"Documento original en mal estado"

IN VITRO BIOASSAY

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1 - INTRODUCTION:

To handle radioactive material or to use radiation producing machines it is necessary to use regulatory and administrative measures and follow technical radiation protection procedures in order to minimize radiation exposures to the workers as well as to the general public. In radiation protection one has to bear in mind two main aspects: (a) The protection of the radiation worker: and (b) The protection of the general public.

More stringent precaution for the protection of the general public is necessary as compared to that of radiation workers, because of the large number of individuals involved and the impracticability of providing large scale radiation protection and medical supervision.

In vitro bioassay is indirect method used to estimate the internal contamination level by the analysis of excreta and other biological samples such as breath, nasal discharge, sputum, saliva and sweat, either of workers handling radioactive materials or of exposed members of public, in accidental situations. Intake of the radioactive material may occur by inhalation, ingestion, absorption through skin or entrance by a skin's wound. After the intake the radionuclide gets into the bloodstream, where it is distributed into the body according the preferencial organ or tissue of the radionuclide. A part of that stays deposited in preferencial organ or tissue and the remainder is eliminated by the urine or feces according its biological half-life.

By a routine bioassay program it is possible to detect an acute intake of radioactive material that has not been suspected and to follow any slow accumulation of radioactive material in the body resulting from chronic low-level exposure. The results of the bioassay programs may indicate that the environmental conditions of the workplace has altered or other monitoring procedures have failed to warn to worker of unfavorable trends (ACGIH1). These measurements can also be used to establish if the treatment to enhance elimination is appropriate, to determine its effectiveness.

2 - METABOLISM OF RADIONUCLIDES:

An understanding of the metabolism of radionuclides and their compounds is necessary for estimating radiation doses received by the body and its component parts from radionuclides taken into the body. This is especially important for interpretation of measurements of body radioactivity and the results of analyses of excreta.

2.1 - ENTRY OF RADIONUCLIDES INTO THE BODY

In discussing this subject it is necessary to have a regular terminology. As far as possible the following words will be used with the meanings given:

Intake: The amount entering the nose or mouth

Uptake: The amount absorbed into extracellular fluid.

Deposition: The amount present in the organ of reference.

Radionuclides may enter the body by inhalation, ingestion, or through intact or wounded skin.

2.1.1 - INHALATION

Radioactive gases, liquids or solids may enter the body by inhalation. Water-soluble gases, e.g. those containing tritium or iodine, are rapidly absorbed from inhaled air and appear in extracellular fluid within a few seconds. Liquid or solid radioactive substances inhaled in the form of aerosols have a number of possible fates depending on their physicochemical properties.

2.1.2 - INGESTION

When a worker ingests a radioactive substance, he will usually do so over a short time, and this constitutes a single intake to him. Although he may have a series of such intakes, it is best, in order to keep the discussion simple, to consider the consequences of one intake only.

If the material is non transportable, most of it will traverse the gastrointestinal tract and emerge in the faeces. If the material is transportable, a significant fraction will be absorbed into extracellular fluid, mainly during its passage through the small intestine. The actual magnitude of this absorbed fraction will depend on the metabolism and nutrition of the individual as well as the chemical compound in which the radionuclide is ingested.

2.1.3 ABSORPTION THROUGH SKIN

Intact skin provides an effective barrier against the entry of most radioactive materials into the body. Exceptions of practical importance are the absorption through intact skin of: tritium oxide either as liquid or vapour, iodine as vapour or in solution and iodide in solution.

2.1.4 ENTRY BY A WOUND

When skin is broken, punctured or abraded, radioactive substances can penetrate to subcutaneous tissues and thence to extracellular fluid, rapidly in the case of transportable compounds, and slowly in the case of non-transportable compounds. In the latter case, if the wound is not decontaminated, the time course of systemic contamination may resemble that resulting from chronic exposure.

2.2 TRANSLOCATION AND DEPOSITION

Extracellular fluid is the chief vehicle by which transportable materials are transferred from one part of the body to another. Two examples of other methods of transportation are the movement of insoluble plutonium oxide from lungs to lymph nodes and the attachment of radionuclides or their compounds to red blood cells. A portion of the radionuclide in extracellular fluid will be excreted by kidney, liver, intestine, skin, or lung, and the remainder will be deposited in any organs or tissues for which it has a special affinity. Soon after systemic contamination by a transportable radionuclide, there will be a net transfer of it into the organ(s) of deposition. This will continue until the concentration in extracellular fluid falls (due to excretion and deposition) to a level which results in a net transfer of radionuclide from the organs of deposition back into extracellular fluid.

2.3 ELIMINATION

The rate of elimination of a systemic contaminant by all routes combined will follow its concentration in extracellular fluid. Bioassay programs consist of measuring rates of elimination at various times after contamination, and calculation from these the amounts retained in the body, and when possible in the various body organs, at all times until the radionuclide has disappeared from the body.

Exhalation is an important route of elimination. Sweat generally will contain any radionuclide present in extracellular fluid. Radionuclides excreted in urine can come only from extracellular fluid. In the course of excretion certain radionuclides may deliver enough radiation to the kidney. Radionuclides excreted in feces may come from ingestion or inhalation of a non-transportable radioactive compound or by passage of a transportable radionuclide into the gastrointestinal tract, either directly or by way of the bile.

3. SAMPLING

One of the more difficult aspects of a bioassay program is the development of a sampling plan that will provide adequate screening to detect accidental exposure or significant chronic exposure. Once an exposure is known to have occurred, the following procedure can be used to develop an effective sampling plan. All excreta are collected and all samples are analyzed until the results show that spaced samples are sufficient.

Most installation consider urinary sampling as the body screening procedure. Planning for a bioassay screening program must be based on the technical requirements, but capital and manpower cost are also considered. The value of the overall program must be favorably balanced with cost. Sampling, especially in a screening program, should not be so burdensome that cooperation is difficult.

The type of sample, the sampling frequency, and the personnel to be sampled are clearly related to the degree of risk to which an individual is exposed. Not only the amount of radioactive material he handles but also his work conditions (in the open, in a fume hood, or in a glove box) and the material (large or solid, powder or liquid) must be considered. The degree of hazard can be ascertained only by experience and by considering the danger of the actual operation performed. This decision normally should not be made by the personnel in charge of the bioassay laboratory but by someone in close daily contact with the individual and the work. Of course the sample program can not be established without reference to the bioassay laboratory team, and it may, indeed, be a major factor in the direction the program takes..

After deciding who to sample, the type of sample must be selected. Regardless of technical consideration, a urine sample is the easiest biological specimen to obtain. It also has an analysis advantage unless the specimen is to have only its gamma spectrum measured. Arriving at a sampling frequency is no easier than deciding who to sample and what to sample, how should the sampling frequency of a survey program relate to the biological and radioactive half-life of the material, whether the sampling frequency have to be related to the general shape of the excretion curve. High-frequency sampling may give a sensitivity gain for single acute exposure since these values will fall on the steeper part of the excretion curve. This frequently may be justified for persons routinely exposed to high risks, but people exposed to low-level chronic hazards may not benefit from high-frequency sampling.

In general, the shorter the period between samples, the better the estimate on body burdens. The bioassay laboratory resources set a very real limit on the number of samples that can be handled. Since all workers cannot be sampled continously, the workers with the highest potential exposures should be sampled at least weekly and the others less frequently, based proportionately on their potential exposure. Weekly sampling is

probably the starting frequency, and frequency may then be decreased as experience dictates. Samples taken at the beginning of the working week fix any exposures to the preceding week. Maintenance workers who enter high-risk areas should be placed in the high-risk sampling group. Most often they are in the high-risk areas at times when the equipment is not working properly, and thus high exposures are more probable.

4 - METHOD OF COLLECTING SPECIMENS

Even though personnel may be classified correctly under a bioassay program and even though the appropriate sampling frequency may be selected to estimate internal exposure of personnel the results will not be meaningful if the sample is not collected properly. Correct collections of specimens include the following three factors: proper selection of containers, adequate treatment of containers, and effective procedure for collection of specimens.

Plastic containers are frequently selected for the collection of urine specimens since plastic bottles are unbreakable and easier to handle than glass. Feces samples are usually collected in 500 ml ice cream containers. Large polyethylene bags taped to a standard toilet seat are also used to collect feces specimens. The bag is closed by means of tape or a rubber band. If a specimen of blood is collected 10 ml is adequate for the measurement of radionuclides in the specimens. Blood specimens are not collected frequently, but, when they are they should be collected in a vacuum tube.

Breath samples may be collected by inflating large balloons and deflating the balloon into a chemical solution or by exhaling directly and repeatedly through flasks with inlet and exhaust stopcocks. The effectiveness of the large balloons to hold the radioactive gases and the efficiency of the chemical reaction as the gas is transferred from the balloon or directly from the breath to the flask should be determined. Breath measurements of radium compounds of bromide or chloride that are incorporated in the body can be meaningful since radon can be released from cristal matrix of the insoluble compound but the sulfate compounds of radium do not release radon.

Treatment of containers includes the use of preservatives, refrigeration carrier isotopes and reacting compounds. Preservatives should be added to the sample to prevent losses due to ph changes the alkaline side on standing. Urine specimens can be stored up to 1 month if a 1 % volume mixture of hydrochloric and nitric acid is used. If refrigerated, the sample can be retained in essentially its collected state until the analysis is made.

Carrier isotopes (stable isotopes such as I-127) may be added to the containers to prevent the uptake of significant quantities of the radioactive counterpart of the carriers (such as I-131) on

the walls of the containers. In the case of I-131 collection in the container, sodium hidroxide pellets may be added to the container to ensure retention of the iodine in the urine solution prior to analysis.

Effective procedures must be established to ensure the collection of a representative sample of potential activity in the urine of an employee. Such procedures should contain the following requirements.

a) Exceptional care and cleaniness is required to avoid invalidating the analyses by the inadvertent introduction of radioactive contamination from hands, dust, dirty containers, etc.

b) All containers must be clearly marked with identification by name or number of the person submitting the sample, the time of collection and the period of collection.

c) All samples must be submitted in clean areas where no radioactive materials are handled.

d) Compliance with radiation - safety procedures is considered a part of the responsibility of each employee. In some instances compliance with company safety procedure is an explicit condition of employment.

5 - RADIOCHEMICAL LABORATORY

A great majority of radiochemical work is carried out with activities at microcurie levels or bellow. While these quantities of radioactive materials can possibly represent a personal hazard they can be handled without the construction of special laboratories. Special laboratories, particularly the so-called hot laboratories, are beyond the scope of this manual. On the other hand, there are certain criteria of design and operation which will simplify laboratory procedures.

The most likely harmful result of a radioactive spill or accident in a radiochemical laboratory is contamination of equipment, in particular contamination of counting facilities or contamination of samples. As a general rule, a laboratory will become inoperable for technical reasons long before any personal hazard exists and conversely if the operation is technically well executed it is very unlikely that any hazard to personnel will arise.

From the dual standpoints of safety and laboratory operation it is necessary to monitor the handling of radioactive material. Survey instruments of the Geiger-Mueller or ion chamber type, as required, may indicate the personal hazard but are not sufficiently sensitive to indicate contamination problems. Also, even though the level of activity may be insufficient to indicate personal hazard from direct radiation, there is the chance that

sufficient material would become airborne to constitute an internal hazard from inhalation. Therefore, every effort should be expended to see that radioactive material does not spread in the laboratory.

Laboratory design for radiochemical work has two requirements: first, adequate ventilation with ample hood space or glove box space to carry out operations where radioactive material might possibly become airborne, and second, working surfaces that can be readily decontaminated.

Solution handling presents few possibilities of air contamination, and it is only in evaporation and ignition processes that hood ventilation is required. On the other hand, operations with powdered samples can lead to dusting, and the use of a glove box is recommended. If considerable activity is being handled, air monitoring is desirable, even though it will only furnish after-the-fact information on non-routine laboratory operations.

There are two approaches to the problem of working surfaces for decontamination. The first, and most expensive, is to use the corrosion resistant materials such as welded stainless steel. The second is to use surfaces which, if contaminated, can be readily taken up and discarded. Since there are no actually impervious surfaces, it would appear that the second technique is more desirable.

Adequate storage space should be provided for stock isotopes and active samples. That storage should preferably be away from the working area to prevent unnecessary exposure of laboratory personnel. In the storage area there should be work benches and hood space so that large amounts of radioactive material are not necessary to be brought into the radiochemistry area for providing working quantities of these materials. The common requirement is the preparation of working solutions for use as spikes or tracers. The usual millicurie quantity as received should be diluted and carriers or other reagents added in the storage area. Sufficient material may then be transferred to a smaller bottle for use in the laboratory during that day or week. In this way, the amount of radioactivity in radiochemistry areas is always minimal and hazards to personnel or of contamination are reduced .

The handling of most radioactive materials with activities of a millicurie or less is little different than handling other hazardous or toxic chemicals. Naturally, certain radionuclides are more hazardous than others and millicurie levels are not always perfectly safe. As a rule of thumb when considering hazard, quantities of a few maximum permissible body burdens may be handled in the laboratory, but larger quantities should only be processed in the storage area and by competent personnel.

Radioactive solutions should not be pipetted by mouth and in operations subject to spills, surgical or plastic gloves are

desirable to prevent hand contamination. On the other hand, at high radiation levels it is neither necessary or even desirable to attempt remote operation behind shielding. It is very valuable in testing a new operation to make several dry runs without radioactive material. This not only assist in setting up the operation and thus reducing radiation exposure but also may improve the technical quality of the operation.

Depending on the total amount of radioactive material in the laboratory it may be desirable to monitor personnel with pocket dosimeters or film badges. The need for this can be best evaluated by determining the maximum dose rate during operations and the maximum length of time such operations would go on. If this is below 25% of the permissible value for exposure, continuous personal monitoring is probably of little value. Area monitoring of the laboratory with survey instruments will reveal unreported spills and increases in the quantity of radioactive material present.

For the measurement of removable surface radioactive material which might become airborne and present a personal hazard, or be transferred to become a laboratory contamination problem, the best technique is probably the swipe method. In this operation, a piece of analytical filter paper, the proper size for counting, is wiped firmly over a 100 square cm area, rubbing in both directions. The paper is then counted and will give an indication of the amount of activity that might be removable in normal operations.

Attention paid to radiation safety must not be at the expense of standard chemistry laboratory safety procedures. Cuts, acid burns and heat burns outnumber radiation problems by many orders of magnitude. The standard procedures of wearing safety glasses, having safety equipment handy and similar precautions are always necessary.

In summary, the prevention of accidents in a radiochemical laboratory that might lead to personal overexposure or to laboratory contamination can best be controlled by a full knowledge of materials worked with, the processes involved, and the potential hazard.

6 - SAMPLE PREPARATION

When large amounts of organic material are present the first step is usually the ashing of the sample. There is a choice of wet or dry ashing depending on the properties of the sample, such as mass, bulk and physical form, on the oxidation resistance of the organic material and on the volatility of the desired constituent. Dry ashing is simpler but has the disadvantage in that many elements are volatile at ashing temperatures. While it is possible to minimize losses by ashing at 400 to 450 C, the process is prolonged. Many samples ignite to produce local temperatures far in excess of the furnace temperature. Ashing at

a low temperature produces a soft ash while higher temperatures may lead to the formation of refractory residues and may even cause a part of the desired material to combine with the container. Porcelain, silica, nikel and platinum all have an affinity for certain elements at ashing temperatures.

It is not always necessary to produce a pure white ash. Frequently the ashed residue is merely a more convenient form of the sample than the original. Where complete removal of carbon is necessary this may be assisted by feeding in oxygen to the muffle, by ashing after mixing in solid oxalic acid, by moistening the residue with saturated ammonium nitrate and reashing or by breaking up the ash with a mortar and pestle and reashing.

The wet ashing process is more tedious, particularly for large samples, but it is to be preferred unless there is direct evidence that dry ashing is suitable for the particular sample. Wet ashing allows the direct addition of carrier during the ashing process, and volatilization losses occur only with the extremely volatile elements such as iodine, bromine, and the like.

In wet ashing the major oxidizing agent is nitric acid, and frequently the complete oxidation can be carried out this reagent alone. It is generally preferable to add the sample in small portions to a relatively large volume of hot acid. Upon evaporation to a small volume, unoxidized organic matter is indicated by brown fumes and ashing may be completed by repeated additions of nitric acid or nitric acid plus small quantities of perchloric acid or hydrogen peroxide. The best evidence for completeness of oxidation besides the absence of brown fumes is the appearence of the residue on drying. In the absence of the transition elements this should be pure white.

The addition of sulfuric or perchloric acid to assist oxidation is sometimes helpful but may lead to the formation of insoluble compounds such as barium or calcium sulfates or potassium perchlorate. In addition, some elements are more volatile at the higher temperatures reached in fuming with these acids. Kjeldahl treatment of the sample provides rapid ashing where the added sulfuric acid does not present a problem. It is applicable, however, only to relatively small samples of material and does introduce a large volume of a difficulty volatile acid.

When organic materials are not present in large quantities, a reasonable variety of samples will be open to acid attack. In some cases it is not even necessary to obtain a complete solution of the entire sample if it can be shown that the desired element may be removed by leaching. This type of attack can be quite useful as leaching also affords some separation from bulk matrix materials.

Samples which do not yield to individual mineral acids may respond to treatment with mixed acids such as aqua regia. For

samples containing silicates in small amounts, a more vigorous attack with nitric and hydrofluoric acids followed by evaporation with sulfuric or perchloric acids may be of value. This latter technique opens up many silicate minerals and volatilizes the silica present. In all cases, the carrier should be added before or with the acids.

Samples that resist acid attack or residues from acid solutions or ignitions may require fusion to bring them into solution. The high temperature of fusion as compared to acid attack promotes solution and as previously mentioned also may promote exchange with the added carrier. In some cases the fusion may bring about volatilization of the desired constituent and this point must be checked.

There are several types of fluxes available for sample attack. The first is the acid flux represented by potassium bisulfate or potassium pyrosulfate. This is quite effective in dissolving oxides or other basic materials and the fusion usually can be dissolved in water or dilute mineral acid. The alkaline fluxes such as sodium carbonate or the hydroxides attack the acid materials such as silicates. The melt may be dissolved in dilute mineral acid with the release of silica which must then be removed. A third class of fluxes are those whose primary function is oxidation. These include nitrates, nitrites and sodium peroxide. These are frequently valuable for the solution of refractory materials but are otherwise similar to the basic fluxes. Some success has been reported with using a mixture of basic fluxes with fluorides for attack on silicates and volatilization of some silica. All fusions must be considered in light of the attack of the flux on the fusion vessel. Platinum is resistant to most molten fluxes except for the hydroxides and sodium peroxide. These fusions can be carried out in iron or nickel crucibles which are also attacked but are cheaper to replace.

7 - CHEMICAL SEPARATIONS

In the chemical separations required for radiochemical analysis, advantage is taken of practically all techniques. While there has been a tendency to utilize ion exchange techniques very widely, the usual radiochemistry is based on classical precipitation techiques. In fact, even when the more recent methods are used in separation it is found that they are most effective when combined with classical methods.

The general goals of chemical separations are to reduce the mass of the sample for counting, to achieve chemical purity sufficient for recovery measurement and to achieve decontamination of the substance sought from other radionuclides.The degree of separation required is dependent on whether the final method of determination can be carried out in the presence of some bulk material and whether it can distinguish between the desired and contaminating activities.

It should be realized that all separations are imperfect and that the conditions set are a compromise between collecting all of the desired constituent and removing the unwanted materials. In radiochemical analysis it is common to sacrifice the former somewhat to improve the removal process. Again, since no separations is absolutely specific, a combination of two or more different separations are required to isolate the desired element. As in more classical techniques it is usually desirable to remove the desired constituent from the bulk material, particularly when precipitations are involved. Even in the presence of carrier the losses incurred in precipitating a major constituent can be quite high. In precipitating the minor constituent, the small bulk of the material does not carry as much contamination and this may be removed by repetitive separations.

The terms decontamination factor or separations factor are widely used in radiochemical analysis. The factor is merely the ratio of the activity of a specific contaminant to that of the desired nuclide in the original sample compared to the ratio in the result sample. The terms are used either to cover the decontamination in a single step or in an overall analytical procedure.

Since many of the separations in radiochemical analysis are carried out in small volumes the centrifuge is widely applied. It has become common practice to redissolve and reprecipitate rather than to wash precipitates carefully. This may be repeated several times very rapidly and will generally give good decontamination. As noted above, where the element sought and the contaminant have similar properties it is preferable to use two different precipitations, a precipitation followed by an extraction, or any two widely different steps to achieve good decontamination.

One process that is frequently of value is the one called scavenging. This is usually applied to a precipitation carried out to remove contaminating radionuclides after the bulk matrix of the sample has been removed. The process involves the addition of a group carrier, precipitation and discarding of the precipitate. A typical example would be an addition of iron carrier followed by hidroxide precipitation to remove rare earths and other heavy metals in a determination of an alkali or alkaline earth. Frequently the scavenging procedure is repeated to improve decontamination, but it is only rarely that the scavenge precipitate is redissolved and reprecipitated to recover any of the desired constituent that may have been absorbed.

Time may enter into consideration of the separation techniques to be used. In some cases the nuclide sought has a short half-life and determination must be carried out rapidly. In other cases, the nuclide sought may be the parent or daughter in a chain and the timing of the analysis must be adjusted to break the chain in the proper way so that only the desired nuclide is counted.

No attempt will be made to detail the common separation techniques but they are merely listed here as a reminder.

- Precipitation
- Electrolysis
- Electrolytic Displacement
- Extraction
- Distillation
- Ion Exchange
- Chromatography

8 - RECOVERY MEASUREMENT

Most radiochemical analysis are carried out in such a way that losses occur during the separations. These losses are usually necessary in order to obtain adequate separation from the large number of contaminants that may be present. Thus, it is necessary to include a technique for estimating these losses in the development of the analytical procedure.

The least desirable technique is to determine the average recovery for a series of spiked samples (samples to which a known amount of radionuclide has been added) and to use this recovery value for future determinations. This does not make allowances for changes in technique under determinations. This does not make allowances for changes in technique under conditions encountered in the analysis. If it is necessary to use this method, a minimum requirement is that one or more spiked samples be run with each batch of analyses to assure that the estimated recovery is being maintained.

By far, the largest number of recovery measurements are made on the basis of adding an isotopic carrier to the sample and determining the amount of carrier recovered. This is very generally applicable and is capable of yielding excellent results. The final determination of recovery is usually made gravimetrically. While this is the simplest method it is not the most desirable since weight is a non-specific property. It is preferable to determine recovery based on a volumetric reaction or a photometric measurement which can be made specific for the element being determined.

In determining recovery gravimetrically, the compound weighted must have the properties required by a good weighting form for standard gravimetric analisys, that is constant composition, low hygroscopicity, and so forth. After separation, the compound should be pure within the limits of accuracy desired in the analysis. It is also highly desirable that the final gravimetric compound mounted for counting be the same as that used to standardize the initial carrier solution.

In many cases where bulk samples must be analyzed, it will be found that the original sample contains appreciable quantities of the element used as a carrier. In such it is possible to

determine the original concentration of this element and subtract from the for a amount recovery measurement. Another technique is the radiometric measurement of recovery. In this case a different radioisotope of the nuclide being determined is added and the recovery is based on measurement of the added isotope.

This technique, of course, requires that the added isotope have a different emission than the one being determined or a different energy of the same type of emission. The technique is limited by the available isotopes. A typical example is the use of the beta active U-237 for determining the recovery of alpha emitting uranium isotopes. A special application that can be made is the use of Sr-89 in determining Sr-90. In this case the recovery is determined by gamma measurement of the final purified strontium solution. Since the Sr-90 is determined by separation and measurement of its yttrium daughter, the Sr-89 does not interfere in the final counting..

While the measurement of recovery does allow correction of the results for the necessary losses incurred in the chemical separations, it should not be used to allow for losses due to sloppy chemistry. After a number of determinations with a method, the analyst will find a general range of recovery values that will be obtained with good technique. A recovery value appreciably below this should probably be cause for rejection of the analysis since the count obtained will require multiplication by an excessively large factor to obtain the final result. On the other hand, a high recovery, well above the normal range, is likely to be a sign of either faulty analysis or the presence of the carrier material in the original sample. This should likewise be a cause for rejection of the analysis.

9 - COUNTING PROCEDURES

The specific procedures used in sample counting are highly dependent on the equipment available, the sample mounting and the characteristics of the radiation to be measured. In measuring radionuclides we are interested in both qualitative and quantitative analysis. In the majority of cases, both identification and measurement are possible. Quantitatively, the desired answer is usually the disintegration rate of the sample.

Qualitative identification makes use of all the properties of radionuclides. Physical measurements may include type of radiation, energy and half-life or disintegration constant. These properties may yeild positive identification but are not always simple to determine, and radiochemical treatment may be required prior to any measurement.

It is rarely possible to do a complete qualitative and quantitative analysis directly on the original sample when several nuclides are present. Most often,either complete or partial chemical separation must be made. A radiochemist requires a high degree of skill in analytical chemistry with added

knowledge of radiation measurements.

9.1 - COUNTING BACKGROUND AND EFFICIENCY

9.1.1 - BACKGROUND

Any counter will show a certain counting rate without a sample in position. This background counting rate comes from several sources: Natural environmental radiation from surroundings, cosmic radiation and the natural radioactivity in the counter material itself. The background counting rate will depend on the amounts of these types of radiation and on the sensitivity of the counter to the radiations.

As general rule, the larger the sensitive volume of a given type of counter the higher will be the background counting rate. This is to be expected since a large number of natural radiations will be intercepted by the larger volume and a larger area of the counter construction material is exposed to the counter.

Radiation from the environment and soft cosmic radiation can only be reduced by shielding the counter. Both of these sources give penetrating radiation and heavy metals such as iron, lead and mercury give the most compact shields. Any practical shielding does not reduce the intensity of the background count from hard cosmic radiation. If it is necessary to count with an extremely low background, an additional screening procedure is required to reduce the effect of cosmic radiation.

In measuring the activity of the sample, it is necessary to correct count obtained for the background which would appear without the sample in position. If the sample has very high activity the background counting rate will be negligible in comparison and can be ignored. On the other hand, with samples of very low activities the background counting rate may be the controlling factor in the counting statistics. For such samples, therefore, it is desirable that the background counting rate be maintained at as low a level as possible. This not only reduces the counting time required for a given relative counting accuracy but it may enable a sample to be counted that could not be reliably counted otherwise.

The stability of the background counting rate is particularly important when low activity samples are being measured. If a series of measurements on counter background are made over a period of time, the results should lie within the expected standard deviation limits.

In low level determinations it is also necessary to run an appropriate number of blanks to determine the true baseline for suctraction from sample counting results. It is also desirable, however, to run counter background with an empty sample to check the blank determination. Suitable blanks may also be treated by the control chart technique to determine if their variability is

within the proper limits

amount of time devoted to counting of background depends on the level of activity being measured. In general, with low level samples, this time should be about equal to that devoted to counting the sample.

3.1.2 - EFFICIENCY

The quantity in the measurement of a radioactive substance is the number of disintegrations per unit time. It is seldom possible to make an absolute measurement of the disintegration rate, it is necessary to compare the sample with one or more standards. The standards determine the counter efficiency which may then be used to convert sample cpm to dpm. It should be noted that more erroneous analytical results in activity measurements can be traced to improper standardization than any other cause.

In the standardization process, the ideal standard is a known quantity of the isotope to be measured, prepared in exactly the same form as the samples and counted under identical conditions. In this way, the factors of self-absorption, back-scattering, radiation energy, counter geometry and the inherent efficiency of the detector are empirically corrected for.

In some cases of routine counter operation it is desirable to have a counter reference standard which is used merely to check the operation of the counter to test its day-to-day reliability.

3.2 - ALPHA COUNTERS

The relatively high energy of alpha particles is dissipated in a short distance giving a high specific ionization. The ionization produced in air, in other counting gases, in a scintillation phosphor, or in a semiconductor leads to a relatively large electrical pulse as compared to other types of radiation. The large pulse requires a minimum of amplification before going to a scaler or register. In this sense, alpha counting is generally much simpler than beta or gamma counting.

The gas detectors include the ionization chamber, the proportional counter and even the Geiger counter. For scintillation detection, activated zinc sulfide is most commonly used, while both silicon and germanium semiconductor detectors are useful.

The efficiency of a good alpha counter should approach 50% and may reach 52% when backscattering from a metal planchet is included. Efficiencies less than 50% are usually due to self-absorption within the source itself and the greatest problem in source preparation is to produce an essentially weigh-less source to minimize self absorption.

It is possible in many cases to identify alpha emitters and to

determine two or three emitters simultaneously by alpha spectometry. Because of the high self-absorption shown by alpha emitters it is necessary, however, that chemical separations be made to free the alpha emitters from the bulk of the sample. The separations do not have to give high decontamination from other radionuclides but the total mass of sample in the spectrometer should ordinarily be in the microgram range.

Spectrometers are based either on the gridded ion chamber or on solid state detectors. The first type is most practical at this time for large area low level samples since high geometria is maintained. Solid state detectors, however, have overcome most of their early disadvantages while maintaining their advantage of extremely high resolution.

9.3 - BETA COUNTERS

Beta counting is dependent upon the energy of the radiation under measurement. For most preparation a thin mica-endwindow Geiger-Mueller counting tube or a plastic scintillator will suffice. In low level beta counting, recourse must be had to the use of efficiently shielded detectors protected with a multiplicity of other tubes arranged in anticoincidence. By such means, background of 0.1 - 1 count/min may be achieved. For the measurement of very weak beta radiation, such as that from tritium, it is necessary to do internal counting, by counting in a liquid-scintillation system.

9.4 - GAMMA COUNTERS

Gamma-ray spectrometry is used, where applicable, for routine determinations of radionuclides in bioassay samples. By the direct counting of untreated samples, many tedious radiochemical separations are avoided. Many individual gamma rays are detected and their energy measured and recorded by the spectrometer, in such a manner that the operator is provided with a measure of the distribution and intensities of gamma-rays from the source of radioactive material. Since each gamma-emitting radionuclide has a characteristic range of radiation energies it is usually possible to identify and measure the nuclides in the sample quite simply.

Sodium iodide scintillation spectrometers are able to resolve medium energy photons of at least 8-10 per cent difference in energy, providing their intensities are similar. In the presence of interfering activities, the accuracy with which gamma-ray energies or intensities can be measured is limited by this resolving power and by instrument stability. It is usually possible to measure gamma-ray energies to an accuracy of at least 1 per cent and their intensities to that approaching the best standard source available (i.e. 2 to 3 per cent), all these figures being subject to the statistical uncertainties of the counting procedures.

The detector consists generally of a cylindrical 5x5 in thallium activated sodium iodide crystal which is mounted on a photomultiplier, operating in conjunction with a linear amplifier and multichannel pulse height analyzer. The crystal and photomultiplier are contained in a 8 cm lead shield.

Skin and blood samples, nasal swabs and other miscellaneous samples are counted directly on top of the crystal. Urine samples are counted in polyethylene containers of capacities until 2.0 l. Fecal samples can be counted directly in the containers in which they are collected. Usually however, they are ashed and the residue dissolved in a mixture of concentrated nitric and hydrochloric acids, diluted to 200 ml and counted.

The pulse-height spectrum produced is characteristic of a particular radionuclide or nuclides and the components can often be identified immediately by reference to the appropriate literature. If there is doubt about the identification, can frequently be obtained by decay measurement on the sample. In some instances, resort must be made to chemical separations but in such cases the main advantage of spectrometry has been defeated. The gamma-ray energies and intensities corresponding to the total absorption peaks (photopeaks) of the observed pulse-height spectrum, are determined by means of standards of gamma-emitting nuclides of known energy and activity. The intensity of the gamma-ray is measured by the area of the relevant photopeak. For gamma-emitters, the limit of detection for a particular detector depends upon a number of factors, including gamma-ray energy, sample volume and geometry and the time for which the sample is counted.

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