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# ABSORPTION OF PLUTONIUM THROUGH THE SKIN OF THE RAT: RATES OF PENETRATION AND METHODS FOR SKIN DECONTAMINATION

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# ABSORPTION OF PLUTONIUM THROUGH THE SKIN OF THE RAT: RATES OF PENETRATION AND METHODS FOR SKIN DECONTAMINATION

by


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Radiological Sciences Department


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## ABSTRACT

The rates of absorption of various $Pu(NO_3)_4$ solutions through the skin of rats were determined. The amount of plutonium present in the carcasses of rats depended upon the normality of the acid vehicle, the area contaminated, and the length of time exposed. The percentage of the applied plutonium absorbed increased with greater acidity, larger area, and longer period of exposures. The plutonium in the carcasses of rats at various time intervals after application of plutonium to the skin was significantly less in rats subjected to a decontamination procedure than in rats similarly contaminated, but not decontaminated.

The effectiveness of a number of decontaminating agents, administered according to various procedures, was studied. It was found that decontamination efficiency depended upon the concentration of plutonium present, the greatest percentage of plutonium being removed when a relatively large quantity of plutonium was applied. The most effective agents were those that removed the superficial skin layer.



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# ABSORPTION OF PLUTONIUM THROUGH THE SKIN OF THE RAT: RATES OF PENETRATION AND METHODS FOR SKIN DECONTAMINATION

## INTRODUCTION

Methods of introduction of plutonium into the body are by ingestion, inhalation, and absorption through the skin. The first two items constitute hazards which by their nature are generally controllable except in the event of a major accident. It is skin contamination that constitutes a continually present and insidious risk in the handling of plutonium in laboratories.

To protect against skin and wound absorption, Hanford employees exposed to the possibility of plutonium contamination are required to record a minimum of two hand counts per day. The warning limit of contamination is arbitrarily taken as 500 dis/min. Supervision in laboratories employing plutonium is responsible for sending all employees with abrasions of the skin to First Aid. So long as First Aid regards the injury as an unhealed break, the employee is not permitted to work with plutonium[1]. Unless plutonium can be absorbed through the skin in appreciable amounts the warning limit of 500 dis/min is excessively low. Increase of the limit would save much time spent in decontamination. Time lost due to minor scratches is appreciable, and could be eliminated if it could be shown that such scratches cause no increase in the rate of plutonium absorption.

The required information is: What is the absorption rate of Hanford product solutions through intact skin? What is the absorption rate through broken skin? How are these two affected by the use of $KMnO_4$ solution followed by $NaHSO_3$ solution for decontamination? If an injury is sustained while handling product activity, the wound is immediately rinsed in cold water, bleeding is encouraged and suction may be applied. How effective are these measures, and what further steps should be taken? How much time is available after skin contamination to conduct decontamination procedures?



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Numerous publications have appeared describing studies on the absorption through the skin of various inorganic and organic compounds. General reviews on skin permeability and metabolism have appeared over the years and show the trend of thought from the concept of skin impenetrability to the cautious principle that many substances have some ability to pass through the skin[2,3,4]. The most generally accepted routes for substances penetrating normal skin are the sebaceous glands and hair follicles. However, the degree or amount of penetration is limited by the substance which penetrates, the vehicle which effects transfer of the substance, and the skin.

Loeffler, et al[5,6,7,8,9] have reported studies on the degree or rate of penetration of strontium solutions through the skin No thorough study of plutonium absorption through the skin has been reported. The present study was undertaken to evaluate the degree of percutaneous absorption of plutonium that can be expected under various conditions of the tissue and the contaminant. The parameters are many, progress is slow due to analytical difficulties, and only preliminary results are reported in this document.

Concurrent with the work on percutaneous absorption, studies were made of the effectiveness of various procedures for the removal of plutonium contamination. The problem of removal of radioactive contamination has been examined in many laboratories and general reviews on this subject have been published by Sachs[10,11,12]. Techniques and procedures for removal of plutonium from skin are summarized by Wirth[13] and Cox[14]. Experimental studies on removal of plutonium from skin are limited to the work of Nickson[15], who removed plutonium from wounds by a water flush. We have attempted to screen and evaluate some of the more common decontaminating agents and procedures.



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## RATES OF PENETRATION

### Methods

All experiments were performed on rats weighing 250 to 350 grams. Areas of skin to be contaminated were shaved with electric animal clippers 24 hours before exposure. Animals showing any evidence of damage to the skin were rejected for these experiments. Any slight scraping of the skin, not grossly evident, will have healed before 24 hours. The contaminating solutions contained Pu(IV) in nitric acid of various normalities and were applied in microliter amounts on the ventral side of the thighs or on the dorsolumbar areas. Constant areas to be exposed were obtained by blackening the end of a hollow glass cylinder of known diameter and pressing the end against the skin. A circle was thus obtained with a known area and whose boundries could be clearly distinguished. The length of exposure varied from 15 minutes to 168 hours. During experiments involving exposures up to six hours, animals were under Nembutal anesthesia and, except for the area of skin exposed, were covered with absorbent paper to prevent splattered or air borne contamination. With exposures of longer duration, animals were anesthetized for six hours, after which the contaminated area was covered with a layer of "Tydrin", a quick drying, self-sealing fungicidal and bactericidal ointment. (Pitman-Moore Company, Division of Allied Laboratories, Inc.) It was recognized that this ointment might affect absorption, but it seemed necessary to prevent spread of contamination to other parts of the body or to cage surfaces. The contaminated spot in the longer exposures was on the dorsolumbar region, and animals found it difficult to lick or scratch the wound. However, since the absorption of ingested plutonium is less than 0.003%[16] any contamination via this route would be insignificant.



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At the completion of exposure, animals were sacrificed by a blow on the head. The pelt was removed and discarded except for an area of skin about one-inch square, the center of which was the contaminated spot. Livers were removed and analyzed separately from the remainder of the carcass. All three fractions were weighed, digested in concentrated nitric acid on a hot plate, and muffled to a white ash in an electric muffle furnace at 600° C. The amount of plutonium present in the section of skin was determined by dissolving the residual ash in nitric acid and directly counting the residue from a dried aliquot of this solution in an alpha proportional counter. Plutonium in the residual ash from the liver and carcass samples was extracted by standard laboratory procedures[17] and its radioactivity measured. By determining the amount of plutonium deposited in the total carcass, excluding pelt and section of skin exposed, and comparing with the total amount of plutonium applied on the skin, the fraction absorbed was calculated.

## Results

Results of preliminary experiments are summarized in Table 1. Although sufficient data were not obtained to evaluate the individual effects of all variables, it is clear that absorption is slight, except at acid concentrations which virtually destroy the skin. The rate of penetration of plutonium appears to increase during the first hour following contamination. This may be due to a lag period before the skin barrier has been penetrated by the acidic solutions employed. There is also some suggestion that percentage absorption may be greater for more dilute solutions.

Table 2 summarizes an experiment designed to study plutonium absorption as a function of time. Five groups of three rats each were exposed to ten microliters of a solution of Pu(IV) in 10 N $HNO_3$ placed on a 1 $cm^2$ area of the clipped dorsolumbar section of the skin. The deposition



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TABLE 1

Percutaneous absorption of plutonium administered as acid solutions to the skin of rats on the ventral side of the thigh.

| Plutonium Applied (mg) | Normality of $HNO_3$ | Volume (μl) | Area Exposed ($cm^2$) | Time (hrs) | No. Rats | Per Cent Absorbed | Per Cent Absorbed/ $cm^2$/hr |
|---|---|---|---|---|---|---|---|
| 1.3 - 2.1 | 12 | 5 | 0.03 | 4 | 5 | 0.0539 | 0.435 |
| 0.15 | 12 | 5 | 0.25 | 4 | 2 | 0.460 | 0.425 |
| 0.5 - 0.6 | 4 | 100 | 1.1 | 1 | 6 | 0.000935 | 0.000823 |
| | 4 | 100 | 1.1 | 0.5 | 5 | 0.000073 | 0.000131 |
| | 4 | 100 | 1.1 | 0.25 | 6 | 0.0000475 | 0.000175 |
| 0.009 | 2 | 100 | 1.1 | 1 | 2 | 0.0016 | 0.0014 |

TABLE 2

Percutaneous absorption of plutonium nitrate solution as a function of time. Ten microliters containing about one microgram of Pu(IV) in 10 N $HNO_3$ was applied to 1 $cm^2$ of the dorsolumbar section of skin of each rat.

| Duration of Exposure (hrs) | Plutonium Recovered (Per Cent of Administered Dose) | | | |
|---|---|---|---|---|
| | Skin | Liver | Carcass | Total |
| 1 | 89 | 0.00028 | 0.006 | 89 |
| 6 | 89 | 0.017 | 0.12 | 89 |
| 24 | 89 | 0.1 | 3.0 | 92 |
| 72 | 55 | 0.7 | 5.3 | 61 |
| 168 | 57 | 1.15 | 7.8 | 66 |



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in the carcass and liver was determined at the end of one hour, 6 hours, 24 hours, 72 hours, and 168 hours. These results are shown graphically in Figure 1. Absorption during the first 6 hours is relatively slow, followed by a markedly increased rate of absorption over the balance of the first 24 hours. The rate falls off gradually over the remainder of the 7 day period. The last two exposure groups showed a poor material balance, where approximately 40 per cent of the applied plutonium was not recovered No concern is felt over the possible ingestion of this material by the rats since, in the case of these prolonged exposures, even if all the plutonium had been consumed it would not have significantly affected results.

Although we found, as will be described later, that by swabbing the skin with a mixture of saturated $KMnO_4$ plus 0.2 N $H_2SO_4$[18] most of the plutonium applied to skin could be removed, an experiment was performed to determine whether this method actually prevented the further deposition of residual plutonium present in the skin layers. Five groups of 3 rats each were used in these studies. One $cm^2$ of clipped dorsolumbar skin was exposed to a solution of Pu(IV) in 10 microliters of 10 N $HNO_3$ for 15 minutes. Decontamination involved swabbing the spot with cotton balls using a mixture of saturated $KMnO_4$ and 0.2 N $H_2SO_4$ for 2 minutes, water for 1/2 minute, $NaHSO_3$ for one minute, and liquid soap for 1/2 minute. This procedure was continued for a 10 minute period.

Rats were sacrificed at 1, 24, 72 and 168 hours following decontamination, and the amount of plutonium deposited in the various body fractions was determined. Results of this experiment are presented in Table 3. and shown graphically in Figure 1. When these data are compared with results shown in Table 2, it is seen that the total deposition of plutonium in rats which had been decontaminated was much smaller. The only exception is during the first hour of exposure where it was observed that the decontaminated rats deposited slightly more plutonium than the untreated animals

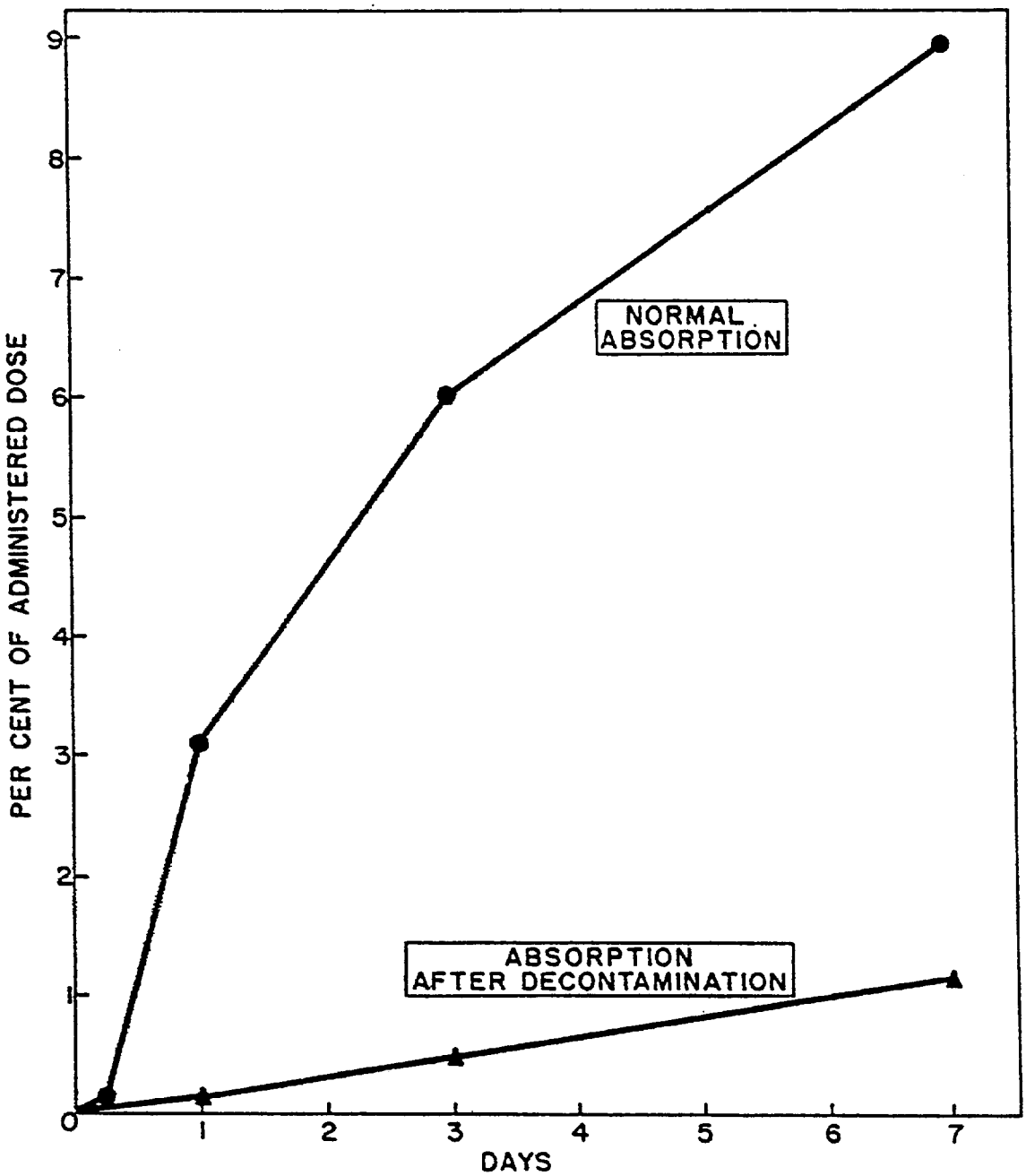


FIGURE 1

Deposition of plutonium in total carcass (carcass plus liver) of rats after dermal exposure to a solution of plutonium nitrate. Normal absorption and absorption after decontamination are shown as per cent of administered dose.



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TABLE 3

Percutaneous absorption of plutonium as a function of time after decontamination. The dorsolumbar section of the skin of rats was exposed to ten microliters of solution containing about one microgram of Pu(IV) in 10 N $HNO_3$ for 15 minutes before decontamination.

| Duration of Exposure (hrs) | Plutonium Recovered (Per Cent of Administered Dose) | | | |
|---|---|---|---|---|
| | Skin | Liver | Carcass | Total |
| 1 | 9.1 | 0.003 | 0.05 | 9.15 |
| 24 | 5.7 | 0.02 | 0.15 | 5.9 |
| 72 | 7.4 | 0.06 | 0.43 | 7.9 |
| 168 | 5.0 | 0.082 | 1.1 | 6.2 |

## DECONTAMINATING PROCEDURES

### Methods and Results

The decontamination studies were designed to evaluate and compare removal of plutonium by irrigating the contaminated area, by swabbing off the contaminant, or by a combination of irrigating and swabbing, using various decontaminating agents. The contaminant was Pu(IV) (500 μg) in 5 microliters of 10 N $HNO_3$ and was applied to a 0.03 $cm^2$ area of the clipped skin of rats. Fifteen minutes after application of plutonium, decontamination was started either by irrigating with three liters of solution for four minutes, or by swabbing with cotton balls for ten minutes, or by an irrigating-swabbing combination lasting fourteen minutes. The irrigating solutions were passed through a tubulated, open ended, cylinder shown in Figure 2, which was held against the skin confining the solution to the contaminated area. The solution was collected after passing over the contaminated area. Following decontamination a one-inch square area of skin containing the exposed area was removed

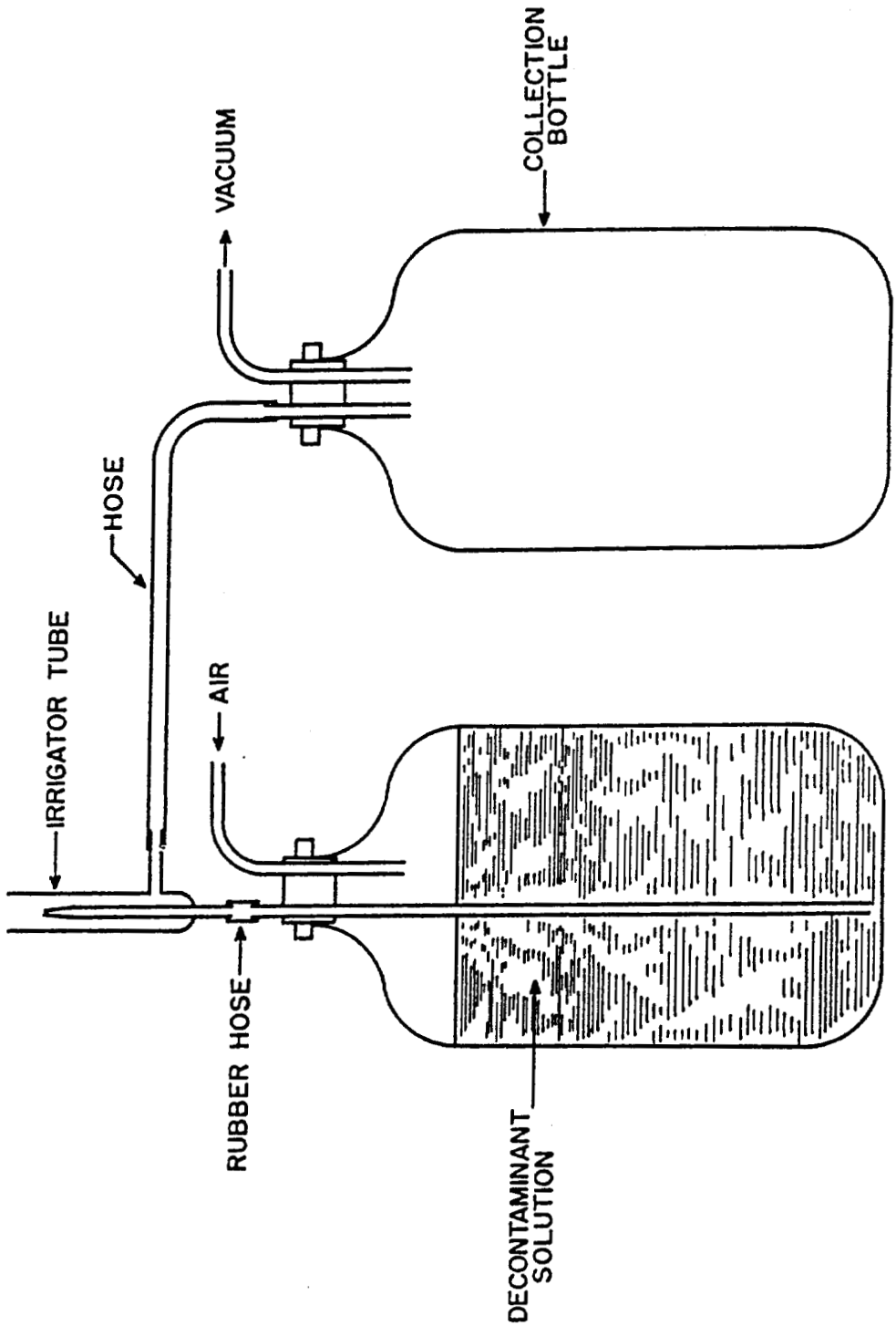


FIGURE 2

Diagram of irrigation apparatus used in washing high level plutonium contamination from the skin of rats.



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and its plutonium content determined. The amount remaining as compared to the amount applied was taken as the criteria for determining the effectiveness of one procedure over another and one agent over another. A description of the decontamination agents employed is given in Table 4. The methods used and results obtained are shown in Table 5.

Additional studies were made, using swabbing procedures only, in which a much smaller concentration of plutonium was applied (10 $\mu g/cm^2$). The exposure time was 15 minutes after which swabbing was carried out for 10 minutes. Efficiency of decontamination was determined as in the previous experiment. Results are shown in Table 6.

## DISCUSSION

The results from the experiments on rate of penetration of plutonium nitrate solutions, as shown in Table 1, seem to indicate a marked dependence of absorption on acid strength, and indicate that absorption may be roughly proportional to area exposed. The small percentage of plutonium that enters the carcasses during the first 15-30 minutes after exposure is evidently limited by the time necessary for the acid vehicle to penetrate the skin barrier and carry significant amounts of plutonium to the underlying capillary system.

The absorption of plutonium through rat skin, over long periods of time, as shown in Table 2, continues to occur throughout the seven day exposure period. The most rapid absorption occurs during the first 24 hours.

It is evident from Figure 1 that decontaminated rats absorbed significantly less plutonium than the undecontaminated rats except for the one-hour period immediately following the application of plutonium. This increased early absorption in the decontaminated rats could be due to the



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TABLE 4

Description of reagents and procedures used for decontamination of plutonium nitrate contaminated rat skin.

| Reagent | Composition and Procedure |
|---|---|
| Cupferron + gluconic acid | 10% solution of cupferron applied alternately with gluconic acid. |
| EDTA | Calcium disodium salt of ethylenediaminetetracetic acid. |
| EDTA + $H_2O_2$ + Tide | 8% solution of EDTA, applied alternately with a 3% solution of $H_2O_2$ and with a 10% solution of Tide. |
| $KMnO_4$ + $H_2SO_4$ | Swab with solution composed of equal volumes of a saturated solution of $KMnO_4$ and 0.2 N $H_2SO_4$ for 2 minutes; rinse with warm water; swab with 5% solution of $NaHSO_3$ for 2 minutes; wash with liquid soap and water; dry[18]. |
| $La(NO_3)_3$ + $H_2O_2$ | 12% solution of $La(NO_3)_3$ applied alternately with a 3% solution of $H_2O_2$. |
| Modified $KMnO_4$ Procedure | Swab with liquid soap for 2 minutes; dry; swab with $KMnO_4$ (4% soln.) for 2 minutes; swab with $NaHSO_3$ (5% soln.) for 1 minute; dry. Continue for 10 minutes. |
| Physo-Hex | Trade name for an antiseptic detergent. |
| Picric acid + $H_2O_2$ | Saturated solution of picric acid applied alternately with a 3% solution of $H_2O_2$. |
| Tannic acid + $H_2O_2$ | 10% solution of tannic acid applied alternately with a 3% solution of $H_2O_2$. |
| Tide + $H_2O_2$ | 10% solution of Tide applied alternately with a 3% solution of $H_2O_2$. |
| Tide, Ponds Cleansing Cream | Trade names for reagents used only as a representative of a class of compounds. |
| Titanium dioxide | Paste prepared by mixing precipitated titanium dioxide with a small amount of lanolin[18]. |
| Radiacwash | Trade name for a solution containing a chelating agent and soap. |

TABLE 5

Comparative efficiencies of decontamination agents and procedures in removing high level contamination from rat skin. A minimum of two rats was used to obtain an average value for the per cent Pu remaining after decontamination (500 μg Pu(IV) in 5 μl of 10 N $HNO_3$ applied to area of 0.03 $cm^2$)

| Reagents Used | Per Cent of Applied Pu Remaining Following Decontamination | | |
|---|---|---|---|
| | Swabbing | Irrigation | Irrigation ($H_2O$) + Swabbing |
| Acetic Acid 10% | 0.8 | 2.4 | 0.78 |
| Acetic Acid 25% | | 3.0 | |
| Citric Acid 4% | | 9.0 | |
| Citric Acid 10% | 0.9 | 3.8 | 2.3 |
| Cupferron + gluconic acid | 11.0 | | 8.9 |
| EDTA 8% | 4.0 | | 2.5 |
| EDTA 10% | | 7.8 | |
| EDTA + $H_2O_2$ + Tide | | | 2.7 |
| HCl 6 N | 0.6 | 4.0 | 0.5 |
| $KMnO_4 + H_2SO_4$ | 0.3 | | 0.65 |
| $La(NO_3)_3 + H_2O_2$ | 4.6 | | |
| Liquid Soap | | | 3.8 |
| Modified $KMnO_4$ Procedure | 3.8 | | 2.3 |
| $NaHCO_3$ 10% | | 3.6 | |
| Picric Acid + $H_2O_2$ | 2.9 | | |
| Ponds Cleansing Cream | 3.7 | | 2.6 |
| Tannic Acid + $H_2O_2$ | 2.4 | | |
| Tide 10% | 3.9 | | 1.9 |
| Tide + $H_2O_2$ | | | 1.9 |
| Titanium dioxide | 3.0 | | 2.5 |
| Water | 1.1 | | 1.25 |

TABLE 6

Comparative efficiencies of swabbing agents in removing low level contamination from rat skin

(10 μg Pu(IV) in 10 μl of 10 N $HNO_3$ applied to area of 1 $cm^2$)

| Reagents Used as Swab Solutions | Per Cent Applied Plutonium Remaining on the Skin (Values are averages from a minimum of of two rats) |
|---|---|
| KOH 40% | 1.0 |
| $KMnO_4 + H_2SO_4$ | 2.7 |
| Radiacwash | 25.0 |
| Water | 28 |
| Tide 10% | 29 |
| EDTA 20% | 32 |
| Acetic Acid 10% | 34 |
| Titanium dioxide paste | 35 |
| Physo-Hex | 37 |
| Citric Acid 10% | 38 |
| HCl 10% | 38 |

stimulation or irritation caused by the decontamination process. It should be noted that the decontamination had apparently no effect on the subsequent absorption of plutonium remaining on the skin.

The decontamination reagents examined covered a wide field and were used to screen many classes of compounds. The data of Table 5 indicate no superiority of irrigation over the more commonly employed swabbing procedure. It is seen that very large "decontamination factors" were obtained. This is probably due to the large concentrations of plutonium employed. Under such conditions much of the contaminant is held



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loosely and the removal of large amounts of it is greatly facilitated. As shown in Table 6, when relatively smaller concentrations of plutonium are employed, the decontaminating efficiency of many methods decreased markedly.

The most effective decontamination agents were keratolytic materials which actually remove some of the layers of skin. Such a mechanism is not an ideal one since injury to the skin may increase the rate of absorption of plutonium not removed. It is interesting to note that water alone was more effective than many of the agents which have been recommended for plutonium skin decontamination.

The conclusions here reported should be considered only tentative, since the carefully controlled experiments investigating all factors involved have not been completed. Such experiments are in progress at the present time.

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