Chemical treatment of Radioactive wastes.

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Chemical Treatment of Radioactive Wastes

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FOREWORD

A number of techniques have been developed for treating radioactive liquid wastes to remove the radioactive components from the bulk material, which can then be discharged to the environment as waste water. It is sometimes difficult to determine what is the most effective and economical method for a particular nuclear establishment. To give developing Member States some guidance on this problem, the International Atomic Energy Agency has commissioned books on the three principal techniques used in concentrating radioactive liquid wastes, namely chemical precipitation, evaporation and ion exchange. The present manual deals with chemical precipitation by coagulation-flocculation and sedimentation, which is commonly called "chemical treatment". This technique is particularly suited to low-activity wastes. The other two books in this series, "Operation and Control of Ion-Exchange Processes for Treatment of Radioactive Wastes" and "Design and Operation of Evaporators for Radioactive Wastes", were recently published as No. 78 and No. 87 in the IAEA Technical Reports Series.

The manual was organized by Mr. L.H. Keher, who also contributed a large part of Section 2, while he was an IAEA staff member. Mr. Frank N. Browder, as the succeeding Project Officer, provided the technical editing and assistance for the completion of the manual.

CONTENTS

1. INTRODUCTION

This manual is a review of coagulation-flocculation and sedimentation processes, commonly called "chemical treatment", as they are applied to decontamination of low-activity wastes.

Its purpose is to present the subject in a condensed form and to summarize results recently published, especially those given in the proceedings of the IAEA symposium on treatment of low-activity wastes [10]. This manual does not present results obtained by all treatments performed in various centres or institutes (a fairly comprehensive bibliography is given after Appendix II), but provides the essential information from most well-developed processes.

Although the major topic of this manual is coagulation-flocculation and sedimentation, it has been deemed necessary to review briefly several related techniques (pre-oxidation, filtration, sludge conditioning), which directly affect the utilization of the chemical-treatment process.

The various chapters have been arranged to provide the prospective user of this decontamination method with practical information to assist in the design and utilization of a plant without making it too large or including any unnecessary luxury, at the same time observing the national and international standards governing releases to the environment.

2. PRINCIPLES OF COAGULATION-FLOCCULATION AND SEDIMENTATION AND ASSOCIATED PROCESSES

2.1. Purpose and application to removal of radioactivity

Chemical treatment of low-level activity wastes by coagulation-flocculation and sedimentation is carried out at most nuclear establishments. This is because the insolubility of the hydroxides, carbonates and phosphates of many cationic fission products allows chemical precipitation to achieve at least a partial removal of activity, particularly if coagulants and carriers are added to give bulk to the precipitate. The object of the chemical treatment is to concentrate radionuclides into a small bulk of insoluble sludge, reducing the activity level of the large volume of low-activity liquid wastes sufficiently to permit their dispersal to the environment.

Sometimes chemical treatment is used for liquid wastes of higher activity level if other methods of treatment such as ion exchange or evaporation are to follow. The object of this chemical treatment essentially is to prepare the liquid for the subsequent treatment.

For removal of some specific isotopes, chemicals which are not true coagulants may be added to the water, causing a precipitate to form. These precipitates are trapped in the floc formed by the coagulant.

The main advantages of treatment by coagulation-flocculation and sedimentation are low cost, the ability to handle a wide range of inactive constituents in the liquid and the use of equipment and procedures for which considerable experience exists.

2.2. Low-activity liquid wastes[1]

2.2.1. Specification for radioactivity

The upper activity level specified for low-level liquid wastes varies from establishment to establishment, the value depending on three factors:

(a) The type of radioisotopes present;
(b) The treatment facilities and the Decontamination Factor (DF) obtained with the treatment; and
(c) The activity level at which effluent can be released to the environment.

The activity levels at which effluent is released to the environment are related directly or indirectly to the Maximum Permissible Concentrations (MPC) in water recommended by the International Commission on Radiological Protection (IRCP). Owing to the diversity of radiation properties between the different isotopes and the varying effects on man, the MPC in water or air vary considerably. These values are shown in Table III of reference [3] and are particularly useful when only one or a few isotopes are present. Where a wide variety of materials is present, e.g. mixed fission products, the analytical problem to determine the concentration of each radioisotope is so great that it makes the use of individual levels almost out of question. Therefore, account has been taken of the fact that a partial analysis may be made to show the absence of the more hazardous isotopes and thus a more relaxed level may be used. These values are reproduced in Table I.

Generally, sufficient dilution exists in the immediate environment to enable a factor to be applied that permits higher levels of discharge. This requires careful study and reference should be made to the IAEA manuals covering disposal into the sea, fresh water and ground [4, 6, 8].

The extent of treatment and the DF obtained will influence the activity level at which waste can be accepted for treatment. DFs of 10 are common for most coagulation processes and DFs of 200 and higher are obtained for some specialized processes. Often one specific coagulation or scavenging process may be operated in series with another specific process. These may also be followed by an ion-exchange process or evaporation to increase the DF.

Consideration of the above factors will enable an establishment to set an upper activity level for low-level liquid wastes. It is not wise to set a value too high as this may cause handling problems which, although they can be overcome, may make the system too expensive.

As a general guide, the upper activity level for liquid wastes to be treated by a coagulation process will approximate a 100 times the MPC in water. The upper value, therefore, is in the range 10^{-3} to 10^{-2} μCi/ml.

It should be noted that, if this value is adopted as the upper level at which waste can be placed in the low-level liquid system, allowing for dilution by liquid waste containing negligible activity, the average

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[1] Low-activity wastes in this manual correspond to liquid wastes of Category 2 (10^{-6} to 10^{-3} μCi/ml) as proposed by a panel convened by the IAEA in November 1967, composed of experts from nine Member States and three other international organizations. The recommendations of this panel will be published in the IAEA Safety Series.

TABLE I. MAXIMUM PERMISSIBLE CONCENTRATION OF UNIDENTIFIED RADIONUCLIDES IN WATER FOR CONTINUOUS OCCUPATIONAL EXPOSURE

$(\mathrm{MPCU})_w$ Values[a]

Limitations	μCi/ml of water[b]
If none of the radionuclides ^{90}Sr, ^{126}I, ^{129}I, ^{131}I, ^{210}Pb, ^{210}Po, ^{211}At, ^{223}Ra, ^{224}Ra, ^{226}Ra, ^{228}Ra, ^{227}Ac, ^{230}Th, ^{231}Pa, ^{232}Th, and Th-nat is present, then the $(\mathrm{MPCU})_w$ is:	3×10^{-5}
If none of the radionuclides ^{90}Sr, ^{129}I, ^{210}Pb, ^{210}Po, ^{223}Ra, ^{226}Ra, ^{228}Ra, ^{231}Pa, and Th-nat is present, then the $(\mathrm{MPCU})_w$ is:	2×10^{-5}
If none of the radionuclides ^{90}Sr, ^{129}I, ^{210}Pb, ^{226}Ra and ^{228}Ra is present, then the $(\mathrm{MPCU})_w$ is:	7×10^{-6}
If neither ^{226}Ra nor ^{228}Ra is present, then the $(\mathrm{MPCU})_w$ is:	1×10^{-6}
If no analysis of the water is made, then the $(\mathrm{MPCU})_w$ is:	1×10^{-7}

[a] Each $(\mathrm{MPCU})_w$ value is the smallest value of $(\mathrm{MPCU})_w$ in Table III. of Ref. [3] for radionuclides other than those listed opposite the value in the above table. Thus, these $(\mathrm{MPCU})_w$ values are permissible levels for continuous occupational exposure (168 h/wk) for any radionuclide or mixture of radionuclides where the indicated isotopes are not present (i.e., where the concentration of the radionuclide in water is small compared with the $(\mathrm{MPC})_w$ value for this radionuclide). The $(\mathrm{MPCU})_w$ may be much smaller than the more exact maximum permissible concentration of the material, but the determination of this $(\mathrm{MPC})_w$ requires identification of the radionuclides present and the concentration of each.

[b] Use 1/10 of these values for interim application in the neighbourhood of an atomic energy plant.

level will generally be much lower (10^{-5} to 10^{-4} μCi/ml) and in many cases the waste can be released to the environment without treatment or with minimum treatment.

2.2.2. Segregation into different origin types

The person producing waste prefers to have a single waste system into which he places all his waste. At nuclear establishments this results in a large volume of slightly contaminated waste to be treated. Conversely, the waste processor prefers wastes to be segregated at origin into many classifications by physical state and concentration. This results in the treatment of smaller volumes of waste at a somewhat higher activity level but with a large part of the waste able to be released to the environment either without treatment or with minimum treatment. Either of the above philosophies, if taken to extremes, wastes effort and is uneconomic, and the solution lies somewhere in between.

Even when segregation is extensively practiced and all liquid wastes with higher than the upper level set for the low-level activity category are placed into containers especially provided for the purpose, it is advisable to further segregate the low-level liquid wastes into several different categories as follows:

A. Liquids arising from laboratories and working areas where radioactive work is performed and the liquid will probably contain small amounts of activity;
B. Liquids arising from laboratories and working areas where no radioactive work is being performed and the liquid will probably contain no activity;
C. Sewage.

It is considered generally advisable to provide different collection systems for the above three categories provided the quantities warrant this action.

All the above categories may require some form of treatment to remove undesirable non-radioactive contamination before release to the environment, but generally radioactive contamination will occur only in category A. However, as it is possible for radioactivity to be accidentally transferred into the other categories, it is essential that their activity be monitored before discharge and if necessary they can then be given the required treatment.

Category-B effluent, after simple treatment to remove non-radioactive contaminants such as oils, greases and suspended matter (e.g. passage through a grease trap and settling tank and a high-pressure sand filter), is available to dilute treated category-A effluent and thus lower the activity level.

Category-C waste may be discharged to a municipal sewage system if available. Most of the larger nuclear establishments, because of location and also as an added safeguard against accidental release of radioactivity to the municipal system, have set up their own sewage-treatment plant. The effluent from this plant can also be used to further dilute treated category-A effluent.

2.2.3. Segregation from materials having adverse effect on process

One of the main reasons why coagulation processes are popular for the treatment of low-activity liquid wastes is their ability to handle a wide range of non-radioactive constituents. However, there are some materials which interfere with the process and although it is not practical or economic to absolutely prevent these materials from entering the collection system, it is necessary to ensure that only minimum amounts are allowed to enter. This is done in two ways:

(a) By segregating, as far as possible, liquids containing these materials and placing them in containers provided for the purpose;
(b) By co-operating with the waste producer to ensure that firstly only the minimum amounts are used and secondly that alternative materials are used which do not affect the waste-treatment process.

The latter is most important and good co-operation must exist between the waste producer and waste processor. It is not satisfactory if the waste processor first learns of the presence of these interfering materials after the treatment process has been adversely affected. The waste processor should know details of all the experiments and processes being performed at the establishment and should then take action as outlined above to ensure that only negligible amounts of materials, which adversely affect the treatment process enter the collection system.

The following materials can interfere with the collection system, or adversely affect the process and should not be allowed (except in negligible quantities) to enter the collection system:

(a) Solid wastes such as paper, glassware and cloth fibres cause blocking of pipe-lines and pumps, clog pump filters and can affect the process if they become wrapped around agitators.

(b) Oils, greases and solvents are generally not amenable to treatment and form scums which can interfere with the process. In pipe-lines, sumps, pumps, lift stations and storage tanks, pockets of inflammable solvents can form and thus create an explosion hazard. Heavy solvents, such as trichlorethylene, may cause leakage by diffusing through plastic pipes and storage vessels.

(c) Highly acidic solutions will either corrode pipe-lines, pumps, etc. or require too much alkali for neutralization. Since most treatment processes are carried out in alkaline conditions, the same restriction does not apply generally to highly alkaline solutions.

(d) Detergents, especially those containing alkyl benzyl sulphonate (ABS), will, at a concentration greater than 5 ppm, interfere with the efficiency of most processes employing flocculation. A concentration in excess of 15 to 20 ppm can by excessive foaming prevent flocculation and completely ruin the process. The effect of detergents can be somewhat overcome by massive doses of coagulating and precipitating chemicals, but this leads to a large volume of sludge and increases the costs of the processes. It is much better practice to limit the amount of detergent in the waste. It is worth noting that the quantity of a detergent needed to provide efficient cleaning is quite low and many laboratory workers and janitors use excessive quantities. If these people are taught the proper amount to be used, generally no excessive amount will be found in the liquid waste.

(e) Complexing agents, e.g. the sodium salts of ethylene diamine tetra acetic acid (EDTA) and the polyphosphates, have an effect somewhat similar to that of detergents on flocculation but slightly higher concentrations are usually required to seriously affect the process. Complexing agents are used extensively in decontamination of equipment and if correct decontamination procedures are used, it is possible to place the resulting small volume of liquid into special containers. If common sense is practiced with the use of the above materials, it will usually be found that very little trouble to the waste-treatment system will be caused by their use for normal purposes.

2.2.4. Non-radioactive contamination

Often the low-level liquid waste at nuclear establishments may have a radioactivity level so low that the waste does not require treatment. However, the presence of non-radioactive contaminants may necessitate treatment and this applies particularly where wastes are discharged into fresh or estuarine waters. Most countries have found it necessary to set limiting values for a wide range of non-radioactive contaminants to reduce pollution of rivers and lakes. Thus it may be necessary to adjust the pH or to reduce the B.O.D. (biochemical oxygen demand), the amounts of total or suspended solids or the concentration of specific ions such as chromium, nickel, ammonium, cyanide or nitrate. The coagulation processes used for removal of radioactivity will generally also remove most of the non-radioactive contaminants. However, if large amounts of a particular contaminant are occurring, these wastes should be segregated from the main waste-collection system and given treatment to reduce the amount of the contaminant before being placed back into one of the main collection systems. For instance, this should be done with wastes arising from plating and pickling processes.

2.3. Coagulation and flocculation

There are three steps in the process of coagulation and flocculation.

Firstly, the coagulating chemicals are added to the liquid. To ensure that the chemicals are distributed evenly and promptly throughout the liquid, rapid agitation or mixing must occur. It is extremely important that rapid or flash mixing of the coagulant chemical occurs, otherwise the coagulant would slowly disperse and the initial chemical reactions would be limited to that point at which the coagulant was introduced. This may produce reactions other than those intended.

Secondly, coagulation, i.e. complex chemical and physicochemical reactions and changes, occurs, leading to the formation of finely divided precipitating solids.

Lastly, by means of gentle agitation of the liquid, flocculation takes place, i.e. the finely divided particles contact and adhere to one another and form progressively larger floc. This insoluble flocculent precipitate will then settle out, carrying with it the colloidal materials present in the liquid. It may be assumed that the floc traps the colloidal matter in three main ways:

(a) By simple mechanical entanglement with the floc;
(b) By adsorption of the colloid on the floc; and
(c) By neutralization of the positively charged colloidal particles by the negatively charged colloidal floc first produced.

Floc formation and coagulation are influenced by the chemical impurities present (both anions and cations), pH, ionic strength, degree of mixing and flocculation, temperature and the presence of suspended matter which can act as nuclei. The pH range in which a given coagulant functions best and the concentrations of other ions present are interrelated to some extent. For example, the type of water that is most difficult to coagulate is one high in colour but low in turbidity and dissolved solids concentration. The pH range in which coagulation in these cases occurs is very narrow. In the use of alum for instance, the effective pH range may be broadened with an increase in the amount of sulphate present. Usually, however, the optimum range for coagulation of soft, coloured waters with alum is around pH 5.0 and even lower with iron salts. The range for alum may extend to pH 7.5, depending on mineral concentrations, and iron salts will coagulate at a pH above 9.0.

Mixing and flocculation periods must permit time for the reactions to take place and in the flocculation stage provide opportunity for gentle motion to permit the growth of the floc. Generally, the higher the mineral content or the higher the coagulant dosage, the less time required for mixing and flocculation. Reaction times, of course, vary inversely with temperature, and usually more coagulant should be added when lower temperatures are reached. As an illustration of the effect of temperature, it has been found with the lime-soda process that at 96°C the flocculation is more complete in 10 min than after 24 h at 10°C [8]. The complex interrelationship between added chemicals and the constituents of the liquid being treated and the period and degree of agitation, make it virtually impossible to determine the coagulant dose from the results of analysis of the water. In practice, therefore, the jar or coagulation and flocculation test (Section 6.1.) is widely used for determining proper coagulant dosages. The zeta potential test (Section 6.2.) is now being used in many water-treatment works and in several nuclear establishments to determine the best coagulation and flocculation dosages. The period available for the initial coagulation followed by flocculation is related to the required dose of coagulant and the degree of agitation, lower doses being effective with longer periods of agitation and generally greater velocity of agitation. Usually the period and degree of agitation are fixed by plant characteristics, so the dosage is varied to meet changes in water temperature, turbidity, pH, etc.

2.4. Pre-oxidation

Pre-oxidation, generally chlorination, at any stage before discharge of effluent after sedimentation is sometimes of advantage with coagulation and flocculation processes. In domestic water treatment, pre-chlorination is often performed for control of bacteriological pollution, but in the chemical treatment of radioactively contamined wastes this is usually not the reason. Pre-chlorination, besides achieving effective disinfection, will also oxidize iron and manganese, bleach colouring matter (frequently containing iron), and improve coagulation and flocculation by destroying bacterial and algae growths and slime formations which interfere with the coagulation and flocculation process; in addition, the dead cells of these organisms are more readily coagulated.

Ozonization is another way to eliminate bacteriological materials and oxidizable organic matters. It has the same action in the coagulation of iron traces and for bleaching of the colouring derivatives. Ozone also makes possible the destruction of EDTA [26].

If the biological pollution is too great, a bacteriological pretreatment may be given before chemical flocculation [22, 57]. It generally consists in aeration of the wastes by blowing air into agitated tanks or by using oxidation beds (trickling filters) or oxidation ponds in which bacterial microflora and algae are growing. The treatment comprises two steps:

settling (primary treatment), then oxidation followed by settling (secondary treatment). The different aeration methods may be combined and the resulting zoogleal sludges partially recirculated.

The purpose of these treatments is to stabilize the effluent by eliminating the components responsible for the biochemical oxygen demand (B.O.D.). The radioactive decontamination observed is often low, but the sludges are sufficiently radioactive for their disposal to be performed under control or after a further treatment.

2.5. Sedimentation

The purpose of sedimentation is to permit settleable floc to deposit after coagulation and flocculation have taken place In conventional water-treatment plants, where the main purpose of sedimentation is to reduce the load of suspended solids that must be removed by filters, sedimentation generally takes place in a basin separate from the tank where coagulation and flocculation have occurred, with the exception of cases where suspended-solids contact units are used. The practice generally adopted with coagulation-flocculation and sedimentation processes for treatment of low-activity liquid wastes is for the three steps to occur in one tank. This can be either a batch process where, after coagulation and flocculation, agitation of the liquid is stopped and the floc is allowed to settle out, or a continuous process where the use of suspended-solids contact units or sludge-blanket clarifiers permit the steps to be performed in one tank. The lime-soda process, however, uses a separate sedimentation chamber [8].

The theory of sedimentation is complex and of little practical value since floc is not uniform and its basic sedimentation properties cannot be given quantitative value and also because the effect of eddy currents in the tank cannot be predicted. However, the main factors that influence sedimentation and which must be evaluated by experience are:

(a) Characteristics of the floc: Floc has a large bulk in relation to its weight and a large portion of the bulk represents space occupied by water. Because of its low specific gravity, floc will not settle rapidly and therefore it is desirable that a dense floc be produced rather than a feathery floc.

(b) Viscosity and temperature of the liquid: The viscosity of water has a significant effect on the sedimentation rate, e.g. at 30°C the rate is 2.3 times greater that that at 0°C. Therefore, with cold waters it is absolutely essential to produce a dense floc.

(c) Period of sedimentation: Well-formed floc settles at a rate of about 3 m/h (10 ft/h), but it is generally wise to adopt a rate of 0.75 m/h (2.5 ft/h) to allow for periods of poor flocculation. The area and depth of a tank determine the time required for sedimentation. Area is of much greater importance than depth but the depth must be great enough to prevent the settled sludge from being picked up when the supernatant liquid is removed. The area should be as large as possible. For most batch processes a sedimentation time of 3-4 h will prove more than necessary. For continuous processes the flow rate of water through the tank, as expressed by the surface overflow rate, which is dependent solely on area and flow rate, should not exceed half the sedimentation rate. The surface overflow rate is expressed in m/h or in unit volume per unit area per day. This rate, when related to the depth of the tank, will then give the detention period, i.e. the period required to fill the tank at the normal rate of flow.

The amount of suspended solids left in the treated liquid after sedimentation should not exceed 5 ppm. If this figure is being exceeded it may be that insufficient time is being allowed for sedimentation but usually it is more often caused by poor flocculation.

2.5.1. Suspended solids contact units

Attempts have been made to facilitate coagulation by mixing a portion of the settled floc or sludge from a sedimentation tank with the water to be treated, a material which forms the nuclei for subsequent coagulation and floc formation. If the liquid being treated is relatively uniform in non-radioactive and radioactive constituents, the re-use of a certain amount of the settled sludge will give slightly better DFs and also lead to a decrease in the total amount of sludge to be disposed of. However, if there is an appreciable difference in the characteristics of the waste being treated, it is possible to achieve DFs of less than one from the re-use of settled sludge. This is apparently caused by the replacement of non-radioactive ions by radioactive ions previously adsorbed into the floc.

However, the re-use of sludge has led to the development of a new coagulation-flocculation sedimentation technique commonly referred to as the Suspended-Solids Contact or Sludge Blanket Clarification Process. Water-treatment plant manufacturers in several countries have developed units which are basically similar in that chemical mixing, coagulation, flocculation, sedimentation and manual or automatic sludge removal are performed in a single tank. In all of them the coagulated water is introduced at or near the bottom of the sedimentation compartment so that upward flow occurs. The water to be treated may enter near the top of the tank, in which case coagulation takes place in a central compartment having downward flow or it may have chemicals added first and then be introduced directly into the already coagulated water in the bottom. As the rate of rise of water in the sedimentation compartment is lower than the rate of settling of the heavy floc, the floc remains in suspension to a depth of several feet, through which the rising water passes. This so-called sludge blanket provides contact between the settling floc moving downwards and the smaller residual floc moving upwards with the water. This favours the formation of larger floc through the agglomeration of the fine material. The scavenging action of the previously formed floc leads to the precipitation of insoluble compounds, as well as to the effective removal of fine suspended particles which would not be settled otherwise. The purpose is to secure a floc that will settle at a rate twice that of the upward flow of water, so that separation occurs and the floc does not rise much above a level of 1.5 m (5 ft) below the effluent outlets.

The volume of the tank necessary for the complete process is such that a gross detention period of from 1-2 h is provided. This is considerably less than that provided with more conventional units. Often coagulant aids are required for satisfactory operation with the low detention periods that result, especially when the water temperature is low. As these units are small, i.e. have short detention periods, the safety factor is reduced, and careful control is even more essential than with the more conventional units if satisfactory results are to be obtained.

The selection of such units in preference to more conventional water-treatment plants should be based on the ability of the operating staff, variation in characteristics of the water and its flow, ease of flocculation, the relative cost of extra chemicals which may be needed to secure rapid sedimentation and the relative costs of construction and maintenance.

2.6. Processes in use

The lime-soda, alum and phosphate processes are developments of water-softening and sewage-treatment methods, but the ordinary techniques used by municipalities have generally not proved very successful in treatment of radioactive liquids [14, 28]. It has been pointed out [13], however, that although removal of soluble and particulate fission-product debris from weapon-testing fall-out averages only 50-75% (for gross beta activity) with standard municipal water-softening processes, an improvement can be obtained by close control of the processes, and in some circumstances the degree of purification obtained would be significant.

Very variable DFs have been reported for the same isotopes using the same coagulation and flocculation processes [2, 11, 14, 15, 28] and it is sometimes difficult to reconcile or explain these differences. It is, however, apparent that the main reasons for the differences are:

(a) The concentration of the major non-radioactive constituents present;
(b) The concentration of the radioactive isotopes present;
(c) The chemical or ionic form of the radioactive isotopes;
(d) The concentration of precipitants and coagulants used;
(e) The pH at which the treatment was performed.

It is unwise to assume that because a process has proved satisfactory at one nuclear establishment it will be satisfactory at another. Care should therefore be exercised in selecting the process to be used, and methods for doing this are explained later.

It is not proposed in this manual to fully describe the various processes used at all nuclear establishments. The following brief description gives details of the main coagulation and co-precipitation processes in use.

2.6.1. Lime-soda process

A standard method for the removal of water hardness on an industrial scale consists of the addition of lime and soda-ash to precipitate carbonates and basic carbonates of all cations except the alkali metals.

Where only temporary hardness (i.e. bicarbonate) is present, sufficient lime is added to precipitate carbonates including calcium carbonate, but if permanent hardness (i.e. sulphate) is present, excess soda-ash must be added as well to ensure complete removal of the magnesium and calcium as carbonates; the doses of lime and soda-ash are calculated from analyses for temporary and permanent hardness.

In permanently hard waters soda-ash is first added equivalent to the permanent hardness, followed by lime to give the pH necessary for optimum treatment. At high concentrations of magnesium (> 60 ppm) excess lime is required to precipitate magnesium hydroxides at a pH greater than 10.5 and the final effluent then has a higher total solids content than that from waters containing little magnesium. The chemical equations are:

$$Ca(HCO_3)_2 + Ca(OH)_2 = 2CaCO_3 \downarrow + 2H_2O \quad (1)$$

$$Mg(HCO_3)_2 + 2Ca(OH)_2 = 2CaCO_3 \downarrow + Mg(OH)_2 + 2H_2O \quad (2)$$

$$MgSO_4 + Ca(OH)_2 = Mg(OH)_2 \downarrow + CaSO_4 \quad (3)$$

$$CaSO_4 + Na_2CO_3 = CaCO_3 \downarrow + Na_2SO_4 \quad (4)$$

Strontium is also removed by this treatment; the degree of removal is greater that that predicted on the grounds of its solubility product alone, which suggests the formation of mixed crystals of calcium carbonate and strontium carbonate. The removal of strontium parallels hardness removal and is more effective in the hot process, e.g. at 96°C its precipitation is more complete in 10 min than after 24 h at 10°C. Greatly increased removals are obtained if an excess of soda-ash is used above that equivalent to the permanent hardness.

The plant at Oak Ridge has been operating successfully since 1957. It has been described in detail [30] and account has been given of its performance [31, 81] (see Table II). The procedure resembles that described above. It depends essentially on the precipitation of calcium carbonate with sodium carbonate at a pH of 11 from solutions of fairly high calcium content. Removal of caesium is increased by addition of clay containing a high proportion of illite. The overflow from the settling basins is usually acceptable for dispersion into public waters, although trouble has occasionally developed with feed solutions containing complexing agents or excess acid. The removal of gross beta activity has been 53-87%; 80-90% of the strontium has been removed but the caesium decontamination has only reached about 55%. If the residual calcium concentration in the water is kept below 15 ppm, more than 90% of the combined α- and β-activity will be removed, i.e. a DF greater than 10 can be achieved.

Since 1960 the plant at Marcoule has been treating its low-activity effluents (< 1 μCi/ml) with the soda-lime process instead of the calcium-phosphate process used before. The over-all DF is slightly higher (12 vs. 10) and the DF for strontium has increased to an average of 100 [32]. A modified process involving inactive strontium carbonate precipitation after soda-lime treatment has been tested on an industrial scale. The final amount of sludge is three to six times less than that obtained with the soda-lime method.

2.6.2. Aluminium and ferric hydroxide precipitation

A conventional method of water treatment consists in adding aluminium or ferric salts and then raising the pH by addition of lime, soda-ash or caustic soda to precipitate the hydroxides of these metals. These methods have been applied to decontamination of low-activity liquid wastes. The reagents used include alum, sodium aluminate,

TABLE II. LOW-LEVEL WASTE TREATMENT AT OAK RIDGE [81]

	Characteristics of treated waste				Average percentage removal of radionuclides						
Period	Flow-rate (litres/min)	Radionuclides in treated wastes Ci/m^3 × 10^4			Period	$^{89+90}$Sr	^{137}Cs	^{106}Ru	^{60}Co	TRE	Gross beta activity
		$^{89+90}$Sr	^{137}Cs	TRE							
					Sep. 1957 - Feb. 1957	77	--	--	--	79	--
					Mar. 1958 - Aug. 1959	84	--	—	--	86	--
Sept. 1957 Sept. 1959	1020	1.5	1.7	1.8	Sep. 1957 - Sep. 1958	--	21	49	65	--	66
1960	1160	1.5	8.3	5.2	Oct. 1958 - Aug. 1959	--	86	76	78	--	88
1961	900	0.51	0.11	0.53	Jan. 1960 - June 1961	72	81	72	42	81	--
1962	1150	0.34	0.23	0.15							
1963	1250	0.27	0.098	--	July 1961 - Dec. 1964	74	75	75	73	71[a]	--
1964	1320	0.37	0.15	--							
Jan. 1965 Aug. 1965	990	0.22	0.097	--	Jan. 1965 - Aug. 1965	86	81	b.	67	--	88

[a] Average through December 1962

[b] Infrequently detected in untreated waste

ferric salts or crude chlorinated copperas (ferrous sulphate oxidized by treatment with chlorine). The complex reactions [33] which dictate the use of any one of these coagulants are beyond the scope of this manual. However, the basic reaction which results from the use of any of the above coagulants depends upon the presence of alkalinity in some form and the floc that is produced is either aluminium or ferric hydroxide. Hydroxides and basic carbonates of many polyvalent cations are co-precipitated and these tend to be absorbed by the flocculant precipitate; only the alkaline metals and to some extent the alkaline earths are unaffected. In addition to acting as a weighting medium in solutions of low solids content where otherwise the total quantity of precipitate might be too small to settle, the bulky aluminium or ferric hydroxide precipitate also clarifies the final effluent by filtration if the system is so designed that the treated effluent is drawn off through a blanket of the floc. When formed under alkaline conditions the floc is negatively charged owing to adsorption of hydroxyl ions and will therefore adsorb positive ions but not negative ones such as iodide ion. Positively charged suspended matter is also efficiently removed, the suspended particles acting as nuclei for growth of the floc particles. The efficiency of removal is improved by the presence of sulphate ions at turbidities greater than 5 ppm. Sodium aluminate is not used alone but with alum to obtain some special results, e.g. the failure of some very cold waters to coagulate with alum alone may be overcome by adding about 3.4 ppm sodium aluminate with the alum [16].

Generally with alum, soft waters of low mineral content coagulate best over a narrow range of pH values between 5.8 to 6.4, whereas harder waters coagulate quite readily at pH values of 6.8 to 7.8. Ferric hydroxide coagulation can be used at higher pH values. A comparison of different flocs and coagulants, bearing in mind that the efficiency of coagulation varies from water to water, shows that ferric hydroxide flocs usually give better removals than aluminium hydroxide ones because of the higher pH at which the former can be precipitated; soda-ash gives somewhat better results than sodium hydroxide because of a slight removal of strontium and increased removal of other elements as basic carbonates. Phosphate removals are better in hard waters than in soft ones because of precipitation of Ca_3PO_4. The addition of clays such as kaolinite or bentonite to soft waters often improves maximum removal.

At Windscale, United Kingdom, the coagulation method used is to raise the pH of the liquid with soda-ash to 8 after the addition of alum (20 ppm Al) [34]. A similar process is used at Lucas Heights, Australia, where the coagulation is generally carried out at a slightly lower pH and with a lower concentration of alum, depending on the results shown by jar tests. With a suitable design of equipment, the DF obtained is about 10. In the USSR [35, 36], as part of a complex treatment process, coagulation is carried out with ferric sulphate (100 ppm Fe) and by raising the pH to 10 with caustic soda. The ferric salt is found to be particularly useful for effluents containing soap and suspended matter and gives good removals of approximately 90% for the rare-earths, zirconium and niobium, but poorer results for ruthenium and strontium and virtually no removal of caesium. When iron salts are used, it is considered that the multivalent cations, yttrium, cerium, promethium and ruthenium are absorbed throughout the volume of the sludge while strontium and calcium are taken up on the surface only. In consequence, the alkaline

TABLE III. DECONTAMINATION RESULTS FOR VARIOUS WASTES AFTER COAGULATION AND FILTRATION AT THE MOSCOW STATION [57]

	Characteristics of waste before and after treatment											
	N° 1			N° 2			N° 3			N° 4		
	B	A	DF	B	A	DF	B	A	DF	B	A	DF
Gross beta activity												
Ci/m^3 × 10^3	2	1.8	1.1	6	5.2	1.15	6	0.4	15	16	1.2	1.3
^{90}Sr " × 10^5	5	4.1	1.2	1	0.8	1.2	2.5	2.1	1.2	3	2.7	1.1
^{90}y " × 10^5	1.4	0.42	3.3	0.7	0.07	10	0.4	0.08	5	0.9	0.13	7
^{137}Cs " × 10^5	2.8	2.6	1.08	2.1	2.0	1.05	1	0.95	1.05	2.1	1.9	1.1
^{144}Ce " × 10^5	20	2.3	8.7	18	0.9	20	1.3	0.11	12	1.7	0.16	10.7
^{95}Zr+Nb " × 10^5	13	0.12	108	1	0.03	75	1	0.015	67	4.1	0.024	166
^{106}Ru " × 10^5	2.5	0.27	9.2	2	0.25	8	2	0.27	7.4	2.7	0.36	7.5
^{131}I " × 10^5	11	9	1.2	1.5	1.3	1.15	0.3	0.25	1.2	0.6	0.5	1.2
^{35}S " × 10^3	0.6	0.55	1.1	5.6	5.2	1.08	0.39	0.37	1.04	1.2	1.1	1.1
^{14}C " × 10^3	1.2	1.1	1.09	0.1	0.08	1.25	0.2	0.05	4	0.17	0.08	2.1
Dissolved total solids (g/l)	0.6			0.5			0.7			0.6		
pH	7.5			8.5			7.8			7.5		
COD mg O_2/l	55			100			70			60		
BOD mg O_2/l	100			120			85			78		

B: Before treatment
A: After treatment
DF: Decontamination factor

earths tend to be released to a much greater extent than are the multivalent elements. (See Table III.)

When plutonium is the principal contaminant, the addition of iron salts aids decontamination. At Los Alamos([37], and pp. 140-143 of [15]) liquid wastes are treated with ferric salts and lime. The ferric hydroxide co-precipitates, adsorbs the plutonium and binds it to a form very resistant to leaching by water. The process together with filtration gives sufficiently low residual plutonium content to allow direct release of the effluent to the environment. A similar process has been studied and applied in batch operation at Livermore [38]. A more complex treatment for plutonium, in two stages, is used at Rocky Flats [39]. In the first step, neutralization and ferric hydroxyde lime precipitation occur; in the second step, ferric hydroxyde precipitation is provided by ferrous sulphate oxidation with calcium hypochlorite in the presence of excess lime.

2.6.3. Phosphate precipitation

Three sodium orthophosphates, NaH_2PO_4, Na_2HPO_4 and Na_3PO_4, have been used for boiler water conditioning as also have sodium pyrophosphate and sodium metaphosphate (the latter two are converted rapidly into the orthophosphates in the boiler).

Coagulation with phosphate has been very satisfactory at a number of nuclear establishments for treating low-activity liquid waste. Generally trisodium phosphate has been the coagulant but in some places monosodium phosphate has been used. At Harwell the low-activity liquid waste is treated by raising the pH to 9.5 with caustic soda and then adding excess trisodium phosphate. With more active wastes, the pH is adjusted to 11.5 with caustic soda, then tri-sodium phosphate (and calcium chloride if calcium is below 50 ppm is added so that the ratio $Ca^{2+}/(PO_4^{3-})$ is 5/8. To produce a denser floc, ferric sulphate (or ferrous but oxidized at elevated pH) is also added as a conditioning agent to give a concentration of 40 ppm Fe^{3+} (see p. 124 of [12], pp. 143-47 of [15], and [41, 42]). Higher doses for calcium (300 mg/l) and phosphate (ratio $(PO_4^{3-})/Ca^{2+} = 5/1$) have been investigated in the USSR [43].

It is probable that rather than the reaction:

$$3CaSO_4 + 2Na_3PO_4 - Ca_3(PO_4)_2 \downarrow + 3Na_2SO_4,$$

a hydroxyapatite is formed of probable composition $3Ca_3(PO_4)_2 \cdot Ca(OH)_2$. There is evidence [40] that this compound has considerable cation exchange capacity and further that certain cations, notably strontium and plutonium, are incorporated into the apatite lattice.

The phosphate process can remove more than 99% of α-activity and about 90% of the β-activity from wastes containing mixed fission products. The removal of ruthenium varies according to the ionic form in which it is present and the removal of caesium is generally poor.

2.6.4. Specific precipitation processes

Several radionuclides occurring in low-activity wastes should be removed. The coagulation-flocculation treatments are often not efficient enough to attain MPC values, or more generally, a gross β DF allowing discharge of the treated effluent to the environment. Among the radioactive elements that pass through the effluent treatments detailed above are:

$$^{137}\mathrm{Cs},\ ^{103/106}\mathrm{Ru},\ ^{131}\mathrm{I},\ ^{32}\mathrm{P},\ ^{35}\mathrm{S},\ ^{60}\mathrm{Co}.$$

A special case is that of wastes produced by uranium and thorium mining and milling plants, whose effluents frequently contain radium isotopes to such a level that they must be treated before their release.

2.6.4.1. Ruthenium removal: One of the most troublesome nuclides to remove from liquid wastes is ruthenium, since it can be present in the cationic, anionic or non-ionic form. Apart from ion-exchange methods, the best removals have been obtained by co-precipitation with copper sulphide in acid solution. Since it is not always possible or desirable to work at low pH values, one treatment employed at Harwell (see pp. 147-54 of [15] and pp. 211-15 of [41]) is the formation of a ferrous sulphide precipitate by the addition of ferrous sulphate and sodium sulphide at pH values above 8.0. The optimum dosages are 20 ppm Fe^{2+} and 20 ppm S^{2-}. With certain effluents it is necessary also to add a calcium salt (20 ppm Ca^{2+}) to obtain a satisfactory precipitate. This process normally follows the phosphate treatment and is valuable, since it gives an increase of approximately 5% in the over-all removal of the β-emitters. The efficiency for ruthenium removal is variable, depending on the form present, but the process is particularly useful in removing radiocolloids.

In the Marcoule plant, a treatment used on a pilot-plant scale is particularly efficient for nitrato and nitroso-ruthenium complexes. It mainly consists, after preoxidation in acidic medium, of a co-precipitation of lead paraperiodate [44]. This process provides a DF for ruthenium five times better than the ferrous hydroxide precipitation used before at Marcoule.

In Japan, another treatment is under investigation with "organoclay complex". Tertiary and quaternary amine cellulose derivatives are used with bentonite flocculation. Removals of 95 to 99% in neutral condition are reported [45].

2.6.4.2. Caesium removal: Caesium is best removed by ion-exchange methods using natural or synthetic zeolites or synthetic organic ion exchangers. An alternative method is the addition of clay during the normal coagulation process [46, 47].

Some success has been obtained in caesium decontamination by co-precipitation of metal ferro-cyanides [48]. For example: by the addition of 40 ppm of potassium ferrocyanide and 25 ppm of copper as copper sulphate, then, after mixing, adjusting the pH to 8.0 by sodium hydroxide and dosing the mixture with 100 ppm of ferric chloride, one may achieve more than 98% removal [9]. Some other metals are currently used: iron, zinc, cobalt and, more often, nickel. Above pH 6 and 8, respectively, ferric ~~and~~ copper ferrocyanides lose their effectiveness. For ferrous ferrocyanide (partially efficient for ruthenium removal) the useful pH range is between 4 and 9. Cobalt and nickel ferrocyanides may be used up to pH 10 (p. 47 of [9]).

2.6.4.3. Strontium removal: Strontium is generally removed to a sufficient level by the softening step of effluent treatment. Nevertheless, some investigators have studied precipitation of inactive strontium compounds to give better radio-strontium decontamination.

A strontium phosphate process has been described, which is more efficient at elevated pH (p. 47 of [9]). A strontium carbonate process, giving higher strontium removal and up to six times less sludge, has also been studied [32].

2.6.4.4. Other artificial radionuclides: Barium and lead chlorides have been used to precipitate sulphates and tellurates, followed by ferric sulphate to remove the excess barium and lead and to act as a flocculating agent (pp. 68–75 of [2]). Radioiodine, normally present in the anionic state, can be removed to some extent by the use of silver salts, activated charcoal or anion-exchange material (p. 159 of [22], and [43, 49]). ^{60}Co and ^{51}Cr, often present as anionic species, may be removed by ferrous sulphate addition followed by ferrous hydroxide precipitation when the pH is raised [50].

2.6.4.5. Natural radionuclides: When milling uranium ores, mine water is generally contaminated with uranium and radium isotopes leached by the acid biodegradation products of natural sulphides. Another very important stream of wastes containing uranium and radium is that released from damp storage of neutralized barren solutions and leached ore. Some treatments using baryte or baryum sulphate precipitation have been proposed [51, 52]. The rapid sedimentation of baryum sulphate with the aid of straight-chain fatty acid coagulant has been used on an industrial scale [53].

For liquid wastes from thorium factories, only treatment with naturally occurring lead has been used after urano-thorianite nitric leaching and solvent extraction [54].

2.6.5. Combined processes

When the waste stream is variable in nature, either in radioactive or non-radioactive content, a single treatment process may be inadequate. Thus, frequently coagulation-flocculation precedes another treatment technique such as ion exchange or evaporation. At Oak Ridge, for example, the "Scavenging Ion-Exchange Process" uses, after alumina stripping of polyphosphates, an iron and clay flocculation before ion exchange [46, 47, 55]. The plant at Mol, Belgium, uses multi-stage mixing-flocculating equipment before cationic exchange in the sodium form [56]. The same kind of complex process is used in the Moscow Station [57] and in the Juan Vigon Center [58].

Double and multi-stages for co-precipitations are often operated, combining any of the processes described above. At Saclay and Cadarache, a double treatment comprising, first, caesium removal with nickel ferrocyanides and, second, soda-lime treatment, is generally applied. But the equipment allows the addition of other reagents for special treatments if necessary. Marcoule treats its liquid low-level and intermediate wastes with: paraperiodate for ruthenium, followed by nickel-ferrocyanide and soda-lime softening. In Saclay, DFs are usu-

TABLE IV. EXAMPLES OF DECONTAMINATION RESULTS OBSERVED IN 1965 AT THE SACLAY WASTE-TREATMENT PLANT (From CEN-Saclay internal report)

	Waste characteristics							Treatment	Decontamination results			
Tank No.	Dissolved solids at pH 7		Ca^{++} (equiv.) (mg/l)	Gross activity		Specific activity		Fe Cy Ni = nickel ferrocyanide LS = lime soda	Gross β-activity		Gross α-activity	
	Total (g/l)	Soluble (g/l)		β Ci/m³ ×10³	α Ci/m³ ×10³	^{137}Cs Ci/m³ ×10³	$^{89+90}Sr+^{90}Y$ Ci/m³ ×10³		Residual Ci/m³ ×10⁴	DF	Residual Ci/m³ ×10⁵	DF
561	7.8	7.0	36	9.8	3.3	0.64	2.6	Fe Cy Ni + LS	3.0	33	1.4	235
361	5.9	5.4	30	6.4	5.6	0.58	0.51	Fe Cy Ni + LS	5.3	12	0.66	850
5141	5.6	5.3	15	32	2.8	1.0	--	Fe Cy Ni + LS	5.2	61	0.28	1000
2261	3.2	3.1	40	9.0	0.33	0.47	1.4	Fe Cy Ni + Pb^{++}	8.2	11	<0.1	>330
132	4.8	4.25	50	1.3	2.0	0.58	2.8	Fe Cy Ni + LS	4.3	30	0.26	770
3102	5.3	4.85	20	3.3	1.8	1.1	2.2	Fe Cy Ni + LS	2.0	17	0.26	690
193	6.6	6.45	20	31	0.88	2.1	6.0	Ba^{++} + Fe Cy Ni + LS	9.4	33	<0.1	>880
374	4.8	4.65	70	19	0.52	12	3.2	Fe Cy Ni + LS	2.8	68	<0.1	>520
3284	7.8	7.25	60	0.66	8.5	--	--	Fe Cy Ni + LS	0.9	7.4	0.7	1200
3315	4.8	4.3	40	2.4	3.8	--	--	Fe Ni Ni + Pb^{++}	2.5	9.6	0.18	2100
3246	1.57	1.28	15	6.6	0.1	--	--	Fe Cy Ni + LS	2.3	29	<0.1	>100
297	8.65	6.30	28	6.0	0.08	--	--	Fe Cy Ni + LS + Ba^{++}	0.8	75	<0.1	> 80
1278	4.5	4.15	45	22	0.06	11	0.22	Fe Cy Ni + LS	0.96	122	<0.1	>60
52010	3.2	2.8	10	9.5	0.23	8.1	--	Pb^{++} + Fe Cy Ni	1.2	80	<0.1	>230

ally of the order of 10 to 60 with a concentration factor of about 60 (see Table IV).

2.7. Chemicals and materials used in coagulation and flocculation processes

2.7.1. Aluminium sulphate or filter alum is usually assigned the formula $Al_2(SO_4)_3 \cdot 18H_2O$ but its composition with respect to the water of crystallization is somewhat indefinite and as produced commercially the formula is $Al_2(SO_4)_3 \cdot 14H_2O$. It is available in lump form and as granules and powder. It is readily soluble in water and is easily applied as a solution or as dry material. If the treatment plant is located within transport distance of an alum-producing works, which are generally situated near paper-making plants, it may be found economical to purchase liquid alum rather than crystallized alum. Liquid alum is generally a 49% solution of dry alum (17% Al_2O_3) so that on a dry basis it is 48.8% $Al_2(SO_4)_3 \cdot 14H_2O$. Liquid alum is of little value to small plants. The pH of a 1% solution of alum is 3.4.

2.7.2. Sodium aluminate ($Na_2Al_2O_4$) consists of chemically combined aluminium oxide and sodium hydroxide. It is highly alkaline and the commercial form contains sodium hydroxide and sodium carbonate. It is available as a crystal in bags or drums with the approximate composition 15% Al_2O_3, 35% Na_2O, 5% excess NaOH, or alternatively as a solution with the approximate composition 32% $Al_2O_3Na_2O$, 10% NaOH. It is generally not used alone but in conjunction with aluminium sulphate. It may be applied by conventional chemical feeders but hopper agitation is generally required for dry feed.

2.7.3. Ferrous sulphate, commonly known as "copperas", is a granular acid compound ($FeSO_4 \cdot 7H_2O$) and is available in bags or barrels. It picks up water in storage and for that reason a product with slightly less water of crystallization is preferred. The alkalinity of most waters is too low to react with ferrous sulphate to form the desired ferric hydroxide floc because the reaction involves oxidation by the dissolved oxygen in the water, which does not occur unless the pH value of the water is between 8.5-11.0. At this value the ferrous hydroxide released will be oxidized to precipitate ferric hydroxide. Either lime or sodium hydroxide is used to raise the pH. While ferrous sulphate is an acid salt, it is readily applied by either solution or dry-feed equipment, although because of its hygroscopic nature there is a tendency for the material to become moist and to block the hoppers of dry-chemical feeders.

2.7.4. Chlorinated ferrous sulphate. To form ferric hydroxide from ferrous sulphate the ferrous sulphate must be oxidized and this oxidation takes place only at high pH; hence the addition of alkali mentioned above. However, chlorine will achieve the same oxidation irrespective of the pH value. Thus, chlorinated ferrous sulphate is more usually used in water treatment than "plain" ferrous sulphate and is obtained by adding ferrous sulphate to water which has received a strong prior dose (of the order of 10 ppm) of chlorine. This results in the formation of ferric sulphate and ferric chloride, both coagulants. Chlorinated ferrous sulphate is especially suitable to conditions requiring prechlorination. As ferrous sulphate is more readily applied to water than the ferric coagulants, chlorinated ferrous sulphate treatment may be used with conventional chemical-feeding equipment without modification.

2.7.5. Ferric sulphate is available in either the anhydrous or the partially hydrated granular form and may be transported and stored in bags or drums. The dry form has a tendency to pick up moisture and should be in air-tight packages if stored for long periods. The material will dissolve easily in about two parts of warm water to produce a solution of about 40% strength (as $Fe(SO_4)_3$ anhydrous), so a special solution pot must be used with chemical feeders. It can be used as a coagulant over the pH range 3.5-11.0.

2.7.6. Ferric chloride is available in the crystal form ($FeCl_3 \cdot 6H_2O$) in wooden barrels, as anhydrous $FeCl_3$ in steel drums or as a liquid in glass carboys or rubber-lined drums. Ferric chloride is very corrosive and should be handled and stored in contact with inert materials. The crystalline form contains about 60% $FeCl_3$. Anhydrous ferric chloride usually contains about 98% $FeCl_3$ and is free from water of crystallization, remaining non-corrosive to the drums as long as moisture is excluded. The drums must therefore be left sealed until use and their entire contents must be dissolved in water at one time. The strength of the liquid ferric chloride varies from 30 to 45% $FeCl_3$ by weight, depending on the temperature. Because of its corrosive nature, ferric chloride is a very difficult material to handle. It acts as a coagulant in the pH range 4-11.

2.7.7. Trisodium phosphate is available as $Na_3PO_4 \cdot 12H_2O$ in three crystalline forms: coarse, medium and standard and can be transported and stored in bags or barrels. It is not very soluble in cold water but has a high solubility in warm or hot water. The pH of a 1% solution is 11.9.

2.7.8. Sodium carbonate or soda-ash is a white powder very soluble in water. It is used to provide alkalinity to allow coagulation to take place. It is also used as a softening agent for removing non-carbonate hardness. Solutions of this chemical do not clog the piping or orifices of dosing equipment and unlike lime suspensions need not be stored after they have been made up. The reaction of alum and soda-ash produces about half the amount of CO_2 formed when the natural alkalinity of water reacts with alum. No CO_2 is formed when an excess of soda-ash is used because the excess combines with the CO_2 to produce sodium bicarbonate. The sludges produced when soda-ash is used are generally easier to handle than those produced when lime is used. Soda-ash gives somewhat better decontamination results than sodium hydroxide because of a slight removal of strontium and increased removal of other elements as basic carbonates. Soda-ash is practically pure sodium carbonate but should be purchased by specification to contain at least 98% Na_2CO_3. It is obtainable in bags and barrels and can be fed either in the dry form or as a prepared liquid. The pH of a 1% solution is 11.2.

2.7.9. Sodium hydroxide or caustic soda is used to provide alkalinity. It is very soluble in water and is available as flakes, lumps or liquid. The solid forms are hygroscopic and the best commercial grade available contains approximately 99% NaOH. The pH of a 1% solution is 12.9.

2.7.10. Lime is used in coagulation processes to provide artificial alkalinity in liquids to be treated with aluminium or ferrous sulphate and also for water softening. It is used either as quicklime or in the slaked or hydrated form. For small plants hydrated lime is more convenient.

Quicklime (CaO) is available as lumps or granules in bags or barrels. Quicklime in bags will not store over six months without slaking and bursting the bags. Since it varies in quality from 75 to 99% CaO, the quality of the lime must be known before doses can be calculated. It is preferable to use lime with a high CaO content because of the ease with which it may be slaked. Slaking of quicklime must be done carefully. Special slaking tanks are necessary and the minimum amount of water should be used to ensure a high temperature during the slaking period, which is 15-30 min under optimum conditions, but poor grades of quicklime may require much longer. The slaked lime is diluted with water and stored in solution tanks. The diluting water should be cold because calcium hydroxide is more soluble in cold water than in warm water. As the calcium hydroxide form is only slightly soluble, the "solution" is in reality a suspension of the chemical. It is therefore necessary to agitate the contents of the tank continuously to maintain a uniform suspension. Because of the insolubility of the lime suspension, the storage tanks and pipes of the dosing equipment must be cleaned at frequent intervals to prevent clogging.

Slaked or hydrated lime, $Ca(OH)_2$, is a white powder and may be obtained in barrels and paper or cloth bags. It does not deteriorate when stored, does not have to be slaked and contains fewer impurities than most quicklime. It varies in quality from about 85% to 99% $Ca(OH)_2$. It can be fed in the dry form by the use of dry-feed equipment or mixed directly in solution tanks and fed to the liquid being treated. The same precautions must be observed when feeding suspensions of hydrated lime through solution-feed equipment as with quicklime.

2.7.11. Coagulant aids. Coagulant aids, i.e. materials which assist the coagulation process, are used extensively in water-treatment plants, but apart from clays they have found only limited use with coagulation processes for treating low-activity liquid wastes. However, it may be found with particular wastes that the use of a coagulant aid is worthwhile. The most important coagulant aids are described below.

2.7.11.1. Activated silica is prepared by partially neutralizing a dilute solution of sodium silicate with acid or an acid reacting salt or gas; the most common materials used being sulphuric acid, aluminium sulphate, ammonium sulphate, chlorine, sodium bicarbonate or carbon dioxide. This produces minute colloidal silica particles called micelles which grow during the ageing period. Once proper micelle size is reached, further dilution arrests micelle growth and prevents jelling. The reaction must be controlled very carefully so that the alkalinity of the sodium silica is only partially neutralized and the desired colloidal silica is formed without the formation of a gel (water-glass). The use of activated silica is complicated and usually requires close technical supervision. It is desirable that information regarding the process be secured from the suppliers of sodium silicate and the manufacturers of chemical feeders. Automatic equipment for the preparation of activated silica is available from various manufacturers.

Rapid formation of a large, heavy floc is obtained by the use of activated silica with alum and it is also used alone or in conjunction with alum or ferric sulphate to assist coagulation of calcium carbonate in water softening. The dose of activated silica required depends upon the characteristics of the liquid being treated. A solution of activated silica may be mixed with the regular alum solution and applied as a mixture with somewhat reduced dosage (or it may be applied directly to the water) as a fraction of the separate alum dose. The most effective procedure and doses are established by jar test. The improved production of a tough, heavy floc when activated silica is used to aid coagulation is especially valuable in treating waters low in turbidity, when low water temperatures interfere with coagulation or when a heavy floc is needed with high surface-overflow rates as with suspended solids-contact units.

2.7.11.2. Clays. Bentonite, fuller's earth and other adsorptive clays can be used to aid coagulation. However, definite information cannot be given as to when the use of clay would be of advantage, what doses are required, etc. because of the varying characteristics of clay obtained at different locations and their reactions in different waters. It appears, however, that clays assist in coagulation of relatively clear waters by supplying suspended matter around which the floc forms. Some clays swell when added to water and produce a floc when used alone or with a limited dose of coagulant. Their use may improve coagulation and reduce the cost of coagulants. Bentonite is a highly colloidal clay which flocculates physically if there is sufficient concentration of dissolved minerals or any electrolyte. Some clays have ion-exchange as well as coagulating properties. It is advisable to experiment with the use of clay when difficulty is being experienced in the decontamination of relatively clear liquids or when the liquids are polluted with industrial wastes, since some clays adsorb taste- and odour-producing substances, especially oily materials.

2.7.11.3. Calcite or whiting, consisting of limestone (calcium carbonate) pulverized so that more than 90% will pass a 100-mesh sieve, can be used as a coagulant aid. It is especially effective when treating cold, soft waters of low turbidity. It has been used extensively with alum in water treatment generally by the application of about 20 ppm calcite near the point where alum is applied. The powder furnishes numerous nuclei for the start of floc formation. Each granule of powder will gradually dissolve in corrosive waters but remains a solid during the initial period and then contributes alkalinity as it dissolves within the floc, thus favouring both the physical and chemical aspects of floc formation. Calcite is readily handled, stored and applied. The dose needed to secure improved flocculation, and which is not related to the alkalinity of the liquid, is determined by jar test. Usually the calcite is added after the coagulant, but the coagulant solution and the calcite suspension can be thoroughly mixed together before addition to the liquid being treated.

2.7.11.4. Polyelectrolytes. Important advances are being made in the development of polyelectrolyte coagulant aids as patented products of a number of manufacturers. A polyelectrolyte can be described as a material that is both a polymer and electrolyte. While most of them are strictly coagulant aids, there are some which are coagulants in their own right. They are complex organic compounds, usually synthetic. They may be negatively charged (anionic) compounds such as caustic-hydrolysed polyacrylamide, positively charged (cationic) such as polyvinyl pyridinium butyl bromide, or carry both positive and negative charges and therefore be non-ionic. Simply stated, the function of a polyelectrolyte can be regarded as that of a binder. The coagulant reacts approximately as demonstrated by chemical equations. The polyelectrolyte is introduced at a point where flocculation first occurs. It coats the floc particles, tending to bring about coalescence of the floc. Thus floc growth is speeded up and floc, presenting a large surface area, is exposed to the turbidity in the water for a longer period. Polyelectrolytes are effective in aiding coagulation even with doses in the range 0.5-4.0 ppm and some of the newer polyelectrolytes can be used with even smaller doses. There are probably up to 30 polyelectrolytes available and new materials are being added regularly. Sometimes the polyelectrolyte is mixed with bentonite clay to give increased effect.

While polyelectrolytes are being used in coagulation and flocculation processes at several nuclear establishments in the United States of America, United Kingdom and France, their use is not widespread at present. However, it is possible that research now being carried out on polyelectrolytes may revolutionize conventional coagulation and flocculation processes and their use may become more common.

2.7.12. Other chemicals which may be used in coagulation and flocculation processes essentially fall into two classes, precipitants and carriers.

Included among the precipitants are lead chloride to precipitate phosphates, silver salts to precipitate iodine, barium chloride to precipitate sulphates and tellurates and potassium or sodium ferrocyanides reacting with nickel or iron sulphates to precipitate caesium. These precipitants are then carried down by the floc formed by the coagulant.

Carriers are sometimes used to assist decontamination. They consist of an amount of the inactive salt of an element, which when added to the solution, increases the decontamination of the radioactive element, present in a much smaller amount, from the liquid. For instance, the addition of inactive strontium salt will increase the efficiency of radiostrontium removal in coagulation and flocculation processes where phosphates or carbonates are precipitated.

There is no need to use chemically pure compounds of these precipitants and carriers in coagulation and flocculation processes, but it is wise to choose the purest commercial grade available unless the cost is too high. In this case a less pure grade can be used.

2.8. Processes associated with coagulation-flocculation and sedimentation

2.8.1. Filtration of waters issued from sludge removal

Regardless of the type of equipment used to separate flocculent sludges from the treated effluent, this latter still contains small quantities of suspended materials even when clarified. These materials carry co-precipitate radioactive elements and must be removed before the release of the effluent to the environment or to a subsequent processing stage (ion exchange, for example).

Two types of filters are used: sand filters (or other granule materials such as anthracite) and precoat pressure or vacuum filters. The function of these filters is not limited to the straining action of passing only particles smaller than the free spaces between granules. Superficial adsorption mechanisms can complete the removal of the finest particles. The filtering precoats do not lead by themselves to a chemical separation but some studies have investigated their replacement by materials having a dual mechanical and chemical action (utilization of ion-exchange filters). For the selective removal of some radionuclides, the importance of the living layer of microorganisms (Schmutzdecke) frequently present on the top layers of sand filters must be emphasized.

2.8.2. Removal of sedimentation sludge

Sludges from the coagulation-flocculation and sedimentation process have a high water content (> 95% and often > 98%). After being pumped from sedimentation equipment, they are sometimes centrifuged, or physically treated (freeze-thaw process at Harwell) or stored in vaults. More often, thickened sludges and sometimes the whole quantity of the flocculated effluent [59] are processed by filtration.

The equipment most commonly used is the rotary filter [59, 63] with or without a precoat to assist a progressive build-up of the filtering layer and holding of the filtration rate. The resulting increase of sludge volume is usually not excessive compared with the gain in filtering surface.

Precoat filtration tests combining separation and removal of sludges have been carried out successfully at Karlsruhe [68] and at Berlin [69].

After this previous concentration step, the carrying liquid is recycled and the sludges are conditioned for long-term storage (see Section 4).

3. PROCESSES AND EQUIPMENT

3.1. Handling and storage of chemicals

3.1.1. Storage

The storage requirements for chemical reagents depend upon the size of the coagulation-flocculation and sedimentation facilities used. For large units with high flow rate some liquid chemical products (concentrated nitric acid and caustic soda used for pH adjustment) may be stored in tanks, which are supplied from tank-lorries. For these same large units, quicklime may also be supplied from lorries specially equipped for pneumatic transport of pulverized materials; this quicklime is usually stored in silos.

For other reagents, generally supplied in bags or drums, it is advantageous to provide packages having a maximum weight of 50 kg to ease handling in feeders. Delivery of these packages on pallets allows easy unloading and handling by elevators and manual or power trucks. These packages may be stored near the dissolving tanks or feeders in the case of small units. (For safety, care should be exercised to avoid moistening reagents by water.) For larger facilities, if the delivery time depends upon contracts with delivery schedules, a dry and temperate storage place must be provided, if possible in the upper part of the process building to permit the discharge of reagents by gravity into hoppers supplying the feeders or dissolving tanks. These hoppers should be provided with screens for retention of possible foreign matter. In all cases weighing-scales should be provided to measure the reagents used.

3.1.2. Dissolving tanks

These tanks, formerly often of wood, are more and more made of plastic material (PVC or reinforced polyester), which needs no maintenance and shows a total chemical inertia to reagents currently used. They are provided with an agitating device. If the solid reagents are highly soluble and free of significant impurities, they are discharged directly into the tanks. Those which frequently contain insoluble impurities (alum, for instance) are placed in a perforated box acting as a strainer and immersed in the tank. In most cases, a drain should be provided on the bottom of tanks for the removal of settled materials. After completion of reagent dissolution an(when the mixture is homogenized to the required titration, the contents of the tank are conveyed to a proportioning feed tank. The contents of this tank are measured by an external liquid level system, or better, by a float controlling an indicator through an external linkage.

When specifying concentrations, care should be exercised to express them in terms of dissolved reagent weight in relation to the final volume of mixture to be obtained, thus avoiding the common error of adding to a known weight of reagent a volume of water considered representative of its percentage in the final mixture.

3.1.3. Feeders

Clear reagent solutions are now more and more distributed by means of dosimeter pumps, especially in large facilities. Smaller units may use cheaper distributing devices: flow-rate measured through a calibrated orifice from a constant-level tank, Mariotte vessel, etc. One original proportioner consists of an adjustable control of the lowering rate of a liquid pick-up at the surface of the feed tank (pp. 88-89 of [17]). Suspensions of low-solubility reagents are distributed from a feed tank that is constantly stirred by pumps or a mechanical agitator. The distribution may also be performed through dosimeter pumps; in this case the pumps are of the pulsating-hose type (i.e., finger pumps) or by means of a calibrated orifice from a constant-level tank (not recommended because of sedimentation). Distributors formed by tilting cups fitted on a wheel or belt, the speed of which may be adjusted, are a satisfactory system frequently used.

Solid, pulverized reagents are distributed by means of devices including a hopper that is vibrated magnetically or mechanically to avoid blocking and either a vibrating strainer or a rotary sole-plate. These distributors may be provided with continuous weighing equipment having an accuracy of ± 1%. In designing a powder proportioner, hygroscopicity of some reagents (sodium carbonate, alumina sulphate) and dust removal must be taken into account.

Proportioning devices must be checked and calibrated at regular intervals and especially when changing the quality of solid reagents. Solution distributors must be cleaned and checked periodically; the orifice of the inlet piping must be located sufficiently high above the bottom of the feed tank to prevent suction of possible deposits.

3.2. Pre-oxidation

3.2.1. Pre-chlorination

In addition to destruction of bacteria and algae, and through the oxidation it imposes on oxidation-reduction products, the addition of chlorine or hypochlorite has a chemical action which favours the coagulation as well as decolouration and deodorization through bleaching of organic matters. It is advantageous to leave in the treated effluent a small residual free chlorine content which prevents any further growth of bio-organisms. A pH increase requires higher doses of chlorine and increased contact times (pp. 134-71 of [16] and pp. 193-213 of [17]); this fact is based on the evolution of chlorine itself: below pH 5, chlorine is in the molecular form; between 5 and 6, in the form of hypochlorous acid HClO, and at pH > 10 in the form of hypochlorite ions ClO^-.

(a) Utilization of gaseous chlorine. Liquid chlorine is supplied in plain steel containers, dry chlorine is inert to iron. The net weight of chlorine cylinders is 50 kg approximately, the internal pressure being a few kilograms per square centimetre. (Liquefaction pressure: at 0°C = 2.66 kg/cm^2; at 100°C = 41 kg/cm^2.) Cylinders are generally fitted with fuse plugs that allow chlorine discharge above 65°C. Consequently, and also because of possible leakage, the storage room must always be ventilated, especially in the lower part, chlorine being a gas 2.5 times heavier than air. The tolerance in air for continuous exposure is 1 ppm; it may be sensed at 3 ppm and irritates the throat at 15 ppm. Leaks may be detected by fumes produced in the presence of ammonia. In areas where chlorine is used, it is advisable to provide gas-masks and anti-chlorine potion as a first aid in case of severe poisoning. In this latter case the sufferer must be sent to the nearest hospital, taking care to avoid chilling.

Two methods of using gaseous chlorine are known: "direct feed" and "solution feed".

In a direct apparatus, the gas is applied through some type of diffuser, usually a carborundum stone which divides the gas into very small bubbles making it easily absorbed by the water. In the solution feed application, which is most widely used, the gas is dissolved in a small flow of water under a slight pressure and the solution containing chlorine is dosed with the treated effluent. There are many types of chlorinators commercially available, some of them delivering as low as 0.05 kg chlorine per day fitted with a chlorine flow recorder and, in several cases, with oxidation-reduction potential meters. When using chlorine, attention must be paid to the temperature of the cylinder. The outside temperature must not drop below 10°C. For this reason, it is wise not to use more than one-third of the total load of a cylinder in 24 h to avoid a drop of temperature by too fast evaporation.

(b) Using hypochlorites. Hypochlorite solutions prepared from high chlorine content hypochlorites are used for small plants. The reagents used are generally calcium or sodium hypochlorites prepared by diluting with water high-strength solutions (about 70% for calcium hypochlorite) to 0.5 to 1.0% (expressed as free available chlorine). In France, the solution is rated in chlorometric degrees, each degree representing an oxidizing power equivalent to 3.17 g of chlorine per kilogram of solution.

The hypochlorinator most used is a small motor-driven diaphragm pump, capable of easy variation of the stroke length or stroking rate (or both), which is connected to the effluent feed pump to start and stop in step with it.

(c) Other chlorinating reagents. Apart from chlorine and hypochlorites, only chlorine dioxide has received attention and has been used in industrial water treatment. It is a very powerful oxidizing reagent.

3.2.2. Ozonization

Ozone (O_3) is obtained by flowing thoroughly dry air or oxygen between two electrodes, the potential difference of which is such that electrical discharge occurs through ionization of air.

Ozone is a very efficient sterilizing and oxidizing agent featuring many advantages; in particular, it does not leave prejudicial decomposition products and allows destruction of complexing and chelatant agents [26]. It is not very soluble in water. This solubility, which cannot be determined practically, because of the fast decomposition of ozone in water, decreases with an increase of temperature and pH. This latter tendency facilitates oxidizing reactions. Ozone features the same coagulating, bleaching and deodorizing effects as chlorine and its by-products. One exception: in the presence of manganese traces, the ozone causes a colouration (pp. 134-35 of [16] and [70]).

Ozonized air or oxygen (if oxygen is used, the efficiency and O_3 concentration are increased) may be diffused by a porous body as in the case of chlorine. It may also be introduced by means of a mechanical turbine injector assuring fast dispersion (to be avoided in the case of highly foaming effluents). For the best use of ozone, it is preferable to inject it into tray towers countercurrently to the treated liquid.

The ozone content in air is severely limited: 0.2 ppb. Even then, the premises where it is used must be ventilated, and leakage is to be avoided. Residual ozone in the treated effluent eliminates itself in 30 min by decomposing into molecular oxygen (pp. 134-35 of [16]).

The use of ozone is thus very attractive. Only the high cost of the equipment limits its utilization and makes pre-chlorination, which is cheaper, often preferable.

3.3. Coagulation and flocculation

3.3.1. Equipment

Coagulation and flocculation are two steps in the formation of floc. In the first step (coagulation) reagents are quickly mixed with the whole quantity of waste to be treated. Then the floc is submitted to a conditioning, with possible addition of flocculation aids to give it good sedimentation properties. Such reactions may be performed successively and discontinuously in the same tank fitted with a variable speed agitating device; but more generally, the process is continuous and requires successive apparatus in series.

(a) Coagulation. To ensure the best reagent distribution, the agitation after addition of coagulants must be performed thoroughly at the point of discharge during the shortest possible time (flash mixing) to avoid breakdown of the flocculus elements which appear when the floc formation is started.

The coagulation may be performed through the use of static or mechanical systems. Among the static systems, some establishments use mixing basins fitted with baffles which impose on the waters a long and sinuous path, downwards and upwards alternatively, thus creating eddies to ensure the mixing action.

Mechanical devices are more commonly in use: either direct injection of reagents into the stream of liquid upstream from the pumps, ensuring movement towards the flocculation vessels, or, more often, owing to highest efficiency, mechanically agitated tanks. Agitators, driven by electric motors may have variable-speed shafts fitted with helices or turbines. In the case of helicoid agitators the tanks are frequently fitted with baffles to avoid cavitation by creating swirling currents which improve homogenization. The same results may also be obtained by off-centring the agitator in the tank.

(b) Flocculation. This second step is intended to develop the flocculus initiated during the previous step by increasing the size of particles through slow stirring to ensure their contact with all precipitable elements of the liquid. This step must be performed immediately after coagulation, at slower agitation, slowing progressively, if necessary, as a function of the floc development: and the step must be of sufficient duration to allow complete flocculation, in other words to prevent later precipitation in the liquid carrying the precipitates.

Flocculators may also be classified into two types, static or mechanical. Baffle basins appear among the static flocculation apparatus, but practically, they are not used for treatment of radioactive wastes.

Mechanical flocculators are highly preferred and, in most cases, consist of variously shaped tanks fitted with hydraulic or mechanical agitating devices. In the latter case, the agitating shafts may be either vertical or horizontal. The main characteristic of this device is to be slow and, if possible, adjustable. The extreme displacement speed of blades or helices ranges between 0.2 and 0.6 m/s, the maximum displacement rate of the treated liquid being half this rate.

Some flocculators use compressed air as the direct source of agitation energy; but in this case, not much recommended, since there is a risk of scum or foam formation. Furthermore, this technique does not ensure a holding in homogeneous suspension.

The flocculator-settler, in the form of suspended-solids contact units, is described in section 3.5.

3.3.2. Operation and controlling factors

Important factors for coagulation, in addition to the admission point and the nature of reagents, are the duration and intensity of agitation. These factors are determined by laboratory tests peculiar to flocculation (jar tests) as a function of pH, coagulant dosage, effluent temperature and the shape of the tanks and the type of agitation. The time needed for mixing the coagulants varies from a few seconds to 3 or 4 min, but is usually about 2 min.

The same parameters are taken into account for flocculation; but the agitation velocity, its duration and type are of primary importance. The velocity and duration of agitation are somewhat related. If the velocity is relatively high, the duration is short; however, a sufficiently long flocculation period (15 to 30 min) is always preferred, especially if effluents are at a low temperature, this latter factor limiting the diffusion rate and consequently the development of the floc. Of course, this duration increase involves an increase in the capacity of the flocculators and a compromise must be investigated between their size and the construction requirements of the unit.

3.3.3. Corrections and aids to control

Difficulties that might be experienced when operating a given flocculation facility may result:

From a wrong selection of coagulants or their erroneous dosage, or from changes in the waste during the process in respect to the representative sample submitted to laboratory tests;

From the fact that the facility is not adapted to a particular treatment.

In the first case, it is easy to resume the laboratory tests by modifying all the variables to obtain a correct procedure. On the other hand, in the second case, where reproducibility of laboratory test results is not observed, an attempt should be made to re-create in the laboratory the physical and mechanical conditions of the process unit; in particular, agitation velocities must be determined by measuring the velocity and the shape of currents (for instance, by dispersing small objects or by adding dye solutions). Assuming these conditions are satisfied, the effect of the various coagulant doses must be studied again with variable flow-rates, thus acting on the mixing duration and on the growth rate of germs which may serve as incipient flocs.

The routine survey of flocculation efficiency is aided by some observations or simple tricks which are a very useful complement to laboratory tests:

The appearance of a well-formed floc during sedimentation (sampled before feeding the settler) must be such that the supernatant is absolutely clear and without any milky or cloudy aspects and without non-settled particles;

The velocities and the forms of eddy currents as well as the presence of dead zones in the flocculators may be determined by throwing into the vessel, filled with water to the normal level, a handful of confetti or thin pieces of coloured plastic and visually estimating their dispersion patterns.

It is also possible to use a dye material such as fluorescein, but this method is useful only so long as the incoming liquid stream remains perceptible by its dilution of the dye material.

3.4. Sedimentation

3.4.1. Equipment

The purpose of sedimentation is to ensure the settling by gravity of suspended solids contained in the floc and so to clear the carrying liquid as much as possible. The efficiency of this separation is the controlling element of the whole coagulation-flocculation-sedimentation process, frequently called "chemical treatment". In fact, because the radioactivity concentrates essentially within the sludge to be settled, traces of sludge swept along will have a direct effect on the decontamination obtained in case of direct release or on the efficient duration of runs in case of subsequent filtration.

Sedimentation equipment applicable to treatment of radioactive wastes may be divided in two classes: static settlers and dynamic settlers. Static settlers, generally with large surfaces, are frequently termed "horizontal settlers"; while dynamic settlers in which streams move continuously in the vertical direction, are termed "vertical settlers" or "accelerated settlers". These latter, due to the reciprocal movements of sludges and flocculated effluents are described in section 3.5.

The static settlers, whatever their shape (rectangular or circular horizontal section), have a low height/ surface ratio. The effluents from flocculation apparatus are admitted into the centre of circular settlers through a small section of perforated cylinder; in rectangular settlers, the effluents are admitted on the side through perforated baffles. Sludges, settled by gravity in counter flow with cleared liquids, accumulate on the bottom and are mechanically scraped [64] by devices which may be coupled to scum removers (p. 113 of [17]). The sludges collected into pits are evacuated by pumping, while cleared liquids are removed by overflowing.

The design of a static settler takes into account the following elements: shape, surface (length and width), depth, flow velocity, retention time, permanent volume of sludge, design of flocculated effluent inlet and design of cleared liquid outlet overflow, and considers the characteristics peculiar to flocculated effluents. Current mean data are the following: ascension rate or so-called "overflow rate" of cleared effluents - 0.5 to 1 $m^3/m^2/h$; retention time ranging about 2 to 4 h; horizontal flow-rate 0.5m/min.

Static settlers must be designed with sufficient allowance for operation during periods of poor flocculation, but oversizing must be avoided because, in addition to investment increase, the volume of retained sludges and waters increases rapidly and may become excessive.

3.4.2. Operation and controlling factors

Preferably, static settlers must operate in a regular manner. Flow-rate variations cause eddies which are prejudicial to the stability of settled sludges. Even small temperature variations cause convection movements which may produce an upward motion of sludge. The presence of tensio-active elements associated with gas releases within the sludge (caused by fermentation, chemical decomposition of carbonates from pH lowering and by so-called "air bubbling" measurement devices) may also produce convection motions and in addition may form at the surface stable scums mixed with sludge.

The flocculated effluent must be distributed in a uniform manner and at a slow rate; it must be uniform in order to distribute the treated effluent over the whole sedimentation surface and thus prevent irregularities in vertical and horizontal flows; the slow rate is required to avoid the breakdown of flocs and the formation of eddies which generate short circuits. However, the inlet velocity of the flocculated effluents must be sufficient to prevent any premature sedimentation in the inlet channel from the flocculator. Then, this channel or pipe must be of sufficient size and free of major roughness. In the case of soda-lime softening, special care should be paid to scaling, particularly at seal locations. When entering the settler, the velocity of flocculated effluent flow must be reduced through perforated baffles or rings. The area of these perforations must be such that the velocity through the holes ranges about 0.05 to 0.1 m/s. On the other hand, the installation of partitions in the settler is not recommended owing to the increase in cleaning and maintenance difficulties they entail.

Another important factor is the linear overflow rate of cleared effluents. It is expressed in terms of volume by linear metre of overflow. Some recommend flow rates of 20 $m^3/h/m$, but such a rate can cause a hydraulic suction of the sludges being settled. Values of 5 to 10 $m^3/h/m$ are preferred. Even if the outfall is fitted with V-shaped weirs, care should also be paid to the correct horizontality of the overflow plane.

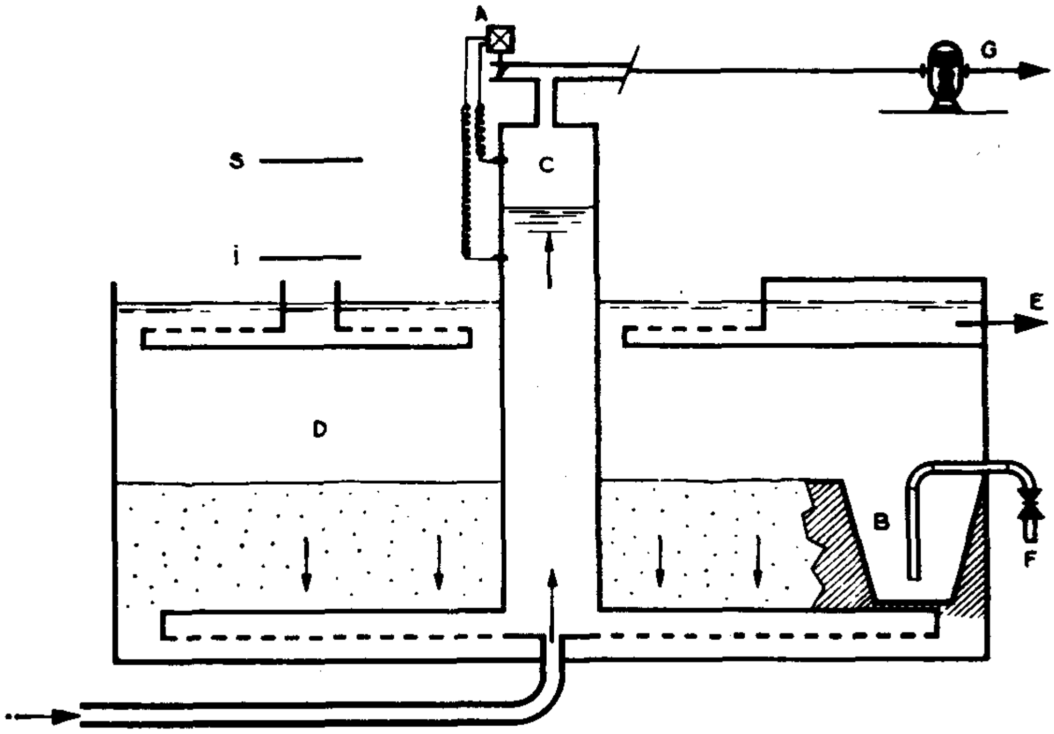
When a static settler is used intermittently, the cleared effluents may be removed by pumping at low flow-rate from a tap immersed below the surface to avoid suction of scums. A photoelectric system (or a sight-hole) installed on the suction pipe, allows the pump to be stopped as soon as the first sludge traces appear. Since the pipe is slanted towards the settler and the pump drains when shut off, the pipe is automatically rinsed.

Several methods may be used to measure the level of sludge in the settler. A transparent calibrated tube may be used as a pipette. Another system is frequently used: a board is secured at right angles to the end of a graduated rod of suitable length; this gauge is sunk into the settler and it is possible either to observe the disappearance of the board into the floc or to sense the change of density owing to the highly settled sludge. In the same way, the disappearance of an immersed luminous source indicates the level of the upper layer of sludge.

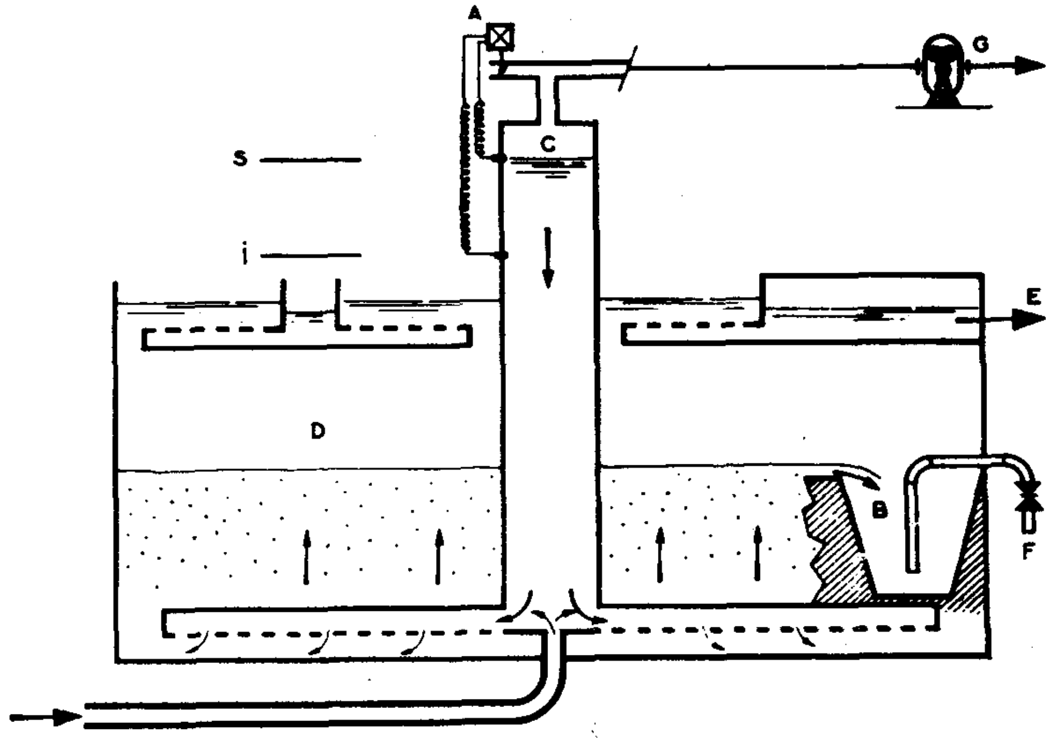
3.5. Suspended solids contact basins

3.5.1. Equipment

This category includes all equipment for handling static or moving sludge layers. For a long time it had been observed that a partial recycling or preformed sludges was beneficial both for accelerating flocculation and for improving separation. Likewise, in a static settler, it may be easily imagined that counter motions of sludges in free fall and of interstitial liquids interfere and have an unfavourable effect. Thus it becomes obvious that feeding a settler by the bottom through the sludge layer in falling-stationary equilibrium offers a dual effect: to complete the flocculation and, acting as a filter, to ensure a more homogeneous sedimentation with only the largest grains falling to the bottom of the settler. These observations gave rise to the design of equipment of various types which ensure both flocculation and sedimentation of effluents previously admixed with



1. First half of cycle: Air valve A is closed. The water rises in the vacuum chamber C. The water in the clarifier D is at rest. The sludge settles.



II. Second half of cycle: The water in the vacuum chamber C reaches level S and the air valve A opens. The water in the vacuum chamber C enters the clarifier D. The sludge in the clarifier rises with the water. The excess sludge enters concentrator B. The clarified water flows off at E. When the water falls to the level i in the vacuum chamber C, valve A closes. The compacted sludge in concentrator B is evacuated via automatic valve F. (This Figure is reproduced by courtesy of Société DEGREMONT, France, [17].)

FIG.1. Operational diagrams

the major coagulants. Included in this equipment applied to the treatment of radioactive effluents, the following may be mentioned:

"Pulsators" (pp. 127-31 of [17], and [71]) consisting of a flat-bottom basin the base of which is fitted with a bundle of perforated pipes, which allow introduction of the effluents to be treated after homogeneization with the coagulation reagents. They are provided in the upper part with an identical bundle of perforated pipes which allow the recovery of cleared effluents without disturbing the vertical stream. The sludges are recovered by overflowing at the upper level of the settling sludge layer. The operation occurs in two steps. In the first step (5 s approximately), the liquid to be treated is sucked by the vacuum in a bell connected to the settler through the distributing systems located at the base of the "Pulsator". This is the settling phase. In the second step (15 to 20 s), the vacuum is broken, the bell is vented to atmosphere, the effluent flows by gravity and crosses the sludge layer in which it is filtered and the cleared liquids overflow into the upper drain. As the sludges accumulate, they overflow during the second phase into a pit, the edge of which is flush with (and controls) the level of the sludge. This pit is mechanically emptied by a periodic pumping. See Fig. 1.

The operation of "Pulsators", which act more as sludge settlers than as flocculator-settlers, is highly influenced by the sludge cohesion.

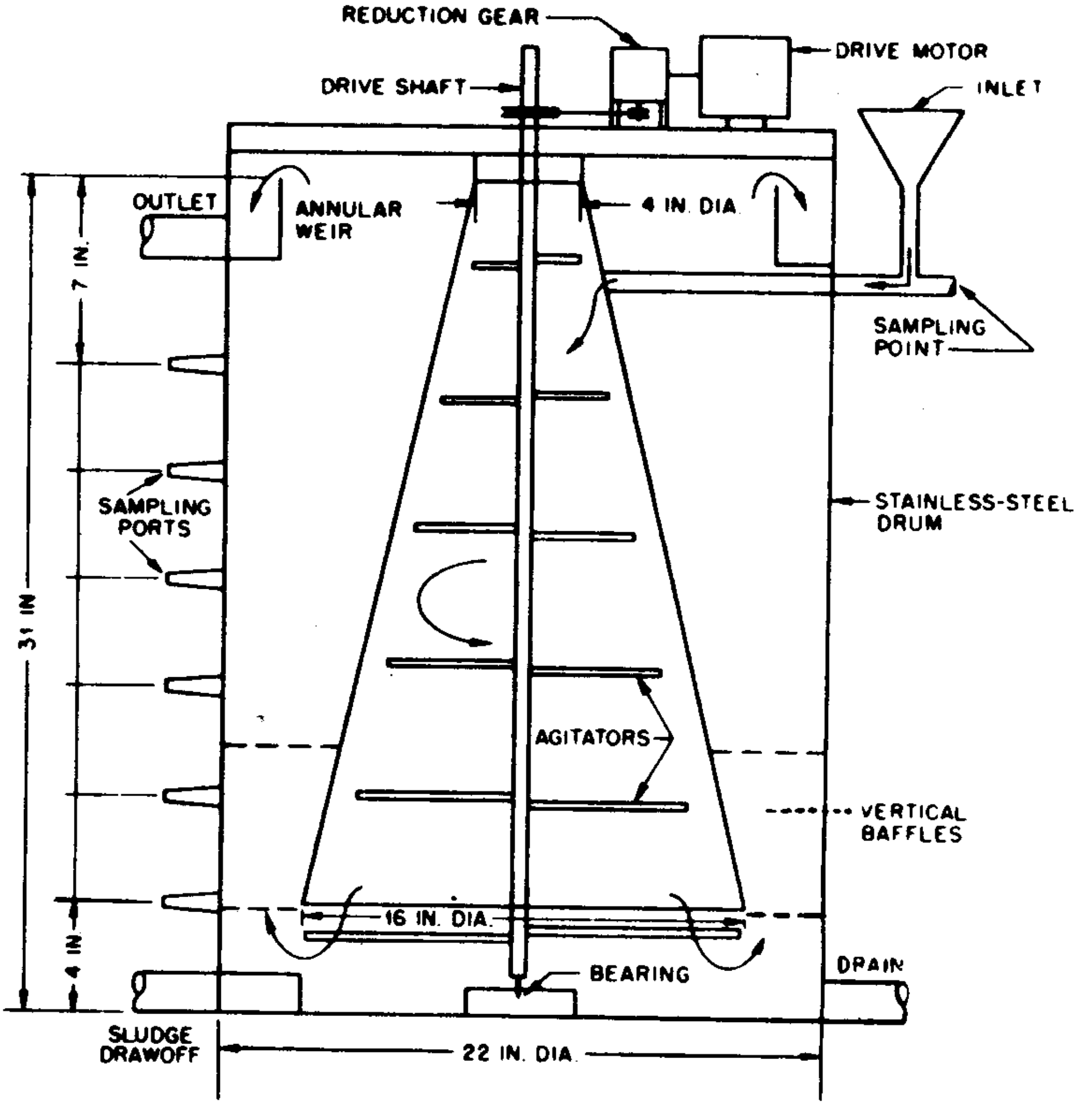


FIG. 2. Spaulding precipitator. Pilot unit.

All "sludge circulation settlers" use an action, generally mechanical, which combines flocculation with sedimentation through a sludge layer.

"Sludge blanket clarifiers" [71, 72] receive the effluents admixed with coagulants in a flocculator section agitated at slow velocity; effluents leave this section and reach a settler section in the same apparatus, in which the water rises and clears through a sludge layer, the level of which is maintained by periodic evacuation.

The "Spaulding Precipitator" (pp. 174-76 of [16]) has a special structure. The effluent to be treated, admixed with coagulants, first comes down into a cone agitated by a paddle stirrer. The widening effect of the horizontal section of the cone from top to bottom is very favourable to the elaboration of well-formed flocs. Then the flocculated effluent flows upwards externally between the cone and a cylinder across a sludge layer in equilibrium. See Fig. 2.

Among the other types used, we may mention the "Clariflocculators" used at Trombay [73], the "Accelerators" used at Mol (pp. 68-75 of [2]), and the "Circulators" used at Marcoule [63]. See Figs 3 and 4.

All these devices, the names of which are peculiar to manufacturing firms, differ in the means used for the generation of hydraulic motions, introduction of liquids and reagents, and removal of sludges and cleared effluents. Some equipment (circulators) may operate in a closed vessel under pressure and are particularly suitable for soda-lime softening.

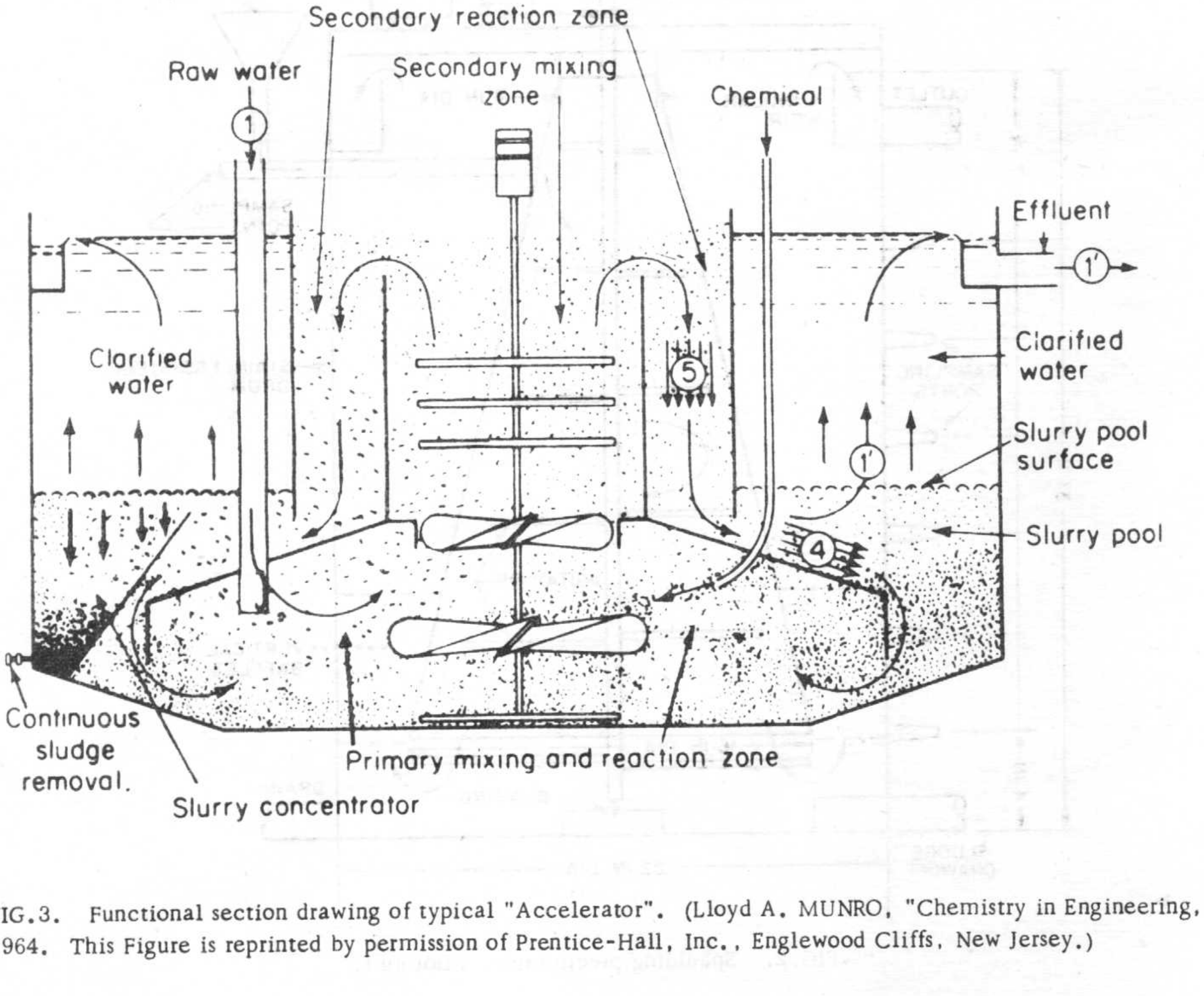
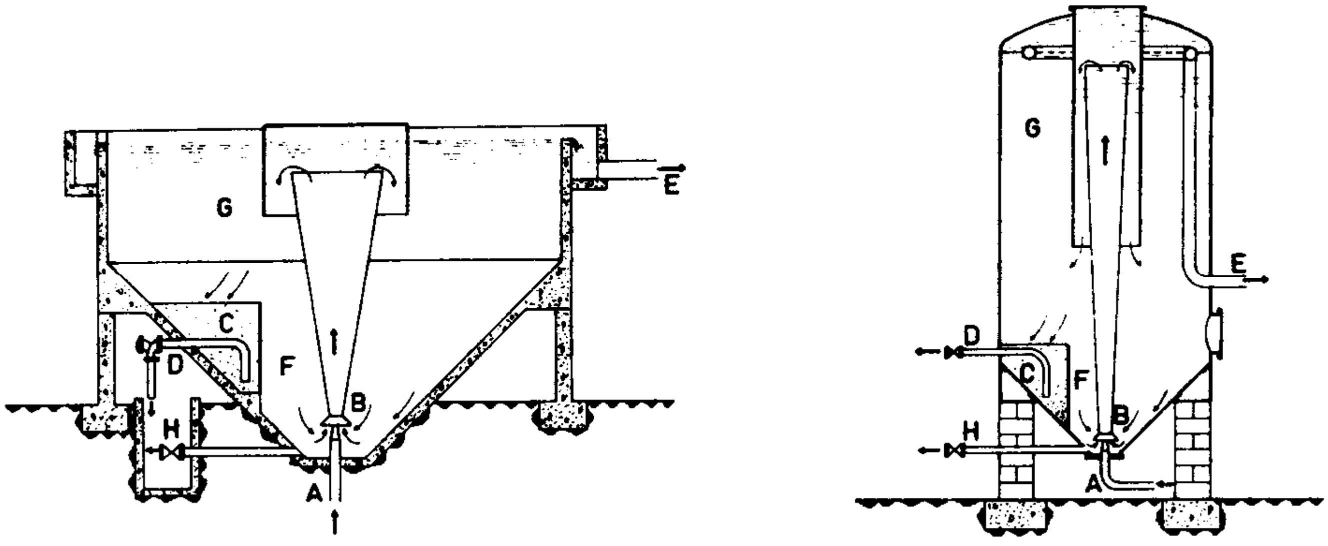


FIG. 3. Functional section drawing of typical "Accelerator". (Lloyd A. MUNRO, "Chemistry in Engineering, 1964. This Figure is reprinted by permission of Prentice-Hall, Inc., Englewood Cliffs, New Jersey.)



I. Open "circulator".

II. "Circulator" under pressure.

A - inlet of influent
B - Circulation ejector for flocculus
C - Sludge concentrator
D - Sludge extraction
E - Outlet of cleared effluent
F - Flocculation section
G - Sedimentation section
H - Drain valve

(This Figure is reproduced by courtesy of Société DEGREMONT, France [17].)

FIG. 4.

3.5.2. Operation and controlling factors

These compact devices combine flocculation and settling. In the agitated section, the stirring action must be smooth and regular in order to create conditions favourable to a fast and stable flocculation. If necessary, this section in particular receives the lime and the flocculation aids. In the settling section, the ascension rate of cleared liquids may reach 3 to 5 $m^3/m^2/h$ if sludges have sufficient cohesion. The height of clear liquid above the sludge must allow the holding of a sufficient margin (frequently from 1.5 to 1.8 m). Sludge removal must be performed regularly and may be controlled automatically through a photoelectric device. The design of the flocculator section must enable fast intervention to correct the dosage of reagents in case of poor flocculation. The residual turbidity of cleared liquids must remain below 10 ppm; 5 ppm is a permissible average.

The correct operation of suspended solids contact basins is also ensured by laboratory control. But, in addition to the tests defining the treatments (jar tests), it is advisable to provide for complementary measures, such as the measurement of turbidity reduction of cleared effluents (by subsequent settling during 5, 10 and 60 min and photometric examination). The efficiency of sedimentation is thus controlled. It is also advisable to measure the concentration factor of sludges in relation to initial effluent, the compressing rate and the density and cohesion of sludge. This latter factor is also important if the sludges are subsequently treated by filtration (pp. 134-36, IV of [18]).

3.6. Removal of sludge

The removal of sludge from the settlers must be performed regularly in order to avoid prejudicial phenomena such as putrefaction. and excessive compression. The various types of settlers are fitted with mechanical devices for the removal of sludge. In static settlers the sludges are scraped from the slanted bottom by scrapers with adjustable tilt and depth. These mechanisms attack the sludge layer at a velocity of about 1 cm/s.

Sludges are discharged into a pit from which they are periodically evacuated by pumps, generally of the suction diaphragm type. Preferably, the pumping operation should be performed frequently and during short periods to avoid clogging of the pipes.

Vertical dynamic settlers and suspended solids contact basins may use the same system, but the removal of sludge may frequently be achieved by gravity through direct-flow automatic control valves. These valves are often controlled by the sludge level.

In spite of all precautions to minimize sludge deposits on the vertical walls of settlers, these must be cleared periodically. Generally, such deposits are not due to chemical sludge but to the growth of living matter such as algae and bacteria. Pre-chlorination and the internal coating of the settler with fungicide paints partially remedy this difficulty.

Finally, for the design of devices intended for removal of sludge it must be kept in mind that the sludge contains nearly all the radioactivity from the treated wastes, with concentration factors ranging from 25 to 200. Consequently, it is advisable to ensure the biological protection of the personnel by taking the necessary measures on the apparatus as well as around receiving vessels.

The sludge from sedimentation may have three destinations: long-term storage in large underground tanks, subsequent conditioning treatment, or mixing with stabilizing elements for immediate disposal. These different processes are reviewed in Section 4.

3.7. Filtration of supernatants - equipment and operation.

3.7.1. Sand filters

This term includes all filters containing a thick layer of filtering material: quartzite sand, anthracite coal, activated coal, etc. They are divided in two types according to the filtering rate of the treated liquids:

(a) Low sand filters have a low filtering rate of the treated waters (2 to 5 $m^3/m^2/d$). They are efficient, economical and easy to manufacture with locally available materials, but their over-all dimensions are such that they are rarely used for applications involving high-flow radioactive effluents. Unless the liquids are chlorinated, sand filters are subject to proliferation of algae and bacteria.

(b) Rapid sand filters are sometimes designed as open tanks or vats. Called "gravity filters" or "mechanical filters" (because of the presence of a mechanical scraper for cleaning), their use is rare and limited to effluents which are hardly contaminated by radioactivity.

The most widely used rapid sand filter consists of a closed vessel in which is deposited the filtering layer and through which the liquid to be filtered flows under pressure. This flow is generally downwards. However, a technique called "reverse flux" is often used, in which the effluent to be cleared passes through layers consisting of grains of decreasing sizes. Some advantages are claimed by the manufacturers: more uniform filtration, decrease of washing volumes, increase of useful duration of filtration, etc.

In rapid sand filters, the filtering velocity for wastes ranges between 3 and 20 $m^3/m^2/h$ and is often near 5 $m^3/m^2/h$. The pressure upstream from the filters is generally supplied by centrifugal pumps and varies from 3 to 10 kg/cm^2 (50 to 150 lb/in^2). Apart from the chemical composition of the filtering material, two main factors govern the efficiency of the filtering medium: effective particle size and uniformity coefficient. The layers consist of a bed of gravel in the drainage device surmounted by filtering layers of decreasing granule size (from 1 to 0.4 mm, according to the case). The present tendency consists in obtaining a layer of almost uniform grain size. The operation is governed by the pressure drop. When it becomes excessive, the filtration is stopped and the filter is regenerated. Thus, if a continuous filtration is desired, at least two filters should be provided, one being regenerated or standing by while the other is in operation. Regeneration is performed by washing countercurrently with or without simultaneous air injection or mechanical stirring. This action enables the reciprocal friction of the grains to free each other from the adsorbed products. If the regeneration is not correct, it will result either in short circuits which decrease the useful time of filtration, or in the formation of mud balls which disturb subsequent filtration. Filtering layers must be systematically inspected to check their condition either for mud balls or for possible coalescence of calcium carbonate on the grains which will eventually cause a mechanical jamming.

In this latter case, after laboratory tests for determining doses, treatment by recirculation of acidic waters (very dilute HNO_3, HCl, 2% SO_2 solution) will generally restore the proper condition of the filtering layer.

Rapid sand filters (or anthracite coal filters) are widely used following combined treatment of coagulation-flocculation-sedimentation (pp. 164-65 of [22], and [41, 59, 60, 61, 63]), especially if the initial activity of the treated effluent was high. In the case of very low activities (often the case for high flow-rates) the wastes are sometimes released directly after sedimentation [64].

3.7.2. Precoat pressure filters

These filters are generally used to separate more efficiently the effluents which must be submitted to a subsequent ion-exchange or adsorption treatment. They have specific flow-rates similar to those of sand filters (5 to 15 $m^3/m^2/h$). They consist of a closed vessel internally fitted with a porous medium (candle-plates) intended to receive a filtering layer capable of being removed by reversing the direction of the fluid stream when the pressure drop becomes excessive. If necessary, a permanent addition of filtration aid during the operation enables the maintaining of an acceptable filtering rate (in the case of traces of gelatinous or colloidal precipitates).

Precoat pressure filters offer the following advantages: they ensure a finer filtration (down to 1 μm), have smaller dimensions and for the same filtering surface the volume of regeneration effluents they require is less than that of rapid sand filters. On the other hand, they require a precoat preparation circuit, their pressure drop is higher and their useful filtration duration is lower. In addition, the formation of the filtering precoat requires accurate setting conditions, and if the pressure drops during filtration the precoat is lost. However, these filters are used in the great majority of facilities before ion exchange [62, 65-67].

3.8. Auxiliary equipment and plant

This Section deals with auxiliary equipment most commonly used for "chemical treatment" of radioactive effluents: piping and connections, valves, pumps, flowmeters, level indicators, pH-meters. It takes into account the experience gained from various difficulties experienced during their use and the cost of this equipment.

Storage tanks for use before and after treatment are not described; we only mention that it is better for these tanks to be of closed design (to prevent animals from entering, penetration of debris carried by the wind, and solar irradiation, which is the main cause of algae growth).

3.8.1. Piping and connections (pp. 698-712 of [19])

Where pipelines are used to transfer liquid wastes, it is essential that there is no possibility for cross connections with any other pipeline system, particularly those for potable water distribution. Therefore, firstly all pipelines carrying wastes should be easily identified, and secondly no possible fittings should be used or practices allowed which will permit back-siphonage. All fixtures which have water inlets below their overflow levels may, if the water pressure is off and the fixture full of water, permit siphonage into the supply pipes. This defect is frequently found in lavatories, baths and sinks and can also occur with priming connections to pumps handling wastes, and connections to water cooling and condenser systems. It is not satisfactory to rely on gate or check valves, as these invariably leak sooner or later. Modern plumbing fixtures, which will not permit back-siphonage, are not available in some areas, so it is essential that good working practices be followed. Otherwise, not only may larger volumes of liquid become contaminated and thus require treatment, but a safety hazard may be created for persons drinking or using water from the potable system.

Conventional drain piping made of cast-iron, concrete or asbestos-concrete should not be used for carrying radioactive wastes. Even if the radioactivity of these wastes is very low, their chemical aggressiveness may lead to damage of such piping. Therefore, piping used for chemical treatment of wastes is designed according to their chemical composition as well as for decontamination possibilities and ease and safety of maintenance, particularly in regard to connections. Materials most frequently used for waste-system piping may be:

(a) Glass (usually Pyrex). This offers numerous advantages: resistance to corrosion, transparency, lightness, easily decontaminable surface and resistance against thermal shock. Its disadvantages are: breakage risks by mechanical shock and, in acidic medium, attack by fluoride ions (generally negligible at low concentrations and ambient temperature).

Coupling between glass piping fitted with tapered and hardened end fittings is achieved by means of metal flanges and seals (teflon, teflon plus asbestos or rubber base, rubber, various elastomers, etc.) This piping is available in nominal diameters with all accessories: elbows, 3-way unions, 3-way reduction unions, glass/stainless-steel connections, etc. Although relatively expensive, glass piping is easily maintained and replaced and is especially adaptable to small plants.

(b) Plastics are divided into two classes: thermoplastics and thermo-setting plastics.

(i) Thermoplastics

Polyethylene, a material of olefinic hydrocarbon base, is one of the most used. It is cheap, light, flexible, resistant to shock even at low temperature and shows an excellent chemical inertness (except to some chlorinated solvents). Coupling between elements is achieved means of flanges and seals as well as by heat sealing. This material is now in competition with polypropylene, which shows the same qualities and is slightly more expensive but shows a better behaviour at higher temperatures and especially a better resistance against solvents.

Polyvinyl chloride (PVC), unplasticized, is the most rigid of the thermoplastics. It is also the one which shows the best mechanical characteristics, the best dimensional stability as well as the best resistance against aging. It features an excellent chemical inertness (except in the presence of plasticizing solvents such as tributylphosphate, of which traces in solution are sufficient to bring PVC quickly out of use). Unfortunately, it shows a low resistance to shock, especially at low temperature, whatever its manufacturing process. Attempts have been made to replace it by polyvinyl dichloride (PVDC) which shows equivalent properties for the same low cost.

Other thermoplastics may be used but few have been applied in the treatment of wastes: acrylonitrile butadiene styrene, the chemical inertness of which is near that of rubber, cellulose acetate butyrate, which may be transparent, chlorinated polyether, polyvinylidene chloride (SARAN), fluocarbons, etc.

(ii) Thermo-setting plastics

These plastics are usually made of two ingredients: a monomer resin added to curing agents or hardeners to a binder (asbestos, graphite, glass-fibre, etc). They differ by the quality of the resin. This class includes phenolic resins, with low resistance to shock. Polyesters and epoxies are both highly resistant to shock and show good mechanical and chemical resistance as well as good behaviour against aging. However, they are not recommended for use in the presence of pure or highly concentrated acids.

In this category, the design of an original connection should be pointed out: the pipes are supplied with sleeves including the elements required for heat sealing by means of electrical heating through a built-in resistor.

(c) Stainless steels, which feature excellent mechanical characteristics, are frequently used in installations having maximum variations in chemical composition of the wastes. This is the case in plants for reprocessing irradiated fuel elements. Stainless steels are highly resistant to corrosion unless the chloride ion concentration becomes excessive and they are practically indifferent to solvents. For this latter reason, and in spite of some corrosion risks, they are widely used for the various elements of coagulation-flocculation processing plants. The most used grade is an 18.8 stainless steel containing molybdenum or, better, a very low carbon content.

Coupling is very easily achieved by bolted flanges, welding (under inert atmosphere), hammered collets, metal collapsing seals, quick-disconnect connections, etc.).

Selection of an optimum pipe material is determined by the experience gained in the use of a material and by several requirements: chemical resistance, working pressure, availability of accessories and coupling methods. These various factors are often highly influenced by local design conditions.

3.8.2. Valves (pp. 1021-1032 of [19])

Valves are intended to allow or stop a flow as well as to control this flow by constriction of the fluid stream. As far as design material is concerned, valve selection is determined by the chemical nature of the fluid. The choice of valve type is made as a function of the physical nature of the fluid and of the operation required. It should be noted that the use of plastics for valve bodies is often limited to the coagulation reagent circuit.

In the chemical treatment of radioactive wastes, valves are used for clear liquid circuits, either corrosive or not (water and reagents), liquids containing suspended solids, either active or not (effluents in course of processing, lime milk), and sludges.

The most commonly used valve types are:

Ordinary plug valves and globe valves, used for clear water circuits or with compressed air; they allow rough adjustments.

Lubricated plug valves, frequently used for chemical reagents in clear solutions. Accurate flow-rate adjustment is often performed by means of needle valves.

Gate valves, the tightness of which is rarely complete, are on-off distribution valves. They are used on large cross-section piping carrying low solid-content liquids.

Ball valves, the ball of which usually moves inside an elastomer seat, are particularly useful for the distribution of loaded liquids and sludges. Numerous models have been designed, especially as far as maintenance and replacement ease is concerned. This type of valve is frequently used on piping carrying radioactive sludges. They allow only rough control.

Diaphragm valves, the variable displacement diaphragm of which allows sufficiently accurate control, are among the most frequently used. They cause a very low pressure drop and their maintenance, through diaphragm replacement, is very easy. They allow distribution of effluents even when highly loaded with suspended solids.

A variation of this type of valve is the pinch valve (formed by a rubber or elastomer sleeve constricted by means of a clip). However, in spite of its low cost, risk of leakage limits its use to inactive liquids or suspensions.

Butterfly valves, which contain a movable disc rotating about a pin inside a rubber (or elastomer) seat, appear among the most simple and require a low operating torque. They are economic in design and are adapted for liquid or sludge circuits. They may have a flow-rate control function but only a rough one.

Flush-bottom tank valves are a particular case. They are installed at the lowest point of the tank bottom and allow drainage to break any scale or deposit.

Other valves used less currently are: non-return valves (swing or lift-check valves), safety valves, pressure reducing valves, etc.

In the case of a sufficiently important plant, automation and visualization of valve control is often achieved by grouping the control components of valves together with those of pumps and level indicators on a schematic or graphic panel in the plant control room.

3.8.3. Pumps

The types of pumps most commonly in use may be divided into two classes:

(a) Positive displacement pumps are used to move or displace a definite volume of liquid at each pumping stroke. They are distinguished from centrifugal pumps by two characteristics: the quantity of liquid displaced per revolution of the shaft is practically constant and the pressure developed is a function of the load imposed on it.

Among the pumps from this class used for the chemical treatment of radioactive wastes are:

Diaphragm pumps, which are variations of the piston pump. The valve arrangement is the same but the piston replaced by a flexible diaphragm which creates a volume change inside the operating chamber. These pumps are easy to put into service and maintain and are specially used to displace suspensions and sludges.

Gear pumps and internal screw pumps are used to displace loaded liquids and suspensions. They differ by their sensitivity to abrasive products; these latter may be moved only by the second pump type.

Vane pumps, in which the liquid is trapped between sliding vanes that project from a hub rotating to the pump casing; and Moyno-pumps, with a metallic rotor of helical form rolling eccentrically in a fixed resilient stator, elliptical in section with double internal helix, are other types of pumps often used.

Controlled-volume pumps and metering pumps have axial piston pump bodies often fitted in pairs on the same shaft, electrically driven and provided with cams which regulate the stroke of each individual piston. They are used for metering the coagulation reagents as well as the flocculation polyelectrolyte solutions. Special care has been observed in the design of valve boxes to ensure tightness (generally ball-bearings on seats); two of them are often installed in series.

(b) Centrifugal pumps create a fixed pressure head of liquid for given rotational conditions of the rotor. If the pump does not deliver, the mechanical energy is transmitted to the liquid stirred inside the pump body and this latter may heat dangerously.

The most frequent form of these pumps is the volute type consisting of an impeller revolving in a casing. The liquid admitted through the centre along the rotation centre-line is thrown to the periphery by rotation of the vanes and then is delivered through a discharge pipe.

Turbine type pumps are more efficient than the volute type.

The characteristics of a centrifugal pump are expressed in terms of flow-rate per hour as a function of the corresponding static delivery head. The total head is the sum of positive lift (or negative suction head), plus delivery head, plus all friction losses in the pump, pipes and valves.

An important factor governing the choice of a pump is its operating suction capacity. For safety purposes in case of leaks, it is often wise to avoid installing the pumps with positive suction head relating to the feed tank. To meet this situation, self-priming pumps are often used.

A self-priming pump consists of the pump proper, a trap on the suction side and a separator on the discharge side. The separator contains a liquid reserve which, when the priming starts, will create a liquid ring in the pump chamber, thus pumping air from the suction pipe as an emulsion. The air then escapes from the liquid in the separator and, as the suction pipe is gradually freed from air, the pump is primed when liquid coming from the feed tank reaches the inlet of the pump. One disadvantage of centrifugal pumps is stuffing box leakage. Mechanical packings have been designed for normal centrifugal pumps and, for self-priming pumps, studies have been carried out to obtain excellent dynamic tightness when the pumps are operating. This allows a system that has no point of piping or feed tank at higher level than that of the pump inlet.

(c) Other devices to move liquids. Liquids or light suspensions may be moved by air-lift systems or by combined action using vacuum and compressed air (vessels must be vacuum- and pressure-proof). Another means, frequently used for high flow-rates, is the steam ejector.

3.8.4. Flow-meters

Rotameters are made of slightly tapered glass tubes with the narrower end down; inside that tube, a float or bobbin is free to float, taking a position which is dependent upon the flow-rate of the particular fluid flowing through the rotameter. A calibration chart is usually prepared for each special case. The bobbin must , of course, be made of a material not corroded by the fluid. Rotameters are only usable for clear liquids but are used over a wide range of flow-rates and are easily inserted in a pipeline network.

Orifice meters are made by placing a plate having a small calibrated central orifice into a pipeline through which a fluid flows, causing a differential pressure from the upstream to downstream sides of the orifice, that pressure being related to the rate of the fluid flow. This pressure drop is usually measured by a manometer. A calibration chart is prepared for each special case. The position of these orifice meters must be studied on the pipeline to avoid abnormal turbulence. They are very useful, especially for main streams; nevertheless they are not so convenient as rotameters, and like these, are only applied to clear or nearly clear liquids.

A venturi meter uses a carefully streamlined orifice mounted in a special device. Its accuracy is higher than that of an orifice meter and loss of head is lower. They are not used as often as orifice meters of cheaper and simpler design. Automatic flow-rate control of liquids is often achieved through motor-operated valves that use the orifice principle.

Mechanical meters using several systems have been designed to measure flow-rates in piping. We will mention only one example: volume-counters with oval wheels (a kind of gear pump body containing two splined, elliptical wheels). These systems are generally sensitive to deposits or adhesion of solid particles and require the installation of a protective filter upstream.

3.8.5. Level indicators [74]

These may be classified into two groups according to the measurement method: direct or inferential.

Direct methods: Sight glass tubing, cable and float devices, conductivity and heat-transfer methods.

The first method is only rarely used (due to shock risks). The cable and float system is the most frequently used for reagent stocks. The two remaining methods are especially used to detect a high level, indicating a single position through a sudden electrical resistance change in a circuit.

Inferential methods: Hydrostatic pressure, float and hydraulic pressure, pneumatic pressure, float and pointer, differential pressure gauge, magnetic level gauge and capacitance gauge are the principal systems used for detection of levels in tanks.

Of these methods, one of the most common is that using a float and hydraulic pressure variation. It allows, should the case occur, the use of remote electrical or elctromagnetic transmitters.

3.8.6. pH meters

Determinations of pH are made on aqueous solutions or on precipitate suspensions (flocculated effluents). The measuring device normally used consists of the following associated apparatus: an elctrode couple of glass and saturated calomel electrode connected to a d.c. amplifier with a very high input impedance. Control and regulation as well as continuous recording of pH variations are frequently combined with pH measurement. Frequent calibration (e.g. weekly) of the electrode couple is necessary. Sometimes the detection assembly consists of a dual-junction electrode and includes a temperature compensator probe.

When designing the pH measurement circuit, the following rules should be kept in mind: the sensing cell must be placed so as to ease inspection, and electrodes must always remain immersed, even when not in service.

Despite these precautions, pH determination is often disturbed. Because of electrical disturbances (magnetic fields, etc.), erroneous pH measurements may be taken; these errors are often due to scale deposits over the electrode surface. Periodic rinsings are necessary and are carried out before recalibration. Certain manufacturers suggest electrode sets that include automatic cleaning.

Degassing occurs frequently in the pH measuring pot (owing to a reduction in pressure following movement through centrifugal pumps); this accumulation of air may lead to the electrodes being set out of the liquid; it is advisable to avoid this phenomenon by allowing air to escape from the top of the measuring pot.

In practice, the pH measurement is made at a tank overflow point or in the piping. One device frequently used consists of a by-pass circuit that contains a small vessel in which the electrodes are installed; the liquid flows back to the tank (or to the overflow drain). Such a circuit may be timed, in other words it may operate with off and on periods at automatically controlled intervals. In this case, the flocculus may settle and the measurement is then imposed. For pH control, a system with two, or better, three ranges, allows regulation of the chemical reagent distribution (by control of metering pumps or mechanical feeders for pulverized products).

By the possibilities they offer and because coagulation-flocculation reactions must be performed with accurate pH measurement, pH-meters are essential aids for chemical treatment of wastes and their purchase should not be guided only by economical considerations.

3.9. Further treatments

Before release or admission to another processing phase, the treated and filtered wastes are stored in delay tanks. After their characteristics have been determined by various analyses, they are, according to the case:

(a) Either directly released to the environment with or without dilution. In the case of no dilution they are brought back to a pH ranging between 6 and 9 before release. They may also be submitted to a post-chlorination treatment;

(b) Recycled through a new type of coagulation-flocculation treatment if the previous one did not give expected results; or

(c) Sent to further finishing treatment: ion exchange on organic resins or mineral absorbers.

In the case of hypothesis (a), reference [6] gives necessary information for release into fresh water; reference [8] for release into the ground, and references [4] and [74] for release into sea water. In case of hypothesis (b), details of treatment can be found in the IAEA guide book, reference [75].

3.10. Examples of design of plants for specific treatment volumes

The design of a radioactive-waste decontamination plant for a coagulation-flocculation process is guided by the average daily volume of product submitted to treatment.

Examination of units designed in various countries leads to four classifications of plants according to the specific throughputs: first, small treatment units which are not described in the literature, because of their small capacity; they have treatment capacities lower than 500 litres/d. Then, for capacities ranging between 2 and 3 m^3/d we find plants in nuclear research centres that possess reactors associated with various utilizations of the radioisotopes produced. Larger plants are industrial size units. The treatment stations of many nuclear research centres can process between 25 and 100 m^3/d. Only a few centres, collecting their effluents through pipelines, process over 500 m^3/d in plants, the treatment methods of which are very similar to those applied for producing drinking water.

For small plants, operations are performed to treat in one working day (daytime) the contents of a tank of sufficient capacity to supply a volume equal to that of decontaminated waste. This leads to fewer controls and makes laboratory tests more able to predict the suitable coagulation treatment. This so-called "batch" method is often used in nuclear research centres when waste collection is controlled in the production areas and isolated for selective treatment.

For large plants, wastes are collected in large volumes in equalization basins and the treatment station operates continuously 24 h/d at a high rate.

Some examples of plants of various specific throughputs are described below. Where possible, these examples are based on existing plants.

3.10.1. Plants with throughput not exceeding 500 litres/d

For such units, wastes are collected in flasks, bottles or drums limited to 30 litres capacity for ease of handling. The treatment concept may be guided by two points of view:

Either accumulation of a large volume of effluent to be treated, the decontamination of which is not frequent;

Or daily treatment of the volume produced per day.

In the first case, a higher investment cost will be balanced by savings on analyses, controls and tests for treatment orientation leading to an important labour decrease. In the second case, the argument is reversed. The choice between the two concepts depends both upon local conditions and the nature of the wastes (those that are non-miscible owing to risks of corrosion, toxicity, presence of foaming agents or chelatants, etc.) In these plants, the treatment is performed by the batch method.

Thus, in the Kjeller Institute [76], effluents to be submitted to a decontamination treatment are accumulated until they fill a 10-m^3 tank. This latter, designed as flocculator, receives reagents (alumina) allowing a coagulation-flocculation treatment. After the mixing action is stopped, the sedimented sludge is pumped and drained before being incorporated into concrete. Clear effluents are filtered and flow over an ion-exchange device. (See Fig. 5).

A plant may also be designed for a specific volume of 500 litres/d to be treated. (See Fig. 6).

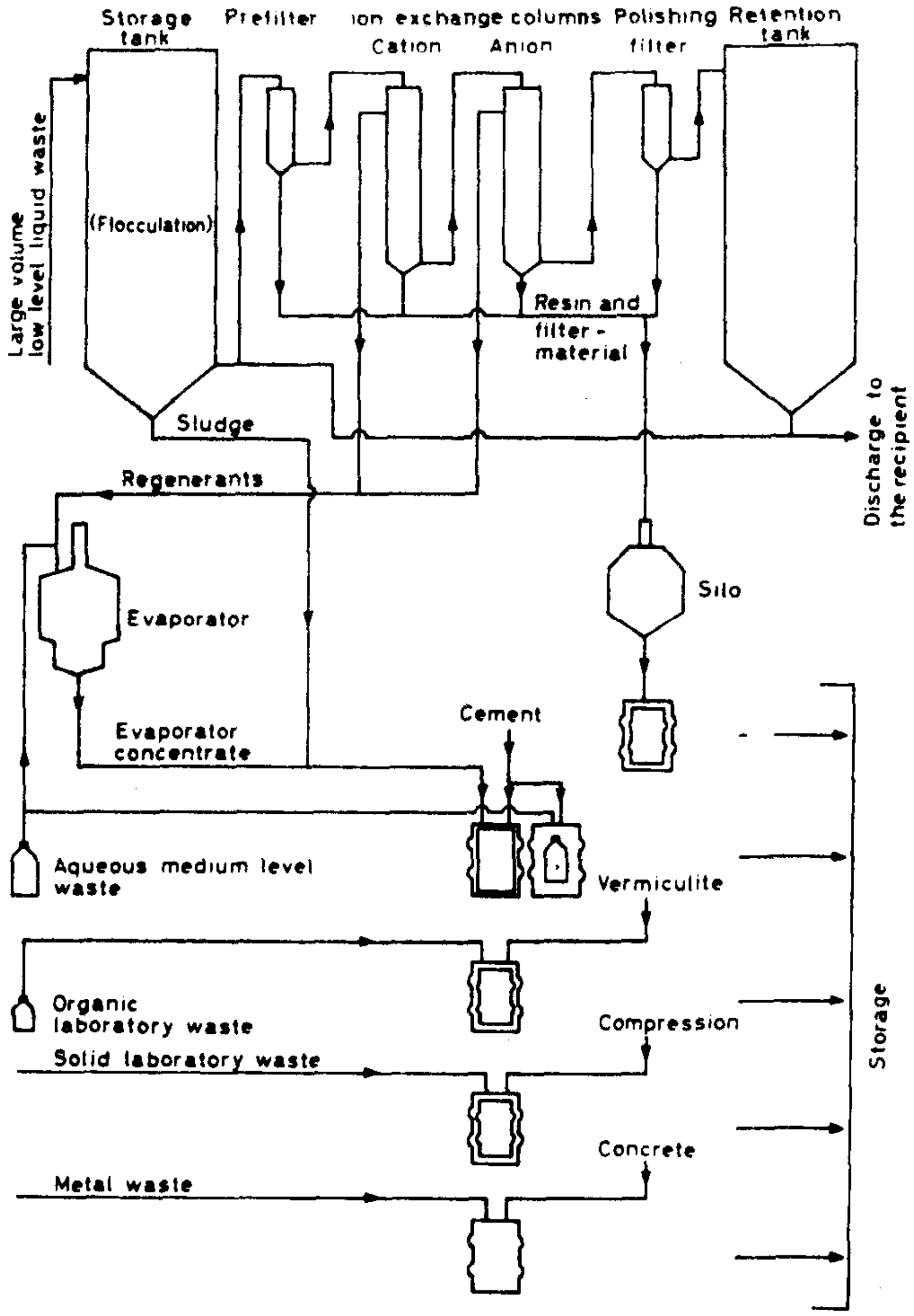


FIG. 5. Flocculation pre-treatment at the Kjeller Institute (Norway). This Figure is reproduced from Practices in the Treatment of Low- and Intermediate-Level Radioactive Wastes, IAEA, Vienna (1966) 151 [10].

Bottles of wastes are emptied into a sink fitted with a strainer (to remove papers and various solid debris) and a device for washing these latter with tap water. Through a siphon in stainless steel, rigid PVC or other plastic piping the wastes are sent to a 500-litre capacity vessel (No. 1) made of stainless steel or glass fibre reinforced polyester. This vessel, provided with a high-level alarm and a cover, is fitted with a fast turbine mixer. It is first intended for homogenization of 500 litres of waste to allow sampling for control and laboratory tests. (A simple sampling device consists in a roller pump squeezing a plastic tube, the movement being manual.) Then coagulation reagents are added to the vessel, titrated in graduated test tubes sampled from a stock of solutions prepared in the laboratory and stored in plastic bottles.

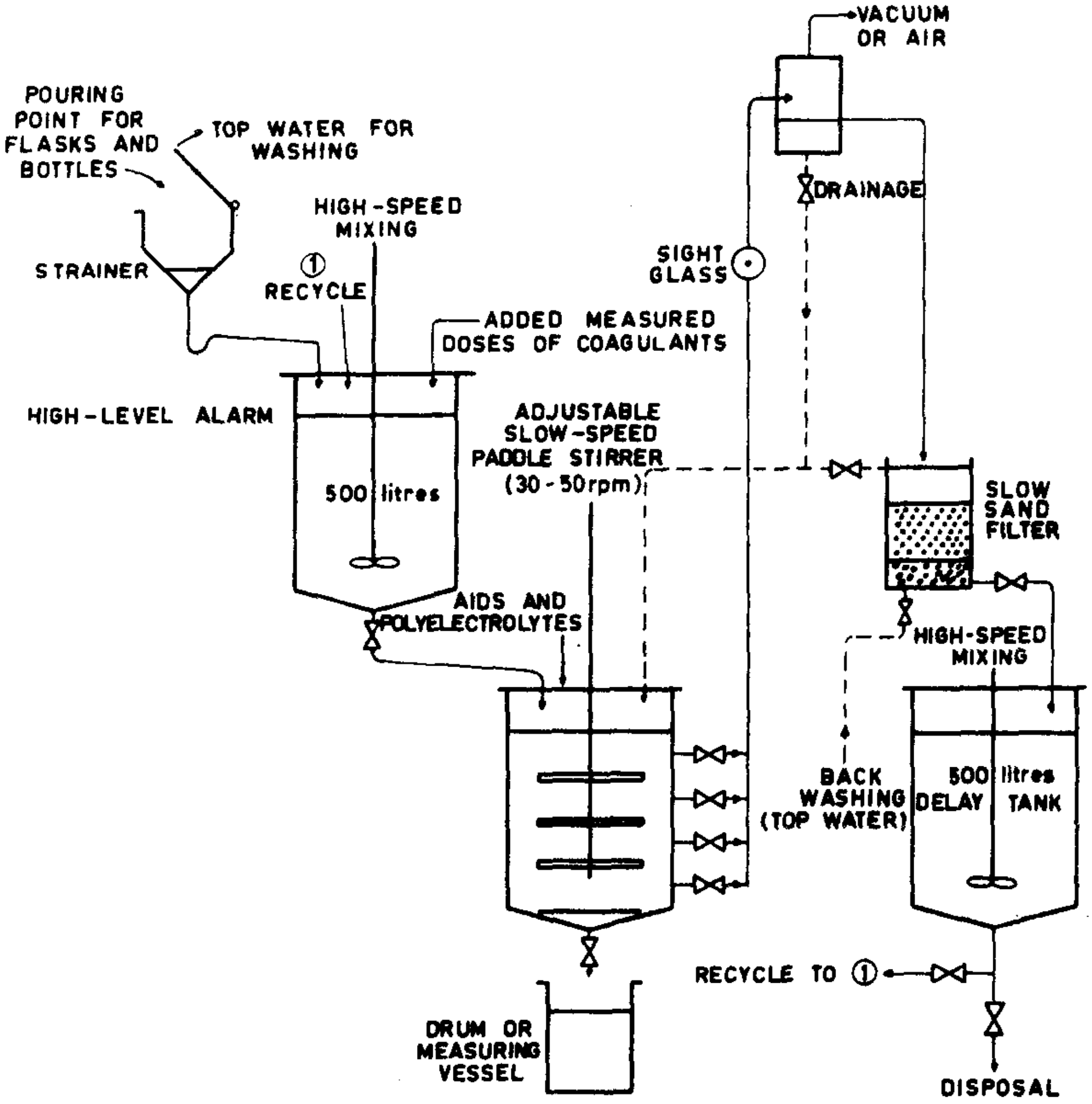


FIG. 6. Schematic design of proposed 500-litre batch-treatment unit.

Mixing is stopped after 3 to 5 min; the waste is then quickly transferred by gravity to vessel No. 2 by means of a pipe of large cross-section. This vessel, the same size and material as the previous one, is provided with an adjustable low-speed paddle stirrer (30 to 50 rpm). In this vessel are added the necessary flocculation aids (activated silica, clays polyelectrolytes). Stirring is maintained during the period required to complete the floc formation (10 to 20 min); then it is stopped and the vessel is used as a settler.

After a few hours (eg. 3 to 4), the clear liquid is sucked by vacuum from lateral piping of the vessel; it flows through a sight-glass and comes into a separation vessel, which serves as the gravity feed tank for a sand filter (containing if necessary other filtering material). This filter may be washed counter-currently with clear water, and water resulting from this washing overflows into vessel No. 2.

Sedimentation sludges from this vessel are drained through the conical bottom fitted with a large cross-section valve, and sent to their final conditioning (incorporation into concrete or concrete + vermiculite).

Clear liquids from the filter are sent into a 500-litre vessel (No. 3) identical to the previous ones, in which the waste is checked before release or recycling through a new treatment.

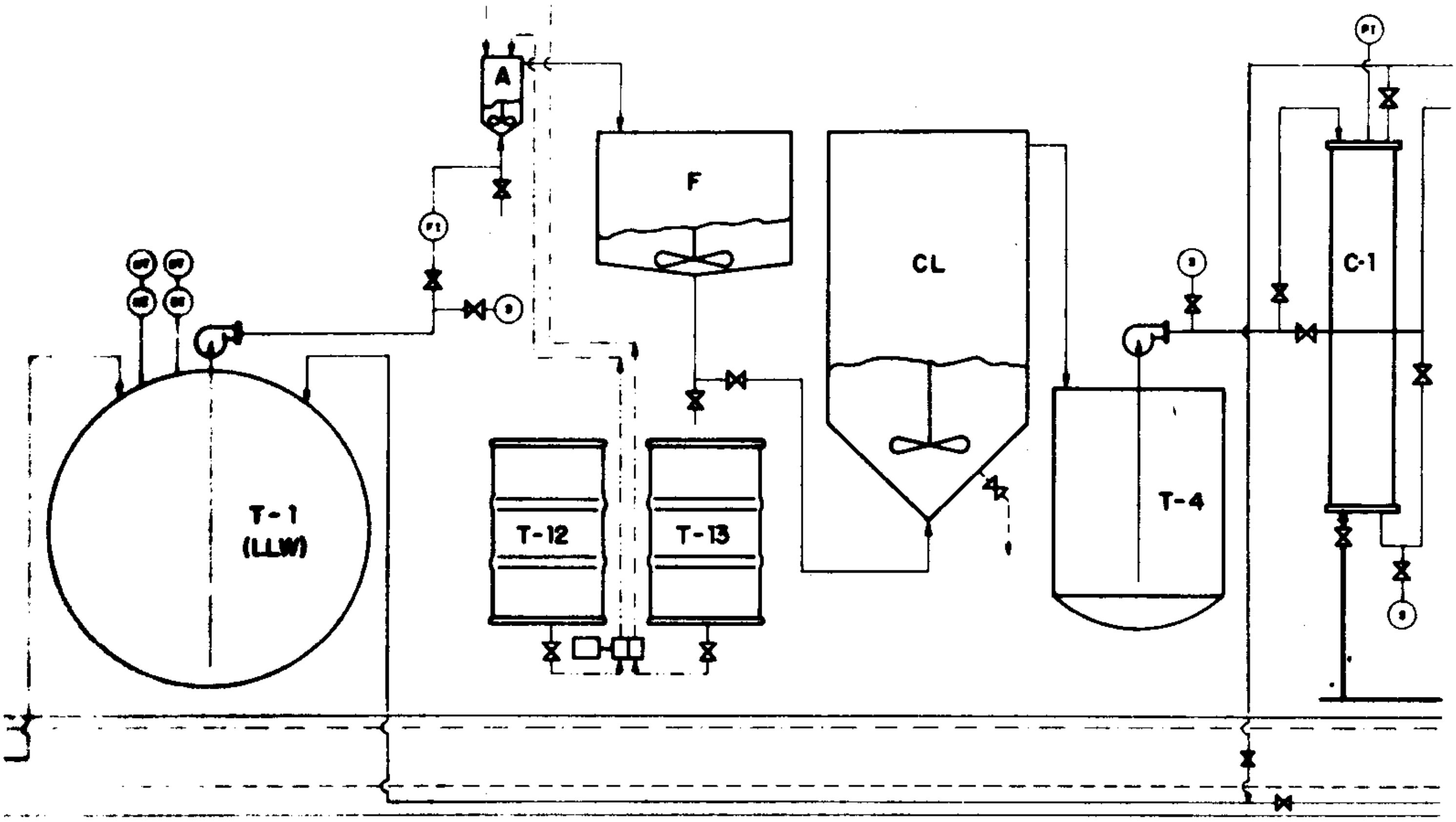


FIG. 7. Flocculation pre-treatment pilot plant of J.E.N., Madrid. This Figure is reproduced from Practices in the Treatment of Low- and Intermediate-Level Radioactive Wastes, IAEA, Vienna (1966) 408 [10].

3.10.2. Case of 2- to 3-m^3 daily throughputs

Here again, the wastes to be treated are collected mainly by means of special bottles and flasks. Hypotheses assumed in Section 3.10.1 remain applicable, although the plants are frequently oversized to cover any further extension of needs. In this case, the plant treats in a few hours the daily production of wastes.

Two designs are typical of these specific throughput levels:

(a) Waste-treatment pilot plant of J.E.N. at Madrid [78]. This pilot plant includes a coagulation, flocculation and sedimentation chain preceding a battery of ion-exchange columns. It allows decontamination of various wastes with a treatment capacity of 2.5 to 3 m^3 during five effective working hours. See Fig. 7.

Low-activity wastes are received in tank (T-1) provided with remote control level and density detectors. They are removed by a pump through a flow-meter and directed to a flash mixer (A) where they receive coagulants (aluminium sulphate and activated silica).

The mixer speed is 1500 rpm. Coagulants are distributed by means of metering pumps from two storage drums, T12-T13. The wastes remain in the flash mixer for 2 min. From there they are sent to a flocculator (F) where they are kept for 1 h, the stirring speed being 1rpm. Then, the flocculated liquids flow into the "sludge blanket" type purifier (CC). They remain in this purifier for 3 h and are stirred at an adjustable speed of 1, 2 or 3rpm. The sludges, periodically drained, are sent to a bitumizing installation. Clear liquids overflowing from the purifier, flow into an intermediate storage tank (T4) from where they are pumped and filtered in an activated charcoal column (C1).

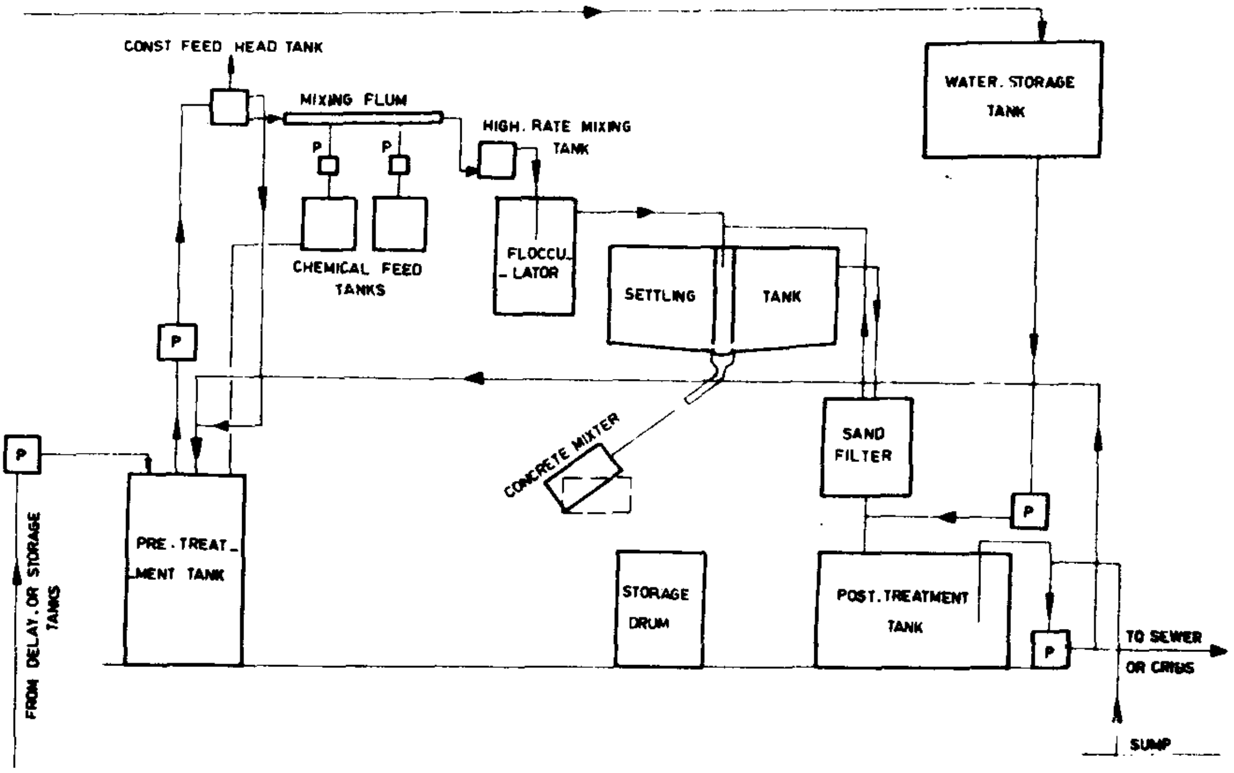
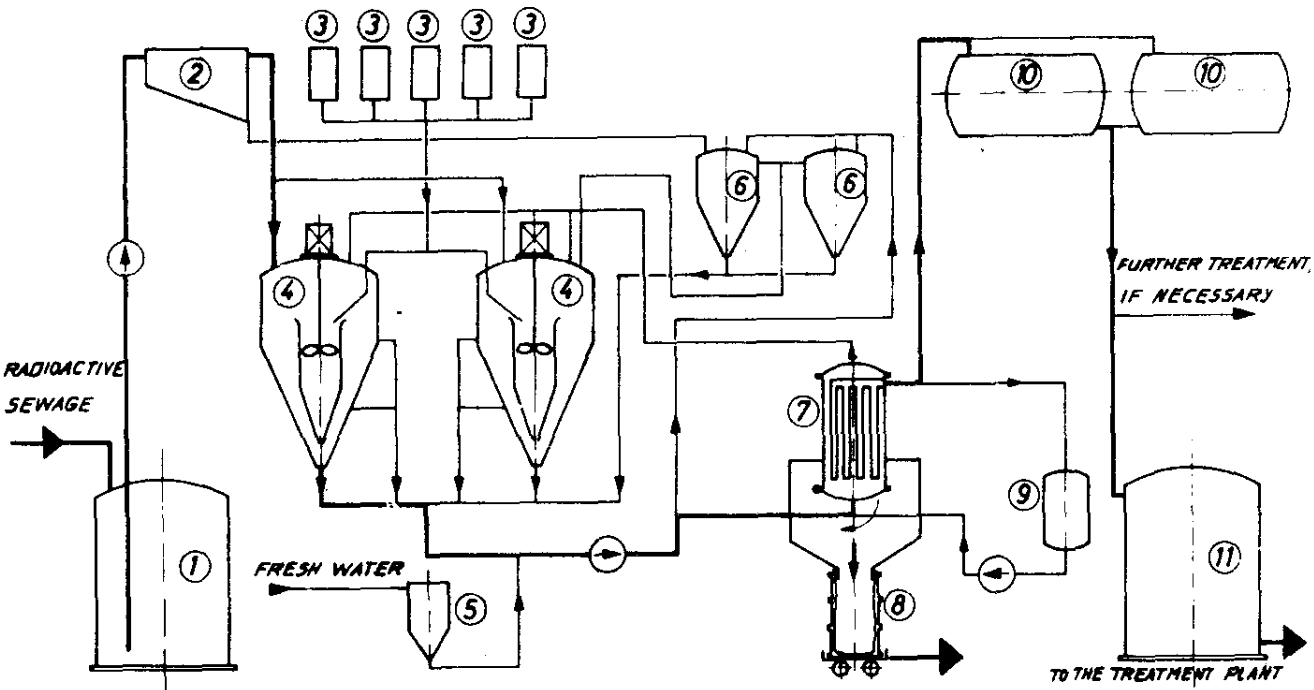


FIG. 8. Flow diagram for radioactive liquid-waste treatment plant of U.A.R. Atomic Energy Establishment. This Figure is reproduced from Practices in the Treatment of Low- and Intermediate-Level Radioactive Wastes, IAEA, Vienna (1966) 415 [10].

(b) Treatment plant of U.A.R. Atomic Energy Establishment [79]. This plant has been designed to treat during the first 5 yr 125 m /yr, and during the next 5 yr 250 m^3/yr. The disposal of de-activated wastes is into the ground, the site conditions being favourable.

The coagulation-flocculation and sedimentation unit has been sized to allow treatment in 2-m^3 batches with a maximum flow-rate of 500 litres/h. The treatment of the contents of one tank is thus performed in a normal working day. See Fig. 8.

Wastes from storage tanks are received in one of two pre-treatment tanks where their pH is adjusted. These tanks, of 2-m^3 capacity each, are made of stainless steel. They are provided with a mixer, level gauges, high-level alarm detectors and filtered vents. From these tanks, a pump raises wastes up to a constant feed head tank from which they flow by gravity to a flash mixer via a mixing flume with baffles. In this latter are added coagulation reagents (ferric chloride, calcium chloride, trisodium phosphate) delivered by metering pumps. From the flash mixer (20 litres) wastes flow into a flocculator (500 litres) with a low stirring speed adjustable from 10 to 60 rpm. Flocculation liquids overflow by gravity into a static settler (2300 litres) where sedimentation sludges are removed through a sludge-measuring vessel into a concrete mixer. Clear effluents overflow by gravity from the settler, flow through a sand filter and are admitted into two post-treatment storage tanks (identical to the pre-treatment tanks). After monitoring, wastes are directed either to a disposal crib through a pipeline for injection into the ground or recycled to pre-treatment tanks. A clear water storage tank allows the sand filter to be washed countercurrently by a pump. Washing waters are sent to the settler. The plant is built over an impermeable tray forming a retention basin. Leaks are recovered in a sump and pumped back to the pre-



1. STORAGE TANK FOR RADIOACTIVE SEWAGE
2. SLUDGE AND GREASE SEPARATOR
3. CHEMICALS DOSING CONTAINERS
4. PRECIPITATION TANK
5. FEED VESSEL FOR FILTER AUXILIARIES
6. SLUDGE TANK
7. PRESSURIZED CANDLE FILTER
8. CASK LOADING
9. ACID TANK FOR FILTER WASHING
10. FILTRATE COLLECTION TANK
11. TANK FOR DECONTAMINATED WATER

FIG. 9. Precipitation and filtration equipment at Karlsruhe. This Figure is reproduced from Practices in the Treatment of Low- and Intermediate-Level Radioactive Wastes, IAEA, Vienna (1966) 321 [10].

treatment storage tanks. Sampling devices are provided at each process stage. Piping and tanks are made of stainless steel; only those with a diameter smaller than 40 mm are made of rigid PVC.

3.10.3. Case of 25- to 100-m^3 daily throughputs

This is the case for many nuclear research centres where waste collection is controlled at the production source to allow segregation. Contaminated liquids are transported mainly by means of tank-lorries, rarely through pipelines.

Two plants are described here: one performing a chemical treatment before ion exchange, the other performing only a chemical treatment.

(a) Treatment of effluents at Karlsruhe [66, 67]. The decontamination chain is designed for discontinuous treatment of wastes by 10 m^3 successive operations. The average daily capacity is 25 m^3. The main coagulation-flocculation processes used are: phosphate-ferric hydroxide, carbonate-ferric hydroxide and nickel ferrocyanide. See Fig. 9.

The wastes received into a storage tank are pumped through an oil and grease separator to one of the conical precipitation vessels of 10-m^3 unit capacity. These cylindro-conical vessels are made of steel lined with rubber. They include a fast mixing zone in which are introduced coagulation reagents. Mixing action is stopped after 5 to 10 min and flocculation effluents settle during several hours. Clear liquids are then drawn off laterally through piping located at different levels and sent to the purifying filter. This latter, of precoat type and operating under high pressure, has a filtering surface of 7 m^2 formed by 12 graphite candles. Sludges are

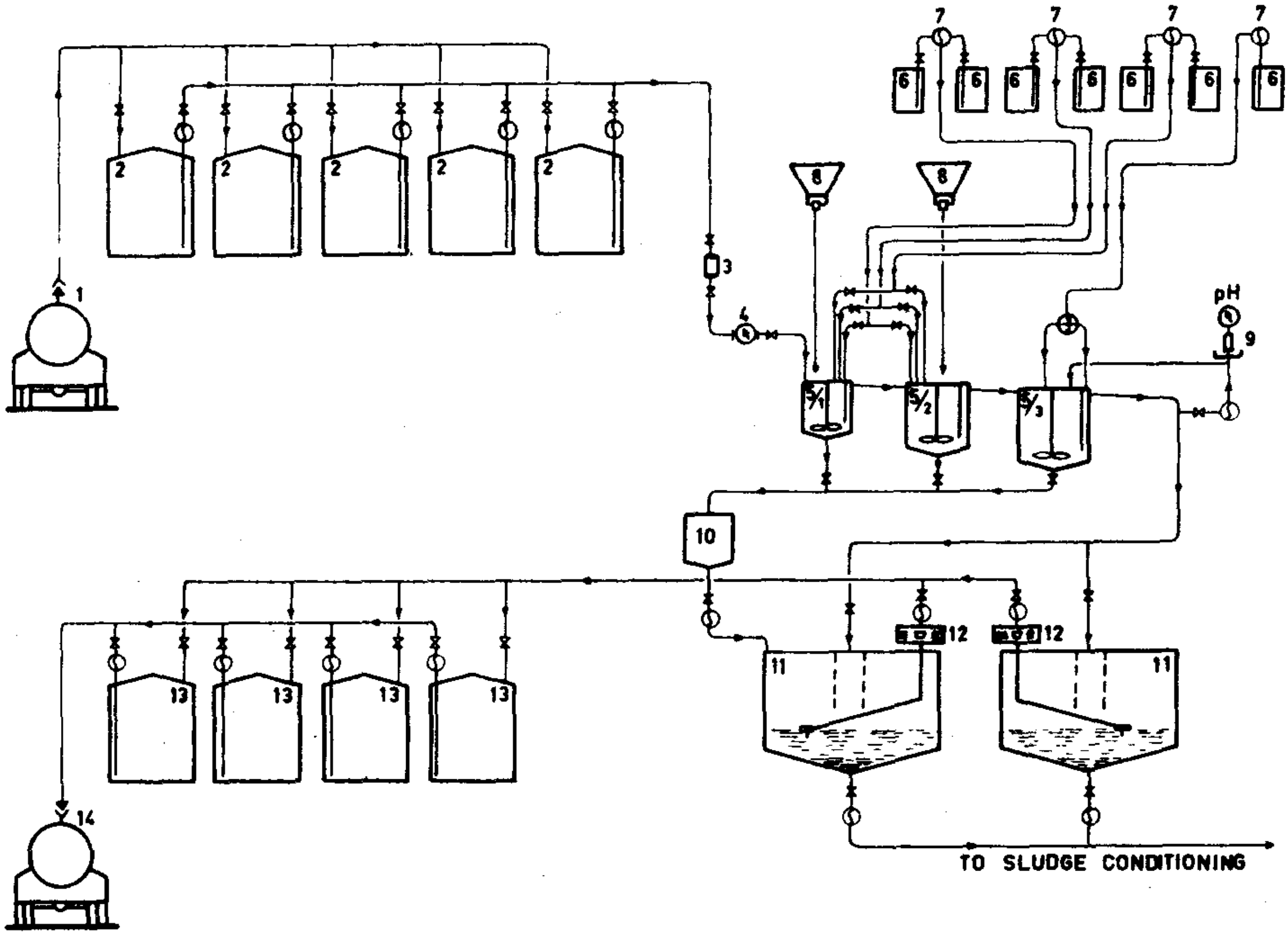


FIG.10. Radioactive waste-treatment plant at Saclay. Flow diagram.

1. Inlet of radioactive effluents
2. Storage tanks before treatment (50 m^3, stainless steel)
3. Grit strainer
4. Rotameter
5. Coagulation and flocculation vessels (0.250, 1.300, 2.0 m^3).
6. Tanks for preparation of coagulents and chemical reagents
7. Metering pumps
8. Dry hoppers
9. Measuring pot for pH
10. Drainage receiving tank
11. Static settlers
12. Turbidimeter
13. Storage tanks after treatment (50 m^3, mild steel)
14. Unloading of treated effluents for disposal

removed from the flocculators only after every three or four operations. They are also sent through the filter. When the pressure drop becomes too high, the liquid is removed from the filter by means of compressed air, thus causing partial drying of the sludge; the air flow is then reversed, thus unclogging the filter. Sludges are drained at the filter base through a tilting movable bottom.

Clear liquids, tested after sampling from one of the two post-treatment tanks, are either recycled through a new chemical treatment, or sent to the ion-exchange unit.

(b) Treatment of wastes at Saclay (See Fig. 10). Wastes to be treated are received in 50-m^3 capacity stainless-steel tanks stirred by recirculation through pumps (self-priming type). These tanks are provided with remote-control level gauges and high-level alarm detectors. Treatment is carried out in 50-m^3 daily batches at the rate of 3 to 6 m^3/h. After the contents of one tank have been homogeneized, a sample is taken for laboratory control and test purposes to determine the nature and doses of coagulants to be used. After a choice of treatment has been made, wastes from the tank to be treated are pumped (stainless-steel pumps and piping) through filters to remove debris. The flow-rate is controlled by rotameter. Wastes are first adjusted in pH in two neutralizers where they also receive a part of the coagulants. These latter are distributed by means of dosing pumps or vibrated feeders according to whether they are in solution or powder form. The neutralizer (5/1) has a 200-litre capacity; mixing is ensured by an adjustable speed turbine from 0 to 1400 rpm. From this vessel, wastes are sent to a reactor (5/2) of 1500 litres capacity, mixing speed from 750 to 1500 rpm, where they receive the remaining coagulants. Then they overflow by gravity into the flocculator (5/3) of 3200 litres capacity, stirring speed 40 rpm. Addition of polyelectrolyte is partially done in this flocculator, as well as in the piping supplying one of the two 70-m^3 capacity static settlers [96].

These circular settlers are 7 m in diam.; they are provided with a mechanical sludge-scraping device on the conical bottom and a sludge-removal system to rotary filters using diaphragm pumps. Operation of these settlers is alternate; while one is fed through the coagulation-flocculation chain, the second is drained or in the course of prolonged sedimentation. After complete settling (e.g. overnight), clear effluents are pumped from beneath the liquid surface through a hinged pipe to avoid pumping scums. A photo-electric turbidimeter installed in the liquid path allows the pumping action to stop as soon as the first sludge traces appear; rinsing of the piping is automatically ensured by the volume of clear liquid which flows back to the settler because of the slope effect.

Cleared liquids, not filtered (although the installation of a filter is foreseen) are stored in 50-m^3 capacity tanks for monitoring before release or re-treatment.

After filtration in rotary filters without precoat (but this precoat may be applied), sludges are incorporated into concrete [80].

3.10.4. Examples of higher flow-rate plants

(a) Chemical treatment plant for wastes at Trombay [73] (See Fig. 11). This is a treatment plant with a capacity of 225 m^3 for eight working hours. It includes mainly pre-treatment storage tanks, a coagulation-flocculation section with mixer and clariflocculator, a filtration and ion-exchange section and vacuum filtration of sludges before their incorporation in concrete.

After pH adjustment in pre-treatment tanks, wastes are pumped to a mild-steel-lined concrete flash mixer where they receive coagulation reagents; the mixing speed is 60 rpm. After this addition, wastes flow to the clariflocculator. This latter includes a main cylindrical body, 7.30 m in diameter and 3.35 m high, made of mild-steel-lined reinforced concrete; the flocculation zone, hung concentrically, includes a cylindrical section 3.60 m in diameter and 1.80 m high, made of mild steel. Retention time for flocculation is 40 min approximately and 4 h 30 min for clarification.

Liquids issued from the clariflocculator are submitted to continuous control to estimate the decontamination obtained; then they are passed

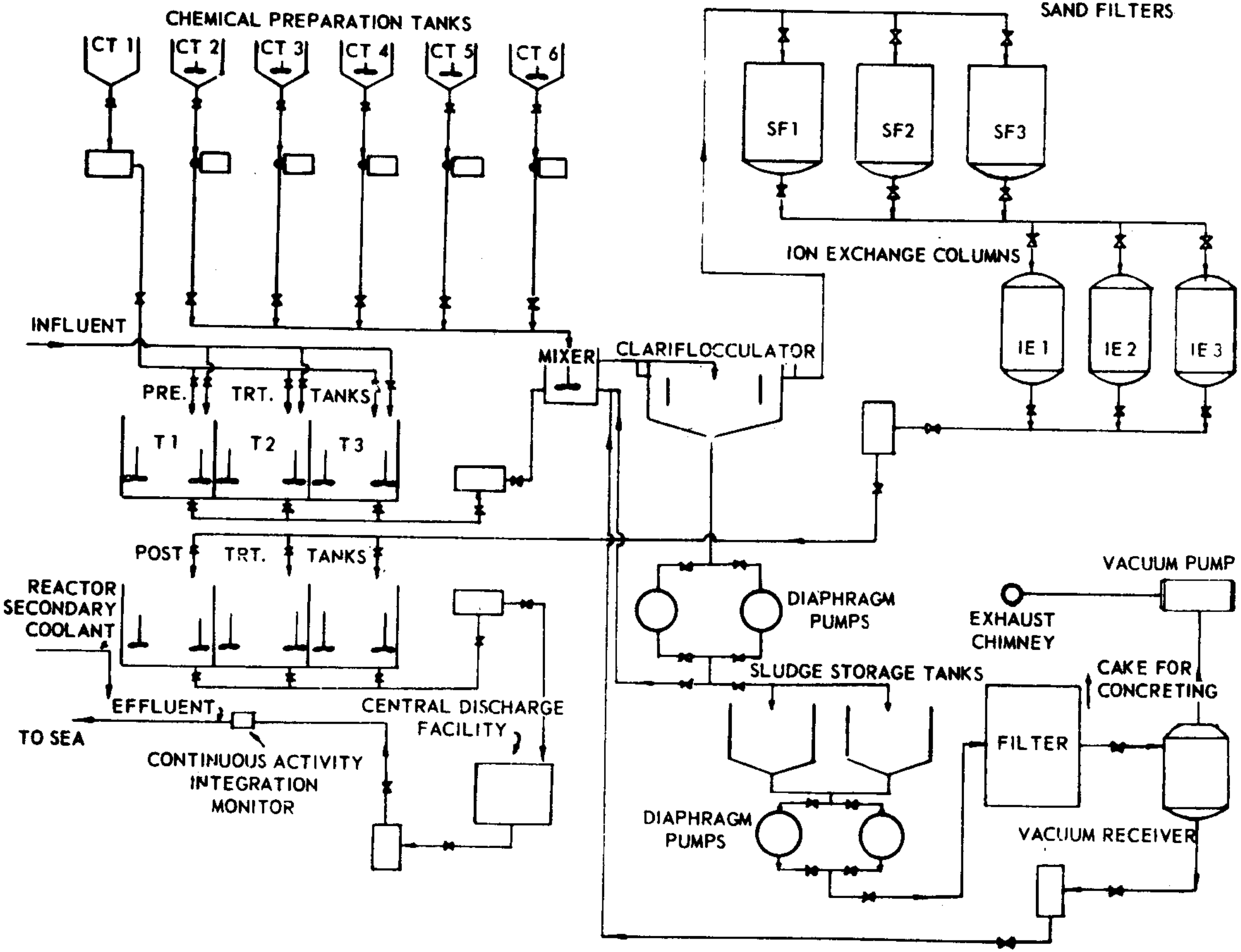


FIG. 11. Chemical treatment plant flow-sheet at Trombay (India). (ThisFigure is reproduced from 3rd. Int. Conf. peaceful Uses atom. Energy, (Proc. Conf. Geneva, 1964) 14, UN, New York (1965)264.)

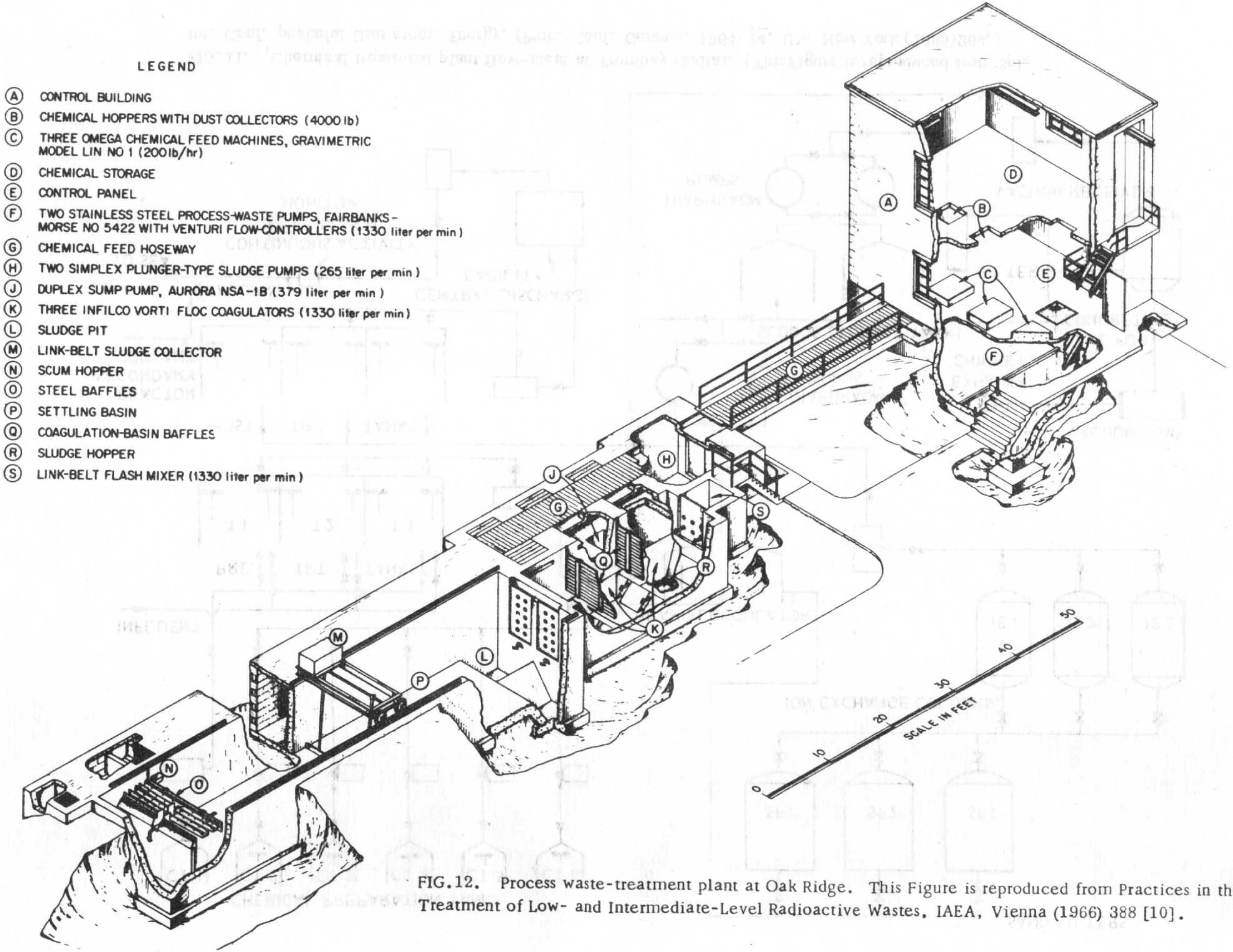


FIG. 12. Process waste-treatment plant at Oak Ridge. This Figure is reproduced from Practices in the Treatment of Low- and Intermediate-Level Radioactive Wastes, IAEA, Vienna (1966) 388 [10].

through sand filters and, after pH readjustment, through ion-exchange columns filled with vermiculite.

Sludges, pumped from the clariflocculator by diaphragm pumps, are stored and then filtered through a precoat-type vacuum drum filter. Filtrates are sent back to treatment. The cakes detached from the filter are received into mild-steel-lined concrete drums. Once filled, these drums are closed with a concrete lid and sent to storage above ground.

(b) Process waste treatment plant at Oak-Ridge. This plant, which has operated since 1957 has been often described [64, 81]. (See Fig. 12).

It is a conventional horizontal-flow water-softening plant using a lime-soda process, or alternatively, lime phosphate (for removal of ^{90}Sr) with clay addition (for removal of ^{137}Cs). The treatment capacity is 1900 m^3/d.

Wastes, previously homogenized in an equalization basin of 3800 m^3 capacity, are pumped at a rate of 1.3 m^3/min by centrifugal pumps, the flow-rate of which is controlled by venturi flow-meters. They are delivered into the flash-mixing basin where they receive coagulation reagents supplied by three gravimetric feeders (unit supply 90 kg/h). Contact time in the flash mixer is 1.3 min.

Then, the wastes cross the coagulation basin, which is divided into three sections by wooden baffles. Contact time is 31 min. Flocculated wastes leave this basin through perforated baffles and flow to the upper section of a settling basin where they are retained for an average of 129 min. These baffles are intended to slow down the waste flow and to supply them evenly at the end of the settling basin. The overflow rate is about 0.5 m/h.

Sludges are removed by means of an automatic Link-Belt collector and then temporarily stored in a 61-m^3 capacity concrete tank before discharge into a pit in a local shale formation.

Cleared liquids are then discharged, after control, into the environment, after having been previously submitted to an additional settling in a 5700-m^3 capacity basin. Feeding pipes from the equalization basin are made of rubber-lined mild steel; pumps, venturi flow-meters and valves are made of stainless steel. These latter, of plug-valve type, are fitted with rubber-faced plugs. Equipment of the unit (flash mixers, coagulators, sludge collectors, etc.) are lined with at least two coats of a protective thermoplastic material (polyethylene) after manufacture.

The general design of the unit offers a flexibility allowing recycling of treated waste and sludges as well as reagent addition at any time during the coagulation phase.

4. CONDITIONING AND DISPOSAL OF FLOCCULANT SLUDGE

4.1. Conditioning of sludge

Sludges issued from coagulation-flocculation processes are sometimes stored for a long term in the same condition as when produced. More often, they are submitted to a treatment allowing a volume reduction through removal of all or part of the water they contain; this treatment is often associated with solidification of the sludges, which confers on them important mechanical and chemical resistance to facilitate their disposal.

A panel of experts convened at the IAEA headquarters in May 1966 studied the technical, economical and safety aspects of sludge conditioning. Detailed information can be found in the proceedings of this panel [24].

4.1.1. Primary concentration

Chemical treatment sludges are concentrated through various techniques: evaporation, filtration, and centrifugation. The 1 to 10% initial solid concentration then can become 20 to 50%.

(a) Evaporation is not usually applied to radioactive sludges from chemical treatment but solar evaporation of such sludges has been used (pp.134-36, IV of [18]): The sludges in open basins are submitted to sun radiation and the moisture evaporates into the adjacent air, which disperses it by atomospheric convection. The method is known to apply only to favourable sites (the yearly average evaporation must at least equal 750 mm) and to well-formed, relatively dense and low-activity sludges. The dried sludges are removed manually and stored in drums.

(b) Filtration. Various filtration methods are used:

(i) Natural filtration, using gravity alone in a horizontal sand filter, is applied at Harwell for low specific activity sludges. Dried sludge is removed manually. This method requires large filtration surfaces, well-formed and low-activity sludges.

(ii) Pressure filtration. Three examples have been described: At Harwell, a horizontal-tank, vertical-leaf filter is used (Sweetland filter). Cakes are formed under 2 to 3 kg/cm^2 pressure and dried by a compressed air flux. This flux, when reversed, allows easy removal of the cakes.

At Karlsruhe and at Berlin (Hans Meitner Institute), vertical candle filters are used, including precoat and cake removal by compressed air. This technique allows handling of higher radioactivity sludges; the residual humidity of the cakes is among the lowest obtained. However, one disadvantage is the risk presented by the use of pressure, which may spread contamination in case of leakage.

(iii) Vacuum filtration is the most frequently used technique. Generally, precoat-type vacuum-drum filters are used (Marcoule, Los Alamos, Trombay, etc.) The filter assembly is compact, hence easily protected and the use of vacuum improves safety. Residual moisture of the cakes may reach 40 to 50%.

(c) Centrifugation appears to be applicable only to precipitates which do not show any thixotropy, therefore excluding a certain number of colloidal suspensions. It has been used for the waste-treatment station at Moscow.

(d) Freeze-thaw, which does not constitute a concentration technique in itself, enables the further solid-liquid separation to be improved. It has been studied in particular at Harwell and has been used before filtration (on a belt filter) and before centrifugation. This treatment does not seem to be justified if sludges are subsequently treated by incorporation into bitumen.

4.1.2. Elaboration of sludge-conditioning schemes

The same types of sludges may be treated according to various methods. When taking into account the safety conditions, experience gained, operating difficulties of the various techniques and economy of processes, it is obvious that a choice may be made as a function of local conditions to select a particular conditioning scheme. The previously mentioned proceedings [24] gives a table listing the advantages and disadvantages of various treatment techniques for preparing sludges for disposal.

4.1.3. Insolubilization of active sludges

Two main techniques are used: insolubilization in bitumen or in concrete.

(a) Insolubilization in bitumen: Sludges are dispersed into hot bitumen or a cold bitumen emulsion. After removal of moisture by heating, the mixture containing 55 to 65% solids is poured into metal drums. Through its water-repelling action, the bitumen reduces considerably the leachability of radioactive materials during long-term storage.

Three methods of using the bitumen may be applied:

(i) Introduction of sludges into very hot bitumen (180-250°C);
(ii) Introduction of sludges at lower temperature with addition of surface-active agents; and
(iii) Cold mixing of sludges with a bitumen emulsion.

The first two methods are used on an industrial scale. The first is used at Mol and Harwell; the second is under continuous operation for the 1-m^3/h flow of sludge at Marcoule; the third is under study in various pilot plants (Los Alamos and Oak Ridge).

This treatment leads to a substantial reduction in the volume of sludges, thus paying for the treatment cost through the savings realized in storage. The leachability has been reported to be about 100 times lower than that for incorporation into concrete and to approach that of glass.

(b) Incorporation into concrete: Sludges are mixed with cement, sometimes with vermiculite added. Where the sludges are especially radioactive or contain hazardous materials that create a risk of serious air contamination, the mixing must take place in a closed container and transfer of the mix to the concrete-lined storage or disposal drums must be made by means of vacuum. The mixture solidifies in the drums, often shrinking to leave a substantial void, but it is often cheaper and safer to waste the void space than to risk opening the drum to make an additional transfer. Incorporation into concrete is used at Cadarache, Trombay, Shippingport, Leningrad, Moscow and other sites. There is no volume reduction (even a large volume increase often occurs) and leachability of the product may not be negligible if the incorporated waste has a high ruthenium or caesium content. However, the process presents some advantages: it is relatively cheap and simple to use, self shielding by the concrete-lined drums occurs (also the containers offer some protection against leaching), and there is a possibility of designing plants to incorporate other contaminated wastes (evaporator concentrates).

4.2. Disposal of sludges

Long-term storage of sludges resulting from chemical treatment depends upon technical, economic and local legal aspects. Various methods are used which may or may not involve a pre-conditioning of the sludges.

4.2.1. Disposal without conditioning

For diffusion into the ground of radioactive ions they contain, sludges present a much lower potential danger than that from liquid wastes.

Their direct injection into deep geological formations has been used or proposed – into shale layers [81] and salt beds [83]. Impermeability between the selected and adjacent layers, the absence of fossil water circulation and the probable negligible risk of geological movement are major conditions governing the choice of a site.

Sludges are sometimes stored in tanks buried at shallow depth. The materials used for tank construction (stainless steel, concrete) are selected to ensure sufficient life (Zagorsk, The Hague).

4.2.2. Disposal after conditioning

More frequently, after incorporation in concrete or bitumen, solidified radioactive sludges are stored under the protection of their concrete or metal containers in covered trenches, galleries or other natural or artificial shelters.

Studies are being made in various nations as well as on an international scale for the creation of burial grounds enabling centralization and monitoring of long-term storage.

Disposal into marine deeps has been used (United Kingdom, United States of America). The study of this disposal method is proceeding (experience of ENEA in the Atlantic) and has been the subject of international conferences convened by the IAEA [4, 75, 98].

5. SAMPLING

In the treatment of radioactive wastes, sampling is an operation which requires special care. A sample conditions the value of analytical work to be performed on it and the resulting interpretation. Raw radioactive wastes, and more especially wastes under treatment, are never clear and homogeneous liquids. They result from mixtures of water and various solutions previously stored in more or less dirty tanks and they contain, in addition to elements in solution or colloidal and non-miscible oils and solvents, solid debris or precipitates of variable density. It is impossible to obtain a thorough homogenization; a mixture of aqueous phases, as they are admitted for treatment, constitutes the original waste.

A sampling scheme must be planned according to the continuous or discontinuous mode of treatment, to the selected chemical treatment process and to technological requirements (sedimentation, filtration). This sampling scheme must allow the user to know the chemical, physical and radioactive compositions of wastes before and after treatment. This will lead to the determination of the decontamination efficiency, the observance of disposal standards and the possible conformity with previous laboratory tests. In the course of time, the treatment results will allow a better orientation of investigations and the need for new investments.

The sampling method often depends upon the size of the treatment plant.

For small plants, the liquid is sampled, after homogenization, by means of a manual pump or directly from a pipe through a small-diameter valve (needle valve). The sample is recovered into a clean (or better, new) flask, preferably made of plastic material such as polyethylene.

When the sampled volume is low or highly active, a device is used which includes a needle fed by the circuit to be sampled and a previously evacuated flask.

For larger plants, sampling centres are sometimes provided, where special piping supplies the liquids sampled by vacuum from each sampling point of the plant (Saclay). In this case, cross-mixing or pollution must be prevented by long and abundant rinsing of the piping before sampling; also, this piping must be of sufficient diameter to avoid clogging or scaling.

The frequency of sampling, in the case of continuous treatment, is guided by possible fluctuations in activity and chemical composition. Generally, large units are preceded by homogenizing tanks which minimize these variations.

The sample volumes must be sufficient in order to allow, after analyses and measurements, a fraction to be retained as records for a limited time. Upon sampling, the flasks must be labelled and checked for correct closure. In the case of quick changing liquids (presence of bacteria, oxidable salts, unstable chemical solution) as well as in the case of suspensions, special care must be taken: sterilization, acidification, etc. The analytical laboratory should be provided with all information concerning the operations performed that affect the initial condition.

Sampling of sludges before or after concentration is practically insignificant with regard to their chemical and radioactive composition. However, it is sometimes performed to measure viscosity, density and moisture content.

As a general rule, every sample must be marked in a precise and indelible manner with its description (origin, date and, if necessary, nature and initial aspect) and the determinations to be performed, so as to alleviate a possible loss of the accompanying document; this latter shall enclose additional data intended to ease the task of the analyst.

6. LABORATORY WORK [121]

6.1. Jar tests – flocculation behaviour

6.1.1. Purpose and definition of jar tests

It is not possible to infer from the analysis of a raw waste the doses of coagulants and flocculation aids that will ensure a good radioactive decontamination together with an acceptable sedimentation. Doses of reagents and of polyelectrolytes and, if necessary, the stirring rates and their duration, must be determined by experiment for each waste type; sometimes it is possible to be guided by composition analogies of treated liquids, but the approximation often remains coarse, especially in regard to radioactive decontamination.

Thus, it is necessary to submit a representative sample of a particular lot of wastes to a series of tests performed under identical mechanical conditions with increasing reagent doses; these doses are selected around values suggested by previous experience. Tests are performed in jars (graduated if required) with a capacity of about one litre; tests are run simultaneously in series of four, six or more. It is often necessary to perform several test runs; the initial sample must then be of sufficient volume (20 to 25 litres).

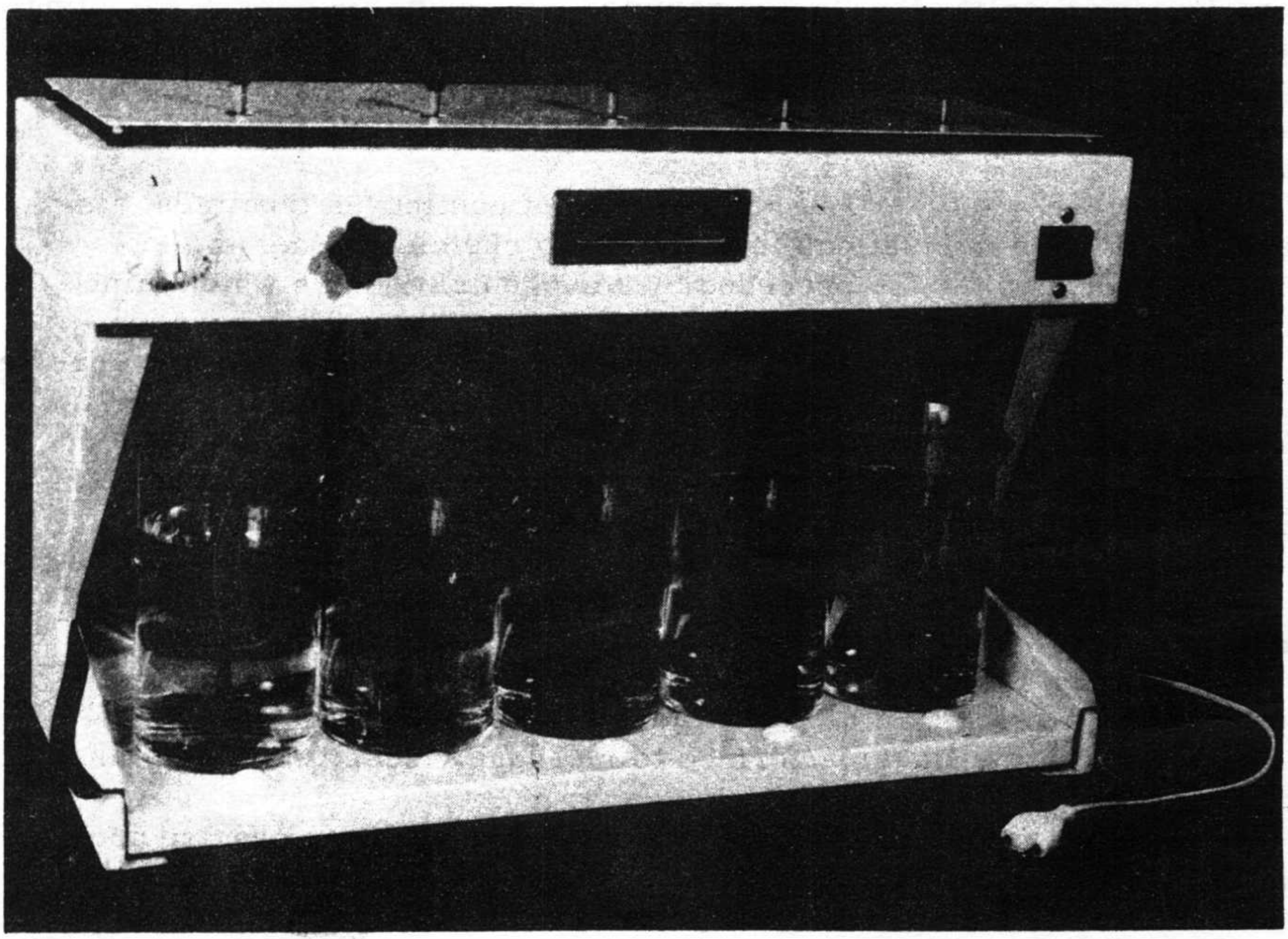


FIG.13. Jar-test equipment. (This Figure is reproduced by courtesy of Société DEGREMONT, France.

6.1.2. Equipment (see Fig.13)

The equipment required (in addition to dosing, measurement and control laboratory equipment) consists of an adjustable speed mechanical stirring system. It allows paddle stirrers to be set in motion at the same speed (between 20 and 100 rpm). This equipment may be purchased from a supplier or fabricated in the laboratory; in the latter, care should be taken to maintain the paddle area lower than one-quarter of the vertical cross-section of the stirred volume.

Provisions must be made to stop the stirring action quickly and it is often useful to provide two speed ranges in order to take into account flash-mixing and flocculation conditions.

6.1.3. Procedure

The first determination concerns pH, turbidity, total hardness and total α-, β- and γ-activities. If required, other analyses are performed: acidity or alkalinity, biological and chemical oxygen demands, specific activity for some elements, gamma-spectrometric detection of γ-emitters.

The waste is then adjusted to the pH selected for the treatment under test. Aliquots are poured into each jar and stirring is started at low speed.

Increasing doses of coagulant are introduced as quickly as possible and stirring is adjusted to a speed corresponding to flash mixing for the required time (1 to 5 min).

Stirring speed is then decreased to the rpm corresponding to the flocculation phase for the designed time (10 to 30 min), flocculation aids being added in due time. The appearance of flocculus and the aspect of its formation are observed during this period.

Stirring action is then stopped and stirrers removed from the jars. Sedimentation lasts for 1 to 5 h. The sedimentation aspect, its speed, the percentage of sludges formed, the degree of clarity of supernatant solution are observed.

After sedimentation, the supernatant is siphoned off and filtered through filter-papers of determined porosity; the clear filtrate is then submitted to the same controls as the initial waste.

If required, the operation is resumed using other coagulant doses, changing their nature if necessary, until optimum treatment conditions are obtained. These tests should be performed using the same reagents as those used in the industrial unit and with the same concentrations.

6.1.4. Treatment of results

Results obtained from each test series are entered on a document, a record of which is retained; this document contains the results of chemical and radioactive analyses as well as physical measurements and visual observations.

These documents, together with that defining the initial characteristics of the waste, allow the waste manager to infer directly the optimum dosages for operating the industrial plant.

Accumulation of these results often allows further tests to be made rapidly by direct analogy; it also enables, should the case occur, an investigation into the cause of treatment irregularities (eg. influence of complexants).

6.1.5. Improvements in using jar tests

The tests summarized above leave much to rules of thumb with regard to selection and association of doses in the case of the addition of several reagents. Optimizing tests by factorial experiment methods have been undertaken.

The response surface method, and especially the path of steepest accent method, has been the subject of studies which have produced appreciable improvements in the determination of optimum doses of coagulant mixtures [84].

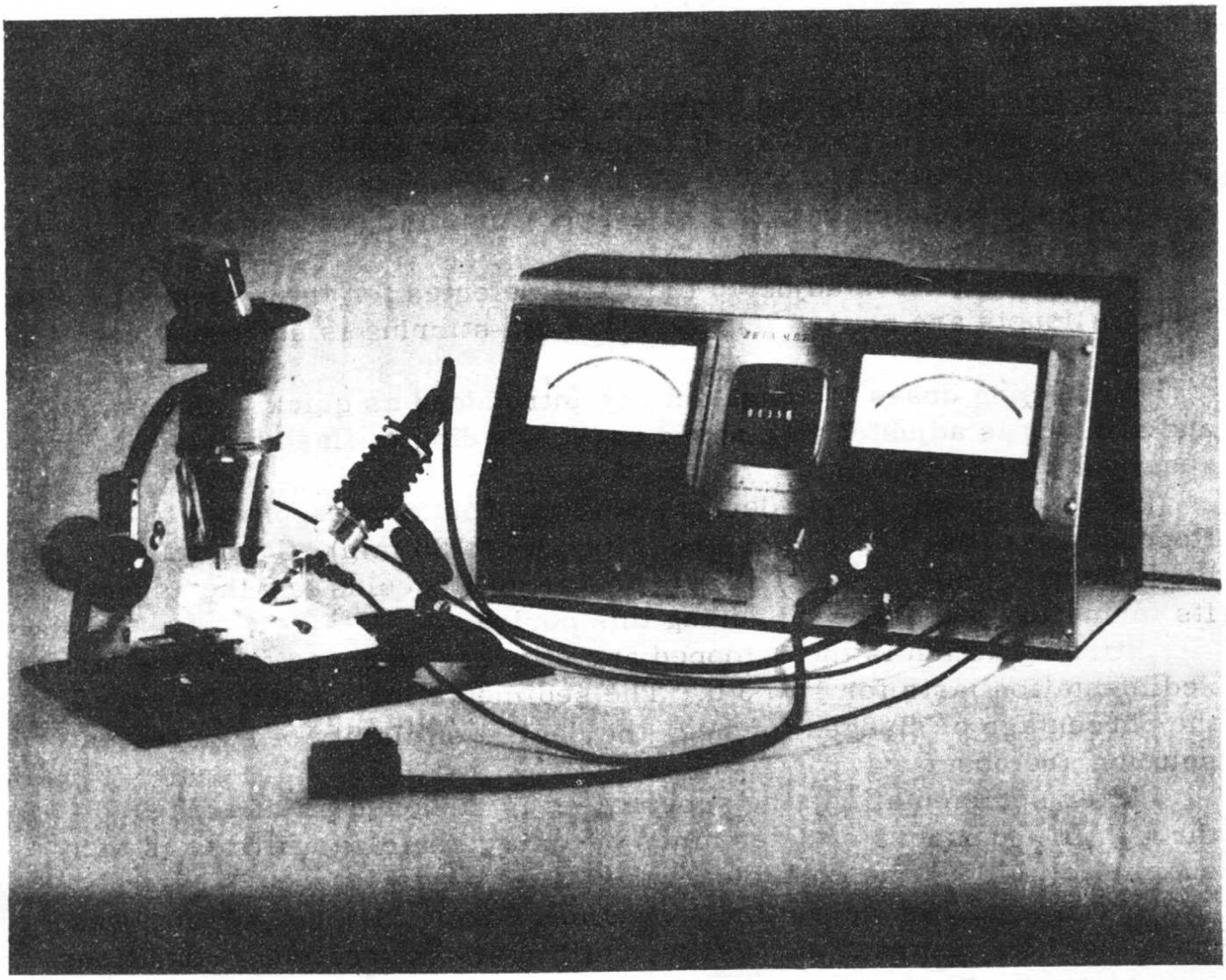


FIG.14. Zeta potential measuring equipment. (This Figure is reproduced by courtesy of Zeta-Meter Inc., 1720 First Ave., New York, N.Y. 10028.)

6.2. Zeta potential tests

6.2.1. Definition – object

Zeta potential (Z P) is a measurement of electro-kinetic charge (in mV) surrounding each solid particle suspended in an electrolyte solution. This potential is generally electro-negative for colloidal materials of raw waters and wastes; it is sufficiently high to allow mutual repulsion between particles to maintain them in suspension.

If the electrical charge of colloids is brought back to zero, for example through addition of colloids carrying positive charges and with moderate stirring (causing numerous collisions without breakdown of formed agglomerates), the respulsing forces are nullified. Particles blend and collect into a flocculus of sufficient size and density to enable sedimentation to occur at an acceptable rate [85-88]. Zeta potential determination of raw and treated wastes allows the measurement of colloid removal.

6.2.2. Equipment – measurement of zeta potential (see Figs 14 and 15)

Measuring equipment for Z P consists of a direct-current generator with variable voltage from 0 to 500 V and a micro-electrophoresis cell.

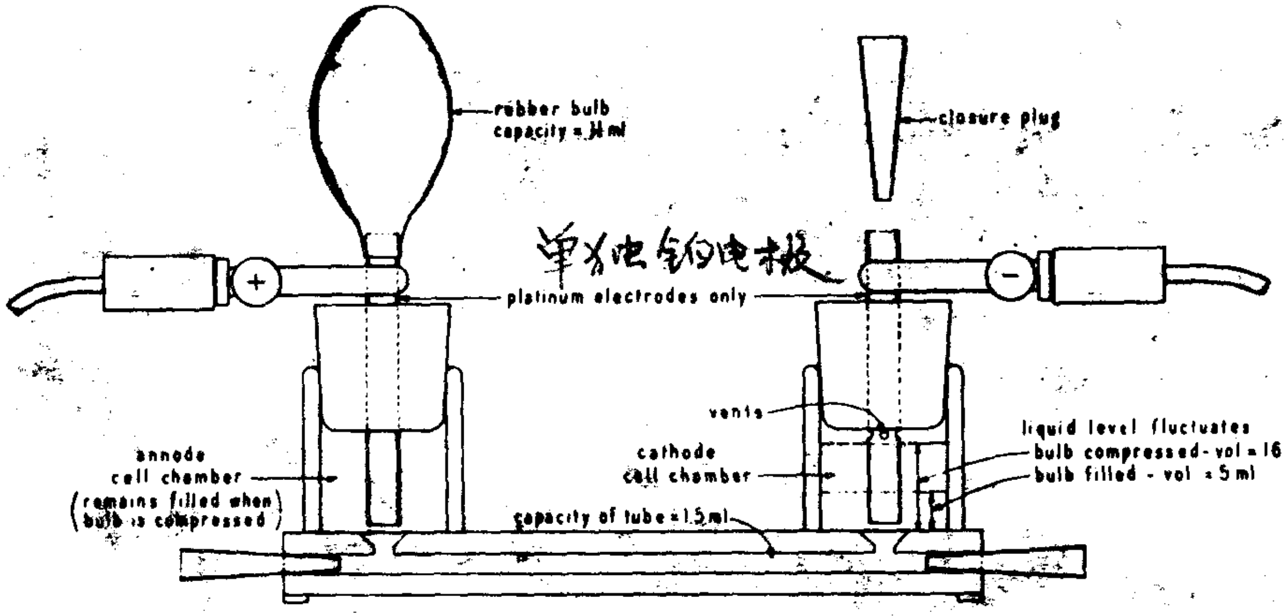


FIG.15. Zeta potential measuring cell. (This Figure is reproduced by courtesy of Zeta-Meter Inc., 1720 First Ave New York, N.Y. 10028.)

This cell includes two chambers containing electrodes connected by means of calibrated tubing of a transparent material. One of the chambers (anodic) is closed, the other (cathodic) is vented to the atmosphere. Electrodes are generally made of platinum if the conductance of the medium is < 2000 µmho. An illuminator, emitting an intense beam of blue-white light, illuminates by reflection the particles moving in the electrophoretic field. A stereoscopic microscope fitted with a micrometric-grid eyepiece allows the velocity of the particles to be measured.

Z P measurement is in fact the measurement of electrophoretic mobility (E M) of colloids and suspensoids of the medium examined. This latter measurement is related to Z P through the Helmoltz-Smoluchovski relationship: $Z\,P = E\,M\,4\pi(Vt/Dt)$ where Vt is the viscosity in poises, Dt the dielectric coefficient of the liquid at temperature t, with $E\,M = F/H$, F being the particle velocity in cm/s and H the difference of potential per centimetre of displacement. The displacement direction of particles determines the polarity of their charge.

Measurement is practically performed by measuring Z P for a certain number of particles (20 to 100). A distribution curve results allowing one to know the mean value of Z P. This determination requires about 30 min to perform. An improved device using automatic sample transfer to reintroduce fresh samples at short intervals has been investigated and is now available. It eliminates the problems of thermal effects and particle settlement (see Fig. 16).

Flocculation jar-tests are performed as a function of Z P as measured on the settled solution.

6.2.3. Results

Zeta potential measurement to improve jar tests, the performance of which were often previously guided solely by visual observation, is frequently used for water treatment. It results in an appreciable improvement in sedimentation and also of subsequent treatments such as filtration

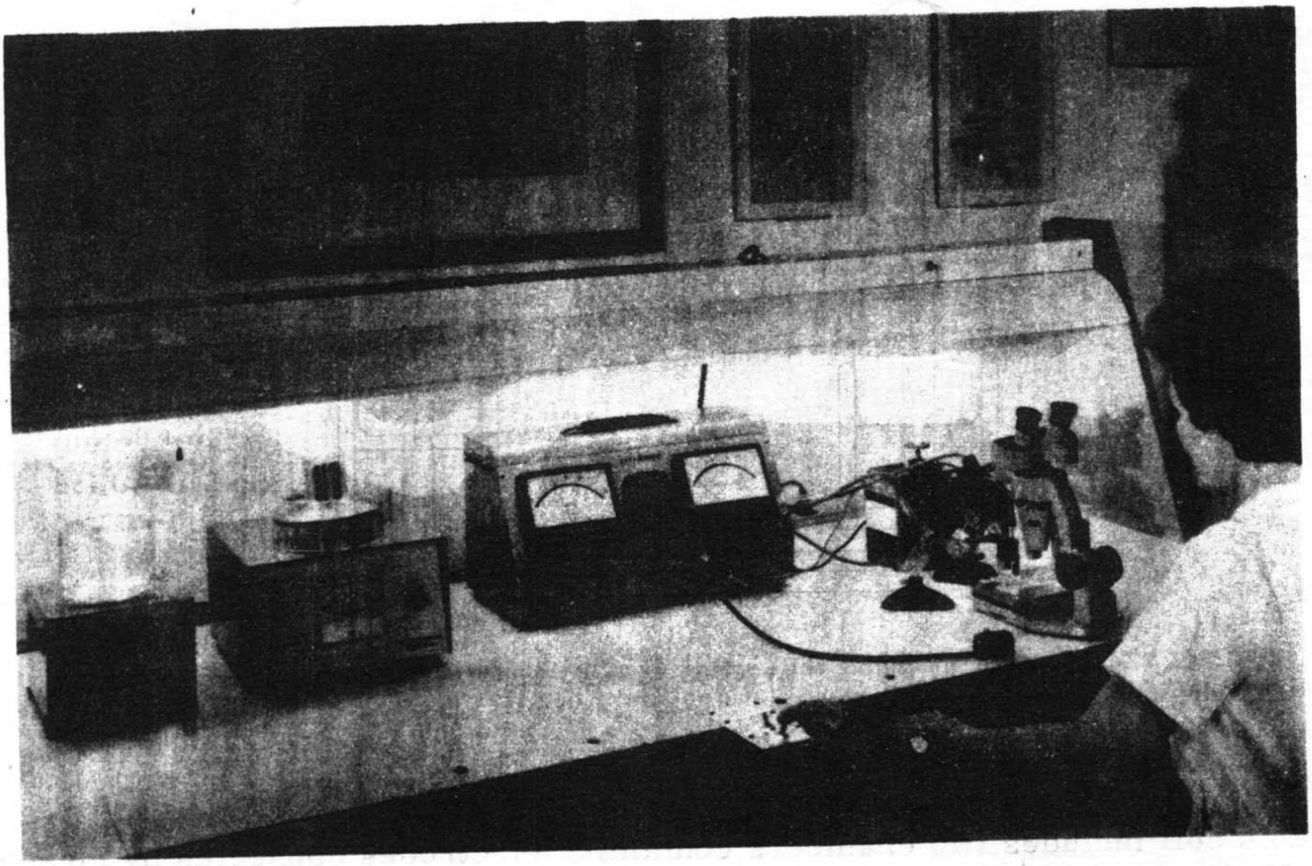


FIG.16. Zeta-meter and automatic sample transfer. (This Figure is reproduced by courtesy of Zeta-Meter Inc., 1720 First Ave., New York, N.Y. 10028.)

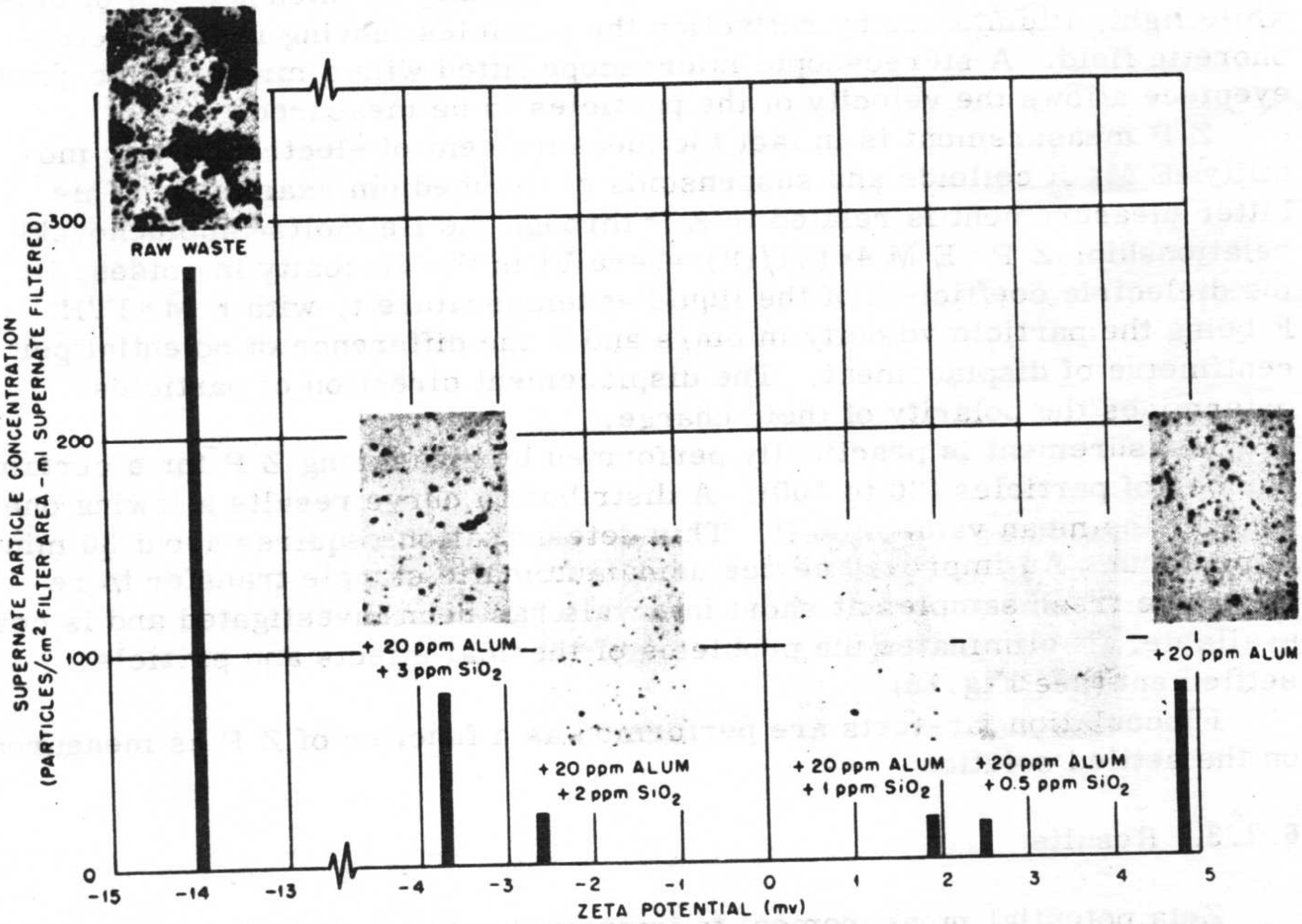


FIG.17. Solids filtered out of treated waste supernatant on to a membrane filter at a Zeta potential of +2.5 mV. Jar-test conditions: 30 min of flocculation followed by 30 min of settling. (This Figure is reproduced by courtesy of the USAEC from the publication "Low-Level Radioactive Wastes", TID-20978 (see also ORNL-3830).)

and ion exchange. Reagents most in use are alum, activated silica, ferric iron, cationic polyelectrolytes.

Application to the treatment of radioactive wastes has been made at Oak-Ridge [55, 89]. (See Fig. 17). For a raw waste with a Z P of -14 mV, the combined addition of 20 ppm of alum with 0.5 ppm of activated silica leads to the best clarification and a Z P of +2.5 mV. It was observed that the best procedure consisted in approaching null Z P at ± 5 mV by the addition of alum and adding a little activated silica to complete the dosage. The use of cationic polyelectrolytes (Primafloc C-3) is very efficient at very low doses.

6.3. Characteristics of influents and effluents

Radioactive wastes must be submitted to a certain number of laboratory tests before and after treatment which specify the characteristics of the wastes. These determinations may be physical, chemical or radioactive; some of them are frequently performed, some others only on occasions. This Section is intended to outline the measurements performed, their relative frequency and their principle. For procedure details it should be helpful to refer to treatises on analysis; as a matter of fact, although they are standardized in several countries but in a different manner, these latter are often modified.

6.3.1. Physical characteristics

(a) Frequent determination

Dry extract represents the non-volatile elements contained in the waste. Water and other volatile substances are removed by evaporation and the operation is completed by a constant-weight drying of the evaporation residue in a stove at 110°C.

Moisture is determined by the same method for thick suspensions and sludges: (i) The dry extract is expressed in g/litre of initial effluent; and, (ii) Moisture is expressed in per cent of initial sludge weight.

(b) Occasional determinations

Turbidity – colour. These checks are performed either by visual comparison with a range of reference samples, or in a more rational manner using a photometer.

Oxidation-reduction potential is determined using a potentiometer and a couple of platinum and reference electrodes. It is expressed in ± mV.

Zeta potential. (see Section 6.2)

Suspended solids are separated by filtration through a cellulose filter or a sintered glass of determined porosity. The weight is estimated, as for dry extract, by constant-weight drying at 110°C.

Fixed residue. Once the dry extract is determined, it is sometimes heated to 750°C in an electric muffle furnace. The constant-weight residue is the "fixed residue".

6.3.2. Chemical characteristics

(a) Frequent checks

pH – acidity – alkalinity: pH may be determined roughly by different indicators, but the universal laboratory method consists in using a pH-meter and a couple of glass and saturated calomel electrodes. Acidities and alkalinities are determined by titration in presence of pH-indicators or, better, by using a pH-meter of the recording type.

Acidities or alkalinities are expressed in terms of normalities.

Total hardness: (Calcium + magnesium sum). The effluent, freed from carbon dioxide and heavy metal ions, is titrated with a solution of EDTA. The end of reaction indicator is Chrome Black T (Erichrome Black T, Pantachrome Black TA, Solochrome Black WDFA, etc.) [93].

Results are expressed in mg equivalents per litre.

B.O.D. (Biochemical Oxygen Demand): The titration is based upon the oxygen consumption at the fixed temperature of 20°C during a determined period (usually 5 d) in various dilutions of the studied effluent with a pure oxygenated water. The operation is performed in the presence of pure water reference samples. Dilutions are selected (1/4, 1/10, 1/25, 1/50, 1/100, etc.) so that in one case the oxygen concentration reduction ranges about 50%. This residual oxygen being titrated, as well as that of the reference samples and the B.O.D. may be calculated. It is expressed in mg of O_2 consumed per litre.

C.O.D. (Chemical Oxygen Demand): Two methods are used: the Kubel and Tieman method (pp. 505-07, IV of [18]) (using potassium permanganate) and potassium dichromate method [83]. In both cases, the excess of oxidizing agent is back-titrated by a standard solution of a reducing agent. Concentration is expressed in mg of O_2 consumed per litre. For B.O.D. and C.O.D determinations, see References [91-94].

(b) Occasional determinations

Anionic constituents: Cl^-, SO_4^{2-}, F^-, PO_4^{3-}, CO_3^{2-}.

Cationic elements: Fe, Al, Ca, Mg, U, etc. Uranium is often determined by fluorimetry. Concentrations are expressed in grams of elements per litre.

Toxic agents: CN^-, CrO_4^{2-}, etc.

Anionic detergents (Alkyl benzene sulphonates) are titrated according to the Longwell and Maniece method [95]. A coloured complex is formed by addition of methylene blue extracted with chloroform and titrated by spectrophotometry.

Concentrations are usually expressed in grams of Manoxol OT equilvalent per litre [99].

EDTA: This method is based upon the very high stability of EDTA-cobalt complexes. The optical density of the rose-violet complex obtained is titrated by spectrophotometry. Concentrations are obtained in g/l.

6.3.3. Radiometric measurements

The results of radiometric measurements performed on liquids or low solids suspensions are conveniently expressed in μCi/ml or Ci/m^3 (For instance, 3×10^{-5} Ci/m^3).

Comparisons and the expression of radioactive decontamination factors (DF = initial activity/residual activity) are thus made easier.

When the counting checks are performed, it is of special value to use well-defined standards to calibrate the counting apparatus in the range of the estimated activity of the sample. The geometry and the material of the detector and the counting chamber must also be properly selected to provide the best counting conditions.

(a) Frequent checks

Gross α-activity: The solution sample (about 2 ml) is evaporated in a cupel. The dry residue is submitted to counting either in an air ionization chamber, or more often, by means of a scintillation detector with a screen of silver-activated zinc sulphide.

Total β-activity: The solution sample is evaporated as above and submitted to counting by various types of Geiger-Müller counters. The activity is sometimes measured by means of counters immersed in the liquid, but this procedure is generally reserved for stream checks.

Total γ-activity: Counting of a few-millilitre fraction is performed using a thallium-activated sodium-iodide crystal as detector. So-called "well" detectors, including a sample holder drilled in the crystal centre-line, are preferably used.

γ-spectrometry: Qualitative and semi-quantitative investigation of the various γ-emitters in wastes is often performed by gamma-spectrometry. This method is based upon the classification of detected pulses as a function of the energy and activity of photopeaks of the various γ-emitters by means of a selector. A recording is obtained which enables one to detect the presence and, roughly, the abundance of γ-emitters having an energy higher than 120 keV under usual measurement conditions.

This method also allows, provided accurate sampling and mathematical analysis of results are used, the sufficiently accurate determination of components of a mixture of γ-emitters.

(b) Occasional checks

Sometimes it is necessary to check with more accuracy the concentration of one or several radioactive emitters. In this case, chemical separations with the use of carriers are necessary.

$^{89+90}$Sr: The dual precipitation method with $Sr(NO_3)_2$ carrier in fuming nitric acid is frequently used.

^{137}Cs: Several methods are used: insolubilization in perchlorate medium, precipitation of silico-tungstate.

$^{103+106}$Ru: Sometimes RuO_4 distillation is used, or anion-exchange techniques.

$^{95}Zr + Nb$, $^{141+144}Ce$, ^{140}Ba, ^{51}Cr, ^{60}Co and numerous other emitters are sometimes evaluated after isolation by chemical methods, the efficiency of which must be checked.

In all of these determinations, it must be taken into account that short-lived radioactive elements may be present and it is advisable for check purposes to perform radioactive decay measurements.

Details of analytical procedures for most determinations needed in the chemical treatment of radioactive wastes are given in Reference [93].

7. FACTORS GOVERNING THE SELECTION OF PROCESSES [99]

When designing a radioactive waste-treatment plant, it must be kept in mind that radioactivity is not destroyed by any treatment process. It is only recovered in the best way possible with regard to health, safety and economic considerations and concentrated in residues that must be subsequently treated to allow their disposal.

Several factors govern the selection of a treatment process. Most of them are inter-related and it is difficult to study each of them separately. However, two major considerations, related to one another by safety and economic consequences, affect the initial selection with regard to the required degree of purification (decontamination factor). The first consideration is the determination of disposal possibilities for the particular site, the second is the characteristics of the waste to be treated.

7.1. Economics and safety

In waste treatment the factor of health and safety demands first attention, but the economic factor is so basic that it must be considered a close second in importance.

Trying to achieve treatment that produces a level of radioactive discharge approaching zero would be extremely expensive and is seldom required. In the past, unnecessary restrictions have been placed on discharge levels, which have resulted in high costs for waste management. On the other hand, attempted saving in such costs can result in excessive or needless expenditure in other directions (e.g. expensive monitoring). A balance must always be sought between using the capacity of the environment to receive wastes without detriment and the treatment necessary to ensure that the discharge levels are such that no hazard arises.

Until recently in many countries, little consideration has been given to the economics of waste-treatment processes. Decisions have frequently been based on personal preferences of the authorities for one or another method, or upon assumptions such as, "chemical treatment is cheaper, but its decontamination is low" or "evaporation is the best, but expensive" etc. Moreover, the published data were fragmentary and not derived in the same way by different nations.

For these reasons, in 1965 and 1966 the International Atomic Energy Agency convened two panels on the Economics of Waste Management [25]: The principal objectives of the panel studies were to examine all the elements of waste management and to suggest a system of reporting their costs that would serve as a useful guide to the true costs of managing radioactive wastes.

7.2. Determination of permissible limits for release and disposal

7.2.1. Laws governing disposal

The permissible discharge limits for a particular site may be set by legislation which may bear direct reference to the IAEA publication on Safety Standards [5] or the ICRP recommendations for maximum permissible concentrations of radionuclides (MPC values, see Section 2.2.1.). Conversely, legislation may permit discharge limits which are set indirectly by the MPCs but take into account the usage of the environment and its capacity to receive radioactivity. Annex II of [7] gives extracts of three national regulations concerning the disposal limits of radioactive wastes. These show two different approaches: the USSR and United States of America specify disposal limits of wide applicability whilst the United Kingdom specifies disposal limits case by case.

National and local laws and ordinances regulating the disposal of radioisotopes must be closely studied by managers of radioactive wastes. If laws have not been established, it must be determined what regulations and guides should be used until such laws are prepared.

The radioactivity remaining in the processed waste may be only a small part of the potential hazard. National and local laws or ordinances regulating environmental pollution by other contaminants than radioisotopes must also be carefully considered.

In all cases, a pre-operational survey of the environment should be made, which will further ensure a more efficient (and often more economical) monitoring programme to ascertain that there would be no significant build-up in radioactivity.

7.2.2. Methods of disposal

The locality and environment of the site will decide the type of disposal and the amount of radioactivity which can be released. Thus, the following information must be collected in connection with the possible method of disposal.

For disposal into fresh water: Location of the river in relation to the origin of wastes and distance to an estuary; maximum, minimum and average river flows, upstream and downstream flow regulations; nature and quantity of other usages of water (industrial, municipal, etc.); chemical quality and abundance of suspended and colloidal material; species and abundance of edible flora and fauna.

For disposal into the ground (shallow land burial): Depth of the water table; permeability of the soil; distance to the nearest points of water usage (wells, river); ability of the soil to absorb radionuclides..

For disposal into deep formations: Hydrological situation; geographical situation (physical features of the area, vegetation, climatic changes, precipitation, evaporation); geological situation (geological structure, permeability of formations, chemical and mineral compositions, physical chemistry: absorption and desorption characteristics).

7.3. Influence of raw water and influent characteristics

7.3.1. Raw water

It is of major importance in selecting the type of chemical treatment of wastes to know the composition of the raw water or water intended to supply the various stations of a nuclear centre. As a matter of fact, this water is the major component of wastes that contain various radioactive pollutants.

Total hardness affects the selection of the treatment type. Thus, with a hard water, the phosphate coagulation process may be applied; with a softer water, the alum process might be selected. This calcium magnesium hardness should be eliminated (by sodium carbonate addition with the soda-lime process) when further treatment by ion exchange is to be performed (scavenging-precipitation). If this removal is not sufficient, the radioactive decontamination may be less efficient, especially that of strontium.

The various treatments applied to water before its admission to a nuclear plant must be known: chlorination degree, inclusion of certain additives (copper, anti-corrosion products, etc.), purification process, etc. Thus, the initial water composition affects both the selection and the economy of the waste treatment and the radioactive decontamination.

7.3.2. Influent characteristics

The quantities of each category of waste to be treated should be well defined: yearly volume and average, maximum and minimum flow-rates. Wastes should be segregated, where possible, according to degree of mineralization, presence of detergents or solvents, biological content, etc. (see Sections 2.2.2 -2.2.4).

Their compositions should be specified as precisely as possible in regard to the nature of chemical products contained and the kinds and quantities of radioisotopes expected.

Technical data concerning the operations or processes from which the wastes are issued should also be obtained. Thus, the presence of ruthenium or cobalt is not sufficient information in itself. These radioelements may exist in very different ion forms more or less refractory to usual chemical co-precipitation treatments. For instance, if it is known that effluents contain radioactive solvent scrubbing waters, then it is probable that a part of the ruthenium may exist in the form of nitroso-nitrato anionic complexes; so it is with cobalt anionic complexes in the case of regenerants of ion-exchange resins.

The presence of toxic compounds must be known, whatever be the toxicity level. A special treatment of toxic wastes must always be performed, preferably before the wastes are treated for radioactive decontamination if the toxicity is high (cyanide effluents, for instance). For the other compounds to be surveyed (hexavalent chromium, heavy metals, amines and certain non-miscible solvents), the normal decontamination treatment and the dilution with other effluents are often sufficient to eliminate the potential hazard.

7.4. Other factors

7.4.1. Decontamination factors required

Total decontamination factors obtained by coagulation-flocculation treatments vary according to the activity level and the radioactive nature of the contaminants.

While decontamination of α-emitters is usually easy and widely sufficient, this is not true for most β- and γ-emitters.

The total β and γ DF is often only about 10 for treatments using common water-purifying and softening methods when the initial activity level of the waste is 10^{-3} to 10^{-4} Ci/m . For many sites these values are low enough to allow disposal using the dilution possibilities presented by the non-contaminated waters of the centre.

However, some radio-elements (especially ^{90}Sr, but also ^{106}Ru, ^{137}Cs) are long-lived enough, and present such a biological hazard that they require a specific DF ranging from 100 to 200.

The previous practice of considering the total DF for estimating the total released β- or α-activity is now being modified. Release formulae are under study or being used, which take into account the specific DF for the most dangerous radio-elements (those having the most restrictive MPC values).

8.4.2. Local economic factors

When selecting a treatment process, local conditions must be taken into account for the following factors:

(i) Cost and availability of chemical reagents, structural materials (especially for chemical engineering) and spare parts
(ii) Cost, availability and skill of manpower
(iii) Cost of energy and water
(iv) Possibility of technical assistance for engineering, maintenance and operation of the plant (especially, the possibility of scientific research)

It is customary to gather all these data into an engineering estimate of requirements for capital investment and yearly operational expenses for various alternatives.

GLOSSARY

A.B.S. (Alkyl benzene sulphonates)	Abbreviation commonly used for anionic sulphonic detergents
ABSORPTION	Ability of a porous solid material to hold within its body relatively large quantities of gases or liquids
ACCELERATOR	A trade name for a suspended-solids contact basin providing flocculation and settling in a one tank. (See also CIRCULATOR, SLUDGE BLANKET CLARIFIER, CLARIFLOCCULATOR)
ACTIVATED SLUDGE PROCESS	Bacteriological treatment of sewage through aeration and oxidation with partial recycling of the resulting sludge
ACTIVATED SILICA	Negatively charged colloidal particle made by partial neutralization of the alkalinity in dilute sodium silicate solution
ADSORPTION	Concentration change at the interface between a solid and a fluid by physical or chemical adhesion of a substance
AGGLOMERATION	Coalescence of dispersed suspended material into larger flocs or particles which settle rapidly
ALUM (FILTER-ALUM)	Common name of aluminium sulphate
BACK WASHING	Regeneration by up-stream washing of a porous bed of granular material
BENTONITE	Colloidal hydrated clay-forming gels and viscous suspensions used as coagulent aids and cation adsorbents
B.O.D. (Biochemical Oxygen Demand)	Quantity of oxygen consumed in the biochemical oxydation of organic matter according to specified conditions. The standard B.O.D. is established with the five days and 20°C procedure

BURIAL GROUND	A place for underground disposal of radioactive solidified or conditioned wastes, the earth acting as a shield
CARRIER	Element or substance added to a liquid in ponderable amount to make easier recovery or elimination of trace elements by co-precipitation
CHEMICAL TREATMENT	Common designation for the whole coagulation, flocculation, sedimentation process including sludge elimination
CHLORINATED COPPERAS	A solution of ferrous and ferric sulphate produced by chlorinating ferrous sulphate
CIRCULATOR	See ACCELERATOR
CLARIFLOCCULATOR	See ACCELERATOR
COAGULANT	A material that, when added to an effluent, causes irreversible precipitation, especially for the removal of colloids which become agglomerated in flocculus
COAGULATION	The process of using coagulants in a liquid-waste treatment with prior determination of the most efficient dosages
C.O.D. (Chemical Oxygen Demand)	Equivalent of oxygen consumed for the chemical oxidation of all chemical and organic oxidizable derivatives in the effluent
COHESION (SLUDGE)	Mechanical characteristic of a sludge submitted to a counter-current flow of liquid which, if good enough, ensures the maintenance of settling and clarifying properties in the suspended-solids contact units
COLLOIDS	Very fine electrically charged particles in a liquid medium with size ranging from 10 to 2000 Å which confer on the solution an intermediate character between a true solution and a suspension
CONCENTRATION FACTOR	Ratio of influent waste volume to final sludge volume or weight

CONDITIONING (SLUDGE)	Treatment of fluid sludge by dewatering or solidifying methods to bring it to a suitable form for disposal
DECONTAMINATION FACTOR (DF)	The ratio of the initial amount of a specified activity to the remaining amount of residual activity after the treatment. The DF may be expressed relative to gross activity or for a particular radionuclide
DOSAGE	The amount of a coagulent or polyelectrolyte determined by laboratory procedure to be the most convenient for good coagulation of an effluent
EFFLUENT	Liquid sewage or waste water issued from industrial or laboratory facilities
EFFECTIVE SIZE (SAND FILTERS)	Size of sand grains such that 10% by weight are smaller and 90% are larger
FLASH MIXER	A tank in which is performed rapid mixing by fast agitation of neutralizing reagents or coagulants
FLOC	A light bulky precipitate which exhibits a large surface area and has some ion-exchange properties
FLOCCULATION	After coagulation, flocculation allows the development of the smallest particles of precipitate into floc by gentle agitation which provides sufficient but not too violent contacts
FLUME (MIXING)	An open conduit with baffles where reagents are added and mixed by stream turbulences
FULLER'S EARTH	A special type of clay, somewhat similar to bentonite in its coagulating properties
HARDNESS	A chemical characteristic of water. (Total): it is computed from the amounts of calcium, magnesium and iron or equivalents estimated by chelatometric methods (Temporary): due to the presence of bicarbonates of calcium and magnesium

	(Permanent): caused by sulphates and compounds other than bicarbonates
HEAD	Height of a column or body above a given point causing pressure
JAR TEST	Laboratory procedure using a number of beakers (jars) similarly stirred to conduct investigations under identical conditions with regularly increased doses of reagents
LIME-SODA PROCESS	Process for softening water by using soda-ash and lime
MPC (Maximum Permissible Concentration)	Limit of permissible concentration for a given radionuclide in drinkable water or in the atmosphere
PARTS (PER MILLION, PER BILLION)	ppm, ppb: Parts by weight of an element or a reagent in effluent analysis or dosage (US billion = 10^9)
PERIOD, DETENTION	Theoretical time required to displace in continuous operation the contents of a tank entirely at a given flow-rate
PULSATOR	A trade name for a pulsed sludge-blanket clarifier
SCAVENGING PROCESS	A treatment using a prior elimination (by co-precipitation) of disturbing compounds for a further process
SCHMUTZDECKE	A dirty skin of live material and fine particles of flocculus that forms on the upper layer of sand filters
SEDIMENTATION	(Settling of thickening), Gravitational deposition of suspended solids and subsequent isolation and recovery of the two phases: clear liquid and sludge
SEWAGE	Domestic used water and more generally non-chemical or industrial liquid effluents containing more or less organic compounds
SLIME	Colloidal substances and films occurring along with the biochemical and chemical treatment of wastes

SLUDGE	The accumulated settled solids resulting from the chemical treatment of liquid wastes
SODA-ASH	A common name for anhydrous sodium carbonate
SOFTENING	The process of removing from effluents the substances which produce HARDNESS, or of replacing the hardness cations by sodium ion
SUSPENSOIDS	Hydrophobic colloidal particles that remain in suspension under all conditions and do not tend to combine with the suspending medium
TURBIDITY	Condition of a liquid containing very fine but visible suspended material with a settling rate nearly zero; also, the measure of the opacity of the liquid
UNIFORMITY COEFFICIENT (SAND FILTERS)	Definition of tne uniformity in size distribution of sand grains: ratio of the sieve size passing 60% to the sieve passing 10%
WATER TABLE	Highest level below which the rocks are saturated with ground water
WEIR	A fixed device used on open conduits to divert a part from a stream for measuring and sampling purposes
ZETA POTENTIAL TEST	An electrophoretic measurement of the electrokinetic potential of colloids and suspended solids
ZOOGLEA	An irregular jelly-like matrix developed by bacteria when activated sludge is growing in biological beds

APPENDIX II

Conversion Table

UNITS OF LENGTH

Inch (in.)	0.0254 m
Foot (ft), 12 in.	0.3048 m
Yard (yd), 3 ft	0.9144 m
Micron (μm)	1×10^{-6} m
Angström (Å)	1×10^{-10} m

UNITS OF AREA

Square inch (in^2)	6.4516 cm^2
Square foot (ft^2)	0.0929 m^2

UNITS OF VOLUME OR CAPACITY

Imperial gallon (gal (UK))	4.54596 litres
US gallon (gal (US))	3.78533 litres = 0.833 gal (UK))
Cubic foot (ft^3)	28.3161 litres

UNITS OF FLOW-RATE

Cubic foot per second (ft^3/s)	28.315 litres/s = 101.93 m^3/h
US gallon per minute (gal/min) US	227 litres/h
UK gallon per minute (gal/min) UK	272.76 litres/h
US gallon per square foot per minute (gal/ft^2/min) US	2.421 m^3/m^2/h
UK gallon per square foot per minute (gal/ft^2/min) UK	2.934 m^3/m^2/h
Cubic foot per square foot per minute (ft^3/ft^2/min)	18.29 m^3/m^2/h

UNITS OF MASS OR WEIGHT

Grain	0.065 g
Ounce (oz)	28.349 g
Pound (lb), 16 oz	453.592 g
Long ton (UK)	1.016 t
Short ton (US)	0.907 t

UNITS OF PRESSURE

Pound force per square inch (lbf/in^2)	0.0703 kgf/cm^2
Atmosphere (atm)	1.033 kgf/cm^2

UNITS OF CONCENTRATION

Grain per cubic foot	2.296 mg/litre
Grain per UK gallon	14.2542 mg/litre
Grain per US gallon	17.1184 mg/litre
Grain per cubic inch	3.96 g/litre
Pound per UK gallon (lb/gal (UK))	99.7 g/litre
Pound per US gallon (lb/gal (US))	119.0 g/litre

CHEMICAL DOSE

French degree (1/5 milli-equivalent)	0.2 meq

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