AUTORADIOGRAPHIC TECHNIQUE IN CONTAMINATION CONTROL

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For detecting extremely low levels of contamination, autoradiography offers a simple method which is a hundred times more sensitive than usual survey meters. Used in conjuction with routine survey meters, autoradiography of swipes taken from laboratory areas of contamination will be a useful, sensitive, long-term monitor. The technique can also be used for assessing the efficiency of different decontaminating procedures.

Medical radioisotopic work involves handling of loose unsealed liquid radioisotopes (such as dilution, fractionation and dispensing to the patients) in which chances of spill and radiocontamination are always present. Moreover, patients taking radiomedicines are themselves radiation sources and their bedding, urine, faeces, sweat may all be contaminated; their improper handling may lead to wide spreading of the contamination. Possibilities of radiocontamination also exist in experiments with radioactive animals and radiochemical tagging of drugs.

External contamination may lead to internal contamination through ingestion and inhalation and even minute amounts (a microcurie or less) of radioistope may produce internal hazard. Contamination also raises the background of the area and lowers the sensitivity of radioactivity measurements. Its detection and control, therefore, are of paramount importance.

The normally accepted maximum permissible level of surface contamination is 10^{-4} $\mu c/cm^2$ of beta activity which is difficult to detect with conventional survey meters. This paper deals with experiments in autoradiography and presents a simple and sensitive method to supplement the survey meter measurements in the control of contamination.

AUTORADIOGRAPHY FOR DETECTION OF CONTAMINATION

Autoradiography is used for determining the location and amount of radioactive materials. The technique involves placing a radioactive specimen in contact with photographic emulsion. Radiation passing through the film sensitizes its silver grains which upon development show the location of radioactive atoms in the specimen. It may appear as an area of varying density of blackening easily seen with the naked eye or it may consist of grains or tracks seen under a microscope. Autoradiography is easy to apply, realiable, requires no elaborate equipment and provides a permanent record.

There are many types of films that can be used for autoradiography, depending on sensitivity and resolution. Whereas in radiobiological studies resolution is important (as for instance radioisotope localisation at the cellular level), sensitivity (or the ability to get a record with the lowest activities) is the requirement here. Table 1 gives the characteristics of the different emulsions used in autoradiography.

Out of the many types of films (X-ray, nuclear emulsion etc.) and different techniques (contact, coating and stripping), X-ray films and contact autoradiography were found most suitable for the present investigation. Although contact autoradiography does not give as good a resolution as do other methods, it is the simplest. The film is placed as close as possible to the specimen and left for development.

TABLE 1

CHARACTERISTICS OF DIFFERNET EMULSIONS

Emulsion	Thickness of emulsion (microns)	Halide content %	Grain size μ	Grains per 1000 μ^3	Resolution μ	Back ground fog	Relative sensitivity*
Photographic	10—20	10—15	1—4	6	..	..	..
X—Ray	10—30	10—20	2—6	6	30—60	High visible	1
Nuclear	50—1000	45	0·2	10,000	5—7	Medium	0·1
Stripping	5	45	0·2	10,000	1—3	Negligible	0·01

*(Amount of a particular radioisotope to produce a given blackening).

The time of exposure for autoradiography depends on the half-life of the radioistope and other factors. Most of the radiation energy is given out in the first two half lives. Fogging may occur with time due to chemical effects of environment. The latent image being formed in the emulsion while an autoradiograph is being done, may itself fade with time. For all these reasons, there is an optimum time of exposure (1—2 halflives) provided this is not too long.

MATERIALS AND METHOD FOR DETECTING MINIMUM CONTAMINATION

Experiments were carried out to find out the minimum level of radioactive contamination that could be detected by autoradiography. The radioisotopes $I-^{131}$ and $P-^{32}$ used for this study are most commonly used in nuclear medicine. $I-^{131}$ is a beta-gamma emitter with a half-life of 8 days, having a some-what complex spectrum; the predominant group of beta rays has a maximum energy of 0·61 mev; the most important gamma ray is the 0·36 mev photon. $P-^{32}$ is a pure beta emitter with a half-life of 14 days and a maximum beta ray energy of 1·7 mev.

The method employed for the detection of the minimum amount of activity was contact autoradiography with X-ray films. 'Kodak Blue Brand' X-ray films and Kodak developer and fixer supplied in packed tins were used. The time of exposure was 15 days for both $I-^{131}$ and $P-^{32}$ (No detailed studies were made to fix the optimum time of exposure). During the processing of the film, the temperature of the developing and fixing baths were maintained at 20°C. The films after having been kept in the developing bath for 3 minutes were dipped in an acetic acid bath and immediately transferred to the fixing bath where they were kept for 15 minutes. The films were then thoroughly washed with running water and dried in a film drier.

Particular care was taken in the series of experiments to ensure that there was no background radiation from radiation sources which could decrease the sensitivity of the studies. The experimental room was first checked for radioactive contamination Separate tables and sets of glassware were used for the two isotopes to avoid possibility of cross contamination. All apparatus was cleaned thoroughly and checked before the start of the experiment.

Desired quantities of $P-^{32}$ and $I-^{131}$ were made with specific activities of about 2·0, 1·0. 0·75, 0·50, 0·33, 0·25 and 0·16 μc/ml respectively. The exact activities were calculated from the known specific activity of the parent solution and the degree of dilution.

Three clean and dry glass plates (12″×10″) were employed. One was used for contamination with $I-^{1-1}$ the other with $P-^{32}$ and the third served as control. The glass plates were divided into 6 approximately equal rectangular areas demarcated by a glass pencil. Exactly 1 ml of $I-^{131}$ solution of strength 2 μc/ml was taken into a clean pipette and dropped slowly and carefully on to the centre of the first of the 6 areas of the glass plate; this was allowed to spread naturally and then dried with the help of an infrared lamp. The next area of the glass plate was then contaminated with $I-^{131}$ solution of the next lower strength, 1 μc/ml. All the six areas were similarly contaminated, each with a different strength of activity. The procedure was repeated on the other glass plate with $P-^{32}$ using clean pipettes for each operation.

After the solution had dried the glass plates were covered with thin tissue paper and transferred to a dark room. X-ray film was placed in contact with the tissue paper on top of the glass plate and the whole assembly was securely fastened inside a wooden contact frame to ensure proper contact between the surfaces. The frames were completely wrapped with black paper and kept in a radiation-free place for 15 days. The same procedure was repeated for the control glass plate. At the end of the period the films were developed under the conditions described earlier.

The area over which the active solution had spread was calculated in each case from the contours of the autoradiographic image negative placed over a graph paper. From the areas and the known activity deposited initially, the surface contamination intensity expressed $\mu c/cm^2$ was calculated. In the first set of experiments where the initial activities used were in the range of 2μc to 0·16 μc even the lowest activity gave recognisable darkening. Instead of repeating the experiment with fresh glass plates and weaker activities, the same glass plates were used for the second experiment. The experiments were thus repeated with fresh X-ray films over the same plates, exposing for a fortnight and subsequently developing. This was continued till autoradiographs were obtained in which blackening was indistinguishable (by visual inspection in an X-ray viewing box) and consquently the contours were difficult to trace against the background. Fig. 1 gives some of the patterns obtained.

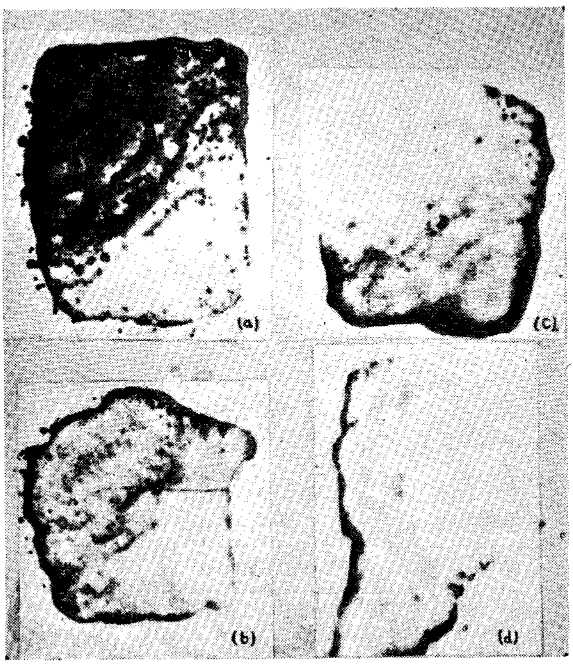

Fig. 1.—Sample autoradiographs I^{131} level of contamination (*a*) **1.4** × 10^{-5} $\mu c/cm^2$, (*b*) (6,5 × 10^{-6} $\mu c/cm^2$, (*c*) **8.3** × 10^{-6} $\mu c/cm^2$ and (*d*) **3.6** × 10^{-6} $\mu c/cm^2$

From these experiments it was concluded that the minimum contamination detectable by the method was about 8× $10^{-6}\mu c/cm^2$ for both $I-^{131}$ and $P-^{32}$ (these correspond to the activities present at the start of the exposure period)

COMPARISON OF SENSITIVITY OF SURVEY METER, CONTAMINATION MONITOR, AND AUTORADIOGRAPHY

The survey meter used in the studies was AEET type GM 29B. This is a battery operated portable survey meter with thin-walled GM counter of diameter 5 cm, length 10 cm and wall weighing 30 mg/cm^2. A metallic window was placed over the detector to cut off beta rays completely. There are 3 ranges, corresponding to 0·2, 2 and 20 mr/hr full scale deflection. The normal background with this instrument corresponded to about 0·02 mr/hr.

The contamination monitor used was AEET Type CM 167, used with an end window GM counter of radius 1·3 cm, length 6 cm, and window weighing 2 mg/cm^2. The normal background of the monitor is about 0·5 cps. Experiments were also carried out using the same probe shielded with about an inch of lead.

The detectors of the survey meter and contamination monitor were kept 1 cm. apart over planchets of diameter 2·0 cm. containing known activities of P^{32} and I^{131} From these the contamination level ($\mu c/cm^2$) which would give a sample reading of twice the background was computed. These were taken to be the minimum detectable levels. Table 2 gives a comparison of the minimum activities detected by the 3 methods.

Apparently the autoradiographic method is far more sensitive than the survey meter or contamination monitor though it does not give an immediate or direct indication of the contamination level as one has to wait for the exposure period and blackening has to be correlated with the contamination level by previous calibration for isotopes.

AUTORADIOGRAPHY FOR CONTAMINATION CONTROL

Swipes are taken from suspected areas of contamination such as table tops, sinks, taps, etc. by rubbing them with tissue paper, which is then autoradiographed. It can then check up whether there is any significant radiocontamination at any spot. By adopting the technique of taking swipes periodically from such areas and testing them autoradiographically it is possible to keep strict control on radiocontamination. This is a useful addition to monitoring of work areas with survey meters.

AUTORADIOGRAPHY FOR STUDYING DECONTAMINATION

Autoradiography can be used to assess the effectiveness of decontaminating procedures.

In this paper contamination with I^{131} on glass, perspex, waxed linoleum, unwaxed linoleum and lead is studied.

Two rectangular pieces (4″ × 4″) from each surface were contaminated with 0·1 μc I^{131} (0·2 ml) at the central portion and dried under an infrared lamp. One plate of

TABLE 2

MINIMUM DETECTABLE CONTAMINATION LEVEL, $\mu c/cm^2$

Isotope	Survey meter	Contamination monitor		Autoradiography
		G.M. Probe unshielded	G. M. Probe shielded	
I^{131}	$3·2\times10^{-4}$	$8·0\times10^{-4}$	$4·0\times10^{-4}$	8×10^{-6}
P^{32}	$5·3\times10^{-4}$	$3·2\times10^{-4}$	$1·6\times10^{-4}$	8×10^{-6}

each surface was kept as standard and the other used for decontamination. The surfaces to be decontaminated were washed with warm water and dried. A check over these surfaces with a survey meter showed that there was some activity above the background.

Normally recommended decontaminating procedures were then applied to the surfaces. Perspex and glass were treated with 10% Nansa solution, lead was treated with dilute hydrochloric acid and linoleum was treated with carbon tetrachloride. All the surfaces were finally washed with water and dried. Survey meter failed to detect any activity on them.

The contaminated and decontaminated surfaces were then kept in contact with X-ray films, and developed after an exposure of 72 hours.

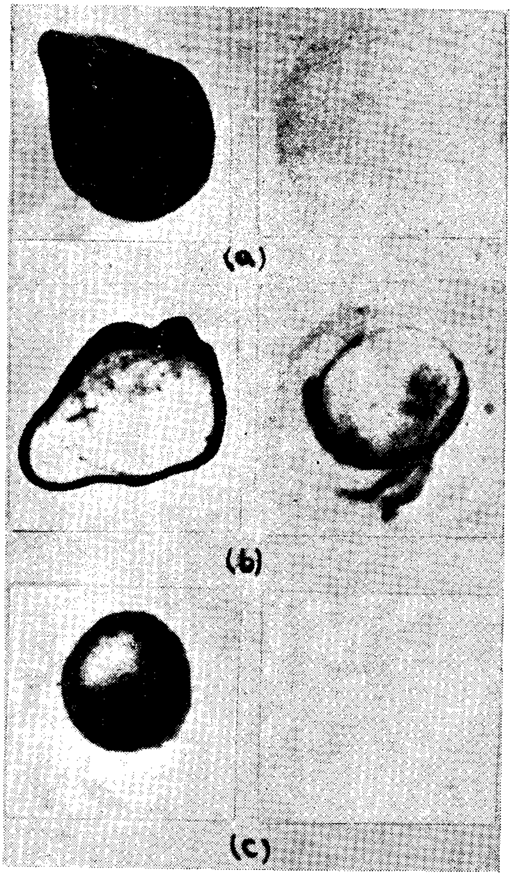

Fig. 2—Surfaces on contamination (left) and after decontamination (right) (*a*) lead (*b*) unwaxed linoleum (*c*) perspex.

RESULTS

The developed films immediately revealed how effective had been a particular decontaminating procedure. (Fig. 2) Perspex and glass surfaces were found to be completely decontaminated. Treatment with dilute HCL could not completely decontaminate the lead surface, although the activity remaining was minimal. In the case of unwaxed linoleum most of the contamination remained on the surface even after decontamination; on the other hand, for waxed linoleum the activity remaining was much less even though some activity did remain.

This is precisely why linoleum flooring, commonly used in radioisotope laboratories, should always be kept properly waxed.

CONCLUSION

Autoradiography is far more sensitive than conventional survey meters for detecting very low levels of radiocontamination that may occur in radioisotope laboratories. Used in conjunction with normal survey procedures, autoradiographs of swipes taken from regions of contamination can be used as an effective monitor for control of radiocontamination.

The technique is also useful for accurately assessing the effectiveness of different decontaminating agents on various radioisotope-contaminated surface combinations.

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