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EUROPEAN ATOMIC ENERGY COMMUNITY - EURATOM

COMPARATIVE LITERATURE STUDY ON THE
PROBLEMS OF THE DECONTAMINATION OF
RADIOACTIVELY CONTAMINATED TEXTILES

by

F. REIFF, K. SCHUSTER, H. SPOOR
(Joh. A. BENCKISER GmbH)

and

M. STOEPLER
(Isotopen-Studiengesellschaft, Karlsruhe)

1967



Report prepared by the Joh. A. BENCKISER GmbH
Ludwigshafen/Rhein - Germany

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SUMMARY

At the request of the Euratom Commission's Directorate-General for Health and Safety, the firm of Joh. A. Benckiser at Ludwigshafen/Rhein, Germany, in collaboration with the Isotopen-Studiengesellschaft at Karlsruhe, carried out a study on the experience and results so far obtained, as described in the literature of textile decontamination. In this study, account was also taken of the possibilities offered by the techniques now available, with a view towards the practical applications and the economics thereof.

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FOREWORD

The decontamination of radioactively contaminated textiles is not only routine practice in large nuclear establishments but is also an important subject of research. The increasing exploitation of atomic energy through the construction of nuclear installations and through the considerable expansion of its smaller-scale applications in laboratories, factories, hospitals, etc., means that, despite all precautions, there is a bigger risk of the radioactive contamination of materials. The decontamination of materials — and especially of textiles, from which protective clothing such as laboratory smocks, among other things, are made — can give rise to a number of specific problems that cannot always be adequately solved by “conventional” or customary cleaning processes. This is due on the one hand, to the wide variety of radionuclides employed and the different chemical forms in which they occur, and on the other hand to the nature of the various cloths and fabrics worn. For also in the field of synthetic fabrics technological progress has resulted in a vast diversification of types and compositions. Consequently, the interactions between all these radionuclides and textile materials can differ greatly, and this in turn can impose special requirements as regards the means of decontamination adopted.

At the request of the Euratom Commission's Directorate-General for Health and Safety, the firm of Joh. A. Benckiser at Ludwigshafen/Rhein, Germany, in collaboration with the Isotopen-Studiengesellschaft at Karlsruhe, carried out a study on the experience and results so far obtained, as described in the literature of textile decontamination. In this extensive study account was also taken of the possibilities offered by the techniques now available, with a view to practical application and to the economic aspects thereof.

In view of the great interest that exists in this problem, and having regard to the manner in which the investigation was conducted and the nature of the information elicited, it was felt that the present publication would bring this report to the attention of a wider readership.

Dr. P. RECHT

COMPARATIVE LITERATURE STUDY ON THE PROBLEMS OF THE DECONTAMINATION OF RADIOACTIVELY CONTAMINATED TEXTILES (*)

I — Description of the known processes for the decontamination of various (natural or synthetic) textile fibres, as well as simple and complex textile tissues which have been contaminated by radionuclides or their mixtures

1 — INTRODUCTION

1.1 General Ideas

In nuclear reactors, hot cells, isotope laboratories, or wherever one has to reckon with the undesired appearance of radioactive substances, there is always a possibility of contamination. According to the type of contamination, different problems result for the cleaning of equipment, surfaces and clothing. The processes for the decontamination of textiles which will be discussed here have been for a long time not only routine work, but also the goal of new research work in the USA and in other countries carrying on nuclear technical and nuclear development work.

1.2 Soilage-Contamination

1.2.1. *Normal Soilage*

The normal soilage of clothing results from the storage of dirt particles on the tissue, and from the accumulation of dissolved dirt on the fibres themselves. The removal of these various dirt particles makes different demands on the washing agents: The stored dirt particles are removed from the fibre by emulsifying and dispersing detergents. The accumulation of dirt on the woven fabric depends on the type and surface of the fibres utilized. Also, the degree of twist in fibres influences the accumulation of dirt: In the case of strongly twisted yarns dirt storage occurs practically only on the surface. The penetration of dirt particles into native and regenerated cellulose fibres is considered to be impossible [1].

In the case of liquid soilage such as oil, grease, organic solvents and dirty aqueous solutions, a penetration into the deepest fabric state, even into the fibre itself, is possible. Then, because of the swelling of the fibre, the more deeply located particles adhere very strongly. In the case of synthetical fibres this type of soilage is particularly difficult to remove causing a so-called graying.

(*) Manuscript received on January 16, 1967.

1.2.2 Contamination

Both types of soilage occur when protective clothing is utilized in areas where there is a danger of radioactive contamination. Furthermore, the clothes may be contaminated by dusty and liquid radioactive substances. Contamination and normal soilage can, thereby, be naturally independent from each other. Normal dirt can, however, markedly influence the adsorption of the radionuclides by the fibres.

Basically, the decontamination of textile fibres corresponds to a normal laundering operation. However, while evaluating such processes one must consider that the extraordinarily small quantities of radioactive substances and the differences of the chemical behaviour of the individual radionuclides often complicate the decontaminating process, and therefore require special procedures.

From the large number of radionuclides and their form of appearance on the one hand, and due to the necessity to treat various types of materials at the same time on the other hand, a great number of chemical and physical problems result. Therefore, the radionuclides in question and their most important properties relevant to this study should be considered first. Afterwards, a survey will be given about the material to be decontaminated, the mechanisms of adsorption, and the most important decontaminating agents, whereby these last will be considered in detail in part II. Finally, a series of decontamination experiments will be compared and discussed.

2 — THE MOST IMPORTANT CONTAMINATING RADIONUCLIDES

2.1 Fission Products

Estimated values for the yield of some long-living fission products resulting from nuclear explosions [2, 3].

Number of Nucleons	Relative Frequency %	Half-life Period	Number of Nucleons	Relative Frequency %	Half-life Period
89 Sr	2.56	53.6 d	129 Sb	1.26	4.3 h
90 Sr	3.50	28 a	131 J	2.89	8.05 d
91 Y	3.76	9.7 h	132 Te	4.24	78 h
95 Zr	5.07	65 d	135 Cs	5.41	2.106 a
97 Zr	5.17	17 h	136 Cs	0.036	12.9 d
99 Mo	5.68	67 h	137 Cs	5.57	29.8 a
103 Ru	5.20	40 d	140 Ba	5.18	12.8 d
106 Ru	2.44	1.0 a	141 Ce	4.58	32.5 d
111 Ag	0.33	7.5 d	143 Pr	4.68	33.4 h
112 Ag	0.240	3.2 h	144 Ce	4.69	277 d
115 Cd	0.227	43 d	147 Nd	2.68	2.67 a
125 Sb	0.290	2.78 a	153 Sm	0.31	47 h
127 Sb	0.540	3.9 d	156 Eu	0.0628	4.7 d
			161 Tb	0.0029	6.7 d

If one does not consider the relatively short-living nucleides, then, in nuclear explosions and in nuclear reactors where essentially the same nucleides are formed, one has to reckon particularly with the radionucleides of the elements Strontium, Yttrium, Zirconium, Ruthenium, Cadmium, Antimony, Iodine, Tellurium, Caesium, Barium, Cerium and Neodymium.

Due to extensive studies which have been previously made, the radiochemical properties of fission products are in part already well known [4-13]. Thus, while elaborating and evaluating the decontaminating processes, one can sometimes make unequivocal predictions in spite of the complex character of the reactions that take place.

2.2 Fissionable Radioactive Material, Activated Products

During the production and the chemical working of reactor fuels one has — in addition to the above mentioned fission products — to work in particular with natural and concentrated Uranium, Thorium, Plutonium and a lot of long-living activated products. Among these last the activated corrosion products (crud) are especially worth mentioning. They are activated in the fission zone of fluid-cooled reactors and then precipitated [14]. The activity is mainly caused by the oxides of the following nucleides :

Co-60	Half-life period	4.95	a
Co-58	Half-life period	72	d
Fe-59	Half-life period	47	d
Mn-54	Half-life period	310	d
Cr-51	Half-life period	27.7	d

When using Zirkonium or Zirkonium alloys as basic material for the covers of the fuel elements, one also has to reckon with

Zr-95	Half-life period	65	d
Hf-181	Half-life period	46	d
W-185	Half-life period	140	d

whereby W-185 is due to welding inclusions. For these radionucleides there are also numerous tracer tests available [15-23].

2.3 Special Fissionable Products and Radionucleides

In the laundering experiments mentioned below, above all, Strontium 89/90, Ce-141, Cs-137, Te-132, and Iodine-131 were tested. Besides, investigations with Ra-226, S-35, P-32/33, Fe-59, and Ca-45 were also carried out. This was done in part because of the importance of these fission products, and in part because of the interesting properties of the other radionucleides which have found a considerable expanse in research and technique [24, 25].

2.4 Chemistry of the Contaminating Radionucleides

The contaminating agent can, occur primarily, as dust or as solution, but also as fallout (nuclear explosions). Thereafter it can also be converted while still on the fibre : the solutions dry up and the activity will then be carried on as dust. Analogously, radioactive fallouts penetrate the earth and are then adsorbed by the silicates which are found therein and carried on as dust, or the dusty fallout activity will be carried on by rain. Because of the effect of rain, the activity which is first adsorbed as dust to the tissue surface can be better adsorbed to the fibre. Therewith, one must consider a number of possibilities. Finally, because of the laundering process itself, there results a solution which in an unfavourable case, can penetrate the fibres again. Hence

the washing and decontaminating agents applied in laundering experiments should certainly prevent such a recontamination. This can be avoided only when the mixture of the contaminating radionuclides is known, since there is no universal decontaminating agent. One must combine the decontaminating agents and the process in a manner that permits an optimal effect to be achieved for the particular case.

When evaluating the laundering processes and their application ranges it is indispensable to point out the most striking properties of the radionuclides which are predominantly in question.

Because textile fibres, in addition to their adsorptive properties, also appear to have an ion exchanging ability, the experiences which have been gathered in recent years by the processing of fallout samples in ion exchangers should be included into the survey [26, 27, 28]. One must assume that primary contamination and secondary reactions may occur in a neutral and weak-acid range, whereas in the case of high acid concentrations the fabric would be destroyed. In the alkaline range hydroxides are formed in the washing liquor. Their precipitation limits can be seen in previous papers [29, 30], the limit values will be referred to, if necessary. The formation of soluble complexes is also important for the laundering process; data will also be given about this. Only the most important valences of the elements which have been mentioned, have been indicated.

2.4.1 *Caesium, Strontium, Barium, Radium (Manganese^{II}, Cobalt^{II}, Nickel^{II})*

These elements form easily soluble compounds up to pH 7. Thereby, they are able to be stored in cation exchanging media. In the case of Manganese^{II} (limit pH value for hydroxide precipitation is about 8.5), insoluble particles of Mn^{II} hydroxide, being available in traces, can be precipitated in neutral solutions by oxidation to a heavy soluble Manganese dioxide hydrate. By fluctuating pH value Cobalt and Nickel — because of the formation of hydroxides with a limit pH value of about 6.7-6.8 — can be adsorbed to the textile surface or to a hydroxide — some Fe(OH)₃ — which can be found in the carrier medium. Since the precipitated oxide-hydrates are mostly positively charged, a rather strong fixation results occasionally.

Investigations of chelating agents with respect to earth alkalies have indicated that the stability of the complexes generally decreases from Calcium to Radium. The chelate formation of the other elements is well known, and uses therefore can be found in analytical and radio-chemical work.

2.4.2 *Manganese^{II}, Cobalt^{II}, Nickel^{II}, Iron^{II}, ^{III}, Chromium^{III}, Yttrium^{III}, Rare Earths^{III} (IV), Zirconium^{IV}, Hafnium^{IV}, Thorium^{IV}, Uranium^{IV}, Plutonium^{IV}, ^{VI}*

The elements above summarized form cations predominantly. However, they are inclined to form hydroxides in neutral to weak acid solutions. Whereas iron may be constantly present as a non-radioactive carrier, the iron hydroxide brings about the additional precipitation of the larger part of the above mentioned elements. In the two works referred to [27, 28] it was possible — by testing the behaviour of fallout samples in aqueous solutions on ion exchangers — to ascertain that a strong adsorption to the cation exchanger at least for zirconium and iron occurred. This adsorption can be attributed to the presence of colloidal oxihydrates. Therefore, one might suppose that this group can also be bound to the electronegative adhesive spots on the textile fibres if the conditions are given. Generally, due to the high molecular size, a deeper penetration into the fibres will be less frequent. In this context it should be mentioned that Thorium^{IV} can be bound very strongly on a cation exchanger — stronger than all other cations [18].

The behaviour of uranium and plutonium is somewhat different. Because of their properties they already stand near to the next group of elements and can possibly still remain in solution. For instance, uranyl salts are prevented from precipitating even by very small amounts

of carbonate ions. Yet due to this they can penetrate deeper into the fibre. In the case of plutonium, the valence of four is the most stable one. However, one also has to reckon with the valence of six. Whereas plutonium^{IV} salts are inclined to be hydrolysed and, therefore, their properties are similar to the ones of the other elements, the plutonium^{VI} salts and the plutonyl salts (which are quite similar to the uranyl salts) have an extraordinary good solubility under the mentioned conditions. Above all, the alkali plutonates are more soluble than the uranyl salts and the precipitation of plutonium by the hydroxides in such solutions is never quantitative [15, 16, 31].

The formation of complexes is easily possible with all elements of this group and is also analytically used. Characteristical examples are the formation of fluoride complexes of zirconium, hafnium and thorium. The last one can also be easily converted into a complex by means of carbonate ions.

Further on, a series of chelates is described. The complex formation of cobalt, nickel, iron and chromium is sufficiently known and is analytically employed, e.g. in complexometric titrations. The complex formation of rare earths and Yttrium has also been explored. For example, by the addition of phosphates and polyphosphates in a sufficient surplus, rare earths can be easily held in solution or brought to solution [7].

In the case of uranium and plutonium, a large number of complexes has been already described. However, Pu^{IV} forms complexes more easily than Pu^{VI} — it also forms easily soluble phosphate complexes.

2.4.3 Antimony^{III, V}, Tungsten^{VI}, Ruthenium^{II-VIII}

Here, the tendency towards the formation of amphoteric compounds predominates under the given conditions. Above all, numerous investigations were carried out with ruthenium. From tests with aqueous fallout samples [26, 28] 20 % to 30 % of the ruthenium results as a cation complex; 0-60 % as an anion complex and the rest probably as an uncharged complex. BARRY was able to show that cationic nitrosyl ruthenium complexes are absorbed to cotton [32]. These compounds formed in nitric acid solutions of fission products, remain stable under certain conditions even when diluted with much water.

These elements, too, are partly adsorbed to hydroxides; however, when dissolved they could also be adsorbed to the fibre, if the pH value is favourable.

Antimony and tungsten can form complexes (i.e. with tartaric and citric acids) and could, therefore, also be removable. In the case of ruthenium the conditions are less favourable, because complexes of varying strength are formed by the simultaneous occurrence of different valences.

2.4.4 Phosphorus, Sulphur, Tellurium, Iodine

Phosphorus, sulphur, tellurium, and iodine are found practically only in the form of anions. A favoured adsorption to textile fibres having the properties of anion exchangers, i.e. having anion exchanger groups, is possible, yet it has been seldom observed. The insufficient degree of decontamination which has been observed only in individual cases has not yet been clarified for individual anions like phosphate and iodine. In the case of iodine an explanation is complicated because of the easy oxydability of the hydro-iodic acid to elementary iodine. More will be said on this in the discussion of the investigations carried out with fabrics and iodine-131.

3 — THE MOST IMPORTANT TEXTILE MATERIALS

3.1 Cotton [1]

Cotton is almost exclusively used for protective clothing in nuclear and radiochemical research institutes, etc., as well as to a large extent in normal clothes. The cotton fibers consist of cellulose with a definite proportion of oxycellulose. The glucose number, i.e. the number of the glucose units per carboxyl group is about 800-2,000 for cleaned and bleached products. The carboxyl groups determine the ion exchanging ability of the fibres as well as the adsorption of the positively charged dirt particles.

3.2 Artificial Silk (Rayon, so named since 1930)

The artificial silk called Rayon is produced according to the well known process of cellulose xanthogenate by pressing the solution of the xanthogenate into an acid spin bath. Chemically it conforms in the main with cotton. It should be considered, that the individual fibres can have a smaller diameter than natural silk. There is a difference between "regenerated cellulose" as viscous silk and copper silk and cellulose acetate silk.

3.3 Wool [33]

Wool consists of framing substances being composed of ceratines, protein compounds containing sulphur and nitrogen; it is an aggregate of cells and consists of spindle and cortex cells which in turn are composed by micro-fibrils held together by cystine bridges, formed by the dehydration of terminal sulfhydryl groups. The molecular weight is about 62,000. Altogether about 19 amino acids participate in the synthesis of the fibre. The elementary analysis shows an average of 50 % carbon, 22-24 % oxygen, 16-17 % nitrogen, 7 % hydrogen and 3-4 % sulphur. The specific gravity is about 1.3.

3.4 Natural Silk [34]

The silk thread consists of a double thread of silk fibroin. It is surrounded by sericin as a cementing substance. Both substances are proteins. The so-called removal of bast, which results under the influence of alkalies etc., removes the sericin. Silk fibroin consists of glycine and alanine up to about 60 % and is practically free from sulphur-containing amino acids. The amino acid sequence is — according to SCHROEDER et al. [35] — Ala-Gly-Ala-Gly-X-Gly whereby X may be serine, tyrosine, valine or asparagine. The value of the molecular weight varies between 33,000 and 84,000. It is noteworthy that silk binds iron, chromium, aluminum and tin salts in cold aqueous solutions. This may be of interest for the charging and colouring of the fibre and also for its behaviour during the decontamination. Nevertheless, as far as we know, investigations with silk of this type have never been carried out.

3.5 Polyamide Fibres (Perlon, Nylon) [36]

Nylon is produced by the condensation of a diamine with a dicarboxylic acid (e.g. Hexamethylenediamine and adipic acid give Nylon 66) (CAROTHERS, 1935). Its thermostability and its high tensile strength result from the hydrogen bonds between the NH- and the CO-groups of neighbouring chains. The melted polymerisate is first pressed through nozzles, then cooled and stretched. This treatment makes the fibres somewhat similar to a silk fibre.

Perlon is produced from caprolactam according to a process developed in Germany (SCHLACK, 1938) and has properties which are similar to Nylon. The melting point is about 245 °C.

3.6 Polyester Fibres (Dacron, Diolen, Trevira) (37)

Since in such materials hydrogen bonds between the chains are not possible, the melting point is relatively low. Pure aliphatic polyesters can thus not be used for textile fibres; one uses e.g. polyethyleneterephthalate as fibre base.

3.7 Polyacrylonitrile Fibres (Orlon, Dralon) [38]

The raw material is produced by the polymerisation of acrylonitrile in solution, emulsion or suspension. The fibre is spun after having dissolved the polyacrylonitrile in an adequate solvent (e.g. dimethylformamide) and has properties similar to silk or wool.

4 — IDEAS ABOUT THE ADHESION OF RADIONUCLEIDES TO TEXTILE FIBRES

4.1 Cotton and Artificial Silk

In a series of investigations with textile fibres [1] it was ascertained that cellulose can act as a cation exchanger; however, the mechanism is still little known. It is interesting, e.g., that SCHÖNFELD and BRODA [39, 40] found in their investigations that the hydrogen ion is much more strongly bound than alkali ions. This does not agree with the results on cation exchangers and can be attributed to the covalent binding of hydrogen. Therefore, when comparing with the usual ion exchangers, caution is requested. In any case it is certain that cations are very strongly bound sometimes, which also has been shown in BARRY's [32] tests with cation complexes of ruthenium. In laboratory tests carried out by LAMBERT [41] with Ca-45 in hard water, a strong adsorption of calcium to the fibres was observed. According to IVANOV, among others, iron is also unusually strongly adsorbed [42]. According to the research work carried out by COLCLOUGH et al. [43] and which will be discussed in detail later on, the surface resistance must be included in the evaluation of these tests. In the case of cotton it is relatively low. Actually cotton is easier to decontaminate than artificial fibres which have a high surface resistance. Hence it follows that the decontamination improves always if the surface resistance decreases.

These considerations are really suitable also to artificial silk.

4.2 Wool, Natural Silk, Polyamides

Because of the similar amphoteric structure of these substances they have cation as well (COOH groups) as anion exchanging (amino groups) properties. By means of sterical effects — i.e. sulphur bridges in the wool — the properties should be less evident as in cotton. However, the behaviour of natural silk differs strongly, sometimes (see also 3.4). HUGHES et al. [44] observed that the degree of decontamination was somewhat higher for wool and somewhat lower for Nylon. Since wool and Nylon differ noticeably in surface resistance, this might be the reason and not the better adsorption of acid groups, as supposed by COLCLOUGH [43].

4.3 Polyester Fibres

Polyester fibres have primarily a neutral behaviour, if one does not consider the charge. However, in ageing processes saponification can occur. The resulting COOH-groups should cause weak cation exchanging properties. Here also the occurring charges should be more important.

4.4 Polyacrylonitrile Fibres

These fibres, also have predominantly neutral properties so that the behaviour can be explained exclusively by occurring charges. In the hydrolysis of the nitrile group first a neutral acid amide is formed and only thereafter the acid. Surely such ageing processes can hardly be of any importance.

5 — DECONTAMINATING AGENTS

5.1 Complexing Agents for Certain Radionuclides

During the contamination by a certain nuclide or with rather simple nuclide mixtures, the use of special complexing agents is often advantageous, as for instance a solution of sodium bicarbonate in the contamination of uranium or thorium salts. In many cases citric acid is also very favourable.

A disadvantage for such specific complexing agents is that only a small effect can be obtained if the radionuclides do not react with the complexing agent or if they cause insoluble precipitations on the fibre.

5.2 Chelating Agents

EDTA (or its sodium salt) is a good decontaminating agent. Disadvantageous, however, are its relatively high costs and especially its strong corrosion of metals. It is also disturbing that it is most difficult to precipitate the chelate compounds again. That can complicate the decontamination of the laundry's waste water.

5.3 Polyphosphates

Polyphosphates, besides their effect as soluble ion exchangers [45], have a good suspending, peptizing and emulsifying power for greasy substances. In addition they can still act as a buffering system. Hence their utilization for the decontamination of textiles is very promising. By the addition of the so-called "wash alkalies" (sodium carbonate or sodium silicate) one can regulate the most favourable pH range so that the polyphosphates have the same decontaminating effect as the salt of EDTA, but they are more economical than those. When polyphosphates are used in the decontamination of the laundry's waste water it is possible to precipitate the soluble calcium or heavy metal polyphosphates by adding surplus calcium or heavy metal cations.

5.4 Detergents (Tensides)

In this group one finds combinations of detergents which can be cationic, anionic, or non-ionic — see the detailed information in part II — for lowering the surface tension with substances, such as CMC which raise the so-called "dirt carrying ability", and with pH regulating alkaline additives. The addition of polyphosphates is a significant improvement on the cleaning effect, and therefore on the decontaminating effect, too.

A disadvantage of synthetic detergents is the fact that in the processing of the waste water they cannot be separated quantitatively, e.g. by adsorption with iron hydroxide.

5.5 Soap

Contrary to tensides, soaps have sufficiently known disadvantages: a smaller ability to dissolve and to disperse fats, in alkaline media precipitation by earth alkalies and heavy metal ions, in acid solutions destruction under separation of insoluble fatty acids. Soap also has some advantages: it emulsifies better and also has a larger dirt carrying ability; nevertheless, this last decreases rapidly with an increasing concentration of inorganic salts.

In the decontamination of waste water one can — by adding slaked lime — precipitate the calcium salts which are insoluble and carry with themselves the larger part of the radionuclides. When treated with steam, such solutions do not foam.

By combining soap with polyphosphates, the washing power and therewith the decontaminating effect is improved [1].

With such combinations one can obtain very good decontaminating effects. Disadvantageous, however, is the fact that polyphosphates are removed already in the first rinsing step, whereas part of the soap still remains adsorbed to the fabric. If a noticeable amount of calcium is present one must, therefore, use soft water for the second rinsing step or add additional polyphosphates in order to prevent the precipitation of calcium soap on the fabric.

6 — STUDIES ON THE DECONTAMINATION OF TEXTILES. LAUNDERING PRACTICE

6.1 Contamination of the Samples

According to the type of contamination of textiles — mostly brought about on purpose — different results are obtained in the washing process. Therefore, in any series of experiments one must know the conditions in order to judge better the mode of operation. Generally, the contamination of the respectively investigated radionuclides predominates in the form of solutions because of the simpler technique [43, 46, 47, 48, 49, 50, 51, 52, 53]. To evaluate the decontaminability in the case of dust contamination, which above all is interesting for military applications and also for the nuclear technique, ingenious apparatus were constructed in order to contaminate the textiles evenly [44, 48]. In part, both possibilities were investigated simultaneously [48, 51].

When utilizing solutions, a stronger adsorption should occur as in the case of dust contamination, because it can be assumed that the radionuclides are already bound in part to the inactive carrier material (silicates, oxides etc.). But it may be that the solutions of the fission products used, or the solutions of uranium, plutonium, radium etc. have a pH value unfavourable to adsorption. An influence of the pH value on the adsorption of ions to textiles has been observed occasionally [1]. In these investigations particular cases shall be discussed later.

6.2 Measuring Techniques

In the US army and in the laundries of the nuclear research centers there are large surface counting tubes in operation which allow an exact control of the contamination and decontamination. In controlling washing tests under conditions similar to those of normal practice it would be favourable to measure the protective clothing, i.e. the contaminated textiles, directly which was actually done in a few investigations [44].

Because of technical difficulties in measuring and because of the high costs for counters and for the contaminating substances in the large scale tests, relatively small samples were measured with counters of normal size in almost all the washing tests (Geiger counters, Müller counters, flow-through and scintillation counters). Thus, in individual cases there resulted considerably more exact measuring values.

6.3 Washing Methods for Laboratory and Large Scale Tests

Tests which closely resembled the normal washing practice were carried out which allowed large pieces of clothing to be contaminated and measured. They also can be controlled in normal washing machines after the various decontamination steps. The large scale tests of military organizations [44] were carried out according to this scheme, and also the control in the nuclear development centers is carried out in this manner, whereby it is also possible to evaluate laboratory washing processes in the practice.

Naturally because of the form of the pieces of clothing considerable differences can occur in the measured values, especially when using soft rays, i.e. less penetrating alpha and beta rays. Since large surface counters of the required construction are normally not available in usual wash or radiochemical laboratories, one has preferred in most cases to wash the textile samples in commercial washing machines whereby the size of the individual pieces was adjusted to the available measuring device [43, 46, 48, 51, 53, 55]. REIFF, SCHUSTER and HEINEN [40] even subjected inactive clothes together with the contaminated clothes samples to the washing processes in a household washing machine. Thus, by a very economical use of the available means, conditions resulted which were close to those of normal wash practice. In this way exact measuring values were gained from the contaminated samples which were fixed to various spots of the inactive clothes.

The extensive and instructive investigations by TALBOYS and SPRATT [52], however, were only carried out with textile strips in shaking apparatus, therefore one cannot compare these results directly with those of other works.

6.4 Decontamination Results with Various Textiles and Radionuclides

6.4.1 Cellulose Fibres : Cotton and Artificial Silk

Including washing tests with textiles of other fibres.

6.4.1.1 Contamination tests by HUGHES et al. [44]

6.4.1.1.1 Contamination

The contamination of pieces of clothing and textile samples of various materials which were used in the test was carried out by an apparatus in which radioactive dust — soil samples from the neighbourhood of the explosion crater of a nuclear test — was fixed to the samples in a controlled manner.

In addition protective suits etc., which were worn by the scientific and technical personnel during investigations of underground nuclear explosions were also examined. The fission products were on the average probably not more than 3-10 days old.

6.4.1.1.2 Washing Process

The washing process was a modification of the US Army standard washing process of the year 1947. The washing was done in large drum washing machines of stainless steel and wood of the type of normal field laundries.

6.4.1.1.3 Modification of the Basic Process

CONFIDENTIAL

Security Information Project 6.7

Thirty suits each of herringbone twill clothing and field clothing were issued to the personnel of Project 6.2 for wear in the "Land Reclamation Program".

A number of Project 6.3.1 test garments worn by persons entering the shot area were decontaminated and returned to that project for their evaluation. (See report of Project 6.3.1 for details and results).

2.7 Operating Supplies

The following detergents and chemicals were used during the decontamination operation :

Citric Acid (commercial crystals)		84 lbs.
Tartaric Acid (commercial crystals)		18 lbs.
EDTA (tetra sodium salt of ethylene-diamine-tetra-acetic acid)		45 lbs.
Oxalic acid (technical crystals)		15 lbs.
Laundry Sour (mixture of equal parts of sodium silico-fluoride and sodium-acid-fluoride)		20 lbs.
Armour Detergent		23 lbs.
Chemical Composition :		
Renex	45.0 %	
Carboxymethyl Cellulose	4.5 %	
Urea	50.5 %	
General Aniline and Film Detergent		5 lbs.
Chemical Composition :		
Antarox (non-ionic)	20.0 %	
Borax	50.0 %	
Carboxymethyl Cellulose	3.0 %	
Sodium Sulphate	26.0 %	
Tamol	1.0 %	
Sodium Hexameta-phosphate		8 lbs.

2.8 Water

Water used in the laundry was taken from the normal water supply at Indian Springs Air Force Base. An analysis of a sample of this water, drawn in September 1951, made by the US Bureau of Standards, is given in Table 2.3.

<i>Load No.</i>	<i>Decontamination Process</i>
3.	Formula 77 A
4.	Formula 77 A — Tartaric Acid sub. for Citric Acid
5.	Formula 77 A — (NaPO ₃) ₆ sub. for EDTA
6.	Formula 77 A — Laundry Sour sub. for Citric Acid

Preliminary tests conducted using the contaminating tumbler indicated that one pound of the sifted dirt per one-third of a laundry load resulted in an adequate level of contamination for evaluation.

TABLE 3.1 — Decontaminating Laundry Formulae

Step	Operation	Water Level (in)	Temperature °F	Time (min)	Supplies
Standard Quartermaster Mobile Field Formula (1)					
1.	Suds	5	90 - 100	5	6 os. Detergent
2.	Suds	5	130	5	3 os. Detergent
3.	Suds	5	140	5	2 os. Detergent
4.	Rinse	8	140	3	None
5.	Rinse	8	120	3	None
6.	Rinse	8	100	3	None
Formula 77 A					
1.	Suds	6	90 - 100	5	6 os. Armour Det.
2.	Acid	12	140	5	4 lbs. Citric Acid
3.	Acid	12	140	5	2 lbs. Citric Acid
4.	EDTA	8	140	5	1 ½ lbs. EDTA (2)
5.	EDTA	8	140	5	1 lb. EDTA
6.	Rinse	12	140	3	None
7.	Rinse	12	120	3	None
8.	Sour	12	Tap	5	1 os. Sour

(1) Source, War Department Technical Bulletin 10-552-2, dated February 26, 1946; however FM 10-16 "Quartermaster Laundry Company Semimobile" Department of the Army dated June 1950 has eliminated the third suds in the above formula. The three suds formula has been tested during previous tests and results have indicated that three suds are imperative.

(2) Tetra sodium salt of ethylene-diamine-tetra-acetic acid.

TABLE 3.3 — (Cont'd) Efficiency of Each Step of Laundry Formula

Step	Operation	Laundry Runs		
		"A"	"B"	"C"
1.	Suds	41.5 %	35.0 %	33.2 %
2.	Citric	39.3 %	30.3 %	28.5 %
3.	Citric	27.3 %	14.4 %	16.4 %
4.	(NaPO ₃) ₆	24.7 %	23.7 %	17.8 %
5.	(NaPO ₃) ₆	18.5 %	38.0 %	18.3 %
6.	Rinse	16.5 %	29.6 %	21.7 %
7.	Rinse	14.3 %	15.8 %	19.2 %
8.	Rinse	2.8 %	7.0 %	9.3 %

Step	Operation	Laundry Runs		
		"A"	"B"	"C"
Formula 77 A with Tartaric Acid				
1.	Suds	47.7 %	36.5 %	36.0 %
2.	Tartaric	52.3 %	45.0 %	31.9 %
3.	Tartaric	23.5 %	23.0 %	19.8 %
4.	EDTA	21.4 %	27.6 %	15.3 %
5.	EDTA	28.8 %	38.8 %	19.7 %
6.	Rinse	27.5 %	26.4 %	24.3 %
7.	Rinse	9.8 %	13.4 %	14.9 %
8.	Sour	3.7 %	14.5 %	10.1 %
Formula 77 A with Sour				
1.	Suds	44.3 %	29.9 %	30.9 %
2.	Sour	40.8 %	24.5 %	19.5 %
3.	Sour	22.7 %	15.3 %	11.6 %
4.	EDTA	34.9 %	17.2 %	14.3 %
5.	EDTA	40.3 %	34.0 %	16.0 %
6.	Rinse	37.2 %	33.4 %	16.1 %
7.	Rinse	24.6 %	20.5 %	9.8 %
8.	Sour	—	7.5 %	5.0 %
Standard Field Formula (Armour Detergent)				
1.	Suds	41.0 %	31.6 %	31.8 %
2.	Suds	29.1 %	31.0 %	27.4 %
3.	Suds	20.8 %	27.7 %	24.8 %
4.	Rinse	23.7 %	26.7 %	21.5 %
5.	Rinse	20.0 %	19.5 %	15.3 %
6.	Rinse	13.8 %	14.5 %	12.0 %
Standard Field Formula (General Aniline)				
1.	Suds	40.6 %	35.1 %	32.3 %
2.	Suds	36.2 %	34.5 %	28.2 %
3.	Suds	28.8 %	27.0 %	25.2 %
4.	Rinse	19.5 %	19.5 %	18.3 %
5.	Rinse	16.6 %	13.4 %	13.7 %
6.	Rinse	7.2 %	8.3 %	10.2 %
Formula 77 A				
1.	Suds	44.6 %	28.9 %	33.2 %
2.	Citric	41.5 %	27.8 %	33.9 %
3.	Citric	19.8 %	16.7 %	19.5 %
4.	EDTA	26.5 %	15.7 %	16.5 %
5.	EDTA	24.2 %	16.8 %	20.0 %
6.	Rinse	21.4 %	22.4 %	24.7 %
7.	Rinse	6.5 %	24.7 %	19.9 %
8.	Rinse	2.5 %	29.7 %	11.3 %

6.4.1.1.4 Trial of the Modifications of the Laundry Formula with Contaminated Textile Samples of Constant Composition.

About 25 kilos of clothing were used for the tests tabulated below. They were contaminated according to 6.4.1.1.1, and consisted of cotton-sateen, Nylon and Rayon.

Composition of Washing Liquor	Average Degree of Decontamination
Standard Field Formula with Armour Detergent	80.5 %
Standard Field Formula with Aniline Detergent	78.6 %
Formula 77 A without changes	87.8 %
Formula 77 A Tartaric Acid sub. for Citric Acid	89.8 %
Formula 77 A Hexametaphosphate sub. for EDTA	87.3 %
Formula 77 A Sour sub. for Citric acid	81.5 %
Pure Water	64.7 %

A table is given below dividing the three washing processes into individual steps as an example of the effectivity of the individual decontamination steps. The respective washing liquors were also measured.

6.4.1.1.5 Investigation of Special Fabrics Which Were Contaminated as Indicated Above

Washing Liquor : According to the Standard Field Formula with Armour or Aniline Detergent

Material	Degree of Decontamination
Cotton (HBT OD 7)	66.6 %
Cotton (Cardet sateen)	83.5 %
Cotton mostly prepared as water repellent :	
Cotton untreated (control value)	74.6 %
Cotton Zelan AP Base	76.2 %
Cotton Norane	35.2 %
Cotton Aluminum, soap and wax	73.8 %
Cotton Permell	82.2 %
Cotton Inorganic Pigments	67.8 %
Cotton Inorganic Pigments, Permell	63.1 %
Rayon (sateen lining, Viscose)	72.2 %

In the case of cotton equipped with a water repellent preparation a lesser decontamination was partly observed as compared to pure cotton.

6.4.1.1.6 Observations, Supplements

The investigations indicated under 6.4.1.1.5 were carried out with the Standard Field Formula, since a price comparison of the substances used for the Formula 77 showed that the SFF was considerably cheaper.

The price data of the year 1952 are given hereunder.

Formula 77 A	\$ 3.55
Formula 77 A with Tartaric Acid	\$ 4.45
Formula 77 A with Hexametaphosphate	\$ 2.42
Formula 77 A with Laundry Sour	\$ 2.37
MFJ with Armour or General Aniline Detergent	\$ 0.20

6.4.1.2 Washing Experiments by TALBOYS and SPRATT [52]

6.4.1.2.1 Contamination

The cloth samples are dipped into the radioactive solution for 30 minutes, then taken out and dried.

6.4.1.2.2 Washing Process

The washing was done in shaking flasks, at room temperature and in the following order : wash 10 min., rinse 5 min., wash 10 min. with a new washing liquor, rinse again 5 min., then dry the samples.

The wet samples were not centrifuged, so that their moisture content was considerably more than 100 % related to the dry sample.

6.4.1.2.3 Degree of Decontamination of Cotton Fabrics

Decontaminating Agent	Contamination with					
	Sr-89	V 91	Ce-141	Fe-59	F-32	J-131
Citric Acid	99.2 %	62 %	94.5 %	93 %	50 %	95 %
Ammonium citrate	100 %	98.2 %	99.4 %	97.5 %	75 %	96 %
Sodium citrate	99.7 %	—	98.5 %	95 %	85 %	99 %
Versene	100 %	99.4 %	99.1 %	96.5 %	96 %	95 %
Radiacwash	99.5 %	—	—	—	94.5 %	95 %
C.P. Cleaner	100 %	99.5 %	99.5 %	—	96 %	97 %
Roccol	—	—	60 %	—	—	—
Durite	62 %	—	—	80 %	87 %	96 %
Nacconol N.R.	98.5 %	78 %	88 %	81 %	84 %	99 %
Igepal CO	91 %	30 %	87 %	79 %	75 %	96.5 %
Calgon	99.9 %	99.6 %	99.5 %	98.5 %	85 %	98.5 %
Rinso	87 %	87 %	81 %	80 %	92 %	96.5 %
Demineralized Water	70 %	45 %	70 %	65 %	82 %	95 %
Tap Water	98 %	35 %	42 %	—	81 %	93 %
Na J	—	—	—	—	—	99 %
Concentration	3 %	3 %	3 %	3 %	1 %	1 %

The authors suggest that "Fe-3-specific" should be taken instead of Versene for radioactive contamination, or rather the substance itself added to Versene.

The samples contaminated with J-131 in an alkaline medium were more easily to decontaminate than those in an acid medium.

6.4.1.2.4 Details about the Used Decontaminating Agents :

- Versene (Na-EDTA).
- Radiacwash (Na-EDTA-containing detergent).
- C.P. Cleaner (Complex forming det.).
- Roccol (Cationic detergent).
- Durite (Soda containing anionic det.).
- Nacconol N.R. (Synthetic anionic det.).
- Igepal CO (Nonionic detergent).
- Calgon (Polyphosphate).
- Rinso (Soap).

6.4.1.2.5 Investigations about the Dependence of the Degree of Decontamination on the Type of the Contamination (Solution, Precipitation on the Fibre, Dust)

Decontaminated with	Degree of Decontamination		
	Fe-Solution	Fe-Precipitation	Fe-Dust
Citric Acid	93.0 %	97.6 %	93.8 %
Versene	84 %	50 %	93.2 %
Calgon	98.7 %	100 %	89.5 %
Soap	66 %	32 %	97.6 %
Tap Water	63 %	21 %	84 %
	Sr-Solution	Sr-Precipitation	Sr-Dust
Ammonium Citrate	100 %	99.9 %	98.1 %
Versene	100 %	100 %	100 %
Demineralized water	72 %	42 %	92.7 %
Tap Water	98.3 %	36 %	95.8 %

6.4.1.3 Washing Experiments by NORWOOD [48]

6.4.1.3.1 Contamination

Place drops of the radioactive solution onto a cotton sample of a 25 by 25 cm surface and let dry.

6.4.1.3.2 Washing Process

The standard washing process will be done in a 19 liter washing drum. The test samples will be rinsed for a 3 minute period each time at the following temperatures : cold, 50 °C, 80 °C. They will then be washed for a period of 15 min with detergents of the previously mentioned combinations. Finally rinsing is done for a 3 min period each time at the following temperatures : once hot, two times at 50 °C, and once cold. All operations were carried out by hand, as well as the wringing at the end. Noticeable are the unusually high concentrations of the decontaminating agents which are given in column 1 of the following table.

6.4.1.3.3 Results of Decontamination in Dependence to the Detergent and the Radionuclides or Nucleide Mixtures

Decontaminating Agent	Concentration %	Degree of Decontamination (%)				
		Pu	Uranium Alpha	Beta + Gamma	Fission Product	Soft Beta rays
Calgon	3					
Hilco Powder	0.75	85			38	44
Soda	0.75					
Calgon	3					
Hilco Powder	0.75	92	99.2	40	92	52
Soap	0.75					
Calgon	0.75					
Hilco Powder	0.75	93				
Versene T	0.75					
Turco 733	2.25					
Turco 4056	1.5	95	79	29		91
Triton X	2.25					
Turco 733	2.25	96	92	88		91
Turco 4056	1.5					
Washed in a household-machine						
Turco 733 and 4056	1.5	99	92	88		91
Triton X	1.5					
Hilco Powder	0.75					
Versene T	0.75	59	84	89	65	74
Duportal	0.75					
Hilco Powder	0.75					
Versene T	1.5	55	89	91	76	84
Soap	1.5					
Repetition of the Process		64	99.4	95	95	93
Versene T	2.25					
Soap	0.75	94	93	79		91
Hilco Powder	1.5					
Pure Turco 4182	2.25	99.6	100		98.3	92.7
Turco 4182	2.25					
Ammonium Bifluoride	0.75	98.5	100		99.3	91.7

6.4.1.3.4 Application in Practice

Since after the washing tests the phosphate containing detergent Turco 4182 turned out to be very favourable, about 6,000 contaminated test samples and other contaminated material were treated in a large scale washing test with this decontaminating agent. Thereby, almost all the treated objects could be released after the second control measurement. Only about 5 (beta-contamination), or 8 (alpha-contamination) of about 6,300 individual pieces had to be rejected.

6.4.1.3.5 Survey on the Decontaminating Agents

1. Ammonium bifluoride (sequestering agent).
2. Brex (high titer laundry soap compound).
3. Calgon (complexing agent - unadjusted sodium phosphate glass).
4. Duponal (high grade synthetic detergent).
5. Hilco powder (heavy detergent - contains 90 % trisodium phosphate).
6. Metso anhydrous (complexing agent).
7. Metso "99" (built silicate compound).
8. Nacconol N.R. (alkyl-aryl sulfonate).
9. Orvis Hi Temp (high titer laundry compound).
10. Soap (Lever Bros. Formula 770 - high titer laundry soap).
11. Soda ash (used as a buffering agent).
12. Sodium bicarbonate (complexing agent).
13. Sodium metasilicate (complexing agent).
14. Triton X-100.
15. Turco 733 (compound of hydroxy acetic acid and non-ionic detergent).
16. Turco 3978 (proprietary amino compound).
17. Turco 4028 (proprietary sodium carbonate based compound with non-ionic detergent).
18. Turco 4056 (glassy phosphate and non-ionic detergent).
19. Turco 4182 (ammonium carbonate compound with glassy phosphate and detergent).
20. Versene T (chelating agent - tetra sodium salt of ethylene-diamine-tetra-acetic acid).
21. Wyandotte 33 (bentonite clay compound).
22. Wyandotte OS (soap builder).

6.4.1.4 Washing tests by SOROM [51]

6.4.1.4.1 Contamination

Cloth samples of a size of 10 × 10 inches taken from clean overalls are contaminated, either by rubbing with a dry insoluble material emitting alpha rays (75 samples), or with drops of a plutonium solution.

6.4.1.4.2 Washing Process

In normal washing machines one washes according to the following cycle : two rinsing steps, each lasting 2 min at 120 °F, two washing steps of 10 min at 180 °F, two rinsing steps of 2 min at 180 °F and 2 rinsing steps of 2 min at 120 °F. The washing process is repeated up to three times in the first series. Nothing is said about the concentration of the decontaminating agent. Therefore, it can be assumed that the concentrations correspond to those of household washing.

6.4.1.4.3 Decontamination of Fabrics contaminated with Plutonium

Decontaminating Agent	Degree of Decontamination (%)		
	1st step	2nd step	3rd step
P & G Hytemp Orvus	98.3	55.1	47
Tide	98.2	38.3	20
Triton X100	98.2	28.2	41
Metso (Anhydrous)	97.3	73.3	
Turco 4324	97.5	47.2	38
Wyandotte-Riddax	97.8	47.2	34
Nacconol N.R.	98.0	39.0	28
Wyandotte OS	96.9	26.8	
Duonal ME	95.5	38.5	45
Turco 4182 A	94.7	21.7	28

6.4.1.4.4 Removal of β -Contamination in an One-step Washing Process with Four Different Substances

Decontaminating Agent	Degree of Decontamination (%)
Hytemp Orvus	99.3
Nacconol N.R.	99.1
Wyandotte Riddax	98.4
Water	96.8

More specific data on this test are lacking in SOROM's paper.

6.4.1.4.5 Composition of the Decontaminating Agents Used by SOROM - Insofar as indicated

Decontaminating Agent	Composition
P & G Hytemp Orvus	Essentially sodium alkylsulphate detergent, granules anionic built, heavy duty type detergent and cleaner
Tide	Cleaning and wetting agent, in 1950 described as follows : Laurylsulphate 12 % High molecular phosphates 45 % Sodium sulphate 15 % Silicate 9 % Laurylalcohol 1.5 % Water 11 % CMC and brightener Available
Triton X-100	Isooctyl phenyl polyethoxy ethanol wetting detergent, liquid 100 %, non-ionic household and industrial cleaner

Metso anhydrous	Complexing agent
Turco 4324	?
Wyandotte Riddax	?
Nacconol N.R.	Alkyl-aryl-sulphonate
Wyandotte OS	Soap builder
Duponal-ME	Technical laurylalkoholsulphate powder
Turco 4182 A	Probably similar to Turco 4182 which is described as an ammonium carbonate compound with glassy phosphate and detergents

6.4.1.5 Washing Tests by REIFF, SCHUSTER and HEINEN [50]

6.4.1.5.1 Contamination

Dipping of the cloth samples into aqueous solution of the respectively used radionuclide. After one hour they are taken out of the solution, dried and measured. Thereafter they can be washed. The individual samples were either of a size of 20 × 20 cm (for the nucleides Cs-137, Sr-90, Ra-226, and J-131) or disks of a 3 cm diameter (for S-35).

6.4.1.5.2 Washing Process

In order to obtain a close approximation to normal practice, the washing was done in a normal household washing machine, and inactive laboratory coats were added to the load. The authors consciously endeavoured — contrary to the experiments of TALBOYS and SPRATT [52] — to work under most complicated conditions. Hence, the contaminated samples were attached to the filling material in the first tests, and later on put into the pockets of laboratory coats in order to complicate the access of the washing liquor. The concentration of the Calgon solutions used was generally 2.5 g/l, in particular cases 5 g/l. The pH value was set at 9, because under this condition the metal ions are already precipitated as hydroxides.

6.4.1.5.3 Summary of the Tests With Cotton Fabrics

Decontaminating Agent	Concentration (%)	Washing time (min)	Degree of Decontamination (%)				
			Cs-137	Sr-90	Ra-226	J-131	S-35
1) Shirting							
Calgon	0.25	15	99.5	96.8	—	—	—
Calgon	0.25	30	—	99.5	96	95.6	99.8
Calgon	0.5	30	—	—	98	—	—
Carbonate-bicarbonate solution			—	64	80	95	—
Water			98.9	47	90	88	97
2) Twill							
Calgon	0.25	15	99.4	75	—	—	—
Calgon	0.25	30	—	88	94-98	97.9	97.8
Calgon	0.5	30	—	—	93	—	—
Carbonate-bicarbonate solution			—	50	63	99.1	98.9
Water			96.9	42	68	95.7	97.9

6.4.1.6 A Survey on the Experiments of Other Authors

BRANCA [56] investigated the decontaminating effect of commercial detergents on cotton. The influence of the concentration of the respective detergents, of the temperature and of the material on the decontaminating effect were stated above all.

CERRE [57] and PAVLOVIC [49] tried without success to decontaminate cotton and other textiles by treating them with supersonic waves.

COOK's AHSB report [58] deals with principles and organizational questions concerning the decontamination of clothes. For the decontamination in commercial washing machines, the AWRE-Detergent No. 10 (Spec. H. R. 378), as well as the detergents Deepio and Horizon — mostly with a small EDTA addition — are recommended.

KANG et al. [46, 55] explored the decontamination with the sodium salt of EDTA and with detergents on fabrics contaminated with uranium and strontium-90.

When uranyl nitrate was used, the contamination was larger in the case of cotton than in the case of wool (proportion about 9 : 1). It was found that the detergents used had practically no effect. Na-EDTA, on the other hand, was rather successful, whereby the optimal concentration of this substance was about 0.5 %.

Cotton took up twice as much of strontium-90 activity as did wool. The decontamination was just as successful with detergents as with Na-EDTA. For an optimal decontamination, a 0.03 % solution of Na-EDTA was sufficient. Optimal effects resulted with solutions of 2 % for Cleaner N, and 5 % for Armour Detergent.

Finally, it should be mentioned that in these experiments the step by step decontamination of cotton resulted in better values than in the case of wool.

MIURA and ISHIDA [59] tested a series of materials in relation to their contaminating and decontaminating properties; pieces of clothes were also among the materials tested.

NETZEL [60] applied activated procedures analytically in order to determine the absolute percentage of dirt on clothes soiled with mud, both before and after having washed them with a detergent. This method could be interesting for the evaluation of decontaminating processes, as applied by HUGHES et al. since it allows to be made a comparison between the dirt remaining on the fibre and the remaining activity.

NISHIWAKI and NISHIOKA [47] studied the contamination of glass, cotton, and foils with strontium-90. On decontaminating with a chelating agent and a surface active substance they found that at least in the case of foils not only the strontium-90 adsorbed on the surface is removed, but also that the strontium-90 being diffused into the foil can be removed to a certain degree.

OHIRAI and YOSHII [61] stated for the decontamination of cotton that the combination of chemical and physical methods is more successful than the application of either one by itself. This also applies to the decontamination of surfaces of different materials.

STEVENSON [32] was able to show that the adsorption of multivalent cations — especially those of cerium — to cotton can be explained as an ion exchange process on the carboxyl groups. Consequently a good decontaminating effect can be expected, especially if the complexing agents added are able to form an anion complex with the respective cations. Since the activation energies of such processes are relatively high, the raising of temperature is always favourable. The sizes which determine the velocity and the limits of the decontamination are probably due to the limited speed of diffusion from the inside of the fibre to the outside (DONNAN-Membrane-Effect).

In another paper [63] — STEVENSON et al. — the behaviour of caesium, tellurium and iodine was examined with respect to cotton. Caesium is only weakly bound. Adsorbed caesium can be removed already with water. In the case of tellurium and iodine they stated that they can easily be removed with water when precipitated on the fibre from a neutral solution. If acid solutions are used for the contamination or the decontamination, both elements are much strongly adsorbed to the fibre. In the case of tellurium, the precipitation of tellurium-dioxide in and on the fibre is supposed. In the case of iodine in an acid medium elementary iodine may be precipitated, since hydroiodic acid is easily oxidized to iodine under these conditions. It appears that iodine is more easily and strongly adsorbed to the fibre than is the iodide-ion. In this case a diffusion of elementary iodine in the inside of the fibre might probably occur, furthermore, it is also possible that the substitution reaction supposed by SIEWERT and SCHIKORA [1] is of importance, since it can cause the strong binding of iodine.

TAKADA et al. (64, 65) studied the decontaminability of a series of surface active substances by altering their concentrations and pH values, primarily with extremely small quantities of radio-isotopes.

The authors came to the conclusion that in almost all cases the decontamination of isotopes without carriers is more difficult than the one of radionuclides with addition of a carrier. In the case of cationic contamination, the decontaminability of anionic surface active substances increased after the critical so-called, "micellae concentration" was obtained. On the other hand, a very good decontaminability was stated for cationic surface active substances within a large concentration range. Non-ionic substances, however, had no effect in the entire concentration range which had been tested. In the case of amphoteric substances, the decontaminability depends markedly on the pH value.

All surface active substances tested proved to be ineffective in the case of anionic contamination.

WADACHI and TAJIMA [66] investigated the mechanism of the adsorption of uranium to cotton and stated a specific linking of soluble uranium ions. In decontamination tests they found that diluted solutions of sodium bicarbonate and ammonium oxalate (each 0.1 mol per liter) were — as expected — especially effective. Furthermore, it was observed that a lowering of the pH value under 3 already removes the uranium ion.

WADACHI and KAWANO [67] tested the decontamination of various types of protective clothings. They used strontium-yttrium-90 as contaminating nucleide. The decontaminating factors for 11 different fabrics and fabric mixtures, i.e. hemp, hemp-cotton, cotton, and hemp-nylon, were divided into three groups: untreated fibres, fibres treated with silicagel, and fibres impregnated with water repellent silicon.

It was stated that hemp and cotton and their mixtures are suitable for decontamination. Especially cotton showed an extraordinary decontaminability. The decontaminating agents used were SDS (this substance was not defined more precisely in the paper), a 1 % solution of hexameta-phosphate, as well as a 0.01 molar solution of Na-EDTA, the phosphate solution being at least equal in value.

6.4.1.7 Patent Literature (Cotton and Mixtures)

BENCKISER D.P. 1,162,695, 1964 [68]

Application of solutions of polymere phosphoric acids, citric or tartaric acids, or rather their salts, for the decontamination of consumer products contaminated with radioactive caesium or with different isotopes combined with anions.

BERSWORTH, F.C. and RUBIN, M., Dow Chemical Co. USP 2,926,576 (1960) [69]

Decontaminating solutions for the removal of radioactive substances from skin, clothes and other objects. Effective substances: EDTA, sodium salts of polyaminopolycarboxyl acids, sodium polyphosphates, etc.

BULAT, T. J., J. Bendix Corp. USP 3,047,434 (1962) [70]

For example a solution is described : Water, EDTA, sodium sulphate, sodium hydroxide manganese dioxide, and "Carbon black".

MORRIS, F. G. USP 3,033,794, (1962) [71]

A tested decontaminating mixture for surfaces and clothes is given as follows : 20-80 % polyphosphate, up to 25 % nonionic detergents, 5-10 % aliphatic monohydroxymonocarboxyl acids, in particular hydroxyacetic acid.

2-5 ounces of this mixture are dissolved in one gallon of water. The results of the experiments with uranium, plutonium and soft beta rays are added.

STEVENSON, United Kingdom Atomic Energy Authority, London. Brit. Pat. 856,377 (1958) [72]

Main ingredients : Besides the detergents, a water soluble polycarboxyl acid with or without the grouping -N (CH₂ COOH)₂. An enclosed graphic representation shows the influence of the pH value on the decontaminating values in the case of cerium and strontium on cotton fabrics.

6.4.2 Wool

6.4.2.1 The Experiments of HUGHES et al. [44]

The following results of decontamination were obtained :

Material	Degree of decontamination (%)
Serge	88.2
Shirting	92.7

The values for wool are always better than the ones for cotton which is in keeping with the above mentioned hypothesis (under 4.2).

6.4.2.2 REIFF, SCHUSTER and HEINEN — Investigations with Woollen Clothing

Decontaminating Agent	Concentration %	Washing time min	Decontaminating factors in % for the Radionucleides				
			Cs-137	Sr-90	Ra-226	J-131	S-35
Calgon	0.25	15	98.2	66-80	—	—	—
Calgon	0.25	30	—	82	97	98	98.7
Calgon	0.50	30	—	—	—	—	—
Carbonate-bicarbonate	—	—	—	40	72 (30 min.)	97	96.8
Water	—	—	95.7	43	64	97.5	98.3

For Cs-137 and the anions J-131 and S-35, treatment with water was completely sufficient from which it can be concluded that the link is extraordinarily loose.

6.4.2.3 Experiments by Other Authors

Once again the experiments of KANG et al. [46, 55] should be referred to the full details of which were given under 6.4.1.6. In particular they showed great differences concerning the degree of decontamination between cotton and wool.

6.4.3 Polyamides (*Perlon, Nylon*)

6.4.3.1 HUGHES et al. [44] obtained an average decontamination of 92.3 % during the testing of about 100 nylon samples

6.4.3.2 The decontaminating experiments on Perlon which are carried out by REIFF et al. [50] are the following:

Decontaminating Agent	Concentration %	Washing time min	Decontaminating factors in % for the Radionuclides				
			Cs-137	Sr-90	Ra-226	J-131	S-35
Calgon	0.25	15	98.8	—	—	—	—
Calgon	0.25	30		91-96	84		100
Calgon	0.5	30			95.9	max. 30	
Carbonate-bicarbonate				77	94.5		99.9
Water			98.4	72	66		97.9

As in the case of wool, the excellent decontaminability of Cs-137 and S-35 is striking. As expected, in the case of Sr-90 and Ra-226 only Calgon is a usable decontaminating agent. At first sight there is no explanation as to why the decontamination is so insufficient in the case of iodine. It is supposed that iodine was irreversibly adhering to the fibres under the influence of mechanisms as discussed under 2.4.4 and 6.4.1.6.

6.4.4 Synthetic Fibres with Antistatica

6.4.4.1 COLCLOUGH's Experiments

6.4.4.1.1 Contamination

The fibres were contaminated according to two different methods:

1. An aqueous fission product solution was sprinkled onto the fibres which were washed after drying (pH value of the fission product solution 6.0).

2. Starting from a solution of the above mentioned fission products calcium phosphate was precipitated.

The precipitation is then washed and spread onto the fibres as an aqueous suspension. After drying it is also washed.

The prescribed formula follows :

Treatment	Time (min)	Temp. (°F)
2 grams detergent 1 gram sodium carbonate	10	100
0.75 grams detergent 0.75 grams sodium carbonate	10	140
2 grams sodium carbonate 0.2 grams detergent	15	212 (cotton and polypropylene) 140 (acrylic and polyester)
Rinse (water)	4	hot
Rinse	4	cold
Rinse	4	cold
Hydro	4	—
Dry	overnight	122

Composition of the detergents :

“Surfactant” (boundary surface)	23 %
Sodium polyphosphate	37 %
Sodium silicate	10 %
Sodium carboxymethyl cellulose	2 %
Fillers, sodium sulphate	25 %
Other ingredients, EDTA, antifoaming agents etc.	3 %

6.4.4.1.3 Purpose of the Tests

It should be investigated in another study to what extent it would be easier to decontaminate artificial fibres treated with antistatica. The lowering of the charge of the textile surface (measurable by the decrease of the surface resistance) could facilitate the decontaminating process.

6.4.4.1.4 Results of Decontamination Depending on the Type of the Pretreatment and the Textile Fibre

Pretreatment	Type of Contamination	Degree of Decontamination (%)			
		Cotton	Polyester	Poly-propylene	Poly-acryl
Untreated	Solution	94.6	85.5	96.8	98.2
	Suspension	81	97.4	99.9	83.5
Cationic	Solution		98.8	92.6	96.9
	Suspension		96.7	99.7	88.5
Non-ionic I	Solution		90.4	97.0	95.2
	Suspension		95.1	99.3	85.6
Non-ionic II	Solution		93.2	98.9	96.7
	Suspension		95.0	99.1	86.3

A considerable improvement with cationic antistatca has been especially obtained in the case of polyester fibres. The surface resistance has been reduced from originally $1.4 \cdot 10^7$ Megohm/sq.in. to $3 \cdot 10^2$ Megohm/sq.in. (In comparison cotton has a surface resistance of $5 \cdot 10^3$ Megohm/sq.in.)

6.5 Influence of the Test Conditions on the Decontamination

6.5.1 Type of Contamination

In evaluating the above discussed experiments carried out by different authors, the type of contamination is not without importance. In the undermentioned tests referring to this subject TALBOYS and SPRATT illustrate the influence of the different methods which were used, as well as the influence of exterior facts such as those treated in detail in the introductory chapters concerning the radiochemistry of the nucleides in question, in the case of two characteristic nucleides.

Influence of the type of the contamination on the results of the decontamination according to TALBOYS and SPRATT [52].

Contamination	Decontamination with	Degree of Decontamination in %		
		A	B	C
Strontium-90	Versene 1 %	100	100	100
	NH ₄ -Citrate 1 %	100	99.9	98.1
	Demineralized H ₂ O	72	42	92.7
	Tap Water	98.3	36	95.8
Iron-59	Versene 1 %	84	50	93.2
	Calgon 3 %	98.7	100	89.5
	Citric Acid	93.0	97.6	93.8
	Soap	66	32	97.6
	Tap Water	63	21	84

A : Dip the cloth samples into an aqueous solution of strontium chloride or of iron-III chloride and let dry.

B : Precipitation of strontium carbonate or iron-III hydroxide on the fibre.

C : Soiling of the laundry by Sr-89 marked ash or by Fe-59 marked infusorial earth.

6.5.2 Tests with Clean or Dirty Laundry

There seem to be no great differences in the decontaminability of clean or dirty laundry. It appears, however, that in the case of repeated contamination, in certain individual cases [52], the decontamination values decrease with the utilization of soap or detergents.

6.6 Influence of the Laundering Conditions on the Decontamination

6.6.1 Concentration of the Decontaminating Agents

According to COLCLOUGH [43], HUGHES [44], REIFF, SCHUSTER and HEINEN [50], SOROM [51] and TALBOYS [52] the common concentrations of the washing practice also suffice for the complexing agents. NORWOOD [48], on the contrary, used considerably higher concentrations. On the average, the usual concentrations of about 1 % or less may be considered sufficient.

6.6.2 Laundering Temperature

A rise in temperature, according to TALBOYS and SPRATT [52] and STEVENSON [62] is always favourable. The acceleration of the speed of diffusion is always aspired for.

6.6.3 Time of Treatment

The decontaminating effect in the drum washing machine used predominantly, clearly tends towards a saturation value, whereas the wear increases with time. It is, therefore, advisable to manage with about 15 to 30 minutes, if one wants to decontaminate rationally and routinely.

6.6.4 Proportion of the Decontaminating Liquor to Wash Load

The proportion of the liquor to the wash load has only little influence on the decontamination so that it should be possible to utilize the normal proportion of 1 : 6.

6.7 Influence of Special Treatment on the Decontamination

6.7.1 Starching of the Laundry

According to results in the Soviet Union [1], the starching of the laundry does not seem to be unfavourable to the decontaminability since the starch film is removed during the washing process and thus contributes to the removal of the activity.

6.7.2 Antistatica

According to COLCLOUGH [43], the antistatic treatment of fibrous material — especially the treatment of artificial fibres with a high specific resistance — is very favourable to the decontamination.

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II — The Physico-Chemical Effect of the Decontaminating Agents on the Decontamination

1 — INTRODUCTION

Decontaminating Agents.

The composition and type of link of the contaminating substances give indications as to which agents come into question for the decontamination. According to their effect they can be classified in different groups as solvents, detergents, complexing agents and ion exchangers.

2 — PHYSICO-CHEMICAL EFFECT OF THE DECONTAMINATING AGENTS

2.1 Water as Solvent

A simple way to remove soluble radioactive substances from a fabric is their displacement through dissolution in water or in other solvents or their mixtures. In this case, the solid particles are dissolved in liquids wherein they are dispersed down to molecular size, so that the solution itself is equally composed over all. Because of the large dilution of the radioactive particles in the solution, the activity remaining on the fabric is reduced to such an extent that it is already under the maximal admissible concentration of radioactive isotopes and elements.

For the solubility of a substance in a solvent, the presence of certain polar or non-polar groups with certain electric properties is required. The dissolution occurs only when the dissolved material and the solvent contain similar groups. If polar and non-polar substances meet, no molecular combination and no solution occurs.

Water is used as the most important solvent for the decontamination of radioactively contaminated fabrics. The polar water molecules contain free energies in certain spots which are saturated through other molecules of similar structure and explain the dissolution.

In the case of substances which dissociate in water, above all, the so-called "Coulomb attraction forces" are decisive for the dissolution in the dielectricum water. Hydrogen bridges as intermolecular forces may also influence the dissolution.

2.2 Detergents (Tensides)

Due to the large number of radionucleides the use of pure water proved to effect a good decontamination degree only in some cases. In most cases, the radioactive contaminations are either insoluble in water, or hard to dissolve. They are adsorbed to the fabric as insoluble dirt particles. Their removal is considerably facilitated by addition of boundary surface active materials or detergents, as they are also contained in common detergents and cleaning agents.

2.2.1 *Boundary Surface Tension*

As to the effect of boundary surface active, wash active substances, also called "Tensides", the lowering of the surface tension of the aqueous solution of the decontaminating agent should be mentioned above all. This is of great significance if the radioactive impurities are hydrophobic. In this case the boundary surface tension between them and the aqueous solution is lowered, and thus, a faster wetting of the textile fabric and of the dirt particles is caused.

2.2.2 *Electrostatic Repulsion*

The wash active anions are adsorbed to the boundary surfaces and enhance the negative charging of both the fibre, and the dirt particles, causing an increasing electrostatic repulsion and a better removal of dirt. As to the distribution of dirt between the liquor and the fibre surface, there occurs a balance between the removal of the dirt and the resoiling of the fibre. This balance is of great significance if one wants to explain the residual activity on the laundered fibre.

2.2.3 *Anionactive and Non-ionic Tensides*

The boundary surface active agents have at least one group with a good affinity to polar surfaces which most often causes the solubility in water. But they also have a group which has little or no affinity at all to water. In solutions of these substances, the concentration on the boundary surfaces is higher than inside. By this the practically significant physico-chemical properties are explained. Very important for the decontamination of radioactive fibres are the anion and non-ionic tensides. The anion tensides mostly contain a $-\text{COO}-$, $-\text{SO}_3-$ or $-\text{OSO}_3-$ group as a hydrophilic group, and outnumber the non-ionics in application. Several ether, hydroxyl and carbonamide or similar polar groups in the hydrophilic part of the nonionic tensides make possible the solubility of the molecules in water. It should be mentioned that the water solubility of the non-ionic tensides is not due to the dissociation of the electrical charge carriers but to the hydrate cover.

The most important groups of the anion and nonionic tensides are the following :

a) Anionactive tensides :

Carbonic acid salts, as available in soaps and modified soaps

Sulphuric acid esters, as for example

 sulphated oils and fatty acids

 sulphated esters

 sulphated amides

 sulphated fatty acid alcanolamides

 fatty alcohol sulphates

Alkylsulphonates

 simple alkylsulphonates

 sulphosuccinic acid esters

Alkylarylsulphonates

 alkylnaphthalene sulphonates

 alkylbenzene sulphonates

b) Non-ionic tensides

 alkyl polyethylene glycoles

 alkyl phenyl polyethylene glycoles

2.2.4 Decontaminability and Tenside Concentration

It was observed that the decontaminating effect increases with increasing tenside concentration up to a certain limit. This is explained by the fact that the surface tension decreases constantly in order to remain constant at a certain concentration characteristic of every tenside (critical micellae forming concentration).

In addition to this, a certain increase of the decontaminating effect was ascertained for anion detergents. That is to say: the micellae can incorporate other substances which are with difficulty soluble in water and thereby they seem to increase their solubility. This effect, called "solubilisation", is applied in the washing process. The proportion of the number of the tenside molecules to the number of micellae depends on the constitution of the tenside molecules.

2.3 Dirt Carriers

It is true that the tensides proved to be very wash active, but used by themselves without addition of auxiliary agents of laundering they do not give any good laundering effects. In particular, the so-called "dirt carrying power" of the tenside liquor is insufficient because the dirt particles removed by the washing process are in part reabsorbed to the fabric. This reabsorption can be prevented by the presence of the so-called "dirt carriers" of which the sodium salt of the carboxymethylcellulose is the best known.

2.4 Organic Complexing Agents

The addition of further laundering agents became necessary by the observation that in the presence of polyvalent metal ions insoluble precipitants occur in the liquor being deposited on the fabric. This disadvantage was eliminated by addition of organic complexing agents, and in the case of the decontamination of these fabrics the decontaminating effect increased considerably. Organic complexing agents form real complex ions. That can be explained by the coordination theory of WERNER, according to which an electropositive central atom is coordinatively saturated if a certain number of electro-neutral and/or electronegative ligands are directly bound to the central atom. If in establishing the complex a chelate is formed — chelating formation — this complex is exceptionally stable. For the decontamination the so-called "complexones" became important, i.e. nitrilotriacetic acid = NTA and ethylene diamine-tetraacetic acid = EDTA. One mol complexon binds one metal atom, independently of the valence and the special structure of the central atom. Being weak acids, the free complexones dissociate by steps, wherefrom can be concluded that the degree of the complex formation depends strongly on the pH value which ranges from the alkali range to pH 2. Stronger mineral acids decompose all complexonates; in alkaline solutions tri- or tetravalent metals are no longer soluble. According to the rule that the stability of the complex compounds increases with increasing valence of the metal ions, i.e. also with increasing number of the ring structures surrounding the central atom, the metal complexonates of EDTA are much more stable than the respective complexonates of NTA. In strong alkaline solutions the complexones form weaker complexes with multivalent metal ions than does tartaric acid. Similarly effective is citric acid. Due to the presence of these complexing agents, the typical reactions of the metals do not occur and insoluble metal precipitations cannot be formed.

2.5 Polyphosphates

The decontaminating effect of the organic complexing agents is frequently surpassed by chain forming polyphosphates. Besides di- and triphosphates, especially the phosphates with 4 or more phosphorus atoms in the molecule are the ones which produce this effect.

2.5.1 *Structure and Chain Length*

In the polyphosphoric acids the PO_4 -tetrahedrons are arranged in a way that they are bound by forming $-\text{P}-\text{O}-\text{P}-$ bridges. The polyacids have two end-standing hydrogen atoms dissociating only weakly, and a number of strongly dissociating hydrogen atoms depending on their degree of condensation. Characteristic of the individual types of the polyphosphoric acids is the proportion of the oxides $\text{Me}^I_2\text{O} : \text{P}_2\text{O}_5$. In the case of the long chain polyphosphates of the general formula $\text{Me}^I_{n+2}\text{P}_n\text{O}_{3n+1}$ the value of this proportion approaches to 1, in the case of medium and short chains it is between 1 and 2. In the case of technical polyphosphate glasses, the chain length can hardly amount to 100. The average chain length of the "long chain glasses" is around 25 and about 10 for the majority of the commercial special glasses. The chain length is decisive for the properties of the special commercial products which are not exactly defined products, but rather selected melted mixtures. These mixtures are adjusted to definite properties. For example, they can have a certain solubility, a definite pH value, a good buffering ability, or a strong so-called "metal binding power". Other products have good wetting and dispersing properties, or an influence on the turbidity point, or a good power to displace adsorption.

2.5.2 *Solubility*

As to the solubility all short, medium, and long chain alkaliphosphates are water-soluble only with the exception of high molecular Maddrell and Currol salts.

2.5.3 *Buffering Power*

Though able to buffer by themselves special commercial phosphates are buffered additionally. Thereby the pH value becomes different, a fact that should be considered when decontaminating radioactively contaminated fabrics.

2.5.4 *Metal Chelating Ability*

In the case of multivalent cations the chelating ability of the chain-forming polyphosphates is known. If one compares this chelating ability to the one of the organic complexing agents, it must be mentioned that the polyphosphates do not form a complexing link as it is in the case of organic chelating agents, but rather according to E. THILO, work as ion exchangers which exchange the sodium ions for multivalent metal ions. The only difference from the usual ion exchangers is the fact that the exchanger is not a solid matter, but a dissolved substance.

2.5.5 *Dispersing and Peptisizing Effects*

The property of polyphosphates to lower the surface tension of the detergents to a greater extent is closely related to their better decontaminating effect. Along with this goes an increase of the surface activity, of the wetting and of the dispersing effect by which the polyphosphates clearly support the cleaning process. If dissolved alkali polyphosphates meet with insoluble metal compounds, a cooperation of the peptisation, of the dispersion, and of the hydration on the one side and of the dissolution through ion exchange on the other side occurs. It depends on each individual case which one of these two processes is primary or secondary. Thus, for example, polyphosphates — through the ion exchange of the sodium ions with multivalent metallic ions — can prevent precipitations caused by the later ones. If, however, the metal ions are present in quantities exceeding those which can be exchanged by the polyphosphates, the peptisizing effect of the polyphosphates can also prevent the formation of coarse precipitations or at least delay it. Most of the radionuclides available only in small concentrations would be carried within the case of coarse precipitation, and be deposited in part of the fabric. The decontaminating effect would thus be a lower one.

In the washing process one also observes that the dirt carrying ability of the liquor increases through polyphosphates. They form multivalent anions which are preferably adsorbed to solid, i.e. suspended, as well as to liquid, i.e. emulsified, particles. This causes a changing of the electrical charge which accordingly becomes effective in a colloid chemical view. This is also a reason why above all polyphosphates with tetra or higher valent ions were used as an auxiliary laundering agent in the rinsing step. By the electrical charging, the repulsion power of the particles increases wherefrom on the one hand, a suspension stabilizing effect and a better dirt carrying ability of the liquor results, and on the other hand, a readsorption is prevented.

2.5.6 Adsorption Displacing Effect

In the course of washing and rinsing processes it was further observed that through the addition of polyphosphates, the fabrics can be better washed out and the particles adsorbed to the fiber can be better rinsed away. This is explained by the fact that the polyphosphates themselves are preferably adsorbed to the fiber and, in doing so, displace the particles adhering to the fiber.

By a subsequent rinsing with water the polyphosphates which adhere but loosely to the fibre are easily washed out, a behaviour which is taken advantage of in order to improve and shorten the rinsing process.

2.5.7 Lowering of the Turbidity Point

It is easy to understand that big particles are easier washed out through the stream of the wash liquor than smaller particles which adhere more strongly to the fiber. The removal of these latter ones is generally supported by a rise in temperature of the wash liquor. Because of the increased kinetic energy of the water molecules, the dirt particles are all the more strongly affected the more the temperature of the wash liquor increases. It is known that generally the removal of dirt is facilitated by hot washing. Delicate fabrics, however, are damaged by hot washing so that one is compelled to wash or rinse only with relatively warm liquors. At these temperatures, however, turbidities and flaws occur in the soap or detergent solutions which reduce the degree of decontamination. Through the presence of polyphosphates the turbidity point is lowered to such a degree that the washing and rinsing with only relatively warm liquors is possible.

Summarizing it can be said that the polyphosphates participate in many ways in the rather complex washing and rinsing processes. The high degree of decontamination caused by the treatment of radioactive contaminated fabrics with polyphosphates can be attributed largely to the different effects of these substances.

2.6 General Conditions in the Decontaminating Process

The approximate range of conditions under which the decontamination of radioactively contaminated textiles occurs will be discussed. Of particular importance are temperature, concentration, proportion of wash liquor to wash load, and time of treatment.

2.6.1 Temperature

The level of temperature depends on the nature of the fabric (cotton, wool, synthetic fibres) and is in the case of materials sensitive to heat between 40 and 60 °C, in the case of boiling resistant materials between 80 and 100 °C.

2.6.2 Concentration

The concentration of the detergents in the washing liquors is generally around 5 to 10 g/l, and of the complexing agents — for example polyphosphates and complexones — around 2 to 5 g/l. TALBOYS and SPRATT (E.C. USA EC NYO 4990, 1954 [52]) also tested higher concentrations, like for example, 1 to 3 %, and even 6 % solutions.

2.6.3 *Time of Treatment*

Higher concentrations as mentioned above are in general not required, unless exceptionally difficult decontamination is to be carried out. Instead of higher concentrations most often a prolongation of the time of treatment from normally 15 minutes to 30 or 45 minutes is preferred.

2.6.4 *Relation of Wash Load to Washing Liquor*

The above mentioned study by TALBOYS and SPRATT [52] shows that the degree of decontamination also depends on the proportion of wash load to washing liquor. A proportion of 1 : 30 was taken in this study. It should be noted that this proportion does not approximate normal practice. Mostly, one uses a proportion between 1 : 5 and 1 : 1. The results of these tests show that the influence of the proportion mentioned above is only small.

3 — LITERATURE

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III — Evaluation of the Difficulties Resulting from the Liquors Used for Decontamination in the Plants Working Up Contaminated Waste Liquors

1 — INTRODUCTION

Processes for the Treatment of Radioactively Contaminated Waste Liquors.

For the treatment of radioactively contaminated waste liquors the following processes primarily come into question : evaporation, chemical precipitation, biological processes, and ion exchange. It is known that difficulties will occur during the working up of the waste liquor containing decontaminating agents. These difficulties shall be discussed here.

2 — DIFFICULTIES IN THE PLANTS

2.1 Detergents in the Evaporators

The laundry liquor to be evaporated usually has a noticeable content of detergents which involve various difficulties in the evaporators. The detergent solutions mainly have an alkaline or a neutral pH range. In this range all solutions incline to foam considerably so that an evaporation is very difficult. Consequently there is much trouble in the separator and in the compressor. On the other side it is known that evaporation is possible in an acid pH range at pH values between 3 and 4. This is also the case if one adds antifoaming agents. Advantageous for the operation in the acid pH range is that neither boiler-scale nor depositions of mud occur in the evaporator [1].

2.2 Organic Complexing Agents and Polyphosphates in Chemical Precipitation

In plants where radioactive sewage is treated with chemical precipitants the following aspects arose : The great influence of detergents, complexing and chelating agents and not at least of polyphosphates was referred to already under I. and II. of this literature study. These decontaminating agents bring about many a difficulty in the chemical precipitation of the sewage

According to KEHREN [2] the wash active substances, due to their high dispersing power, make the precipitation and sedimentation very difficult. Thus in order to obtain an optimal precipitation of iron they require higher pH values which should not fall below the limit value of 7.5. At pH 7,5 part of the iron remains dissolved and could be precipitated in unwanted places. In accordance with this KRAUSE [3] observed that the chemical precipitation was considerably disturbed by the presence of complexing agents and of detergents. On the other hand, KRAWCZYNSKI [4] found that tensides cannot disturb the normal precipitation in the cleaning process of the radioactively contaminated sewage.

The disturbances due to the tensides are caused by traces of complexing agents like tartaric acid, citric acid, ethylene-diamine-tetra-acetic acid, and other chelating agents which are difficult to precipitate. It is known, for instance, that already 28 ppm ethylene-diamine-tetra-acetic acid lower the decontaminating effect at a pH value of appr. 11. This is also the case in the presence of 42 ppm of Calgon. At this concentration uranyl ions are complexly bound, which explains the known fact that especially in sewages with a very low activity, only an insufficient degree of decontamination occurs in the chemical precipitation [1]. Better results were obtained with a two step precipitation. The first precipitation is carried out with potassium cyanoferrat II and nickel sulphate (about 2 kg/10³ m sewage) at a pH value of 9. In the second step the decanted solution is treated with calcium hydroxide up to a pH value of 9.5 and then with a 50 % surplus of phosphate. Subsequently, 7 — 10⁻⁵ % FeSO₄ are added. Finally, a pH value of 11 is adjusted with soda lye. Referring to the chemical precipitation for the elimination of fission products in sewage, a quotation of a lecture by GOODMAN [6] should be finally cited : "The chemical precipitation has been successful only for the concentration of individual radioisotopes. The difficulty is that the fission products in the sewage water are greatly diluted. It appears, however, that this problem can be mastered to a certain degree through an adsorption of the fission materials to solid materials and through a multi-step precipitation."

2.3 Detergents and Organic Complexing Agents in Biological Processes

If the contaminated laundry sewage contains complexing agents which influence the chemical precipitation and other processes, DOBBINS [7] recommends the application of biological processes. Because of the good adsorptive properties of biological suspensions, biological processes appear to be particularly suitable for the removal of very small quantities of radioactive substances from relatively large quantities of sewage. The removal of the individual cationic fission products depends on their valence, on the concentration of the stable isotopes of the element, and on the amount of elements with similar chemical properties contained in the sewage. The pH value also has a significant influence. Furthermore, radioisotopes with a high valence are better retained than those with a lower one.

The difficulties which arise in biological processes by means of detergents and complexing agents are known. For example, the so-called "hard detergents" cannot be decomposed by microorganisms. This applies especially to the nonionic detergents.

If, however, the detergents are taken up by the microorganisms they increase the BOD. Laundry sewages which contain a particularly great number of organic substances are subjected to a special treatment of aeration with activated sludge whereby the BOD of the sewage is considerably lowered. It was established that the Versenes (on the basis of EDTA) are not assimilated by microorganisms but they rather show the tendency to prevent the bacterial activity. Even though a biological process carried out by itself is mostly unsuccessful it can be taken advantage of as a first step to a chemical precipitation or some other similar methods.

2.4 Complexing Agents and Non-ionic Detergents in the Work Up Processes with Ion Exchangers

Finally, the application of ion exchangers for the treatment of radioactive sewage will be discussed. The waste liquor containing decontaminating and washing agents is generally rich in salt so that the application of ion exchangers will not be very economical in most cases. The costs of the process depend on the content of total solid matters. It is reasonable to work up waste liquors by means of exchangers only with a limited salt content. Further difficulties are caused by the presence of complexing agents and non-ionic detergents so that, in this case, other processes should be preferred.

3 — LITERATURE

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IV — Review of the Known Processes In Order to Evaluate the Favourable Processes with Regard to Application and Economy

The processes for the decontamination of radioactively contaminated textiles are based on the washing processes, which are common to commercial and household laundries. A high degree of economy is obtained by applying the usual processes and equipment. No known process is more economical. The composition of the washing and decontaminating agents used in each individual case depend on the type of fabrics and on the type of radionuclides. In order to obtain a decontaminating effect which meets all requirements, the use of polyphosphates is of great practical significance. Since they are available everywhere in sufficient quantities, the decontamination of radioactively contaminated textiles through polyphosphates will be guaranteed in the best possible practical way.

V — SUMMARY

As can be seen from this comparative literature study, sufficient basic knowledge for the removal of radioactive substances from textile fibres has already been gained. In special instances of decontamination several points can be considered as direct hints and can be taken advantage of in practice. Thus the majority of the studies mentioned above show that — in the case of the mostly used textile fibres as well as in the case of the most important radionuclides — very good results could be obtained by decontaminating textiles by means of polyphosphates or organic complexing agents such as hydroxy-poly-carbonic acids or complexones.

For a general procedure on a large scale, however, a number of special cases or rather of special questions have not yet been clarified. They should be settled in further investigations. These investigations should attempt to clarify the special behaviour of certain textiles fibres and the cloths made from them, as well as the specific behaviour of certain radionuclides.

Special attention should be given to the decontaminating agents or rather to their mixtures in considering the adequate conditions of application. In the radionuclides group an elucidation of the behaviour of Iodine 131 would be of particular interest. In the group of decontaminating agents, the cationactive substances should be studied in detail. Adequate methods for special cases exceeding the general instructions can be elaborated only after extensive studies of this type have settled important questions.

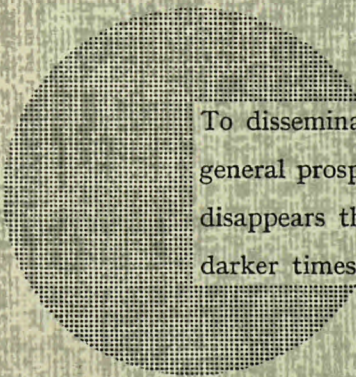
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