrying out the purification by stages in such a manner that at least the major part—and preferably all—of the radioactive components, and most conveniently only the radioactive components, of the gas mixture, accumulate in a single purification stage and thus at the end in a single purifying agent, in particular an adsorption agent, whereas the other non-radioactive impurities or components to be eliminated for other reasons are retained in one or more other purification stages in other purifying agents.

The advantage of this solution of the problem is that the radioactive impurities are collected into the smallest possible space, because an adsorber need only be relatively small if it is to adsorb the radioactive—and not, also, the non-radioactive—impurities or components. Such small structural units can be more easily decontaminated than can larger ones. It has been suggested, for instance, that adsorbers charged with radioactive substances should be left inside the atomic pile unit—which latter is of course shielded from the outside—until the radioactive substances shall have broken down into harmless or less harmful decomposition products, after which the adsorbers can then either be regenerated or their filling material can be taken out and removed. It is also easier to handle a small contaminated structural element than a larger one; thus, in the case of larger structural units, their transportation out of the zone of operation would be made considerably more difficult by their larger dimensions.

The process according to the invention may conveniently be carried out by oxidizing the oxidizable components of the initially-used gas, for instance, hydrogen and carbon monoxide, to substances which are relatively easily congealed by cooling.

According to a special feature of the invention, the system for carrying out the process comprises a final purification stage in the form of an adsorber cooled to —70° C., which final adsorber is preceded by at least one pre-dryer, especially a gel-adsorber, cooled to a temperature between 0° C. and —70° C. The pre-dryer may with advantage be preceded by a stage operating above 0° C., for instance at room temperature, for the elimination of components, especially organic components, which can be condensed at higher temperatures. This stage can in turn be preceded by a high-temperature stage in which the substances which are difficult to remove otherwise—for instance, the aforesaid substances, hydrogen and carbon monoxide—are converted into water and carbon dioxide, for instance by the use of a catalyst or by some other chemical process.

It is, however, also within the scope of the invention that yet other transformations should follow, and that altogether different stages and types of purification be chosen, in each case with the object of securing the elimination of the radioactive substance or substances as completely as possible and the segregation of the radioactive substance in one stage and in one purifying agent. In principle this could even be achieved in a first step, which possibility has the advantage that the radioactive substances would not have to pass through, and hence, in some way contaminate, any of the further purification stages. From the investigations made up to now, however, it appears more expedient to provide for the elimination of the radioactive substances in the last stage, because the radioactive substances which accumulate in atomic piles, especially when the fuel elements of the same are not shielded, are often less easily removed than are the non-radioactive substances and hence only their removal in the last stage can insure, at any rate up to a certain point, that yet other, non-radioactive substances are not removed at the same time, which circumstance would, as already mentioned, involve an increase in the necessary adsorption material or the like.

Another feature of the invention is that—all regeneration within the purification system being avoided—the cooling gas circuit is equipped with separation means, in particular adsorbers, which retain at least the harmful components and are preferably all of the same type, which can be switched into the circuit successively and separately and are of such a size and in such number that the sum of the working-times of all the separation means, in particular adsorbers, is of about the same order of magnitude as is the lift of the atomic pile per se.

Thus the inventive concept consists in providing within the atomic pile unit a series of separation means, in particular adsorbers, none of which is regenerated, but which are switched off successively as soon as they are full, and then remain in the pile unit until the radioactivity they had assumed has diminished to a tolerable or acceptable degree so that they can be dismantled and taken out of the atomic pile unit in order either to be destroyed or to be overhauled by removal of the adsorptive agent or by means of a regeneration process. On the other hand, after this delay, regeneration inside the pile may also be considered, for which the now less dangerous, or harmless, components may be withdrawn from the pile through the usual type of conduit.

In the accompanying drawing, a diagrammatic sketch of the purification system according to the invention is given as an illustrative example.

The cooling gas mixture, consisting mainly of helium and neon and containing radioactive impurities, enters the purification system, according to the invention, at A after having been diverted, on leaving the interior of the pile, from the stream of cooling gas flowing through the interior of said pile. The portion of the total stream of cooling gas which is diverted from the interior of the pile through the purification system represents from about 0.5% to about 5% of said total stream of cooling gas. The diverted cooling gas mixture, especially helium-neon mixture, freed from the impurities, leaves the purification system at B, to be returned to the cooling gas circuit. The gas mixture to be purified is shown by a solid line; the purified mixture is shown by the broken line, while a cold air stream required for the purification process is shown by a dash-and-dot line. The latter enters the purification system at C in the form of liquid air and leaves it again at D, according to the invention, in the form of gaseous air generally at a temperature above 0° C.

The whole system consists of three successive, connected parts, each operating in a different temperature range: a high-temperature part, a medium-temperature or pre-cooling part and a low-temperature part, in which the actual separation, e.g., adsorption, of the harmful active components takes place.

1 represents a catalyst unit in which any hydrogen contained in the gas mixture to be purified is oxidized to H_2O and any carbon monoxide contained therein is oxidized to CO_2. 2 represents a heating conduit for heating the catalyst 1 to a suffifficiently high temperature. In heat exchangers 3, 4 and 5 the gas to be purified is cooled by the purified gas flowing in the opposite direction, after which the gas to be purified is further cooled by means of cold air in the heat exchangers 4 and 5. Between the heat exchangers 5 and 6 is a separator 7 for easily condensed substances, especially water.

8, 8′ and 8″ represent three gel dryers for use in succession, mainly for the separation of water vapor and carbonic acid. The temperature of approximately −60° C. required for this operation is produced by means of a refrigerant of the same type as that designated by the commercial name "Freon," which refrigerant passes through conduits 9, 9′ and 9″, respectively, shown as dotted lines, in such a way that through the chosen supply line for the refrigerant there is no possibility that the refrigerant will become mixed with the gas to be purified, even in the event of a leak.

Further refrigeration takes place in heat exchangers 10 and 11. The necessary cold therefor is produced by an evaporator 12 in which liquid air, which enters at C, is stored and simultaneously the purified gas flowing back is cooled.

13, 13′ and 13″ represent adsorbers filled with activated charcoal, which carry out the actual purification of the helium-neon mixture with respect to the active components, the adsorbers 13, 13′ and 13″ being placed in use, one after the other—in the same way as are the dryers 9, 9′ and 9″—one being used until it has become saturated with impurities. Each adsorber is designed to operate for about 5,000 hours. In view of the very small concentrations met with in this case, this working life can be obtained with small-size adsorbers especially if appropriate non-active substances are added to the active material to be eliminated in particular crypton and xenon, in the manner already suggested. When its working time has run out and the adsorber is fully charged with impurities, the major part of the helium-neon mixture is pumped out of it so that the adsorber can be warmed and can then remain in the storage chamber of the pile

at any desired temperature. The working life of the adsorbers 13 and the gel-dryers 9 as well as the liquid air unit required for the liquid air supply may expediently be adjusted in relation to one another so that the replacement of dryers and adsorbers and the thawing of the liquid air unit can always be carried out simultaneously.

The transfer of cold is effected by indirect heat-exchange in such manner that during heat-exchange between the purified and the as yet unpurified gas, the purified gas acts at the same time as protective gas for the unpurified gas. The purified gas is also used as safety gas for the valves and conduits, in each case being made to flow through a jacket surrounding the stream of unpurified gas.

In the purification system shown in the diagram, the arrangement is such that the purification from traces of water takes place—after the countercurrent coolers 6 and 6′—in the gel-dryers 8, so that no water reaches the adsorbers 13. On the other hand, the radioactive inert gases to be eliminated by them are practically entirely retained in the charcoal adsorbers 13. In the case of the embodiment of the invention shown as an example some CO_2 does enter the charcoal adsorber 13; however, the quantity of charcoal which must be provided for the CO_2 is small in comparison with the quantity—incidentally, also not a very large amount—required for the adsorption of the inert gases.

We claim:

1. Process for purifying a gas mixture containing radioactive impurities and hydrogen and carbon monoxide as non-radioactive impurities, which comprises oxidizing only the non-radioactive impurities of said mixture with the aid of a catalyst mass to convert the hydrogen and carbon dioxide to water and carbon dioxide, separating the water and carbon dioxide from the gas mixture by condensation and adsorption at a temperature within the range 0° C. to −70° C., and separating the radioactive impurities from the gas mixture by adsorption in an adsorption agent at a temperature below −70° C.

2. Process defined in claim 1, in which the adsorption agent is contained in a plurality of adsorbers which are each successively switched into the gas purifying circuit until the end of their working time and the used-up adsorbers at the end of the periods required for the removal of the radioactive impurities with which they are saturated are removed from the circuit and replaced by active adsorbers.

3. Process for purifying a gas mixture circulating in an atomic pile, said gas mixture containing radioactive impurities and hydrogen and carbon monoxide as non-radioactive impurities, which comprises continuously withdrawing from the atomic pile a stream of the impure gas mixture, oxidizing only the non-radioactive impurities of the withdrawn gas mixture with the aid of a catalyst mass to convert the hydrogen and carbon monoxide to water and carbon dioxide, separating the water and carbon dioxide from the gas mixture by condensation and adsorption at a temperature within the range 0° C. to −70° C., separating the radioactive impurities from the gas mixture by adsorption in an adsorption agent at a temperature below −70° C., and leading the resulting purified gas mixture stream back to the atomic pile to circulate therein.

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