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EQUIPMENT DECONTAMINATION
WITH SPECIAL ATTENTION
TO SOLID WASTE TREATMENT -
SURVEY REPORT

J.A. AYRES

June 1971

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EQUIPMENT DECONTAMINATION WITH SPECIAL ATTENTION
TO SOLID WASTE TREATMENT - SURVEY REPORT

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June 1971

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EQUIPMENT DECONTAMINATION WITH SPECIAL ATTENTION
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I. INTRODUCTION

This report is a summary of all pertinent information on methods for decontamination of equipment that has been used at a nuclear installation and has become contaminated with radio-nuclides.

Contamination complicates and limits use of equipment, sometimes to such an extent that it may be of no further value and must be discarded or decontaminated. Contaminated equipment, especially if the activity levels are very high or if the contamination is easily removed, is usually decontaminated prior to storage or disposal, to facilitate handling, reduce exposure levels to personnel, and decrease probability of contaminating the surroundings.

As contamination levels of equipment increase, evaluations must be made to determine whether it is more economical to continue using the contaminated equipment, to decontaminate and reuse, to decontaminate and discard (or store), or to discard (or store) without decontamination. A realistic study of the economics of decontamination and disposal must include a careful consideration of many different factors, including waste disposal, which is assuming an ever greater importance. Final disposal of contaminated solid waste is expensive. Usually it is buried or stored in a controlled area. Any method to reduce the amount of contaminated waste will decrease the overall cost of the operation.

This report attempts to include, along with a discussion of the different decontamination processes, an evaluation of each method from the standpoints of radioactive wastes generated during

the decontamination and the net reduction in volume of solid wastes, with special attention given to those processes that will provide the smallest total volume of solid wastes that must be stored or buried.

No attempt was made to limit the scope of this report from the standpoint of size, design, or materials of construction.

II. SUMMARY

Radioactive waste volumes must be kept to a minimum because disposal of these wastes is very expensive, especially in densely populated areas. One method for reducing volume of solid wastes is to modify or repair equipment rather than discard it. Decontamination may be used to reduce radiation levels on equipment so that it can be reused, modified for a different use, or repaired if necessary for continued use. In addition, a partial decontamination may reduce levels to a relatively low value and permit disposal of the equipment in a simpler and less expensive manner.

The proper choice of a decontamination procedure will depend on many interdependent variables, including: location of the contamination (in deposited dirt, oil, or grease; in the corrosion product layer; or in the material itself), type of contamination, adherence, mechanism of contamination, materials of construction, conditions of contact of contamination (temperature, time), and total history of the specimen.

In choosing the decontamination procedure, one must consider both the material to be removed and the materials of construction. If the equipment is to be reused, the surface must not be damaged to such an extent that the equipment is not fit for reuse or that subsequent corrosion rates are excessive. The procedure must also remove the contamination and prevent it from redepositing. No one

procedure will be effective for decontaminating all equipment, and an installation that relies on only a few procedures for treating all wastes may be the most expensive, least effective, and may create the most solid and liquid waste. Some special procedures may have only limited application, because they require expensive installations or special skills on the part of the operators, but for some jobs may prove to be extremely valuable and worthy of consideration.

Care must be used in designing and equipping the decontamination installation. Areas and facilities must be available for treating all types and sizes of equipment, with separate areas for work with varying activity levels.

Decontaminants may be applied by hand (scrubbing), jetting or vapor blasting, vapor bath, soaking, or soaking with agitation (stirring, moving the work, spraying, or ultrasonics).

If the contamination is in the adherent corrosion product film, the entire film must be removed. Since the radionuclides will be dispersed throughout the crystal structure of the film, and since diffusion rates of large molecules into crystals is so slow, specific radionuclides cannot be leached from the films. The procedure most generally used for removing corrosion product films and scales is by contacting with acids (pickling) in a bath or spray. Agitation accelerates decontamination and reduces possibility of localized corrosion. Electrolysis aids in removing films by scrubbing with O_2 (at the anode) or with H_2 (at the cathode) and by attacking the film at the film-metal interface with some dissolution of the metal (if the work is the anode).

Ultrasonics speeds decontamination, sometimes by a factor of 50 or more, by applying a physical force (cavitation of myriads of bubbles) at the surface of the work. Ultrasonics is especially

valuable for decontamination of intricate assemblies, since the tiny bubbles cause the solution to flow into narrow crevices. The ultrasonic force can be focused in a certain area, and long pieces (e.g., tubes and rods) can be decontaminated by slowly moving them through this area.

Solution volumes can be decreased by diluting the solution with air or gas by foaming, or with steam in steam cleaning or jetting. Large surface areas may be decontaminated with steam jets, strippable films, or gelled decontaminants. Scrubbing provides the greatest concentration of force and chemical or solvent action at the area where contamination is greatest. It also permits decontamination of one or more spots of localized contaminated areas without subjection of the entire equipment to chemical and physical attack, and it generates the least waste volume. On the debit side, each scrubbing is an individual operation and usually requires disassembly of the equipment. It is a very expensive operation since it requires considerable manpower and exposes the workers to high radiation levels. It is generally used only when no other procedure is feasible and when the contamination is localized. The scrubbing action may be accelerated by electrolysis (Johnny Mop technique).

Soaking is the most widely used method of decontamination. It affords the greatest control of variables, such as contact time and temperature, amount of agitation, and solution concentration. It is easily adapted to automation and thus reduces man-hours and exposure levels. Very large pieces cannot be decontaminated by soaking in a tank, since tank sizes and solution volumes would be excessive. Large vessels (nuclear reactors, piping systems, or dissolvers) can be decontaminated by recirculating solutions and rinses throughout the system or by filling, agitating, and draining.

Equipment composed of two or more different metals in galvanic contact cannot be decontaminated by soaking because accelerated corrosion will occur at the galvanic junctions.

Equipment may also be decontaminated by jetting with steam or hot water that may contain detergents or other additives. Steam jetting reduces amount of liquid waste since most of the steam flashes off. If volumes of liquid wastes are no problem, high-pressure water jetting may provide satisfactory decontamination. Abrasive jetting (wet or dry) is effective, especially on flat or nearly flat surfaces (e.g., walls of large vessels), but is generally used as a last resort and usually on equipment that will not be reused. It is relatively slow, removes some metal in addition to the film, causes work-hardening of the outer layer (down to about 30 A° or so), and may cause problems when the equipment is reused (roughened and active surface, imbedded activity, residual abrasive). The amount of metal removed depends on size and type of abrasive, distance from work, angle of jet, air pressure (or centrifugal force), and carrier medium. Abrasive blasting is an excellent procedure for removing contamination that has diffused below the surface. One specific application is the decontamination of concrete by removing one or two centimeters from the surface.

Since disposal of solid wastes is extremely expensive, more emphasis should be given to the development of procedures that will generate low volumes of radioactive residues (spent chemicals, reaction products, etc.). Foams, gelled acids, and strippable films permit decontamination with much smaller volumes of spent decontaminants than may be generated by other techniques. Electrolytic polishing or deplating and ultrasonics increase efficiency and may simplify procedures. Some techniques such as sorption or ion exchange may permit regeneration and reuse rather than

disposal of spent decontaminants.

A field that has received insufficient attention but may prove to be of great value is the development of improved decontaminants. Instead of accepting general purpose reagents for cleaning all types of equipment, the worker should try to formulate new mixtures to improve efficiency or reduce amounts of solid wastes. He has at his disposal a wide variety of chemicals and can prepare any type of solution he may wish: from one that removes only an unmeasurable amount of the underlying metal even after several days to one that can react almost violently and can remove several mils in a few seconds. By choosing the right acids, bases, or salt with additives such as detergents, surfactants, inhibitors, chelating agents, and oxidizing or reducing agents, he can make exactly the reagents that will accomplish the desired results in the most efficient, economical manner.

Use of the wrong type of reagents can prove to be very expensive. Concentrated inorganic salt solutions will generate the largest irreducible waste volumes and should be used only if necessary. Some decontaminants (nitric, formic, citric, hydroxyacetic, gluconic, and oxalic acids, hydrogen peroxide, EDTA, NTA, DTPA, etc.) can be destroyed by chemical interaction ($\text{HNO}_3 + \text{HCOOH}$, $\text{H}_2\text{C}_2\text{O}_4 + \text{H}_2\text{O}_2$), oxidation, radiation, or incineration to give gases and water. Use of such reagents would reduce waste volumes and should be evaluated.

III. ECONOMICS AND SAFETY

Economics of decontamination is extremely complex and difficult to evaluate for two reasons: (1) the required information has so far not been available in published reports, and (2) the ground rules are not clearly stated. The several discussions

of decontamination economics that have appeared in the literature^{1,2} are valuable because they are the only information available and because they show that equipment decontamination has saved considerable money. These discussions, however, are too provincial and therefore are of limited value. The discussions state the net value accrued during the year (dollars saved - dollars spent) but they do not give sufficient detail about the operation. The cost of a decontamination process must include everything concerned with the decontamination: manpower, overhead, cost of the installation, equipment, chemicals, waste disposal, packaging, handling, and storage. The one item that may be the most important and, at the same time, the most difficult to evaluate economically is waste disposal. Therefore, detailed information on waste is required: volume, contamination level, composition, and concentration of liquid wastes; volume, type and level of radioactivity, and composition of solid waste; method of treatment of liquid waste (disposal to crib, evaporation); and ultimate disposal of wastes. If wastes are stored, packaging, shipping, and storage costs and plans for ultimate disposal must be given. Only with all this information available can anyone relate decontamination costs to his own location. For example, disposal costs at Hanford would not be applicable to Pennsylvania or other metropolitan areas; costs of evaporation, storage, and shipping would vary widely. Furthermore, if one is to predict economics of operation for the next 10 years, he must know what the waste treatment costs will be during that period and how possible changes in the ground rules will affect these costs. Some of these changes may make unsuitable a procedure now being used and may make new procedures more attractive. A thorough knowledge of economics is important before anyone can determine the suitability of any decontamination procedure.

If solid waste disposal is very expensive it may be economical to use a complicated procedure to decontaminate a piece of equipment for reuse, even though the cost of decontamination is greater than the cost of replacement. Also, special procedures to reduce solid waste volumes may be worth investigating: purification (by ion exchange, sorption, or scavenging) and recycling of solutions; and use of reagents that are volatile or can be destroyed by chemical reaction, radiation, or heat (HNO_3 , H_2O_2 , $\text{H}_2\text{C}_2\text{O}_4$, citric acid, EDTA).

Somewhat related to the economics of any process are the safety aspects. An unsafe method or one that inherently possesses formidable safety hazards can be very expensive.

With the exception of the detergents, citric acid, and some of the complexing agents, almost all the reagents are corrosive to the skin, some (such as cyanides) are poisonous, and some (H_2 , HClO_4) are explosive. Special ventilation must be provided for processes that produce dust (sandblasting) or vapor (electroplating, ultrasonics, or soaking baths). Carryover, sparking, foaming, gas evolution, and dust generation can all spread contamination.

Safety and efficiency will both be improved by providing suitable facilities. A worker cannot be efficient if he is working under adverse conditions, and inefficient methods are generally unsafe. Tanks should be sized for the work, and work should be separated according to size, method of treatment, and activity level. The facility should have areas for low-level, medium-level, and high-level activity. All these principles are discussed in several articles and books on laboratory design.³⁻¹²

IV. STATUS

An extensive literature on decontamination¹³⁻⁴⁸ has been established during the past 20 years; some of it pertinent to equipment decontamination and some of only minor interest (decontamination of laundry or of personnel). Many of the articles tend to be a recital of successes with no clear delineations of limitations of the procedures tested, but there are many excellent reviews in the professional magazines and books. One of the best sources of information are the publications on chemical cleaning.⁴⁹⁻⁵³ Decontamination and chemical cleaning are identical processes; although the criteria for success may differ slightly. The chemical cleaning industry is more established and ubiquitous. Many of the techniques tried or established in chemical cleaning may be useful for decontamination.

A decontamination procedure must remove contamination from the work piece. The degree of removal is most conveniently expressed as decontamination factor (DF), the ratio of (original activity) / (final activity). It is very important to decide what type or degree of decontamination is necessary. Decontamination to an unnecessarily low level can be extremely expensive, not only because it will require more chemicals, more equipment, and more manhours, but more importantly because it will generate larger volumes of contaminated waste.

The DF required will depend on the purpose of the decontamination. If the equipment is to be cleaned merely to facilitate disposal, perhaps only the smearable activity need be removed, and the DF may be very low, between 1 and 2. If the equipment is to be cleaned to reduce activity levels to permit more efficient operation, the required DF will depend on the contamination level and the degree of access required; any DF will be helpful since it will reduce exposure levels, but usually a DF of 10 to 100

is all that is required. If the equipment is to be cleaned for extended repairs, for removal to a new location, or for disassembly, an even higher DF may be desired; but a DF greater than 100 is often extremely difficult to obtain, especially if the equipment is large or of a complex design. Small, simple pieces of equipment with smooth surfaces can sometimes be decontaminated very efficiently ($DF > 1000$), but most equipment is complex with crevices, convolutions, pitted surfaces, joints, and other complicating features.

A. Mechanisms of Contamination

It is beyond the scope of this report to discuss the mechanisms by which radionuclides contact and become attached to metals, plastics, cloth, or other materials. Many discussions of different aspects of this subject have been published.^{14,52,54-66} Only a very brief summary will be given here.

A typical piece of contaminated equipment will probably have loose residue on the surface (oil, grease, dirt, loose scale, or other foreign material). The contamination may be considered to be located in one of three areas: the loose residue, the corrosion product film (usually an oxide), or the substrate. The contamination may be attached by purely physical forces, may be mechanically trapped, adsorbed, or chemically bound. If it is chemically bound, it will appear in various depths of the corrosion product film and may even be diffused into the substrate. The oxide film is usually crystalline and the contamination may form part of the crystal structure.⁶¹⁻⁶⁷ For example in a magnetite crystal [Fe_3O_4 or $Fe(FeO_2)_2$], the iron atoms may be replaced by ^{60}Co , ^{51}Cr , ^{59}Fe , or some other di- or trivalent ion. On iron alloys (mild steels, stainless steels) exposed to high-temperature aqueous environments, the films are spinels, which are very resistant to chemical attack.⁶¹⁻⁶⁶

B. Decontamination Procedures

If the contamination is loosely adsorbed, it can be removed by washing or scrubbing with a detergent or organic solvent.^{34,55-57} Alkaline detergents* also remove most of the loosely adsorbed material and the included smearable activity, although the decontamination factor is usually low (2 to 4). Washing with dilute detergent solutions has several advantages: the spent solutions can be evaporated to a very small volume; and the solutions are safe to handle and are noncorrosive to the metal substrate.

If the contamination is actually diffused into the oxide layer, as is usually the case, it cannot be washed out or leached out. The only way to remove the contamination is to attack the oxide film (by abrasion or dissolution) and remove it and the contamination from the surface. If radionuclides have diffused into the metal substrate, a high DF can be obtained only by removing a few microns of the metal.

Since the location, chemical state, and mode of attachment for the contamination can vary to such a degree, no one decontamination procedure will be universally applicable. In a decontamination installation, as each piece of equipment is received, someone must decide how this piece is to be treated. The one who makes this decision should have a thorough knowledge of all available decontamination procedures, even though many of these will have limited application.

The design of the decontamination facility should be dictated by the work that is to be treated. If almost all of

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*These reagents are discussed on pages 43 - 45.

the equipment can be decontaminated efficiently by soaking in an acid bath or by sandblasting, the facility may consist entirely of sandblasters and acid baths. However, if many different types of equipment are received, it may not be economical to adopt such a limited approach. The added efficiency, economy, and reduction of solid waste may validate consideration of processes such as ultrasonics, electroplating, steam cleaning, foaming, or use of materials not normally considered.

Decontamination procedures may be considered as modifications of scrubbing, soaking, and spraying.

1. Scrubbing

Scrubbing is the simplest of all decontamination procedures and requires the least outlay for equipment. Equipment in the home and in the laboratory is most often cleaned by scrubbing with a cloth, metal sponge, or abrasive and some suitable detergent or decontaminant.

The principal advantage of scrubbing is that all of the physical and chemical force can be applied to the area that is dirty or contaminated. For example, if a large pump becomes contaminated and tests show the contamination to be concentrated in one or two relatively small areas, this contamination can be removed efficiently by scrubbing, if necessary with relatively corrosive or abrasive agents. Only the dirty or contaminated area is subjected to abrasion or corrosion.

Scrubbing is limited in scope because it is a very expensive procedure. It is applicable only when the contamination is located on a relatively small portion of the overall area. Each decontamination will be a unique operation, which

must be done by hand. Scrubbing cannot be mechanized or done by remote operation and will thus require a large expenditure of manhours. If the radiation levels are high, the high dose rates will require a large number of available personnel to accomplish any amount of decontamination by scrubbing.

An electrolytic modification increases the efficiency or scrubbing.^{68,69} An abrasive or asbestos cloth is wrapped around one end of a metal bar and soaked in a suitable decontaminant, which serves as the electrolyte. The metal bar is made the cathode, the piece to be decontaminated serves as the anode, and the contaminated area is scrubbed with the wrapped end of the bar. The current causes anodic dissolution of the metal right at the contaminated area. This procedure is more efficient than straight scrubbing but is still subject to the limitations noted above. Electrolytic scrubbing (Johnny Mop technique) has been used by Westinghouse^{68,70} and by General Electric Company⁷¹ to decontaminate contaminated areas of large equipment around some of their reactors.

In summary, scrubbing is an effective but expensive method for decontamination and is used primarily as a last resort when other methods are not feasible.

2. Soaking

Soaking is probably the most universal method of decontaminating. It requires the least manhours and the least skill, is most efficient, and is easily adaptable to mechanization and remote control. Once the equipment is in place, many different types of equipment can be decontaminated easily and automatically. Almost all large waste-treatment installations have one or more

soaking assemblies that are completely automatic, some even providing for solidification, packaging, and removal of all contaminated solutions.⁷² Removal of dirt and contamination is principally by chemical action although some physical force may be added by agitation or sprays.

a. Limitations

Limitations or disadvantages to soaking are:

A soaking facility is large and may be complicated. It is difficult or impractical to soak very large pieces of equipment.

Complicated assemblies cannot be cleaned by soaking. Overall treatment time may be longer than for some other procedure such as jetting or blasting.

Any equipment having a galvanic junction cannot be soaked in an acid bath.*

Soaking requires a relatively high investment for equipment and a relatively large work area. If different types of equipment are to be decontaminated and if different types of soils or contamination are present, several tanks may be required to hold the different solutions. Provisions for agitation, temperature control, immersion and removal of equipment, rinsing, drying, addition and removal of solutions, and removal of sludges and waste all complicate the assembly.

The size of the equipment that can be treated in a soaking bath is limited by the size of the tank. An installation with a very large soaking tank may be exceedingly expensive, because the requirements for storage, shielding,

*If the material is to be cleaned to facilitate disposal, relatively high metal losses may be acceptable, and this consideration may be of minor importance.

chemical makeup, heating, and waste removal must all be sized to the tank. Almost all of the equipment requiring decontamination will be relatively small, and provision of a large tank for only an occasional large piece would be uneconomical. Equipment having a very large volume can be cleaned by a modification of soaking; the equipment is filled with the solution and thus serves as its own tank.⁷³⁻⁷⁵ Agitation can be by bubbling, stirring, spraying, or by recirculation of the solution. Nuclear reactors, such as N Reactor, Shippingport PWR, PRTR, the Rheinsberg Power, Reactor, and SENA have been decontaminated in this manner.^{67,76-81} The disadvantage of this method is the very large amount of radioactive waste solutions that are generated and must be treated.⁶⁷ Approximately 90% of the costs may be due to waste treatment.

Soaking is especially efficient for equipment of relatively simple design (such as, beakers, rods, and large-bore tubes). If narrow crevices are present, soaking will not provide adequate decontamination. Diffusion of the chemical into the narrow passages will be so slow that these areas will not be contacted with the chemical for sufficient time to provide adequate decontamination. If the equipment is left in the bath for a longer time to compensate for the slow diffusion rate, some areas may be corroded excessively. Moreover, contamination tends to concentrate in narrow crevices and efficiency of removal is low. Agitation or sprays may help, but are not too effective.

If in any part of the equipment, two different metals are in galvanic contact, localized corrosion may occur to cause deep penetration at the junction. The decontaminant is usually a strong electrolyte at a concentration of 5 to 10%,

and a very effective galvanic cell is formed. The areas can be protected (e.g., by coating) but this increases manpower costs and exposure levels, and is usually not practical for treating contaminated equipment.

b. Modifications

In recent years, several modifications have been tried to increase scope and effectiveness of soaking. These innovations have been used in the chemical cleaning industry for some time and have proved valuable. A real effort should be made to determine to what extent they are applicable to decontamination.

(1) Electrolytic Procedures

The chemical effect can be enhanced by electrolysis. The contaminated equipment is made the anode or cathode and placed in the decontaminant which serves as the electrolyte, and some inert material serves as the other electrode.^{69,82-88} With the passage of current, hydrogen or oxygen gas is generated at the surface of the equipment and provides a lifting action to remove the dirt and scale. Materials that are subject to hydrogen embrittlement should not be made the cathode unless some subsequent operation is provided to remove any occluded hydrogen.⁵¹ Usually the piece to be decontaminated is made the anode, and the surface layer actually dissolves during the electrolysis. Thus not only are the scale and oxide film removed, but also the underlying metal is dissolved to a controlled depth. Materials that are passivated easily (e.g., aluminum) are not usually made the anode, since a

passive oxide layer will form, and the high electrical resistance will effectively stop current flow.⁸⁹ Rates of metal dissolution, cleaning, and gas evolution all increase as current density increases.^{90,91}

Under proper conditions, the rate of attack can be controlled to provide any type of surface from an etch to a polish.⁹¹ If the equipment is to be subsequently discarded or if finish is unimportant, the rate can be high even though an etched surface is formed. Usually, however, it is preferable to set the conditions so that the final surface is smooth. This provides the most uniform removal of material, and also a surface that is less subject to corrosive attack or contamination buildup when the equipment is reused.

In contrast to abrasive methods, electrolytic polishing does not change the character of the surface layers.^{92,93} When contamination is removed by abrasion, the surface layers are left in a work-hardened condition and may be more prone to chemical attack during subsequent use.^{94,95}

The principal disadvantage of electrolytic cleaning or decontamination is the expense resulting from relatively high installation costs, increased handling, and waste disposal. An electrocleaning installation consists of a special bath, electrical connections, direct current source, and special provisions for placement and removal of work. Unless the installation is used routinely, it may not be economical.

The greatest difficulty so far has been in finding a suitable bath composition. Successful electrocleaning requires concentrated solutions, because viscosity of dilute solutions (less than 10% acid or salt) usually is low, and when the viscosity of the electrocleaning bath is too low, throwing power is not sufficient; the areas nearest the cathode become deeply etched and areas further away are not affected at all. In chemical cleaning, a concentrated bath is reused until impurities build up to an excessive level or until the chemicals are ineffective. However, for decontamination of radioactive equipment, if the solution is reused for a considerable length of time, activity levels will build up to such high values that operation will be complicated and restricted. On the other hand, if the solution is not reused many times, chemical costs will be very high and large amounts of concentrated contaminated wastes will be generated and must be treated. Waste treatment and disposal costs could make the process uneconomical. Workers at Karlsruhe⁴⁵ recognized the problem, and in 1967 investigated dilute salt solutions with additions of chemicals that form viscous solutions at low concentrations. So far no practical result of this type of research has been announced. The only electrolytes that have proved useful are those similar to or identical with those reported in the literature for electropolishing or electrocleaning.^{84,91,96}

Still another problem in electrolytic decontamination is control of solution loss. Usually, in handling non-contaminated work, the concentration is adjusted by adding new solution periodically to make up for the loss

resulting from dragout. When handling contaminated material, this is not a desirable method of operation. Dragout, spray, and any other type of liquid loss must be rigidly controlled to prevent spread of contamination. Furthermore, the usual methods of controlling purity of an electrolytic bath (dragout, decantation, skimming) will generate an undesirably high volume of concentrated liquid wastes.

Electrocleaning, as distinguished from electropolishing, utilizes less concentrated solutions (~10%), either alkaline or acidic, and the work may be either the anode or the cathode.⁹⁷ Cathodic electrocleaning has sometimes been preferred because twice as much gas is generated at the work surface and thus provides greater scrubbing action.⁹⁷ Anodic cleaning, or reverse current, is more desirable because, since oxygen is generated, there is less chance of hydrogen embrittlement.^{97,98} Another advantage is that the decontamination may be enhanced by metal dissolution. The current may be reversed periodically, or in some cases, alternating current may be used.⁹⁸

Current density must be regulated, usually by adjusting the voltage.⁹⁹ The current density must be sufficiently high to produce enough gas for the scrubbing effect, but not so high that excessive dissolution and etching occur, especially at areas nearest the opposite electrode. For mild steel or stainless steel, the preferred current density and voltage are 50 to 150 A/ft² and 3 to 6 V.⁹⁹

Inhibitors are sometimes added to the electrolyte to reduce possibility of localized attack and hydrogen embrittlement, but are usually omitted because they sometimes adhere strongly and are not entirely removed by rinsing.^{57,100} Inhibitors may also retard or prevent complete removal of contaminants adsorbed by electrochemical and exchange reactions. Surfactants (surface-active agents) are added to decrease surface tension and produce a foam.^{69,101,102} During electrolysis, gas bubbles formed rise to the surface and cause spattering or spray of the solution. A proper foam blanket traps the bubbles and reduces the amount of spray. Too much surfactant or some impurities that may be present can produce an excessively thick foam blanket. Not only does the foam escape from the tank, but it can cause a residue on the work or can trap hydrogen and oxygen gases and lead to minor explosions.

Stainless steel and lead coupons and pieces of equipment have been decontaminated at NRTS at 100 A/ft².⁶⁹ The samples were made anodic, placed in a 10% sodium hydroxide bath at near-boiling temperature for 1-3 min, removed, rinsed with water, scrubbed (sometimes), dipped in 2N H₂SO₄, removed, rinsed, and dried. In the laboratory, the current was supplied by a 6V storage battery. For decontaminating larger pieces of equipment; up to 4 ft² surface area, current was supplied by welding generators of 200-300 A capacity. The electrolyte was hot (near boiling) 10% NaOH solution with a phosphate added (6 oz NaOH + 1 oz Calgonite^{R*} per gallon of water). Another

^{R*}Registered Trade Mark

solution that appeared satisfactory contained 4 oz NaOH, 2 oz Na_2CO_3 , and 2 oz Na_3PO_4 per gallon of water.

No results are cited for effectiveness in decontaminating laboratory samples, but decontamination of stainless steel mechanisms with lead pigs gave a DF of 1000.

At Saclay,¹⁰³ stainless steel coupons were electro-cleaned in 5, 10, and 20% solutions of HClO_4 , H_3PO_4 , H_2SO_4 , and NaOH. In the NaOH solutions at 75 °C, corrosion was not detectable, but decontamination was inefficient (DF was 2.5 in 50 min and 30 in 4.5 hr). In the acidic solutions, corrosion and decontamination rates varied with time, temperature, current density, and type of acid. Phosphoric acid gave lowest corrosion rates but the decontamination factors were still sufficiently high. For example, at 20 °C with a current density of 24 A/dm², in a 10% solution of H_3PO_4 , the DF after 10 min was greater than 100 and corrosion was 1.6 μ ; in a 5% solution, DF greater than 100 was attained only after 35 min, and corrosion was 4.2 μ .

In electrolytic polishing, the work is made the anode and the conditions are such that high potential areas (projections) are preferentially dissolved to give a level surface. The ions resulting from the electrochemical dissolution accumulate in the immediate vicinity of the anode and then diffuse into the bulk of the solution. A steady-state equilibrium is established between the ions diffusing from the anode to the solution and the ions formed at the anode.^{82,84,92,104} To maintain this equilibrium it is necessary to have at the anode a high-resistance viscous layer. The rate of dissolution of the anode is

controlled by the rate of diffusion of metallic ions through the layer. The solutions used for electropolishing are usually highly concentrated acids (70-100%). One bath recommended for electropolishing copper contains 75% H_3PO_4 , 0.8% Cr^{3+} , 0.3% Al^{3+} , 0.3 Cu^{2+} , balance H_2O .¹⁰⁴ One solution for carbon steel contains 50% HF or HCl , 50% H_2O , 0.2 g/liter gelatin.⁹¹ The baths are usually agitated to keep the viscous layer thin, reduce resistance, and increase dissolution rate. Current density and voltage must be carefully controlled, since electropolishing occurs only when current density is above the value at the critical point.

An electropolishing facility in the waste treatment installation at Saclay⁸⁴ consists of a polishing bath of 700 liters capacity, a rinsing tank, and a selenium rectifier with a rating of 1500 A at 14V; ventilation is sufficient to remove contaminated gases and sprays. The work piece is suspended between two lead or stainless steel cathodes in an agitated solution. The preferred electropolishing solution is a mixture of chromic and phosphoric acids (1300 g. H_3PO_4 and 65 g. Cr_2O_3 per liter) because it is easier to control dissolution rate. Many different types of contaminated equipment (valves, Raschig rings, tanks, pump parts, agitators, etc.) were cleaned in the electropolishing bath at 200 A/ft² for 1 to 20 min. The decontamination factors ranged from 10 to >500; in most cases, no measurable activity remained on the cleaned surfaces.

In summary, electropolishing and electrocleaning are very effective decontamination procedures, especially for pieces of moderate size and relatively simple design. A facility for routine use must be in a shielded area with adequate provisions for ventilation. The manpower requirements for preparation, loading, and removing the work will be high, and the installation will be expensive. However, a well-designed and skillfully operated electrocleaning facility may be a valuable adjunct to a complete decontamination facility.

(2) Ultrasonic Procedures

Ultrasonics is the name given to the vibrations having a frequency greater than the upper limit of audible sound (somewhere between 16 and 20 kHz). The range of interest for ultrasonic cleaning is 20-100 kHz, especially at the lower part of the spectrum.¹⁰⁵⁻¹⁰⁷ Ultrasonics increases the efficiency of a soaking bath in two ways; it provides vigorous agitation and imparts a physical force at the surface of the work.¹⁰⁵⁻¹⁰⁸ Agitation is important during soaking to transport fresh solution to all parts of the surfaces to be cleaned.

Ultrasonic vibrations are produced by providing an alternating current to cause changes in dimension by either ferromagnetic or piezoelectric phenomena. The vibrator may be a barium titanate or lead zirconate block (electrostrictive transducer), a quartz crystal (electrostrictive transducer), or a laminated ferromagnetic sheet or rod wound with wire (magnetostrictive

transducer).¹⁰⁵⁻¹⁰⁸ The transducer placed either in the solution or bonded to the tank produces vibrations in the fluid. These vibrations cause waves of alternate high and low pressures. At the low-pressure cycle, millions of tiny bubbles are formed which break when the high-pressure cycle occurs. This breaking or implosion creates a very high force in a very small area. It is stated that as the bubbles collapse, the pressure can be as high as 10,000 psi and temperature can reach 20,000 °F.^{105,107,109} This cavitation and the resulting agitation of the liquid not only provide a vigorous scrubbing action right at the surface but also force the solution into narrow passages, cracks, and crevices.^{105,106} Thus equipment of complicated design can be cleaned thoroughly, rapidly, and uniformly. In typical examples, cleaning time has been reduced by a factor of 40-50.^{105,106}

By way of caution, it has been stated that ultrasonics will increase the efficiency of a soak bath, but an ineffective solution will remain ineffective even with ultrasonics. Although this is true up to a certain point, ultrasonics can increase efficiency of some solutions to such an extent that they can be used under conditions in which they are normally ineffective. For example, aqueous detergent solutions are very effective when used with ultrasonic agitation at room temperature while they may be ineffective under the same conditions in a soak bath.¹⁰⁶ However, a detergent solution will not remove high-temperature oxide film from stainless steel, even with ultrasonics.

The implosion of the cavitation bubbles occurs preferentially at the surface and forces the soil and scale away.^{106,107} Exposure time and power to the unit must be controlled, because overlong exposures or too much force can produce tiny pits in the metal.¹⁰⁶ Although the pits are very shallow, the surface could become roughened to such an extent that severe contamination problems would occur during subsequent exposure. This may or may not be a problem, depending on subsequent disposition of the equipment.

The transducers are usually flat and can be attached to the tank or can be hermetically sealed and placed in the liquid along the sides and bottom of the tank. The latter arrangement is more flexible since the units can be moved to accomodate special geometries or can be moved from one tank to another as the need arises.¹⁰⁵⁻¹⁰⁷ Small parts may be placed in a glass beaker or jar or in a metal (sheet or wire) basket and placed in the ultrasonic bath. The tanks and baskets are preferably constructed of some hard material, such as metal or glass, since soft materials absorb energy.¹⁰⁶ Metal containers are preferred in industrial baths even though they are less efficient than glass. Thin-walled aluminum or 20-gauge steel baskets can cut the ultrasonic efficiency by 50%, while glass has a much lesser effect.¹⁰⁶ Most commonly, the parts are placed in wire mesh baskets, but the mesh must be finer than 200 mesh or coarser than 1/4 inch. Intermediate screen sizes scatter the ultrasonic waves and reduce efficiency.¹⁰⁵⁻¹⁰⁷

Ultrasonics is used extensively in industry to clean small parts or equipment of intricate design, especially those containing narrow passages or irregular contours,¹⁰⁷

and it seems evident that such a technique can be used advantageously to clean similar components in a decontamination installation.

The use of ultrasonics has been investigated at Saclay,^{110,111} Harwell,¹¹² Chalk River,¹¹³ in several sites in the United States,¹¹⁴⁻¹¹⁹ and in other countries¹²⁰⁻¹²³ with varying degrees of success. It is difficult to evaluate the potential of this method because most ultrasonic decontamination studies have not given the information that is required. From data in published reports, it is not possible to state the optimum conditions (frequency, temperature, power, time, dimensions, geometry, bath composition) for use of an ultrasonic bath and to estimate cost of operation or efficiency of decontamination. Probably the most pertinent information can be obtained from publications of results from non-nuclear installations.

Several books and reviews discussing ultrasonic cleaning have been published^{105-109, 124-133} and these should be consulted by anyone wishing to consider decontamination with ultrasonics. The efficiency depends on several interdependent variables including frequency, bath temperature, solution characteristics, power, tank dimensions, and tank loading.

The ultrasonic cleaning installation consists of 3 principal elements: the generator, which converts low-frequency AC to high-frequency current; the transducer, which changes high-frequency electrical energy to ultrasonic energy; and the bath, containing the cleaner and the contaminated work. The design and size of the

generator determine the power transferred to the transducer. The type of transducer determines the type of cavitation and, especially, the force of the implosion: the tank, solution, and loading must all be designed to provide suitable cavitation force at the work surface. Two types of power generators are used: the rotary electron tube and the (more efficient) solid state. The power output may be half-wave pulsed, full-wave pulsed, or continuous. The transducers, which convert the generator output to ultrasonic energy, may be either piezoelectric or magnetostrictive. The piezoelectric are more efficient converters, but the choice of type will depend on many things, including frequency, type of bonding, and temperature of operation. The cavitation force and bubble size vary inversely with frequency. Therefore it is usually more efficient to operate at a lower frequency, although frequency must be kept above 20 kHz to reduce noise level. The magnetostrictive normally vibrate at a lower frequency (20-22 kHz). The piezoelectric transducers operate at higher frequencies (40-50 kHz) but can be specially designed to operate at a subharmonic (20-22 kHz) with some loss in efficiency. One design generates several different frequencies to provide low frequency for high-energy cavitation and high frequency for production of smaller bubbles for better penetration into crevices.

The size of the tank, the liquid level, type of liquid, temperature, loading and placement of transducers all should be designed for optimum efficiency.^{105-109, 124-133}

When a transducer is placed in a tank filled with liquid, a standing wave is formed, and the cavitation will be greater at certain locations in the tank. If uniform energy distribution is desired, several different transducers must be placed throughout the tank, or specially designed transducers must be used. One special transducer produces the main frequency and frequencies 3 kHz above and below the main frequency. The nodes occur at different areas and smooth out the energy distribution. Conversely, it may be desirable to concentrate the energy at certain areas to obtain greater cavitation force and place the work at these areas. If desired, the energy can be focused at one area, and the work can be moved slowly across this area to obtain maximum cavitation force along the entire length.¹⁰⁶

Temperature has a complex effect on cleaning efficiency. The solvent effect increases directly with temperature, but cavitation force increases with temperature to some maximum usually about 15 °C below the normal boiling point and then rapidly decreases.¹⁰⁵⁻¹⁰⁷ The cavitation force depends to a large extent on the pressure inside the bubble. If the vapor pressure of the liquid is too high, the bubbles break before very high pressures can be attained. Since cavitation force is one of the most important effects in ultrasonic cleaning, the optimum temperature will be close to 15 °C below the boiling point of the liquid.^{105,106}

The average power intensity in an ultrasonic cleaning tank is calculated by dividing the average power of the transducer(s) by the tank cross section and is expressed in W/in^2 (or W/cm^2). At low energies, no cavitation occurs and there is no cleaning. As the energy increases, a value is reached at which cavitation starts. This level is called the "threshold of cavitation" and is a characteristic of the liquid being used and varies with both frequency and temperature. For water, the threshold of cavitation is 2.5 W/in^2 ($\sim 0.4 \text{ W/cm}^2$) at 20 KHz and room temperature; at 400 KHz, ten times as much power is required.^{105,126} Above the threshold, amount of cavitation increases with power up to a limit.¹⁰⁵ At very high power levels, cavitation force is concentrated at the face of the transducer and is, therefore, relatively ineffective.

Each transducer will have a natural resonant frequency and will operate at maximum efficiency if the frequency of the activating voltage matches this resonant frequency. The acoustic resonance of the ultrasonic cleaning tank depends on chamber dimensions, liquid level, loading, liquid characteristics, and temperature, and should match the resonant frequency of the transducer.¹⁰⁵ Any change in loading, liquid level, or characteristics of the liquid may change the acoustic resonance and decrease efficiency. When this occurs (as it often does), the unit should be tuned to bring it back into resonance and increase efficiency. Many installations are equipped with automatic tuning to ensure maximum efficiency.¹⁰⁵

With this information at hand, any investigation of ultrasonic decontamination should start with the system at maximum efficiency: properly tuned system, frequency 20-22 kHz, temperature 15-20 °C below boiling point, sufficient power to operate well above threshold of cavitation, and equipment designed to provide sufficient energy at work surface. The solution must be one that will remove the film or scale with the accompanying contamination. In industrial ultrasonic cleaning installations, the most popular solutions are alkaline detergents and organic solvents. These may be of only limited value for decontamination.

At Saclay,¹¹¹ coupons were exposed in 0.1 N H₂SO₄ for two minutes at temperatures of 25 to 69 °C and at intensities between 2.5 and 40 W/in²; frequencies varied between 16 and 3000 kHz. Ultrasonics increased rates of decontamination of mild steel by factors up to 235 or greater. Similar results were obtained in decontamination of copper and tin in solutions of 0.1 N H₂SO₄, 0.1 N HNO₃, 0.1 N (H₂SO₄ + HNO₃ + HCl), and 0.1 N H₂SO₄ with additions of copper and lanthanum sulfate.

At Harwell,¹¹² irradiated Magnox fuel elements were decontaminated in a citric acid bath with ultrasonic vibration furnished by six transducers operating at a frequency of 40 kHz with a pulsed output averaging 1 kW with a peak of 4 kW.

In the United States, small components placed in baskets were decontaminated in a 250 kW ultrasonic bath using a 300 kHz quartz transducer. Heat exchanger

pipng was decontaminated using: (a) a 120 W quartz transducer operating at 300 to 1000 kHz; and (b) a magnetostrictive transducer operating at 19 to 21 kHz. As might be expected, the magentostrictive transducer was more effective. At another installation,¹¹⁶ the ultrasonic decontamination tank consisted of a large-diameter pipe, closed at one end, and mounted vertically. A ring of transducers placed around the upper end provided the ultrasonic energy. Contaminated fuel elements were lowered into the tank past the transducers so that all parts of the fuel rods were exposed to the zone of maximum energy. Ultrasonic energy was used as an aid in decontamination of stainless steel pressure differential detector wells.¹⁰⁵ The wells were decontaminated first in alkaline permanganate and then in a proprietary solution. Comparative tests showed that ultrasonics decreased cleaning times by a factor of four.

At Chalk River,¹¹³ ultrasonic decontamination tanks are used routinely in the waste treatment and decontamination facility.

In summary, ultrasonic cleaning is a technique that can be used advantageously for decontaminating certain types of equipment. It is rapid and easily adapted to mechanization and remote control. It will handle only a relatively small portion of the total volume, but will simplify cleaning of small and intricate parts. The principal disadvantages are that the installation is relatively expensive and it requires a certain skill for operation.

(3) Foams and Coatings

Vessels and other receptacles of large volume cannot be economically decontaminated by soaking with a solution. Chemical costs, storage and transport of solutions, and (especially) treatment and disposal of the large quantities of contaminated liquid wastes will be too costly. Three modifications of soaking that are useful for treating tanks, pipes, or other equipment with large surface areas and volumes are steam cleaning, foams, and coatings (gels, strippable films).

The volume of decontaminant can be reduced by diluting with air, inert gas (such as nitrogen) or steam. A large vessel may be cleaned by a static procedure in which a small amount of solution is vaporized by heating to an appropriate temperature. Piping systems may be cleaned by a dynamic procedure; the acid (or other solvent) is aspirated into the steam which then travels throughout the piping system and is condensed at the outlet. This procedure has been used to passivate and clean large vessels and long pipe runs.¹³⁴⁻¹³⁶ Pilot plant tests have shown that such procedures effectively decontaminate metal surfaces (DF > 60) with very small solution volumes.¹³⁷

Foamed acids have been used for chemical cleaning for over ten years.^{134,138,139} A patent describes a method of forming and using foamed acids, especially inhibited HCl.¹⁴⁰ A surfactant is added and a gas (usually an inert gas, such as nitrogen) is blown through the liquid to form the foam and push it through the system.

As the foam breaks down, the liquid formed is collected and returned to the generator or discarded. One gallon of inhibited acid can form 20-50 gallons of low-density foam that can be easily transported throughout a piping system or used to fill a large tank.¹³⁸ The foam must be stable enough that it remains on the surface an adequate length of time to accomplish the desired amount of cleaning, but it must not be too stable or it cannot be broken down for disposal.

Some of the earliest attempts to use foamed alkaline and acidic solutions for decontamination were made at ANL in 1960.¹⁴¹ Samples from a loop were conditioned with a foamed alkaline permanganate solution (0.5% KMnO_4 , 10 % NaOH, 0.2% detergent) and decontaminated with a foamed solution of 10% ammonium citrate with 0.2% detergent. Several foaming reagents tested in the dilute alkaline permanganate at 25 °C were sufficiently stable to be of interest (foam half-life was 15-54 min.).

The only extensive study to evaluate foamed chemicals for decontamination was a laboratory investigation at Saclay in 1969.¹⁴² The solution was placed in a large glass vessel and foamed by bubbling nitrogen through at a controlled rate. The gas forced the foam through a vertical column past the contaminated sample and to a centrifuge where the foam was broken down to form a liquid which flowed back into the main vessel. The decontaminants tested were 1 M, 3 M, 6 M HNO_3 , 0.05 M and 0.15 M citric acid, and 1 M NaOH with a nonionic or amphoteric foaming agent at a concentration of 0.6% stabilized with an amine oxide.

With an acidic solution (1 N HNO_3 + 6 g/liter Solusol^R), an airflow at 20 °C was regulated to generate a foam at a rate of 1.5 liters/min. The acid consumption was 20 cm^3/min (foam factor of $\frac{1500}{20} = 75$). The flow past the sample was 3.7 cm/sec and temperature was regulated at 60 °C. Simultaneously an identical sample was soaked in a 1 N HNO_3 solution at 60 °C to compare decontamination rates in soaking and foaming. After 1 hour, soaking was more efficient (DF foaming = 4; DF soaking = 5), but after 12 hours, the decontamination factor for both soaking and foaming was 200. The same trend was noted for 3 N and 6 N HNO_3 except decontamination rates were higher. The decontamination factor increased slightly with increased foam flow rate.

The dilute citric acid solution (10 g citric acid + 6 g Arment^R per liter) was much less efficient: after 1 hour, DF foam = 2.5, DF soak = 3; after 40 hours, DF foam = 16, DF soak = 22; and after 50 hours, DF for both was 25. With a solution of 30 g citric acid + 6 g Arment^R per liter, almost identical results were obtained.

Alkaline solutions were prepared by adding 6 grams Arment^R to a 1 M NaOH solution. After 1 hour, DF soak = 3.0, DF foam = 1.2; after 25 hours, DF soak = 3.3, DF foam = 2.6. Decontamination factors were low, and soaking seemed superior to foaming. The low results were attributed to presence of CO_2 which changed NaOH to Na_2CO_3 .

In a large-scale test, the laboratory conditions were duplicated with larger amounts of foamed solutions to clean a stainless steel tube 2.5 in. diameter by 14 ft long. The pipe was treated with foamed 1 N HNO_3 for 17

hours to give a DF of 4; a fresh solution of foamed 1 N HNO_3 after 8 hours increased the DF to 14. The pipe was then treated with foamed 1 N NaOH for 2 hours to increase the DF to 100. After a final treatment with foamed 1 N HNO_3 for 3 hours, the DF = ∞ (no detectable activity remained).

The results reported so far can be considered only indicative, but they do show that foamed acids can be used to give decontamination factors almost as good as or equivalent to those obtained by the standard soaking procedure. More information is required with use of more efficient decontaminants and for materials other than stainless steel. Since the use of foamed acids might be a simple, economical method for decontaminating large vessels or piping system, additional exploratory work is warranted. Recently many new surfactants have been developed, and it is reasonable to suppose that satisfactory foam solutions can be prepared with the decontaminating solutions that are now being used for soak baths.

Acid or alkali solutions can be gelled by addition of small amounts of a suitable reagent such as hydroxyethyl cellulose, and this gelled solution can be applied to the surface by brushing or spraying.¹³⁸ After a suitable exposure time, the gelled solution can be removed by spray rinsing. This is a standard procedure for cleaning interiors of large tanks or other large surfaces of equipment or buildings. The acid may be applied by spray nozzles that are hand-held or mechanized (robot).¹³⁸ At one reactor site,¹⁴³ a gelled solution containing NaOH and NaCN was used to remove radioactive silver from a stainless steel surface.

Some decontaminating solutions can be applied, allowed to dry, and then stripped from the surface. The reagents contain a filming agent, a complexing agent, an acid or alkali or salt, and a wetting agent.¹⁴⁴ Such a reagent (polyvinylalcohol, 2-10%; EDTA, 1%; Na_2CO_3 , 0.02%; glycerine or ethylene glycol, 1-2%; remainder water)* was used to decontaminate different types of surfaces: PVC, polyethylene, glazed ceramic, aluminum, chrome-plated, steel, glass, resin, and painted wood.¹⁴⁵ The solutions were applied by swabbing or as an aerosol spray, allowed to dry, and peeled off. The films were easily removed from all but the most porous specimens (e.g., wood). Decontamination factors were all about 10 and compared favorably with those obtained by soaking.

In summary, foamed solutions, gelled solutions, and strippable films appear to be promising methods for obtaining satisfactory decontamination factors with small amounts of decontaminants. The volume of liquid waste is only 1 to 2% that obtained by soaking procedures. Gelled solutions and strippable films are also satisfactory reagents for decontaminating hot spots or selected areas. Strippable films are especially attractive since no liquid wastes are generated. The only waste is a solid film that can be reduced to a much smaller volume by incineration, if desired.

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*See Sec. IV.C.1 for description of similar reagents.

3. Spraying

Spraying is an efficient method for transporting the solution or solid to a desired area. A logical differentiation for discussion is: solution spray and abrasive spray.

a. Solution Spraying

All types of solutions can be used for spray decontamination, ranging from water or water + detergent to concentrated acids or alkalis. Almost all soak baths have jets for spraying decontaminant or rinse water or both. Spray not only forces the solutions into all narrow passages and all other parts of the surface but also adds a physical force to increase the solvent effect. Spraying with deionized water is the most efficient rinsing method. In many facilities, the spray jets accomplish a major part of the cleaning.

Solutions can be sprayed at any temperature up to boiling. One modification that has proved useful is steam cleaning with detergent solutions.¹⁴⁶ A hot detergent solution mixed with some steam is forced through a jet under considerable pressure to give a coarse high-volume spray (up to 600 gal/hr) at pressures of 60 to 150 psig. Approximately 90% of the steam vaporizes when the solution comes out of the jet, and temperature drops rapidly. At the work surface, it is approximately 80 °C. A dilute solution (0.04 to 0.08% detergent) is used for a very short time (2 to 10 sec contact time) so not much detergent is required, approximately 1 oz per 100 sq ft of surface. Evaporation reduces amount of liquid waste to a relatively small volume. Both high turbulence on impact and high temperature aid in cleaning. The detergent must provide enough foam to act as cushioning agent.

for recoil of the steam gun but must not produce so much that residual foam dries on the surface and requires rinsing. Many portable and installed steam spray facilities are available.¹⁴⁶

A specially designed high-pressure (90 psig) spray has been successfully used at Chalk River for decontaminating caves, walls, and equipment.^{147,148} The steam gun has a narrow, flat orifice and is held closely against the work to take advantage of the full force of the steam. Essentially the steam gives a concentrated force to scrape off the contamination. The detergent is fed at a rate desired (usually 4 gal/hr) by the operator using a manually controlled valve. Most of the solution vaporizes, so liquid waste volumes are very low.

Water at very high pressures can be used to clean equipment and to remove contamination, especially if the contamination is present in loose scale or crud.¹⁴⁹⁻¹⁵¹ High-temperature films are less easily removed. At Savannah River, jets delivering water at 400 psi and 3000 psi were used to decontaminate parts of a process heat exchanger. All smearable contamination was removed, with overall decontamination factors ranging from 3 to 50. This may be a useful method for some pieces of equipment, if the large volumes of liquid waste do not present a problem. It may be possible to recirculate and reuse the water, since no chemicals are used. Hill discusses equipment, procedures, applications, and limitations of high-pressure steam and water sprays in greater detail.¹⁵²

b. Abrasive Spraying

Abrasive blasting is essentially mechanized scrubbing in

which a stream of abrasive material is driven by high-pressure water, centrifugal force, or compressed air to impinge against a surface at high velocity. The physical force depends on the blast force at the surface of the work and the type of abrasives. For different applications, soft abrasives (cracked nut shells, ground corn cobs), glass beads, metallic abrasives (pellets, grit, cut wire), and sand and mineral abrasives have all been used.^{153,154}

Dry blasting is the simplest, cheapest, and most versatile method. For non-nuclear applications, sand is the most commonly used abrasive because it is cheap and need not be recovered. One disadvantage, especially in cleaning contaminated equipment is the generation of fine dust. Blasting must be done in an area with adequate ventilation and provisions for handling radioactive dust, and workers must be adequately protected. A modification to reduce this problem includes a vacuum system around the jet (Vacu-Blast) to collect the contamination and spent abrasive.^{155,156} Dry blasting is satisfactory when no other method is applicable; e.g., for removing a considerable amount of material from concrete floors, walls, or equipment. Vacu-Blast equipment has been used to remove concrete to a depth of several centimeters after it has been contaminated with radioactive solutions.^{155,156}

Centrifugal blasting, in which the abrasive is propelled by centrifugal force, has largely replaced air blasting.¹⁵⁴ Formerly centrifugal blasting was used only for cleaning smaller pieces, because so much handling and positioning were required to direct the blast to all areas of larger equipment. Recently developed equipment overcomes this

difficulty and the procedure can be entirely mechanized.¹⁵⁷ Centrifugal blasting is more economical, but the equipment cannot be moved easily and, hence, is less versatile.

Wet blasting techniques can be used to remove films and contamination without too much metal loss. If one chooses and maintains proper operating conditions, he can maintain a smooth surface.^{154,158} If greater penetration is desired, this can be easily obtained by adjusting the operating variables. For example, with a 3/16-in. jet with 1 lb 20-40 sand per gallon of water, the penetration rate can be varied from negligible (pressure = 1000 psi, jet standoff = 6 in.) to 0.6 in./min (pressure = 3000 psi, jet standoff = 0.5 in.)¹⁵¹ Dusting is not as great a problem as in air blasting, and ventilation requirements are less rigid.

More detailed descriptions of special equipment available, applications, limitations, advantages, and hazards are found in the literature and in manufacturers' bulletins.^{154,159}

Abrasive techniques are valuable when soaking procedures are not feasible (surfaces of very large tanks, reservoirs, buildings, or installations), when a large amount of material must be removed (concrete floors), when contamination is not easily dissolved away, or when isolated areas must be cleaned and metal loss is no problem. It should be noted, however, that gelled solutions or strippable films may be applicable in some instances and may be preferable to blasting.

The disadvantages of abrasive blasting are:

- o Dust (especially with dry blasting) or sprays may create ventilation problems and hazards.

- o Abrasive material may not be removed completely and create difficulties during subsequent operation.
- o Contamination may be driven into the material, into cracks or narrow passages, or peened over and result in low decontamination factors. Furthermore, the rough surface may make subsequent decontamination very difficult.
- o With rough abrasives, final surface may be etched or rough, and this may favor contamination buildup and complicate subsequent decontaminations.
- o Outer surface layers are mechanically worked and may be prone to accelerated corrosion; etching may also cause accelerated corrosion.

C. Chemicals

Chemicals of interest include those that are used for washing degreasing, metal cleaning, pickling or acid dipping, chemical polishing, etching, electrocleaning, electropolishing, or decontamination.^{91,96,160-165}

Any discussion of suitable decontaminating reagents will be confusing unless the criteria are clearly defined. One may list as possible criteria:

1. The reagent must remove the contaminant.
2. It must prevent the contaminant from resorbing.
3. It must not attack the metal (or other substrate) at an excessive rate.

The first two criteria are obvious. Criterion No. 3 must be further defined for the particular sample that is to be treated.

If the equipment is to be cleaned to facilitate disposal, corrosion per se is not important, but corrosion will deplete the reagent and thus create additional liquid waste. Therefore, corrosion should be kept in bounds to reduce costs of chemicals and waste disposal. If the material is to be cleaned for maintenance, repair, and reuse, corrosion must be such that the piece is not damaged or subsequent life is limited. This will be especially important if some of the equipment has thin walls or if tolerances or surface finish are to be maintained.

A choice of a suitable decontaminant requires a knowledge of the nature of the contaminant, the nature of the contaminating surface, and the history of the sample.

If the contaminant is located in the loose material deposited on the surface (scale, dirt, oil, grease), it can be removed by soaking, spraying, or scrubbing with water or with a dilute detergent solution. If the contamination is located in the adherent film or scale, the entire film must be removed to effect any appreciable decontamination.

For the purpose of this discussion, reagents will be classified as cleaners and film removers. Cleaners are reagents that remove dirt, grease, oil, loose scale, and similar materials but do not attack the adherent corrosion product film or the underlying metal. Defilmers are reagents that chemically attack and remove all film and scale, including the adherent corrosion product film, and may even dissolve some of the underlying metal. Some defilmers (e.g., the chemical polishing agents) are very aggressive and may etch off appreciable amounts of metal surface. These are used in the relatively uncommon situation where the radioactive contaminant has diffused into the metal or is mechanically entrapped below the surface (e.g., as by peening). Aggressive

reagents may also be used for rapid decontamination if metal loss is not an important consideration.

1. Cleaners

In decontamination, cleaners may serve three purposes:

- o As a preliminary treatment prior to defilming to remove grease, oil, and other materials that may decrease efficiency or shorten useful life of the defilming bath.
- o For partial decontamination, especially when most of the undesirable activity is in the deposited foreign material.
- o To remove smearable contamination and thus facilitate handling and storage.

Although the use of a cleaner prior to defilming appears to complicate decontamination, it may reduce the solid wastes. Cleaners usually have a low solid content and may be recycled or reduced to small volumes.

Two types of cleaners that may be useful are organic solvents and aqueous detergent solutions.

Detergents are usually surface active agents (surfactants)*. The mechanisms and factors affecting detergency are discussed by Schwartz and Perry.¹⁶⁶ Essentially, the detergent forms with the dirt or grease a compound that has no attraction for the metal surface. The dirt detergent complex is hydrophilic; i.e., it preferentially goes into the aqueous phase, either dissolved as an ion or suspended.^{167,168} The mechanism is primarily adsorption with an equilibrium being established

*Surfactants are discussed on pages 47 and 48.

between the concentration on the metal surface and the concentration in the solution. A clean surface placed in a contaminated detergent solution will become contaminated, and rate and degree of contamination will depend on the concentration of the contaminant in the bath. Conversely, the rate of decontamination will depend on the level of contamination of the work and of the bath and will decrease with time. Agitation will increase the rate of removal, but will not increase the maximum amount of activity that can be removed. The detergent concentration is usually low (0.01 to 0.05%). Other materials (builders) may be added to increase efficiency by removing certain ions or by providing proper pH. Commercial detergents are complex mixtures and may contain substances that are of questionable value and may actually decrease decontamination efficiency (fluorescent brightening agents, foam stabilizers, blue dyes, corrosion inhibitors).⁵⁴

At the Atomic Weapons Research Establishment, Aldermaston, England, two "built" detergent were prepared:

Detergent N10 ^{144,169}			
Lissapol ^R (non-ionic wetting agent)	1.0	}	Parts by Weight
Sodium carbonate	1.2		
Sodium tripolyphosphate	2.0		
Sodium carboxymethylcellulose	0.1		
EDTA	0.5		

The reagent is used at a strength of 0.5 to 1.0%; pH of the working solution is 9.5.

Solution SDG 3^{169,170}

Comprox ^R A (anionic wetting agent)	1.5	} Parts by Weight
Sodium sulfate	2.5	
Sodium carbonate	0.6	
Citric acid	2.0	
EDTA	1.0	
Water	1.0	

This solution is used at a concentration of 0.8%. The dilute solution is slightly acidic (pH = 3) and is used preferably at 70 to 80 °C. It should probably be classified as a film stripper rather than as a detergent since it can attack the base metal.

Both reagents can be thickened by addition of sodium alginate to form a cream that can be spread on large surfaces, such as vertical walls, and subsequently washed off. A strippable film may be prepared by blending either of these reagents with rubber latex.

The organic solvents are used, either in soak baths or in vapor-phase cleaning apparatus, to remove oils, greases, and other organic matter from the surface of the work.⁵¹ Typical organic solvents are trichloroethane, trichloroethylene, petroleum ether, hexane, perchloroethylene, and tricresyl phosphate.

2. Defilmers and Descalers

The film removers used in decontamination are often complex mixtures containing some combination of the following ingredients: a surfactant, as a wetting agent, foamer, or detergent; one or more acids, alkalies, or salts to dissolve the film; a complexing agent to prevent reprecipitation or resorption; an inhibitor to prevent excessive attack on the

metal; and other special additives, such as oxidizing or reducing agents, gelling agents or foaming agents. Sometimes one component may serve several purposes; an acid such as HNO_3 will also be an oxidizing agent, and a surfactant may be both a detergent and a wetting agent.

The alkaline defilmers are not used to any great extent in decontamination. Alkaline complexing agents are sometimes used alternately with strong acids to clean materials, especially stainless steel, heavily contaminated with a wide spectrum of radionuclides. Some alkaline solutions are used for electrolytic decontamination.

Most of the defilmers used in decontamination are acid solutions ($\text{pH} < 5$), although neutral ($\text{pH} = 6$ to 8) complexing solutions have recently proved valuable and are being used to a greater extent.

The acidic and neutral defilmers preferentially attack the scale and film at the film-metal interface. The film is first removed and then dissolved. For example, a heavily filmed carbon steel sample may be completely clean in 15-30 min, but a considerable portion (10 to 30%) of the film may remain as a residue and not be entirely dissolved until after several hours or even days. If the decontamination is done in a large vat, this may not be important since the undissolved film and scale are dense and will sink to the bottom of the vat as a sludge. If the equipment serves as its own tank or if the decontaminant is recirculated through a piping system, undissolved material may deposit in crevices or other sinks and not be completely removed. Decontamination factors will be low, and the deposited material may cause corrosion problems.

Certain minor constituents (additives) are important in the formulation of an effective defilmer. The principal types of additives are surfactants, complexing agents, and inhibitors.

Surfactants are discussed thoroughly by Schwartz et al.¹⁷¹ and more briefly by Pollack and Westphal.¹⁷² A surfactant is a compound that decreases the surface energy of a solvent to an extreme degree, even when it is present in very small amounts (0.01 to 0.04%). Surfactants usually consist of long hydrocarbon chains with one or more polar groups at or near the end of the chain. The hydrocarbon chain may be alkyl, aryl, or a combination, and the polar group or groups may be carboxyl, sulfoxyl, etc. The hydrocarbon portion is attracted to the organic phase (oil, grease) and is repelled by the water (hydrophobic), while the polar group is attracted to the water phase (hydrophilic). The commonest type of surfactant is soap, which is usually a salt of a fatty acid and has as the hydrophobic group a long unsaturated alkyl chain and as the hydrophilic group a carboxyl ion. Surfactants serve many purposes: they may increase the attraction of the solvent to the surface (wetting agents); they may form hydrophilic groups with oil or dirt (detergents); they may decrease repulsive forces between two solvents (emulsifying agents); or they may act as foaming agents. The structure determines the characteristics of the surfactant, and any one property may be improved by judiciously modifying the structure. The best detergent is not the best foaming agent, and some foaming agents have very poor deterrent properties. A surfactant with a short carbon chain may have good wetting powers but be worthless as a detergent.

Surfactants may be ionic or nonionic; in aqueous solutions the ionic are the most important. Ionic surfactants are further classified as: anionic (carboxylic acids, sulfates, sulfonic acids, phosphates, persulfates, sulfamic acids, sulfonamides); cationic (amine salts, quaternary ammonium compounds, phosphonium compounds, sulfonium compounds); and ampholytic (amino and carboxy, amino and sulfuric ester, amino and sulfonic acid).

Complexing agents are added to prevent reprecipitation or resorption of radionuclides. The most pertinent illustration is the use of a complexing agent with oxalic acid. Oxalic acid readily removes film and scale from iron, but an insoluble ferrous oxalate forms as a result of hydrolysis and precipitates on the surface. Citric acid added to the oxalic acid complexes the iron and reduces ferrous ion concentration to such a low value that the oxalate precipitate does not form. Other agents (e.g., EDTA, DTPA) that form strong complexes with ferrous ion at the operating pH may also be effective.

Certain complexing agents, called chelating agents, have two or more complexing groups that form stable 5- or 6-member rings with metal ions. The best known are the iminodiacetic acids such as NTA, EDTA, DPTA, and TTHA, which form complexes with positive metal ions to form negative metal complex ions:

The metal ion is essentially removed from solution and will not precipitate even when the pH is at a value where the hydroxides are normally insoluble.

The iminodiacetic acids are very important in decontamination because they form strong complexes with ions of many metals used in construction of equipment (Fe, Cr, Ni) and with many of the radionuclides (including Ce, Zr, Pu, U, Th, Co, and La) that form hydroxides with a very low solubility product. Thus, it is possible with the iminodiacetic acids to keep these ions in solution even at a relatively high pH. The stability of the (metal)-(iminodiacetic acid) complex increases with molecular weight. Thus, the following stability series holds:

Since the stability constants for M·TTHA and M·DTPA are much higher than that for M·EDTA, TTHA and DTPA will keep iron in solution at a higher pH and are much more useful for formulating neutral decontaminating agents. Unfortunately, TTHA is not produced in large quantities in the United States,¹⁷⁴ but DTPA is readily available. The M·DTPA complex has more residual COO⁻ groups than has M·EDTA and is more soluble at all pH values.

The chemical decontaminants, and especially the acidic solutions, must be inhibited with some compound that limits the attack on the metal substrate and at the same time does not prevent or excessively slow down the removal and dissolution of the film and scale. Suitable inhibitors for acid solutions include propynol and some of its homologues, formaldehyde, mercaptobenzothiazole, and many proprietary compounds and mixtures. Fontana and Greene list approximately 100 inhibitors with specific applications and references.¹⁷⁵ The efficiency of the inhibitor is usually related to its ability to adhere to the metal, and residual amounts of inhibitors are not removed completely by rinsing with water. Usually this does

not cause any deleterious effect, but the fact should be recognized.

Decontaminating agents may be either prepared by the laboratory personnel or may be purchased as proprietary reagents from some chemical corporation (Wyandotte, Turco, Dow, etc.) that supplies these materials.^{160,161} Purchasing proprietary compounds has advantages and disadvantages; but if proper precautions are taken, it will probably be more satisfactory to buy the proprietary decontaminants rather than attempting to mix your own. Many of the reagents are solids, and trying to blend several solid chemicals to obtain a homogeneous mixtures requires skill and proper equipment. This is especially true when small amounts of one or more chemicals (e.g., inhibitors or surfactants) must be blended with large amounts of others.

The principal difficulty is using proprietary reagents is that too often they are, like patent medicines, designed to cure all ills. It is not economic to prepare special reagents to meet every possible need a potential customer might have, so many of the compounds are standard formulations or perhaps minor modifications. The mixtures may contain some expensive constituent that may not be necessary or even some that may be undesirable. Furthermore, the formulation may be changed slightly from time to time by the manufacturer to improve efficiency or reduce cost. Information concerning these "minor" changes may not be relayed to the customer or may be indicated in some ambiguous or meaningless manner, such as addition of "improved" or "revised" to the product designation. Since the formulation is proprietary, the customer can determine corrosivity and suitability of the materials only by empirical tests, and too often it is

extremely difficult to design laboratory tests that will duplicate results in the plant.

a. Acidic Reagents

For chemical cleaning, pickling, and decontaminating the acid reagents are used in concentrations of 5 to 15 (volume or weight) percent of the normally available concentrated acids.

The reagent that has been used most extensively in the past and is still probably the most popular reagent for cleaning steel systems and equipment is inhibited hydrochloric acid, since it is efficient, easy to use, inexpensive, and does not form insoluble salts with the materials usually found in process equipment.⁷³ Satisfactory inhibitors include propynol, formaldehyde, and proprietary solutions. Hydrochloric acid is so corrosive to mild steel that care should be taken to ensure that the inhibitor is present in sufficient amount. This may require periodic checking if the inhibited acid is reused, stored, or used over a long period of time.

In a German process¹⁷⁶ for cleaning boilers and piping systems, hydrazine is added to the HCl to raise the pH to 8.5 and reduce corrosion rates to less than 10 mg/dm²-hr. The hydrazine acts as a transporting agent for the HCl and also reduces ferric oxide to magnetite. With time at temperature (80 °C), the hydrazine decomposes; the HCl is depleted by reaction with the iron oxides, and is regenerated by hydrolysis of the FeCl₂ formed, so the net reaction is:

$$6 \text{ Fe}_2\text{O}_3 + 3 \text{ N}_2\text{H}_4 + 22 \text{ H}_2\text{O} \rightarrow 12 \text{ Fe(OH)}_2 + 8 \text{ H}_2 + 3 \text{ N}_2$$

and net consumption of HCl is very low.

Hydrochloric acid also causes cracking of stainless steel if stress is present, and its use for stainless steel systems and equipment is limited if the equipment is to be reused. Again, if the equipment is to be discarded after cleaning, stress cracking is of no importance.

Sulfuric acid (or sodium bisulfate), suitably inhibited, is also used for cleaning steel equipment. It does not cause stress cracking of stainless steel, is inexpensive, relatively efficient, and easy to use. Certain sulfates (Ca, Ba, and Sr) are relatively insoluble and may block narrow passages, but this usually will cause no difficulty in cleaning the process equipment at nuclear installations.

Fluoride is sometimes added to remove silica deposits. For example, heat exchangers and boilers are cleaned by recirculating HCl-HF solutions (e.g., 5 to 8% HCl, 1% NaF, wetting agents, and inhibitors) at 60 to 70 °C for 5 hr. Mixtures of HF with HNO₃ or H₂SO₄ are recommended for removing stainless steel scales that have been loosened by some preliminary conditioning treatment (see Film Conditioners).¹⁶³ Typical solutions are: HNO₃, 20%, HF, 30%, H₂O, 50% at a temperature of 50 to 65 °C; and H₂SO₄, 6.5%, HF, 6.5%, H₂O, 87%, Cr₂O₃, 250 grams, at either room temperature or elevated temperature.

The most commonly used complexing agent to keep iron in solution is citric acid. The pH can be adjusted with ammonia or hydrazine; usually the efficiency is still sufficiently high and corrosion rates are reduced considerably if the pH is between 4 and 5. Hydrazine is also a reducing agent and keeps the iron in the more soluble ferrous state. One

recommended procedure consists of recirculating an acid solution (0.9% citric acid, 0.25% NaF, 0.1% inhibitor, 0.04% surfactant, and 0.05% N_2H_4) at boiling temperature for 12 hr with periodic injection of H_2SO_4 or NH_3 and N_2H_4 as necessary to regulate pH to 5.¹⁷⁶

Nitric acid is used to clean metals normally passivated by oxidizing acids (aluminum, stainless steel). Dilute nitric acid and HF (20% HNO_3 + 3% HF) at 55 °C is suggested for decontaminating stainless steel, especially when other reagents are not effective.¹⁴ The efficiency increases with the temperature. The same reagent is recommended as part of a multi-step procedure for decontaminating stainless steel, porcelain, and glass; the HNO_3 -HF treatment is alternated with an alkaline complexing solution at 60 - 80 °C.⁷⁵

Phosphoric, sulfamic, and oxalic acids are used for certain situations although they are more expensive. Inhibited, dilute (5 to 10%) H_3PO_4 is very effective for decontaminating mild steel.¹⁷⁷ It is a liquid and easily handled. Corrosive action can be decreased by lowering the temperature. After extended use at temperature (60-70 °C), it tends to deposit a film on the iron and this may reduce attainable decontamination factors.¹⁷⁷ It is difficult to inhibit attack at galvanic junctions.¹⁷⁷

Inhibited sulfamic acid is a solid, which may be easier to store and handle. It is effective for decontaminating mild steel and aluminum, does not cause reprecipitation, and usually does not cause severe attack at galvanic junctions.¹⁷⁸ Monel and nickel that have been exposed to water at 570 °F or lower can be cleaned by immersing in 25% sulfamic acid for 1 hr at boiling.^{179,180}

Inhibited oxalic acid is relatively corrosive and forms an insoluble oxalate film on mild steel.¹⁸¹ This film formation can be decreased by addition of a complexing agent such as citric acid.¹⁸²

Organic acids (especially citric, formic, and hydroxyacetic acids) have replaced the mineral acids to a great extent in the chemical cleaning industry.¹⁸³⁻¹⁸⁹ Citric acid is the most effective. Hydroxyacetic acid is cheaper and can be used to replace part or all of the citric acid. Formic acid is less effective but is a liquid and may be useful in certain applications such as injection into a steam jet.^{135,136} In typical cleaning operations, the system is filled with ammoniated citric acid (pH = 3.5-4.0), circulated several hours at 140-160 F; the pH is adjusted with NH_4OH to ~ 10 , and NaNO_2 is added to passivate the system.¹⁹⁰

Any of the solutions used for pickling metals may be used for decontaminating the same metal. Many are mixtures of strong mineral acids.

Chemical polishing reagents are usually highly concentrated and have a limited use. However, they are useful for removing contamination quickly and easily to provide a clean smooth finish. For example, copper can be chemically polished in 1-2 min at 60-70 °C by a mixture of equal parts of phosphorus, nitric, and acetic acids.¹⁹¹ Brass can be polished by dipping into a concentrated solution of fuming nitric acid (4 parts acid to 1 part water) for 5 sec at 40 °C.¹⁹²

Some 100% acids (anhydrous H_2SO_4 , H_3PO_4 , or HNO_3) at elevated temperatures are sometimes effective solvents for refractory films or residues. They have been used successfully to remove ceramic fuel residues (PuO_2) from piping systems.¹⁹³ The corrosion rates are not excessive for stainless steel.

Anhydrous H_2SO_4 and $\text{H}_2\text{SO}_4\text{-HNO}_3$ mixtures at 100 °C corrode 304 stainless steel at rates of 1 to 10 $\text{mg/dm}^2\text{-hr}$ (0.01 to 0.1 $\mu\text{/hr}$).¹⁹⁴

b. Acidic Peroxide Reagents

Solutions of hydrogen peroxide and oxalic acid are excellent chemical polishing agents, derusters, and decontaminants for mild steel and ferritic stainless steels.¹⁹⁵⁻²⁰⁰ Solutions containing hydrogen peroxide, oxalic acid, and hydrofluoric acid decontaminate stainless steel effectively.¹⁹⁵ The peroxide acts as a corrosion promoter in lower concentrations and as an inhibitor in higher concentrations. As H_2O_2 is added to 0.3 M oxalic acid at room temperature, filming and corrosive attack of mild steel begins at 0.05 M H_2O_2 ; at 0.2 M H_2O_2 , metal becomes clean and attack is rapid; between 0.3 M and 1.0 M, chemical polishing occurs; and above 1.0 M, the metal is passivated.^{195,199}

A solution of 0.25 M $\text{H}_2\text{C}_2\text{O}_4$ and 0.3 M H_2O_2 (80 ml H_2O , 28 ml 10% $\text{H}_2\text{C}_2\text{O}_4$, 4 ml 30% H_2O_2) polishes mild steel in 15 min at 35 °C.¹⁹⁵ All of the film and some of the metal is removed to give a very smooth surface with a bright finish. The cleaning action and the corrosivity can be controlled by adjusting concentrations of the components (especially the H_2O_2) and temperature. At high temperatures, peroxide is rapidly destroyed by oxalic acid. In one hour at 60 °C, 1 M H_2O_2 oxidizes 0.4 M $\text{H}_2\text{C}_2\text{O}_4$ completely to CO_2 and water.¹⁹⁵ Solutions can be stabilized by buffering to pH of 4-5.¹⁹⁵ One solution recommended for decontaminating carbon steel at 90 to 95 °C contains 0.4 M ammonium oxalate, 0.16 M citric acid, and 0.3 M H_2O_2 . The citric acid complexes the iron ions and prevents formation of insoluble oxalates, and aids in dissolution of the film

and rust.

If fluoride is added, the reagents can be used to decontaminate stainless steel.¹⁹⁵ One solution recommended (0.4 M oxalic acid, 0.1 M HF, and 0.0 to 1.0 M H₂O₂) decontaminates stainless steel in 2 to 4 hrs at 90 to 95 °C. The efficiency (and corrosivity) can be increased by reducing the peroxide concentration to 0.01 M.¹⁹⁵ Similar solutions are recommended for decontaminating Zircaloy.¹⁹⁵ The contamination and black oxide are removed completely with no pitting of the Zircaloy. The corrosivity can be controlled by varying the concentrations of the peroxide of fluoride. Corrosion rates are very low if Al³⁺ is added to repress the ionization of the fluoride.

Reagents containing oxalic acid, citrate, and peroxide can be used for decontaminating mild steel at room temperature if the peroxide concentration is lowered to 0.08 M.

Mild steel may be derusted by use of very dilute solutions at room temperature. The sample is exposed to 0.2% oxalic acid for 5 min; then H₂O₂ is added (5 ml 30% H₂O₂/liter) and the sample is exposed for 5 min longer.¹⁹⁹ The preliminary treatment with oxalic acid apparently changes the character of the oxide to a form easily dissolved by the oxalic-peroxide. (This procedure did not remove magnetite film formed on steel after exposure to high-temperature water.)²⁰¹ Several modifications of this procedure (changes in concentrations, times, temperature) were investigated.¹⁹⁵ Citrate aids dissolution without materially increasing corrosivity. Raising the temperature decreases decontamination time (2 min at 100 °C is as effective as 25 min at 25 °C) but increases corrosivity. At room temperature, peroxide

solutions are much more stable; the only peroxide reaction is that of oxidizing iron. Not enough attention has been given to this type of reagent. They are very easy to prepare; the reactions can be controlled easily and accurately by proper formulation or by variations in temperature; the reagents are relatively inexpensive; and the excess reagent can be decomposed easily so that the amount of solid waste can be kept to a minimum.

A slight modification of this procedure (temperature = 65 °C, pH = 4.0, time = 16 to 20 hr) was used to decontaminate a stainless steel sample to give an α DF of 300 and a β - γ DF of 20.²⁰²

The oxalate-citrate-peroxide reagent was also used for spray decontamination of mild steel.²⁰³ The solution was fed at a ratio of 1:8 into a 15-psig jet held 2 to 24 in. from the surface of the work. Decontamination rates of 500 to 1000 were obtained in 1 hr.

Since the oxalate-peroxide reagents are effective in fairly dilute solutions and since corrosivity can be regulated by peroxide concentration and solution temperature, they may be useful in steam or water jets to replace abrasive blasting. Experimentation may develop a procedure that will give the desired penetration in a relatively short time. The spent solution can be concentrated by boiling with destruction of the oxalic acid and peroxide to reduce volume of solid waste.

c. Neutral Reagents

Ethylenediaminetetraacetic acid (EDTA) and its homologues (NTA, DPTA, TTHA) are general spectrum agents that form stable

complexes with iron, nickel, cobalt, chromium and many other di- and trivalent ions. Complex stability and pH range of solubility increase with increasing molecular weight: TTHA > DPTA > EDTA > NTA.¹⁷³ Mixtures of citric acid and EDTA at a neutral pH (6-8) readily remove high-temperature film and scale from mild steel and the conditioned high-temperature films from stainless steels.^{67,174} The rate increases with temperature, becoming effective at 90-100 °C and much more efficient in pressurized systems at higher temperatures. The pH can be regulated with NH_4OH , but hydrazine is preferred because it also provides a reducing atmosphere, which reduces any tendency to pitting, and increases complex stability. In an unpressurized system, ammonia cannot be used since it will boil off and pH will decrease to some value close to 6. Either hydrazine or sodium hydroxide is suitable for keeping pH between 7 and 8. Hydrazine is preferred to NaOH because it is decomposed by heat and will not form a residue in crevices.

In the fossil fuel industry very dilute complexing solutions have been used on a periodic or continuous basis to remove film and scale.²⁰⁴⁻²⁰⁸ Several reports written over the past ten years describe procedures that have been used successfully in large installations to keep boiler surfaces free of any but a very thin protective film.

At one installation, dilute EDTA solutions were used for continuous internal cleaning of 1325 psi boilers.²⁰⁶ Initially, the EDTA solution was added at a rate sufficient to maintain a concentration of 5 to 6 ppm EDTA to remove deposits already present; subsequently the rate was dropped to maintain 2.5 ppm EDTA. If oxygen was kept from the system, corrosion rates were very low.

In another application, EDTA was added to the feed water for a 900-psi boiler.²⁰⁷ The initial dosage was 30 ppm EDTA to remove deposits already formed. After the boiler was clean, EDTA was fed continuously to maintain a 2 to 5 ppm residual at the economizer inlet. The OH^- concentration was maintained at 25 ppm as measured at room temperature. The corrosion of carbon steel, as measured from coupons, was about 6 μ/year (2.5×10^{-4} in/year).

Russian workers have used chelating acid blends for routine cleaning of thermal power equipment.²⁰⁸ The Mosonegro TP-70 boilers were cleaned with monosodium EDTA (2.8 ppm) and with a mixture of EDTA (3 to 4 ppm), citric acid (1 to 2 ppm), and hydroxylamine sulfate (0.3 ppm). The blend was much more effective, removing about three times as much iron oxides as the solution of monosodium EDTA.

Solutions of EDTA, citric acid, and hydrazine (or NaOH) at a pH between 6 and 8 should be satisfactory for decontaminating equipment. The reagents are relatively noncorrosive and nontoxic. Rate of attack can be easily controlled by regulating the temperature. Dilute solutions (0.5%) are effective at 90 to 100 °C, but efficiency increases with concentration, at least up to 5%. At higher temperatures (200 to 300 °C), very low concentrations (5 to 10 ppm) are satisfactory if the EDTA is present in amounts in excess of that required to complex all the iron present in the film and scale.

If the solution is to be used just once and then discarded, the concentration should be as low as feasible; but if the

solutions are to be used repeatedly, the higher concentrations should be used. Citric acid, EDTA, and hydrazine can all be decomposed by heat if reduction of the amount of solid wastes is a goal. EDTA can also be recovered by precipitation as the free acid.

Citric acid, added to EDTA solutions, appears to exert a synergistic effect. In a series of tests, it was found that the best formulation for decontaminating steel was a mixture of EDTA, citric acid, and hydrazine.¹⁷⁴ Hydroxylamine increased the efficiency, but also increased the corrosion rate by a factor of 10 or greater.¹⁷⁴

d. Film Conditioners

Films on some metals (especially stainless steels and high-nickel alloys) are not easily removed by soaking in a hot, dilute (up to 10%) acid solution, but require more corrosive treatments or some conditioning step to change the film characteristics.

The choice of an effective chemical treatment will depend on the composition and structure of the film. Unfortunately, very little is known about films formed on stainless steels that have been exposed to high-temperature water or steam under varying conditions. Some hypotheses have been forwarded, but they are based on sketchy information. Procedures that have been developed for removing these films are empirical and may be effective for only certain types of films that have been formed under rigidly controlled conditions.

The films on stainless steel after exposure to high-temperature water or steam are composed of substituted magnetite and are essentially crystalline in nature.

Magnetite, Fe_3O_4 , contains both Fe^{2+} and Fe^{3+} atoms, which may be replaced in part by other di- or trivalent ions, especially those having approximately the same dimensions. In stainless steel films, the iron atoms are partly replaced by chromium or nickel atoms. The type and degree of replacement and the crystal structure depend on exposure conditions. Films formed on stainless steel and Inconel after any appreciable exposure to high-temperature water or steam are spinels which are very stable and resistant to chemical attack. On stainless steel that has been exposed to high-temperature water where O_2 content is low (e.g., in a PWR), the corrosion product film is a modified magnetite in which iron atoms are replaced to a large extent by chromium. If the O_2 content is relatively high (e.g., in a BWR), the nickel content of the film is high and chromium content is low (usually less than 2 atomic percent). In some instances the film has been identified as nickel ferrite.²⁰⁹ Other variables, which have not been considered, may also affect film composition. Under certain conditions, both chromium and nickel may replace iron atoms in the magnetite structure, and it is reasonable to assume that by changing exposure conditions, the relative amounts of nickel and chromium atoms could be altered.

In designing a satisfactory chemical treatment, it would be very desirable to know: (1) under what exposure conditions, chromium and nickel replace iron atoms in the magnetite crystals formed during corrosion; (2) how chromium and nickel appear in the crystal lattice; (3) how chromium and nickel change the crystal structure; (4) how ratio of chromium to nickel can be changed (e.g., can conditions be

altered in such a manner as to go from a high-chromium, low-nickel to a low-chromium, high-nickel substituted magnetite); (5) under what conditions (if any) the chromium or nickel can be replaced by iron; (6) how susceptibility to chemical attack can be related to chromium or nickel content; (7) how different reagents attack the different crystal structure; (8) how film composition varies with distance from the metal surface; and (9) how film composition and crystal structure vary with exposure.

Films formed on surfaces of primary coolant systems of nuclear reactors have been investigated at some length, and satisfactory (although not optimum) procedures have been developed for removing these films and decontaminating the surfaces. However, the investigation has been limited to surfaces exposed only to high-temperature, high-pressure water or steam.

Equipment used in the nuclear industry may be subjected to any of a wide variety of operating conditions and atmospheres; solutions in contact with the surfaces may be dilute or concentrated mixtures of salts, acids, or bases, with additives such as chelating agents, surfactants, oxidizing, or reducing agents, and temperatures may vary from room temperature to several degrees above the boiling point. Virtually nothing is known about the films formed under these conditions or the manner in which the radio-nuclides are attached to the surface. Almost all decontamination of equipment has been done in an empirical manner, generally by using successive treatments with progressively more corrosive reagents. Films formed on stainless steel in a high-temperature pressurized water system, such as a

PWR, are usually removed by a two-step procedure. In the first step, the films are conditioned by treatment with a solution of alkaline permanganate (3 to 20% NaOH, 1 to 5% KMnO_4) at or near boiling for 1 to 2 hr.²¹⁰ The permanganate apparently oxidizes the Cr^{3+} in the spinel film to Cr^{VI} , which is soluble in the alkaline solution.²¹¹ The crystal structure is destroyed, leaving a more or less homogeneous layer of disintegrated crystals on the metal surface.⁶⁷ As determined with radioactive samples, the chromium is removed more or less quantitatively, but the weight is not changed much. The film has a high concentration of manganese, probably as MnO_2 from reduction of the MnO_4^- .²¹² The stainless steel films, after conditioning by AP, are easily removed with either a dilute complexing solution⁶⁷ or a dilute acid solution. Removal and dissolution of the film is almost quantitative with no residue. Some stainless steel films, after AP treatment, can be removed by relatively gentle physical abrasion (rubbing with a rubber eraser).¹⁸⁰ Alkaline permanganate also conditions films on Inconel and Incoloy, presumably by the same mechanism, although no information is available about structure of the films on these alloys after treatment with AP. The films on Inconel and Incoloy, after conditioning with AP, are slightly more difficult to remove than those formed on stainless steel, but can still be removed with the neutral complexing solutions recommended for stainless steel films,⁶⁷ with nitric-hydrofluoric acid (25% HNO_3 , 25% HF at 70 °C),¹⁶³ or with a mixture of sulfuric acid (13 oz/gal) and Rochelle salts (13 oz/gal) at 80 °C.

The AP has no apparent effect on the films formed on carbon steel after exposure to high-temperature water. This is logical since the films contain no chromium atoms. Surprisingly, however, the films on carbon steel, with or without conditioning by AP, are much harder to dissolve than the conditioned films on stainless steel or Inconel.^{67,174} The dilute acids and neutral complexing solutions attack at a much slower rate, and the films are readily dissolved only in a solution with low pH.^{67,174} In neutral solutions, the film is quickly and completely removed from the metal surface, but a small portion (5 to 10%) remains as a residue and is dissolved only after prolonged treatment. The residue does not appear to differ from the rest of the film in either composition (including radionuclidic composition) or crystal structure.

Stainless steel films that have a high nickel content (e.g., nickel ferrite) and little if any chromium are not appreciably affected by AP; and such films, before or after AP treatment, cannot be dissolved in hot, dilute acids. These films can be removed by soaking in boiling, concentrated (70 to 80%) H_3PO_4 ,^{213,214} or in a strong reducing solution such as a hot chromous sulfate solution (0.4 M CrSO_4 , 0.5 M H_2SO_4 at 85 °C).^{214,215}

Stainless steel and nickel-base alloys exposed to high-temperature steam can be descaled by heating in a borax bath (see Anhydrous Materials), immersing in hot water to crack the scale, conditioning with AP, and soaking in hot (near boiling) 10% dibasic ammonium citrate solution.¹⁷⁴ Other more exotic methods are described in the literature.¹⁸⁰

In summary, the degree of success that can be attained with any procedure for decontaminating stainless steel or nickel-base alloys will depend on the exposure history; and lack of fundamental information makes it difficult to predict how successful any procedure will be.

e. Alkaline Reagents

Alkaline solutions used in decontamination of steel include: alkaline detergent solutions, alkaline soaking baths, paint strippers, and alkaline oxidizing solutions. The alkaline detergent solutions and one type of alkaline oxidizing solutions (AP) have been previously discussed.

The alkaline soaking baths strip films very slowly even at high temperatures and are effective only for rust and films formed at low temperatures.²¹⁶ High-temperature films can be removed electrolytically in an alkaline bath with either reverse (the work piece is the anode) or periodic reverse current (the current is reversed periodically, e.g., 10 sec. anodic and 5 sec cathodic).²¹⁶ The latter method is preferred because there is less danger of pitting. Some metals (alloys of aluminum, tin, and zinc) cannot be cleaned in caustic solutions unless they are made the cathode or a suitable inhibitor is used.

The alkaline derusting baths are strongly caustic, about 2 lb NaOH/gal (6N) and usually contain some complexing agent such as EDTA, NaCN, gluconate, glucoheptonate, or some combination of these. Dissolution is very slow, but scales are loosened and can be removed by brushing or by some other type of physical action such as alternate immersion and spray rinse. Removal of heavy rust or scale may require

several hours at boiling, and even then may not be complete. The alkaline baths are safer than acid baths because there is practically no corrosion of the steel, no hydrogen embrittlement, and no overpickling.

Alkaline soaking baths also remove dirt, grease, oil, and paint, although cleaning of dirty material will reduce life of the bath. The greatest use of these baths in decontamination is for stripping paint and removing dirt and grease. Aside from the alkaline detergents and paint strippers, the only alkaline decontaminating solution used to any extent is alkaline permanganate. The use of alkaline permanganate to modify films on stainless steels and nickel alloys was discussed previously (see Film Conditioners). Alkaline permanganate is a very strong oxidizing agent and can be used advantageously to oxidize certain ions or metals to a state in which they can be more easily removed. For example, alkaline permanganate solutions oxidize and are effective for removing chromium and ruthenium contamination from surfaces or solutions.

The potential advantages of alkaline cleaners should not be overlooked, and a re-evaluation may be profitable. Many new complexing agents have been developed and have proved valuable for dissolving compounds of iron, nickel, and chromium in alkaline solutions ($\text{pH} > 10$). A systematic investigation might lead to development of efficient descalers and decontaminants that show less tendency to cause accelerated or nonuniform corrosion, even with less rigid control of operating variables.

f. Anhydrous Materials

Molten caustic or salt baths with other additives (usually oxidizing or reducing agents) are sometimes used to modify the scale on steel specimens to make it easily removable in acid. The baths are expensive to install and operate and present several formidable operating hazards.²¹⁶

A molten borax bath at 1475 °C is recommended for conditioning stainless steel and nickel alloys exposed to steam above 1200 °F.¹⁸⁰ The samples are immersed in the molten borax bath for 3 to 5 min and then plunged into hot water to crack the scale.

A molten carbonate bath (60% NaOH, 40% Na₂CO₃) at 1040 °F can be used as an electrolytic bath to condition films on stainless steels, nickel-base alloys, Fe-Cr-Al alloys, Incoloy 800, and Haynes 25 that have been exposed to high-temperature steam (e.g., 1000 °F, 3000 psi).^{180,217} The samples are charged cathodically, immersed in the bath at 30- to 60-second intervals, removed, quenched in 10 vol % HCl, rinsed, and dried. Recommended current densities vary with the alloy treated and range from 15 to 75 A/dm².

A 3-min dip in a sodium hydride bath (99% NaOH, 1% NaH) at 371 °C, followed by a water quench, a rinse, a 30-sec dip in inhibited 5% H₂SO₄ is recommended for conditioning films on iron and nickel alloys exposed to high-temperature (up to 370 °C) water.^{180,218}

Another anhydrous procedure for decontamination of metals is melting. The impurities and contaminants will concentrate in the slag which can be removed in some suitable

manner. Although this method is of limited applicability, it has been used to advantage for decontamination of lead.^{219,220} After melting, a mixture of Al_2O_3 , SiO_2 , and CaO is added to form a slag which is skimmed off. After purification by this method, the contamination level was very low, from 1×10^{-5} to 3×10^{-5} $\mu\text{Ci/g}$.

Anhydrous acids were discussed on pages 54 and 55.

V. CONCLUSIONS AND RECOMMENDATIONS

Utilization of proper decontamination methods in a correctly designed decontamination installation will result in a considerable reduction of the volume of solid wastes.

Empirical approaches, while they may be occasionally necessary for solving problems on a crash basis, may prove to be exceedingly expensive, and should be supplemented by a long-term systematic investigation of the fundamentals, especially studies of film characteristics and mechanisms of contamination and decontamination.

Improved procedures and materials have been studied by different groups, but progress has been slow because the effort has not been sufficiently organized. Often the work is a minor part of another task and has been done on a low-priority basis and ultimately abandoned without clear-cut results or recommendations. Lack of communication or delays in transmittal of information among the groups potentially interested has contributed to the lack of progress.

An evaluation should be made of procedures that may prove to be much more efficient and reduce manpower requirements, exposure levels, and waste volumes. The ones that appear especially worthy of investigation now include: ultrasonics, electrolytic polishing and deplating, foamed decontaminants, gels, and strippable films.

Simultaneously, new decontaminant formulations that will result in low final waste volumes should be evaluated. Especially promising are the organic reagents and others that can be removed more or less quantitatively by destruction, sorption, extraction, or volatilization.

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