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Recommendations for Plutonium Colloid Size Determination

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RECOMMENDATIONS FOR PLUTONIUM COLLOID SIZE DETERMINATION

by

S. T. Kosiewicz

ABSTRACT

This report presents recommendations for plutonium colloid size determination and summarizes a literature review, discussions with other researchers, and comments from equipment manufacturers. Four techniques suitable for plutonium colloid size characterization are filtration and ultrafiltration, gel permeation chromatography, diffusion methods, and high-pressure liquid chromatography (conditionally). Our findings include the following:

- Filtration and ultrafiltration should be the first methods used for plutonium colloid size determination because they can provide the most rapid results with the least complicated experimental arrangement.
- After expertise has been obtained with filtering, gel permeation chromatography should be incorporated into the colloid size determination program.
- Diffusion methods can be used next.
- High-pressure liquid chromatography will be suitable after appropriate columns are available.

A plutonium colloid size characterization program with filtration/ultrafiltration and gel permeation chromatography has been initiated.

I. INTRODUCTION

Since 1969, Group CHM-1 at the Los Alamos National Laboratory has been involved in environmental studies of the fuel pellets from radioisotope thermoelectric generators (RTGs) used as power sources for space satellites. The radioisotope used in the RTGs, ^{238}Pu, is present as the dioxide PuO_2. Earlier studies focused on how different terrestrial and aquatic environments affected release rates of plutonium from the RTGs. During this time, gross radioactive counting was used for determining the amount of plutonium released by the fuel pellets from the RTGs. No attempt was made to identify the chemical form of the plutonium ions, complexes, or colloids.

As this experimental program developed, its objectives evolved. Current emphasis is on obtaining more fundamental and less empirical data.[1] With this change in emphasis, the need for plutonium colloid size characterization was identified. One would expect that with the capability to measure the plutonium species in a system, coupled with thorough knowledge of both plutonium chemistry and chemistry of the environmental system, a predictive capability could be developed. With this capability, predictions of the plutonium distribution in other environmental systems would be possible if their chemical parameters are understood. Knowledge of the distribution of plutonium species could lead to understanding the plutonium transport in terrestrial and aquatic environments as well as in plants. Ultimately, this

knowledge should provide better data for risk assessment analyses.

The objective of this effort is to identify and recommend methods for determining the size distribution of plutonium. Since plutonium probably will be present as a colloid under most environmental conditions, size distribution of plutonium colloid or polymer was suggested as the first phase of characterization. Later work can be directed to determining the complexes that are present and the plutonium oxidation states.

This work, which was done during the last quarter of FY1983, consisted of a literature review, discussions with researchers, [including a trip to Pacific Northwest Laboratories (PNL)], discussions with chemical and equipment manufacturers, and equipment acquisition.

II. LITERATURE REVIEW

A computer literature search for articles on plutonium colloid size determination was accomplished by R. Heaton. From an initial survey, it rapidly became apparent that a concentrated effort of high-quality work had been done by Tom Garland and coworkers at PNL in Richland, Washington. Their work from the early seventies to the present has evolved from characterization studies to predictive modeling (for some elements). A central theme of these articles is plant uptake of plutonium and attempts to identify those variables affecting the uptake. Techniques used in their research for plutonium colloid size determination included filtration and ultrafiltration,[2-8] gel permeation chromatography,[6] diffusion,[3,9] thin layer chromatography, and thin layer electrophoresis.[6]

Several other researchers have done work that provides useful information for plutonium colloid size determination: Haire and Lloyd at Oak Ridge National Laboratory,[10-11] Alberts el al.[12] and Lovett and Nelson[13] at Argonne National Laboratory, and also Kepak in Hungary.[14-16]

Haire and Lloyd[10-11] studied physical characteristics of Pu(IV) polymer that was formed by solution precipitation. Following are several important points from their papers for tetravalent plutonium hydroxides and colloids.

1. Primary particles of hydrated PuO_2 are 5 to 20 Å in diameter, with most about 10 Å. These primary particles do not increase in size on aging. Alberts el al.[12] also report that most plutonium particles in Lake Michigan have diameters less than 30 Å (and they are anionic).

2. Upon aging, the primary polymers change from amorphous to crystalline (based on x-ray diffraction data) with subsequently slower depolymerization rates.

3. Reversible secondary aggregation can occur. This explains the wide range of molecular weights reported for plutonium polymer. Some of the aggregates can be *very* stable.

4. Haire and Lloyd do not believe that Pu(IV) precipitates in solution to form a hydroxide. Rather, they suggest hydrated PuO_2 forms.

This work is of considerable interest because it provides size information on plutonium particles that form in solution. There seems to be a tendency for most particles to be about 10 Å in diameter.

Kepak[14-16] published papers and a review on properties of radiocolloids that include Pu(IV). He cites one study in which the molecular weight of colloidal plutonium was determined to be 4×10^3 Daltons by its self-diffusion coefficient. This is consistent with data reported by Garland et al. at PNL. Kepak discussed a technique for molecular weight determination based on self-diffusion in a capillary tube (see Sec. IV.D). He stresses the fact that transport of a trace radiocolloid may be controlled by some other colloid (e.g., iron or manganese). Hence, if care is not exercised, erroneous diffusion coefficients may be obtained. This is an important point since plutonium transport in environmental systems may be enhanced or retarded by other elements.

Lovett and Nelson[13] determined the oxidation states of plutonium in aquatic environments. Our review focused primarily on particle size determination rather than oxidation state determination since that seemed to be a reasonable initial plutonium characterization step in work to be performed in CHM-1. However, oxidation state characterization should be an area of study in our future program; an exhaustive literature study should be pursued then.

III. COMMENTS ON PLUTONIUM CHEMISTRY

Plutonium can be present in the III, IV, V, VI, and VII (rare) oxidation states,[17] with the first four valences being the most common. However, disproportionation of the IV and V valence states can occur. Consequently, even though all the plutonium in a particular system initially may be in the IV or V valence, through disproportionation other valences may be present. Tendencies for

hydrolysis are $Pu^{+4} > PuO_2^{+2} > Pu^{+3} > PuO_2^{+}$. Some very stable complexes are formed by Pu(IV). The solubility product of $Pu(OH)_4$ is estimated to be about 10^{-56} (Ref. 17).

Researchers at PNL have demonstrated that even though the hydroxide is highly insoluble, plutonium is taken up by plants. Cataldo* discounts the possibility that PuO_2 particles, no matter how small, can pass through a plant's membrane. Therefore, the plutonium must enter the membrane attached to a ligand. Once inside, he hypothesizes, it exchanges and complexes with other ligands that are, as yet, unidentified.

Our project is concerned with plutonium whose origin is $^{238}PuO_2$ from RTGs or radioisotope heating units (RHUs). Garland** suggests that plutonium that originates from PuO_2 will become soluble by first passing through the +5 valence state. From the preceding paragraph, it is apparent that plutonium must go through a soluble form before it can enter the food chain through plants. Two ways that people can take up PuO_2 are through inhalation and ingestion. In the case of ingestion, it most likely would pass directly through the digestive system since the acidity of this system (~1.5) is insufficient to retard hydrolysis of Pu(IV);[7] tetravalent plutonium can be in the colloidal state in the pH range 1.4 to 12.0. Plutonium that is taken up by plants is complexed by some ligand in them and in this form could be more readily absorbed if it is ingested.

Consequently, the importance of determining the species distribution of plutonium (ions, complexes, polymers, particle sizes) is apparent. The species distribution will be highly dependent on the chemical system measured and will provide predictive capability only for systems that are similar chemically. However, once the chemistry of a new system is thoroughly understood, predictions concerning plutonium forms in that system may be possible.

IV. TECHNIQUES FOR PLUTONIUM COLLOID SIZE DETERMINATION

Several techniques appear suitable for plutonium colloid size determination. In this section they will be discussed along with some specific product information and product names. This information should enable a program for characterizing plutonium colloid size to be established. However, several factors may complicate trying to extrapolate data from one chemical system to another. First, ^{238}Pu particles may be in a process of size reduction because of alpha recoil effects. Second, aggregation of primary particles can occur[11] with subsequent apparent increase in particle sizes. Third, the effects of different particle sizes may be mitigated by factors other than the plutonium polymer size (e.g., other cation polymers, adsorption, etc.). However, colloid size determination still seems to be a reasonable first step in plutonium characterization.

Four techniques that seem suitable for plutonium polymer size determination are filtration (including ultrafiltration), gel permeation chromatography, diffusion techniques, and possibly at some future time, high-pressure liquid chromatography. The following section will discuss them separately.

A. Filtration and Ultrafiltration

A basic, accurate way of determining colloid size distribution is through filtering the solution of interest. There is a high probability that sample integrity will be maintained, i.e., the plutonium polymer size distribution will not be altered during the filtration process. However, certain experimental details must be considered to ensure that the size distribution data produced are not erroneous. One potential problem is polarization, wherein larger species that cannot pass through the pores of a filter are retained to form a mat that then retains smaller species. Polarization can be diminished by stirring the solution just above the filter to resuspend the materials that did not pass through the filter pores. Amicon Corporation (Danvers, Massachusetts) and Nuclepore Corp. (Pleasanton, California) sell filtration cells that incorporate a magnetic stirrer suspended over the filtration membrane.

Adsorption onto the filter may be an additional problem. Using inert, nonionic filter materials would decrease adsorption. As a check on adsorption, a filter with large pores that should not retain any of the species could be used.

Finally, filters can reject particles of a particular size while allowing smaller ones to pass through the pores. A measure of this capability is the rejection coefficient, which ranges from 100% for material that is completely rejected by the filter to 0% for material that is freely permeable through the membrane. Although a given filter

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*This information was supplied by D. A. Cataldo, PNL, September 1983.

**This information was supplied by T. R. Garland, PNL, September 1983.

may have a specified pore size or molecular weight cutoff, species larger than the nominal pore size may pass through it. Consequently, in a sequential filtering scheme there may be some larger species retained on smaller filters that should have been retained on the previous filter with larger pores. Different manufacturers use different criteria in specifying the nominal size of the pores of their filters. Amicon defines its cutoff as the approximate molecular weight (of a globular solute) at which the membrane has 90% rejection. Nuclepore states that its pore size distribution is +0 to −20% of nominal; the actual size of a given batch of filtration membranes is available upon request.

Several companies manufacture filters and ultrafilters suitable for plutonium colloid size determination, including Nuclepore, Ultrapore, Millipore, and Amicon. Nuclepore Corp. produces filters that appear to have desirable properties for plutonium colloid size characterization. The pore geometry of their filters is uniform in pore diameter as well as in dispersion on the filter surface. About 90% of the membrane surface is available for particle collection since only 10% of it consists of pores. Because these filters are thin, tortuosity of a given pore is decreased and flow rate is increased. Nuclepore manufactures filters with pore sizes ranging from 12 to 0.015 μm. Previously, they marketed filters with smaller pore sizes, but that part of Nuclepore was sold to Ultrapore Corp. (Long Beach, California). Sample test kits of Nuclepore filters with pore sizes as small as 0.1 μm were obtained for evaluation. Some filter holders, as well as a holder for sequential size filtering, also were obtained.

Ultrapore Corp. filters, whose physical characteristics are unlike those of the Nuclepore filters, more closely resemble Millipore (Bedford, Massachusetts) filters. They have a mat-like structure with a high degree of tortuosity, so they do not have as sharp a cutoff as the Nuclepore filters. This was verified by Davis et al., who compared the two types of filters.[18] From 5.0 to 0.1 μm, the Nuclepore filters had superior performance when 0.005- and 0.05-μm ^{198}Au colloids were filtered. Davis et al. performed the comparative study to enable them to decide which filter would be more suitable for determining the size distribution of ^{99m}Tc-S colloids. However, Ultrapore produces filters with smaller pore sizes than those of Nuclepore. The filtration size of smaller filters is usually expressed as the molecular weight of a spherical (globular), uncharged molecule (e.g., proteins) that will pass through it (see earlier definition of rejection coefficient). Ultrapore produces membranes with molecular weight cutoffs ranging from 1 000 000 to 500. Nuclepore Corp.* reports that there is an approximately linear relationship between pore size and molecular weight in the 0.01- to 0.1-μm range. A 0.015-μm pore size corresponds roughly to 150 000 MW. Ultrapore filters with molecular weight cutoffs ranging from 100 000 to 500 were obtained for evaluation. In addition, some filters were received from Millipore; their size range approximates that of the Nuclepore filters.

Amicon Corp. also markets ultrafiltration membranes that have a thin (0.1- to 1.5-μm) "skin" of fine, controlled pore structure on a thicker (50- to 250-μm) open-celled, spongy layer. The thin skin does the actual filtration while the substrate provides support. Filters are available from Amicon with nominal molecular weight cutoffs ranging from 300 000 to 500. At 500, the water flow rate is only about 0.02 mℓ/cm^2/min with 55 psi applied pressure. Consequently, flow rate is a consideration when choosing sample volumes to be filtered with low molecular weight cutoffs (Amicon filters or otherwise). Amicon filters previously had a limited shelf life (about one year), but apparently that is no longer true.

Amicon also makes cone-shaped membranes that fit into centrifuge cones and use centrifugal force to force the solvent through. These may not be of much use to the present program because they are available only with molecular weight cutoffs of 25 000 or 50 000. However, their ultrafiltration membranes should be evaluated with plutonium-laden waters.

A short, but intense, effort was expended to seek radiocolloids with known particle size distribution to use for independent checks on the performance of the various filters.[19] Apparently, ^{198}Au colloid is no longer marketed for biological use because it delivered too high a radiation dose in humans.** This effort was terminated after a few days because of the limited time available for this study.

B. Gel Permeation Chromatography

Gel permeation chromatography can be used for size determination of plutonium colloid.[6] In this technique, porous beads with controlled pore sizes are used for size separations. By using smaller or larger pore sizes, different size molecules can be separated. If the pore is the

*This information was supplied by D. Peterson, Nuclepore Corp., August 1983.

**This information was supplied by E. Lieberman, Cadema Medical Products, Inc., Middletown, New York, August 1983.

correct size, a molecule can enter into it; those that are too large to enter the pores are excluded and elute off the column first. The larger molecules are followed by the smaller molecules because the smaller ones have a longer path to follow to get through the chromatographic column.

Column materials from Bio-Rad Laboratories (Richmond, California) and Pharmacia Fine Chemicals (Piscatoway, New Jersey) appear suitable for this program. Two of the porous polyacrylamide gel beads that Bio-Rad manufactures could be useful for determining the size distribution of plutonium colloid—BioGel P-2 and P-10. Bio-Gel P-2 has a fractionation range of 1800 to 100 Daltons for globular molecules. Bio-Gel P-10 has a fractionation range of 20 000 to 1500 Daltons. These two column materials should provide the capability to separate plutonium colloid in the size range of interest. Initial experiments will rapidly show if plutonium colloid binds to the Bio-Gel material.

Pharmacia markets Sephadex GPC gels. They are cross-linked dextran gels that swell in water and aqueous salt solutions. Again, it appears that two gels can be used to separate plutonium colloid in the size range anticipated at this time—Sephadex G-10 and G-50. The Sephadex G-10 gel has a fractionation range of 1500 to 700, and Sephadex G-50 has a fractionation range of 30 000 to 1500. Both gels and also columns have been obtained from Pharmacia. Based on recommendations from Tom Garland, ISCO fraction collectors (Foxy Model) and high-pressure metering pumps (Model A30) from Eldex Laboratories, Inc., will be incorporated into the experimental apparatus.

Column materials from both companies will be evaluated. It is expected that only one column material will be used after comparative experiments are completed. Robinson et al.[6] used the Sephadex gels in their work. The anticipated molecular weight range that will be observed for plutonium colloids will be from about 700 to possibly 20 000 equivalent molecular weight (if the colloids were proteins). However, since plutonium is heavier than carbon. the actual molecular weight observed will be considerably higher than the apparent equivalent molecular weight based on size. Garland[3] suggests that plutonium species with a molecular weight of 10 000 to 25 000 would be roughly equivalent in size to globular protein with a molecular weight of 500. Lindenbaum and Westfall[20] state that a globular protein with a molecular weight of 11 700 would have the same size as a $Pu(OH)_4$ particle with a molecular weight of ~200 000.

C. High-Pressure Liquid Chromatography

A preliminary survey was done to determine if high-pressure liquid chromatography could be successfully used for plutonium colloid size characterization. From manufacturers' literature it appeared that a TSK column supplied by Alltech Associates (Deerfield, Illinois) held promise for separating plutonium colloids in the molecular weight range of interest. However, Cataldo* found that the column material had too many active sites so the plutonium did not elute from it. He suggests that Brownlee Labs, Inc., soon may be marketing a column that may be suitable for size characterization of plutonium colloid.

Additional effort is not warranted at this time because column materials are not available. At a future time when high-pressure liquid chromatography is considered, detection of the plutonium species probably would need to involve scintillation counting. Either fraction collection or some flow-through scheme on a scintillation counter may be suitable. Refractive index or uv detectors may not possess adequate sensitivity for the low concentration of plutonium species that would be in solution.

D. Diffusion Methods

Kepak[14-15] and Garland et al.[3] used different diffusion methods to obtain estimates of molecular weights of radionuclides. These diffusion methods are attractive because of the high probability of maintaining sample integrity, i.e., not altering the species of interest during the analysis procedure. The basis of separation in the diffusion methods is that larger colloid particles diffuse more slowly than do smaller ones. Kepak's method used diffusion of radionuclides into a capillary tube. A capillary tube containing an inactive solution was vertically immersed into a solution that had an identical chemical composition and pH and also the radionuclide of interest.

Garland used agar gel in Petri dishes for his diffusion studies for molecular weight determinations of plutonium species. In his experiments, solutions that passed through a 0.01-μm filter were placed in a well in the agar gel at the center of the Petri dish. Samples were taken at various distances and times to determine the diffusion coefficients of the mobile plutonium species. By comparing this

*This information was supplied by D. A. Cataldo, PNL, September 1983.

diffusion coefficient with that of a plutonium species with a known molecular weight, estimates were made of the molecular weights of the mobile plutonium species. Garland also determined that there was negligible interaction between the mobile plutonium species and the gel matrix. He found that the most mobile plutonium species after interaction with soils had a molecular weight of 600 000 to 900 000 (roughly the same size as a protein with a molecular weight of ~10 000). Lindenbaum and Westfall[20] report variations of plutonium colloid size from 4×10^5 (by diffusion) to 2×10^7 (high-speed centrifugation) to 10^{10} (track counting in a nuclear emulsion). They hypothesize that these differences in molecular weight may reflect differences in colloid preparation. However, there is approximate agreement for the molecular weights obtained by diffusion by them and Garland.

V. SUGGESTED EXPERIMENTAL PRIORITIES

The first priority in establishing an experimental program for characterizing ^{238}Pu species will be to identify suitable facilities for work with the radioactive solutions that will be analyzed. The work place requirements will depend upon the level of radioactivity in the solutions. If very low levels of radioactivity will be measured, care must be exercised that the solutions do not get contaminated by the selected work station. With high levels of radioactivity, care must be exercised to ensure adequate personnel protection from a health standpoint. It may be necessary to use more than one work station if the samples to be analyzed have extremely different levels of activity.

Filtration and ultrafiltration should be the first methods used for plutonium colloid size characterization. Because they are the simplest of the methods that appear suitable for size determination of plutonium colloid, they will produce usable data in the shortest time. They can be used to establish a base for other characterization methods and could "standardize" the colloid distribution of various solutions. The most important sizes for filters probably will be those that have an equivalent molecular weight cutoff of 500 to 10 000. Garland and others have a working definition that any plutonium that passes through a 0.01-μm filter is considered "soluble."

Some of the initial experiments on size distribution can be accomplished with stacked filter holders that are "ganged" together in order of decreasing pore size. There will have to be a consideration for the pressure differential required to achieve reasonable solution flow when low molecular weight cutoff filters are stacked together. Results from these preliminary experiments could be affected by polarization as discussed previously. More definitive, but laborious, experiments can be performed with stirred cells to decrease polarization. Appropriate filters, ultrafilters, stacking accessories, and an initial stirred cell are available and will be evaluated. Comparisons should be made between equipment and materials, and the design of an experimental procedure should be accomplished.

After the size distribution of plutonium colloid in a solution can be reproducibly measured with filters, work should proceed with gel permeation chromatography. Perhaps the initial work should be done with the Sephadex gels since Garland has done his work with them and some cross checks might be available. Additionally, he may be able to provide advice on the system. There also will be cross checks available from the data produced by the filtration/ultrafiltration experiments. The experiments with gel permeation chromatography will take more time to set up because it is a more complicated experimental system. It involves properly making the columns, calibrating the columns, matching the columns to the proper solvent and flow, setting up the fraction collector, and ultimately, detection (probably by scintillation counting).

The diffusion experiments can provide good estimates of molecular weight data for ions and complexes as well as for the plutonium colloid. To obtain these estimates, diffusion coefficients of known and unknown species must be compared. Garland[3] used an organic plutonium complex for this purpose. It is not apparent that either the Petri dish method or capillary-tube method has an advantage.

Forecasts for further work could include high-pressure liquid chromatography (if suitable columns are developed), thin layer chromatography, and thin layer electrophoresis. For future characterization, oxidation state determinations should precede identification of unknown complexes.

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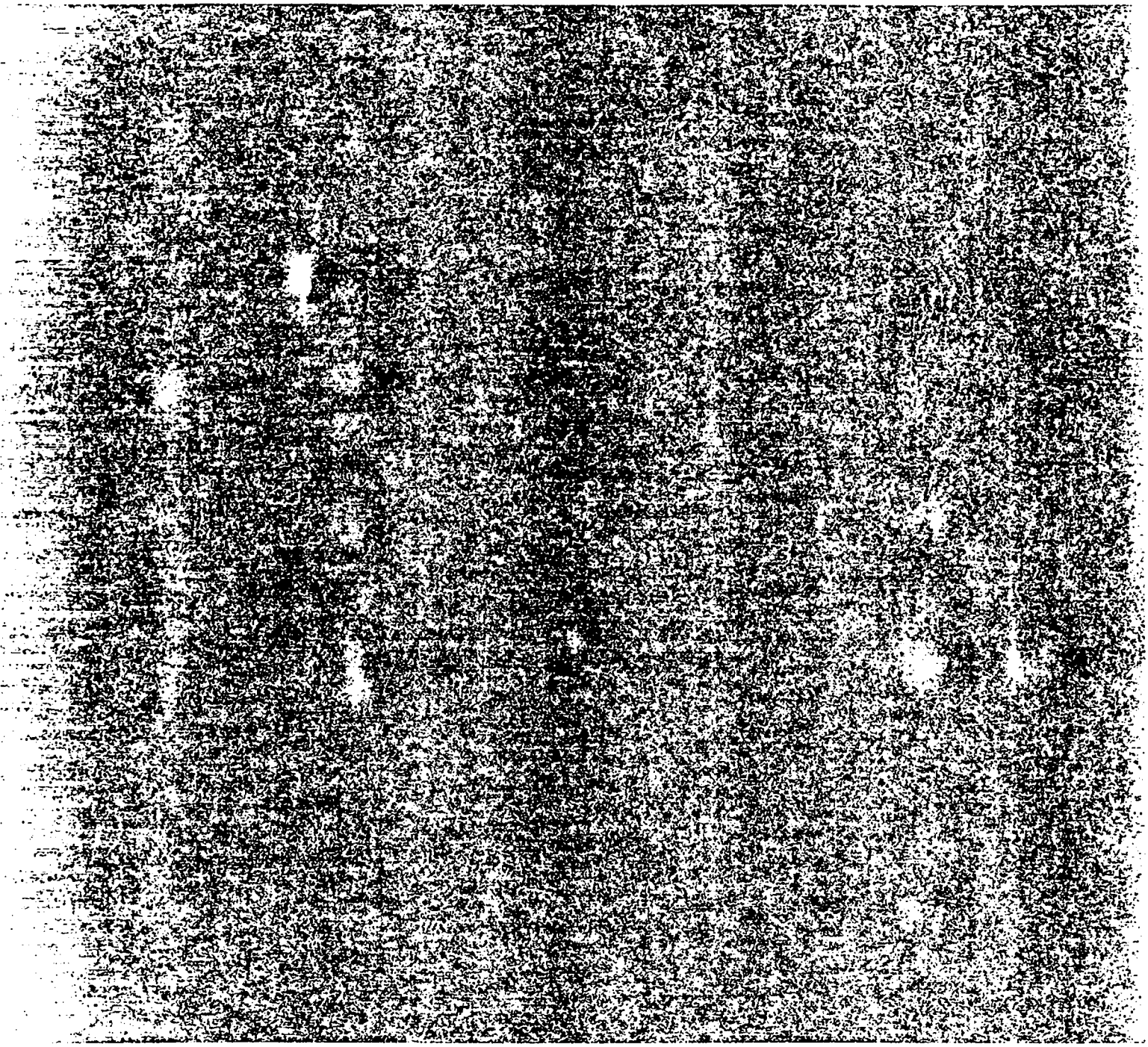

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