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METHOD FOR MITIGATING RADIOACTIVE
CONTAMINATIONJack De Ment, Portland, Oreg., assignor to the
United States of America as represented by the
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3 Claims. (Cl. 134—28)

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This invention relates to colloidochemical methods for the mitigation and lessening of radioactive contamination, applicable from a site removed from said contamination.

It is an object of this invention to provide methods of versatility which have broad applicability to a variety of radioactively contaminated objects and the problems presented associated therewith, whereby military, naval, civilian or other installations may be treated subsequent to radioactive contamination to reduce same.

It is among the objects of this invention to make available methods whereby areas and targets possessing very great and dangerous radioactive contamination levels and degrees may be treated for the mitigation of same at distances which insure safety for personnel engaging in decontamination operations.

In the employment of fissionable metal explosive and/or radiochemical warfare agents and weapons, the target is characterized as being treated and infected by a plurality of toxic radioactive substances, frequently solid particles dispersed in the atmosphere in aerosolic form. Likewise, the target, especially the ground and structures thereupon, but also ships and other structures floating upon water say in harbor or at dock, becomes contaminated with injurious and baneful radioactive matter.

It is therefore a problem of the greatest magnitude to effectively and safely mitigate the spread of such toxic matter, to limit its dispersal to adjacent and nearby areas and targets, and, in general, to remove as far as possible, i. e., mitigate, decontaminate or reduce in radioactivity, these primary products, after-products, by-products and debris and the like inherent in the operation of such warfare weapons. Briefly, the present invention is directed to cooperative mechanical and colloidochemical methods and means, hereinafter set forth in considerable detail, whereby a broadly new class of decontaminative agents, also fully disclosed infra, may be employed to effect decontamination via the agencies of physicochemical and mechanical reactions and processes which involve among others and usually concurrently, chemical reaction, with the formation of a plurality of water-insoluble radioactive compounds, e. g., silicic acid derivatives, heavy metal and other sulfides, but particularly and coincidentally with the aforementioned processes those of adsorption and absorption. That is, the present invention is directed to methods for carrying contaminants such as vapors or gases which may otherwise be reactive or responsive

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and which are present in the atmosphere and upon structures and earth therebeneath by adsorption, absorption and other physical and mechanical processes and means, e. g., dispersion.

Herein, the terms and expressions "decontaminative agent" and the like will be taken to mean all substances which conform to the broad characteristic of "colloidochemical," as do examples of substances and mixtures hereinafter set forth.

The term "colloidochemical" will be taken to mean those substances which upon release into the atmosphere, by virtue of moisture therein, or in the presence of water, react to form adsorptive colloids of an insoluble nature and which substances concurrently exhibit chemical reactivity with a plurality of elements so as to form insoluble derivatives thereof, and/or derivatives which are characterized as becoming adsorbed and absorbed or carried into the colloids coincidentally formed.

Broadly, decontaminative agents or those substances which upon contact with moist atmosphere or in the presence of water exhibit the aforementioned colloidochemical behavior and properties, include, as examples, hydrolysable compounds of silicon, particularly the silicon halides, certain hydrolysable and unstable compounds from the sulfur-bearing chemical compounds, especially certain sulfur halides, and, certain other compounds which may contain a cation which forms an insoluble oxide, hydroxide or hydrous oxide, such as aluminum sulfide, along with colloidal sulfur.

Certain colloids possess a pronounced selective adsorption for many radioactive substances, including solids and gases from elements represented in the periodic system in the so-called alkaline earths (e. g., radium) on up through the heavier metals, and including uranium, and the transuranium elements as well. Silicic acids and sulfur are among those colloids with the most effective action in taking up radioactive substances from solution in liquids and also from dispersion in gases, such as the atmosphere. It is a characteristic of such adsorption processes that the taking up of radioactive substances continues in effectiveness with repetitious application or availability of the colloid. The adsorption and absorption processes function irrespective of dilution of radioactive materials, and can be applied to the most dilute as well as the most concentrated.

As has been disclosed in my co-pending application, Serial Number 738,916, certain reactive silicon compounds, of a hydrolysable or sol-

uble nature and characteristic, are relied upon for the lessening of radioactive contamination, by virtue of the fact that upon hydrolysis complex silicic acids are formed, these having very great powers of absorption and adsorption.

In the present invention, these substances and others which are described more fully herein-after, are transported to the site of contamination, say a naval vessel or a building (which may be of a military nature) and released in suitable form at the said site, thereby to exert their mitigating characteristics.

As an example of colloidochemical materials, it is preferred to employ a silicon halide, say of typical formula SiX_4 (where X is a halogen), to treat the radioactively polluted object or area. The silicon halides are prepared by the direct combination of silicon and the halogens, though the combination is less readily accomplished as the atomic weight of the halogen increases. It is emphasized that not all of the silicon halides of general formula SiX_4 may be useful, though this class of derivatives is herein disclosed as an example of a reactive silicon compound which may be obtained in the gaseous or vaporous state with little difficulty. Hence, silicon tetrafluoride may not be desirable for employment, in view of the fact that it hydrolyses in water and moist atmosphere to give hydrofluoric acid and other complex compounds, e. g., fluosilicates.

I prefer the utilization of silicon tetrachloride, SiCl_4 , which can be made by passing chlorine gas over a mixture of silicon dioxide and carbon, or, by the action of sodium chloride upon silicon carbide (commonly known as "Carborundum"), in which instances the silicon tetrachloride can be manufactured on a large scale at relatively low cost. Silicon tetrachloride is a colorless fuming liquid, having a formula weight of 169.89. Silicon tetrachloride hydrolyses completely and easily, and when it comes into contact with moist air it fumes strongly, producing a dense smoke-cloud which consists of droplets of hydrochloric acid, HCl , in solution, and, small particles of silicic acid, H_4SiO_4 (or silicon hydroxide, $\text{Si}(\text{OH})_4$).

Yet another example of a related compound, which may be stored in a solid or liquid state, and released in an aerosolic state upon employment, is silicon trichloride, Si_2Cl_6 , occurring as leaflets or liquid, and having a formula weight of 268.86. Both of the aforementioned silicon halides act similarly upon hydrolysis in the presence of water or atmospheric moisture.

In addition to silicon halides, such as the SiX_4 compounds disclosed above, as well as mixed silicon derivatives, including silicon oxyhalides of the general formula which denotes the series of oxyhalides from Si_2OX_6 through $\text{Si}_7\text{O}_6\text{X}_{16}$, or represented by the general formula $\text{Si}_n\text{O}_{n-1}\text{X}_{2n+2}$, the sulfides of silicon can be used. As to these compounds it is preferred to employ silicon disulfide, SiS_2 . In general, the chemical behavior of silicon oxyhalides is very much like the silicon halides, in that in the presence of moisture or water there is decomposition and hydrolysis, the ease of hydrolysis increasing with decreasing molecular weight, with the exception of the cyclic silicon oxyhalides which contain eight halogen and four oxygen and silicon atoms in the molecule; other than these compounds which are solid crystalline materials, all are colorless, oily liquids with high boiling points, and are miscible with most organic solvents, including carbon disulfide, carbon tetrachloride and chloro-

form. As to silicon disulfide, which substance is to be recommended, since upon hydrolysis it forms not only adsorptive silicic acids but also colloidal sulfur, the compound occurs as white needles, decomposing in water and ethanol.

Other colloidochemical or decontaminative substances, which are in general to be less preferred than the aforementioned silicon compounds, include certain sulfur derivatives: thus, sulfur monochloride, SCl_2 , proves suitable in that it is an oily liquid which is decomposed by water with the separation of colloidal sulfur, and which is soluble in alcohol, benzene, ether, carbon disulfide, carbon tetrachloride and oils. Also, elemental sulfur is readily dissolved by sulfur monochloride, and, therefore, instead of the sulfur monochloride singly, a saturated sulfur (solute) solution of sulfur monochloride (solvent) is also employable. Yet another sulfur compound is operable, but also less preferred, and this is aluminum sulfide, Al_2S_3 , which is prepared by heating aluminum and sulfur, and which exists as yellowish solid, this being easily decomposed in moist air and in water with the formation of a variety of adsorptive products, including hydrated aluminas and sulfur.

Whereas in the preceding disclosure a number of preferred compounds are set forth as of utility, it is emphasized that the choice is by no means limited to these special and specific substances, and that therefrom other choices of perhaps more suitable substances to be found by others later on can be made, and yet not violate the broad novelty, spirit nor scope of this invention.

As to form of the agent or chemical substance or composition generally liquids or gases under pressure will be preferred, with solids being less preferred.

The decontaminating agent may be applied in any convenient manner taking into consideration the activity of the surface or object which is to be decontaminated. In the event of light radioactivity, simply sprinkling by hand may be in order. However, where objects are dangerously radioactive, other means must be employed to apply the decontaminating agent which will allow personnel sufficient distance from the object so as not to be exposed to radioactivity. This may be done by spraying from aircraft, using a bomb dispersal method, and the like. Other methods should be immediately obvious.

Depending upon the nature, circumstance and characteristics of the decontamination problem, water may or may not be required for subsequent application to expedite colloidochemical action, e. g., hydrolysis of a silicon compound or other decontaminative agent. Many factors will often have to be considered, including climatic factors, though it is pointed out that in most cases of utilization of this invention the natural moisture content of the atmosphere will serve satisfactorily and that an extra supply of water will be unnecessary. For large quantities, e. g., thousands of pounds of decontaminative agent spread over relatively large targets, varying periods (e. g., up to several days) may be required before complete equilibrium will have been attained, e. g., a level reached at which the decontaminative agent has exerted its concurrent chemical and colloidal (sorptive) properties upon the contaminative radioactive matter such that the decontaminative matter, often in the form of pasty or slushy material, containing the

radioactive matter, can be removed should removal be desired.

As to means for the removal of radioactive contaminant entrapped and carried in decontaminative matter, a variety of methods and means can be employed, and the choice will vary according to the type of decontamination undertaking, target, degree of contamination, and other factors, but especially those concerned with the health physics problems which are now well known to those skilled in the art.

Thus, in the case of structures such as ships and buildings which have been equipped with the invention disclosed in my pending application entitled "Method and Means for Protection of Ships and Buildings from Radioactive Weapons," Serial Number 748,632, filed May 16, 1947, that invention may be put into operation and the umbrella of water droplets or rain which is thereby produced or formed and which strikes contaminated and decontaminated surfaces is relied upon to wash away contaminant carried by decontaminant. In the inside of certain structures the usual fire sprinkler system may be put into operation, provided same is intact and operable (which will not usually be the case), so as to effect washing away of said matters.

If the climate becomes so inclined, then natural rainfall may be relied upon to wash away decontaminant carrying contaminant. Otherwise, in large scale decontamination operations, the methods and means disclosed in my application entitled "Method and Means for Reducing Atmospheric Radioactivity," Serial Number 776,864, filed September 29, 1947 (now abandoned), wherein so-called "man-made rain" is produced by application of seeding means to precipitate cloud systems, will function to provide rainfall in many instances, this serving to wash.

In cases of lesser degrees of radioactive contamination, the reliance upon flexible asbestos or rubber water-carrying hoses, or upon water-carrying pipes or other conduits, will serve to bring water into the area wherein this invention has been reduced to operation. Also, in

instances, say in the case of ships, where radioactive contamination is very mild or has been thoroughly reduced to low activities, personnel may then enter and work with brushes and hoses and the like for periods commensurate with tolerance dosage.

I claim:

1. The method of mitigating radioactive contamination of an object which comprises the steps of applying a hydrolyzable silicon compound selected from the group consisting of silicon tetrachloride and silicon oxychloride to said object, exposing said object to moisture whereby said compound is hydrolyzed and removing said hydrolyzed compound and the radioactive contamination adsorbed therein.

2. The method of claim 1 wherein the hydrolyzable silicon compound is silicon tetrachloride.

3. The method of claim 1 wherein the hydrolyzable silicon compound is silicon oxychloride.

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