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PHYSICAL AND RADIOCHEMICAL PROPERTIES OF FALLOUT PARTICLES

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ABSTRACT

A survey has been made of the very large body of literature now existing on the characteristics of fallout particles. An attempt is made to summarize in concise form the ranges of physical and radiochemical properties of particles of the debris produced by devices detonated under various conditions. The results of studies of the leaching action of various solvents on fallout particles are also summarized.

SUMMARY

The physical and radiochemical properties of fallout particles from nuclear weapons do not fall into narrowly defined ranges. Detonations at altitudes sufficient to prevent incorporation of soil into the fireball tend to produce small, spherical, highly active particles with the activity distributed throughout. If soil and other on-site material is incorporated into the fireball, one observes increased frequency of particles with lower specific activity, irregular shape, larger size, evidence of partial melting and agglomeration, and activity concentrated on the surface of the particle. The leaching action of various solvents on fallout particles depends upon the nature of the particles themselves as well as that of the solvent.

INTRODUCTION

As the application of nuclear power expands, the nature of radioactive debris becomes a subject of ever increasing interest. In addition to concern with world-wide radioactive fallout from atmospheric testing, consideration must be given now to venting underground explosions and reactor accidents, and perhaps in the not too distant future to contaminating events produced by the peaceful applications of nuclear explosives and to the safety aspects of space vehicles and satellites powered by nuclear reactors and radioisotope sources.

In the twenty years since the testing of nuclear devices began, an enormous amount of attention has been directed toward characterizing fallout particles. These studies have involved many different observers at many different installations, and a large literature on the subject has accumulated. By far the greatest portion of this literature is in the form of classified and unclassified technical reports to the Armed Services of the United States, although important contributions have been made in the open literature. The diffuseness, classification, and general unavailability of this material make it difficult for the non-specialist to form a clear idea of the general characteristics of fallout.

Nuclear debris resulting from various fission processes will naturally have many similar radiochemical properties. Nuclear debris formed at high temperatures in explosions or excursions and thus consisting primarily of oxides of uranium, plutonium, and structural metals will have many physical properties in common. Recognition of this has led to an interest in the properties of fallout particles for the purpose of permitting valid comparisons with radioactive debris from other sources and also, equally important, to avoid the pitfalls of unwarranted comparisons. This report is the result of a survey of the fallout literature and it attempts to summarize in a concise form the ranges of physical and radiochemical properties that have been observed.

LITERATURE SOURCES

In 1960 the Defense Atomic Support Agency initiated a project to collect, critically select, summarize, and publish the then existing test data relevant to the physical and radiochemical properties of fallout. More than two hundred source documents were consulted, as were pertinent documents covering the testing since 1960 and numerous other sources of information. This investigation has provided the background for the present report.

The characteristics of fallout particles depend to a certain extent on such factors as weapon yield, height of burst relative to ground surface, the nature of the materials in the device, the presence or absence of a tower or other on-site materials for testing and instrumentation and how much and what kind of soil is drawn into the fireball. For this reason it is convenient to summarize the physical and radiochemical characteristics of the particles according to the type of burst which produces the fallout. However, there is some unavoidable overlap in the classification by this method. For instance, the particles produced by low air and by tower bursts may resemble either those produced in high air bursts or those from ground-surface bursts. Furthermore, the classifications are not completely unambiguous, since a burst altitude that is high relative to a low-yield device might be low relative to a high-yield device. The interesting concept of a scaled height of burst, which would properly weight the parameters in such a way as to provide an unambiguous classification, does not yet seem to have acquired a formulation which applies to all device weights.

This summary does not attempt to include the properties (other than leaching behavior) of fallout particles resulting from bursts on and under the surface of the ocean. Many tests of this kind have been conducted, but those properties of the particles which have not been reported elsewhere[1,2] remain too poorly defined to merit inclusion here. Similarly, no information is included on the particles formed in completely contained underground bursts.

The observations which form the basis of this report were made by many workers and involved a variety of objectives, methods, and instrumental biases. In particular, the sampling methods used differed greatly and often are described in weapons test reports only loosely or not at all. In fact, it is not always clear whether a description refers to a complete sample or to some selected portion of it. Where obscurities, contradictions, and omissions have been encountered, no attempt has been made to incorporate the data.

PHYSICAL PROPERTIES

Table 1 summarizes size, shape, color, density, location of activity, and ferromagnetic properties of fallout particles for three different types of burst. High air bursts are those which involve no appreciable amount of soil, while ground-surface bursts involve large amounts of soil. Low air and tower bursts form an intermediate classification which sometimes involves soil interaction with the fireball and sometimes does not.

Only rough limits are shown for particle size. There has been much speculation about the distribution of particle sizes. Unfortunately the test data available do not permit any definitive statements. Very little information is available with regard to particles in the submicron size group. Particles in this size range certainly exist in fallout, but apparently account for only a small fraction of total mass and activity in local fallout and for ground surface bursts. In high air bursts, however, they may be the largest contribution to world wide fallout.

TABLE 1. PHYSICAL PROPERTIES

	High Air Burst	Low Air and Tower Bursts	Ground Surface Bursts
Size	Seldom greater than 20 microns diameter. Distribution unknown. Size varies inversely with yield.[a]	Range from a few microns to several hundred or a few thousand microns. Upper limit depends upon nature and amount of interacting soil, etc.	Same as low air and tower bursts.
Shape	Almost all spherical. Occasionally two spheres are stuck together.[a]	Both spheroidal and irregular. Proportions depend on shot conditions. Small spheres stuck to larger particles sometimes reported.	Irregular particles predominate. Some spheroids observed. Various kinds of agglomerates and partially melted particles.
Color	Colorless, gold-yellow, orange, red, brown, green, black.	Colorless, gold-yellow, orange, red, brown, green, black.	Soil color predominates, but other colors also noted.
Density	3 to 4.3 g/cc	1 to 3 g/cc	1 to 3 g/cc
Activity Location	Particles are active throughout.	Some particles active throughout. Some have activity mainly on surface or outer zones.	Activity concentrated on relatively small proportion of particles, mainly those that appear to have been partially melted.
Ferromagnetism	No data.	Particles may or may not be magnetic. Magnetism may be associated with dark color. In some tower bursts most particles may be magnetic.	Small proportion of particles sometimes magnetic. Depends on soil and materials on site of detonation.

[a]Some large non-spherical particles were reported from a sub-megaton air burst 1480 feet above coral reef. Some large particles were also reported from a high yield air burst 4350 feet over water.

Statements about the shape and color of the particles are essentially qualitative and are based on subjective judgments. The small spherical particles, which are characteristic of high air bursts, become scarcer as more soil interacts with the fireball, but they are observed occasionally even in deeply buried cratering shots. The colors observed in fallout particles sometimes have been ascribed to the presence of specific chemical elements. Such assignments are not very strongly supported by chemical analysis. Figures 1 through 4 show some typical shapes observed in surface and tower shots.

The density values given are for single particles; that is, they are true densities rather than bulk or tap densities. The location of radioactivity within the particles has been studied mainly by autoradiographic methods applied to thin sections or mounted particles. Figures 3 and 4 illustrate this technique. The ferromagnetic properties of fallout particles have not been studied much.

RADIOCHEMICAL PROPERTIES

Table 2 summarizes specific activity, decay, fractionation, induced activity, and deposition and partition of fallout particles. These properties are strongly dependent on whether or not soil interacts with the fireball. The first two classifications of bursts in the table are based mainly on this distinction. A third classification, sub-surface cratering bursts, has been added since some of the radiochemical properties of deeply-buried bursts are distinctive.

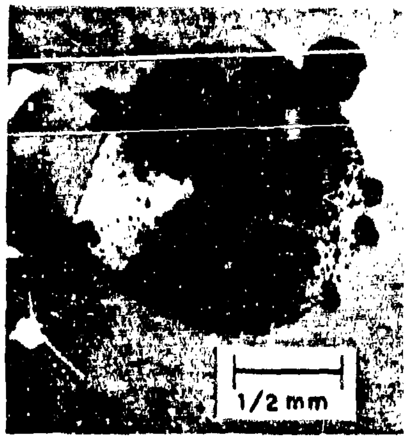

Fig. 1. A typical radioactive fallout particle from a tower shot in Nevada. The particle has a dull, metallic luster and shows numerous adhering small particles.

Fig. 2. An active fallout particle from a tower shot in Nevada. The particle is spherical with a brillant, glossy surface.

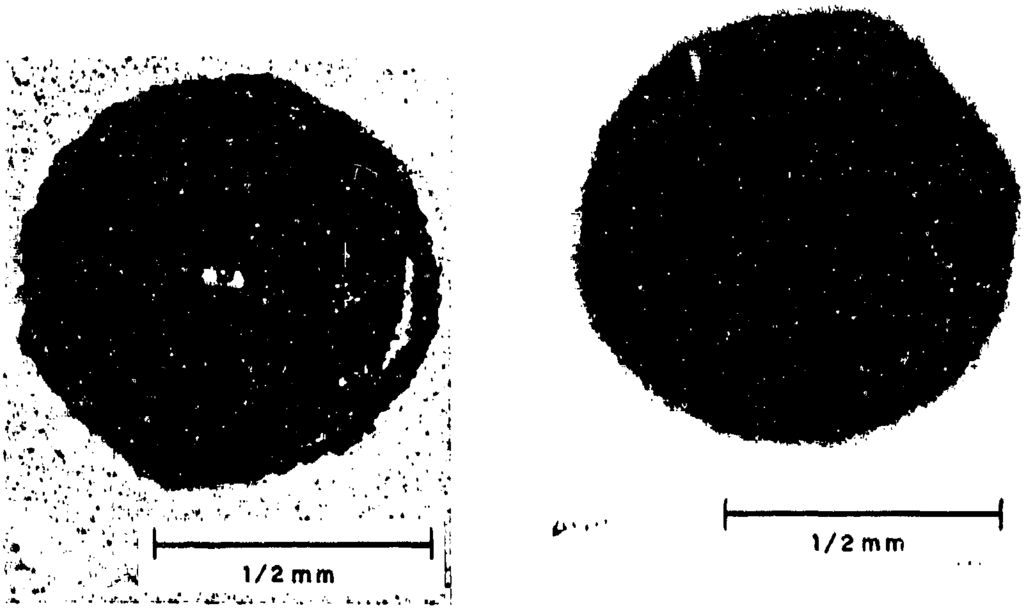

Fig. 3. Photograph (left) and autoradiograph (right) of a thin section of a spherical particle from a ground surface shot at Eniwetok. The radioactivity is uniformly distributed throughout the particle.

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Fig. 4. Photograph (left) and autoradiograph (right) of a thin section of an irregular particle from a ground-surface shot at Bikini. The radioactivity is concentrated on the surface of the particle.

TABLE 2. RADIOCHEMICAL PROPERTIES

	High Air Bursts (No soil enters fireball)	Low Air or Surface Bursts (Soil enters fireball)	Sub-Surface Cratering Bursts
Specific Activity	Volume specific activity (dpm at 25 days per cubic micron) 2 to 750, depending on yield. The number of Zr^{95} equivalent fissions* per gram can be estimated from: $\left(\frac{1.45\times10^{23}\times\text{Yield(kt)}}{\text{Device weight(g)}}\right)$ but this gives high values. Experimental values range as high as 10^{18} to 10^{21} fissions per gram.	Lower than that of high air bursts, depending on amount of tower, soil, etc., interacting in fallout formation. Typical surface shots produce fallout with activity as high as 10^{15} Zr^{95} equivalent fissions per gram.	Shots involving very large amounts of soil may not produce more than 10^{10} equivalent fissions of Zr^{95} per gram.
Decay**	Beta decay exponent ranges from 0.5 to 1.7 with modal value of about 1.1 for large (greater than 1 micron diameter) particles.	Gamma decay exponent approximately 1.2.	Same as low air bursts.
Fractionation	Larger particles (greater than 1 micron diameter) depleted in volatilely behaving mass chains. Submicron particles relatively representative.	Close-in fallout is usually depleted in volatilely behaving mass chains, while world-wide fallout is enriched.	Cratering shots may trap large amounts of refractory mass chains in fallback. In these cases, local fallout is enriched in volatilely behaving mass chains.

Continued

*This is the number of atoms of fissioning material required to produce the Zr^{95} found in the sample. For example, suppose 1000 atoms of Zr^{95} are found in a sample of debris from U^{235} thermal neutron fission. Since in this fission process the fissioning of 100 U^{235} atoms produces 6.26 atoms of Zr^{95}, the sample of debris contained 1000/.0626 or 15,970 equivalent fissions of Zr^{95}.

**The exponent referred to is the quantity x in the equation $A_1/A_2 = (t_2/t_1)^x$ where A_1 is the activity at time t_1.

TABLE 2. RADIOCHEMICAL PROPERTIES (Continued)

	High Air Bursts (No soil enters fireball)	Low Air or Surface Bursts (Soil enters fireball)	Sub-Surface Cratering Bursts
Induced Activity	Numerous induced activities observed. Those of mass less than mass 203 do not appreciably increase the activity at early times. Contribution of U^{237}, U^{239}, U^{240} and Np^{239} are variable and sometimes appreciable.	Remarks on high air bursts apply, except that at early times activity from Mn^{56} (particularly in steel tower bursts) and Na^{24} may be appreciable.	Remarks on high air bursts apply, except that at early times activity from Na^{24} may become appreciable.
Deposition and Partition	Virtually all debris goes into world-wide fallout. If height of burst is low enough, ground-radiation patterns result from induced activity. The radiation contours of these patterns are circular and can reach 1-hr dose rates of 25 r/hr at the center when ground material is not drawn into the fire-ball. Relatively little ground material raises this dose rate to 1000 r/hr by scavenging radioactive material from the cloud.	Close-in radiation fields of high intensity, extending downwind in cigar-shaped pattern. Deposits as high as a few grams of fallout per square foot for typical surface shots. Partition between close-in and world-wide fallout variously estimated at 25%-to-75% close-in; depends on device yield, soil conditions, etc.	Cigar-shaped fall-out fields of high intensity, like surface bursts except mass deposited may be much greater. Large part of activity may be brought down in fall-back and local fall-out. This varies with nuclide, due to fractionation. In one event 12% of Sr^{89} and 0.2% of Zr^{95} reported to have escaped into world-wide fallout.

In the literature many different units have been used for reporting specific activity. One of the most useful measures of activity for fission products is equivalent fissions based on Zr^{95}. This is the number of atoms of the fissioning material required to produce the Zr^{95} found in the sample.

The decay of fission product activity with time has been widely studied.[3,4,5] The early expectation that this would prove to be a valuable method of characterizing fallout samples has met with some disappointment. It is true that the decay curves exhibit considerable variation in overall slope and slight differences in shape, yet it is impossible to draw any reliable conclusions about the radiochemical composition of a sample from the study of its decay curve alone. It is not even possible to explain differences in two or more samples from the same event by comparing their decay curves. The decay measurement is essentially too insensitive to shed much light on the very complicated compositional variables that are of interest. For purposes of predicting dose rates and making shielding calculations, the early prediction of Way and Wigner[3] that gross fission product activity should vary roughly as the -1.2 power of time remains the most useful general rule.

The phenomenon known as fractionation of the fission-product radionuclides is commonly observed in fallout and has been widely discussed.[6,7,8,9,10,11] Fission processes are known to produce the fission products in fixed proportions, yet the proportions observed in fallout samples are often very far from the expected values and may even differ greatly from sample to sample. It is generally believed that this fractionation of the radionuclides is caused by differences in the condensability of the elements composing the mass chains during the very rapid quenching of the fireball. Thus, mass chains which contain Br, I, Kr, and Xe at early times (volatile chains, such as mass-89 and mass-137) may condense at a different time and in a different way from chains which do not contain these elements (refractory chains such as mass-95 and mass-144). A method for the systematic analysis of radiochemical data on fractionated nuclear debris has been developed by Freiling.[9]

Both fission and fusion processes result in the production of activity other than fission-product activity. These activities, called induced activities, arise from the interaction of neutrons with the atmosphere, the unburned core material, the device casing and shielding, and the tower materials and other on-site materials including the soil. The range of nuclides observed in fallout which are attributed to neutron activation is large and includes, for example: Be^{7}, Na^{24}, Al^{28}, Mn^{54}, Mn^{56}, Fe^{55}, Fe^{59}, Co^{60}, Cd^{115}, W^{185}, W^{187}, U^{237}, U^{239}, U^{240} and Np^{239}. The number of neutrons from a fission event that are available for inducing activity is of the same order of magnitude as the number of fissions, and this number must be distributed among all induced activities. It is therefore unlikely that more than one or two of the induced activities will be present in fission-event fallout in sufficient quantity to account for an appreciable fraction of the total activity at reasonably early times after detonation. Since most fission-product activities die out within a few years, the contribution of persistent induced activities, such as Co^{60}, may become relatively important in old fallout.

LEACHING PROPERTIES

The effects of leaching nuclear debris are of particular interest because of their relation to the biological availability of the radioactive species, and considerable effort has been devoted to documenting these effects. Of the numerous solvents studied, by far the most extensively investigated are distilled water (including rainwater), seawater, and 0.1*N* HCl. Distilled water and rainwater are of interest because of their relation to weathering and soil transport. Seawater is of importance in the uptake of radionuclides in the marine biosphere. The 0.1*N* HCl is generally considered to be a simulant for stomach fluids.

The leaching properties of nuclear debris have usually been reported in terms of the fraction of gross beta or gross gamma activity found in a solid, soluble or colloidal state. In these terms the behavior is very complex. Besides depending upon the matrix material of the fallout, its particle size and type, its previous history, the duration of leaching action and the solvent employed, it also depends upon the time after detonation at which leaching begins. Results in terms of individual nuclides are much more significant, but these are relatively scarce. The most desirable data of all, the rates of dissolution of individual nuclides, are virtually non-existent, doubtless because of the effort required. Most workers report some "equilibrium value" of the solubility, which usually corresponds to a levelling off of the curve for leached activity vs. time.

Results in terms of gross beta activity suffer the additional disadvantage of requiring great care for reliable determination and it is seldom clear that this care has been taken. It does appear to have been established, however, that gross beta activity is more easily leached in going from tower-shot debris to silicate-burst debris to air-burst debris. As expected, 0.1N HCl is more effective than water: Water dissolves at most a few percent of the gross beta activity from tower debris; 0.1N HCl dissolves most of the gross beta activity in air-burst debris. Some authors report that the solubility increases as the size decreases. The debris from coral-surface bursts is reported to be highly soluble in water, not only the gross beta activity but the particles themselves. Fresh coral-burst debris is largely CaO and $Ca(OH)_2$ and hence raises the pH considerably when it dissolves (to 10-11).

Measurements of gross gamma activity in samples of debris from coral surface shots show that at 4-8 days only about 8 % of the activity dissolves, but at 20 days some 23% dissolves. These results are considerably lower than those reported for gross beta activity. For surface-water barge shots, at 2-3 days, 60-70% dissolves. In both the island and barge cases the colloidal fraction was relatively negligible.

Radiochemical measurements of the leached material from silicate-surface and subsurface bursts indicate that iodine, cesium, strontium and barium radionuclides are among the principal contributors to the leached activity. In the case of silicate-surface bursts, the increase in solubility with decreasing particle size has been clearly established. Unexpectedly, the superior leaching power of 0.1N HCl over water disappears for particles less than 44 microns in diameter.

Of particular interest is a study on weathering effects which concludes that the effect of rainwater in leaching silicate particles is minor. The physical movement of the soil, caused by rain, is more effective in transporting the activity.

The physical state distributions of individual radionuclides for water surface and underwater bursts have recently been disucssed at length elsewhere.[(2)]

DISCUSSION

The physical and radiochemical properties of fallout particles from nuclear weapons do not fall into narrowly defined ranges. Depending on the characteristics of the device and the shot conditions, particle sizes may range from sub-micron to centimeter dimensions. Almost all colors common to minerals may be observed at least occasionally; specific activity may vary over many orders of magnitude; active particles may have either irregular or spheroidal shapes and be either magnetic or non-magnetic. Activity may be concentrated on the particle surface or uniformly distributed throughout the particle and all degrees of fractionation may be observed.

If the components of the device, the height or depth of the detonation and the nature of the soil and other on-site materials are specified, the range of properties to be expected in the fallout particles can usually be narrowed considerably. Detonations at altitudes sufficient to prevent incorporation of soil into the fireball tend to produce small, spherical, highly active particles with the activity distributed throughout. If soil and other on-site material is incorporated into the fireball, one observes increased frequency of particles with lower specific activity, irregular shape, larger size, evidence of partial melting and agglomeration, and activity concentrated on the surface of the particle.

The leaching action of various solvents on fallout particles is quite complex and depends rather strongly upon the nature of the particles themselves as well as that of the solvent and the experimental conditions.

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