

EVALUATION OF THE KAPL SEPARATIONS PROCESS STACK EFFLUENT

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ABSTRACT

The KAPL Separations Process stack effluent is evaluated. The adequacy and the efficiency of all the sampling instruments are determined. The size of the particles entrained in the stack are studied under both the light and the electron microscope. The mean particle size is less than 0.05 microns. Autoradiographs of the particulate material indicate that the majority of the activity is deposited on these sub-micron particles.

Chemical separations of the material deposited on the Hollingsworth and Vose, H-70 filter papers and the caustic scrubber are made. The rare earths comprise the largest portion of the particulate activity while Ru-106 is given off in relatively large quantities during the Head End Operation.

The relative percentages of the activities given off during the most important phases of the Separations Process are tabulated. The KAPL stack effluent is then evaluated on the basis of the MPC recommended in the Bureau of Standards Handbook 52.

EVALUATION OF THE KAPL SEPARATIONS PROCESS STACK EFFLUENT

An evaluation of the KAPL stack effluent from the operation of the separations process was conducted; to determine the environmental and biological effects of the Pilot Plant operations, to establish maximum permissible limits for discharge of the effluent to the atmosphere, and to determine whether more stringent control of the discharge of activity would be required at higher (gm Pu/ton U) operating levels.

The air monitoring and air cleaning system for the separations process operations is schematically illustrated in Figure KH-9A2403. This stack is approximately 100 feet high and 3 feet in diameter. The gaseous and particulate material emanating from the separations process is passed through a caustic scrubber which takes out some of the volatile components while the CWS-6 filters are over 99 per cent efficient in the collection of most particles. The stack effluent is sampled at the top of the stack after it has been diluted by a factor of approximately 10^3 by the room air. At distances from the stack, constant air monitors are located in selected sites to check the radioactive concentrations at various points near ground level. Vegetation samples are collected and analyzed on a regular schedule to evaluate the accumulation of radioactivity on the vegetation.

The evaluation of the stack effluent required the knowledge of; the total activity discharged, the particle size distribution of the activity discharged from the stack, the isotopic composition of this activity, and the dispersal of the radioactive material from the stack. Each of these requisites will be discussed briefly.

TOTAL ACTIVITY

The determination of the total activity discharged from the stack involved the investigation of; the adequacy of the sampling units, the efficiency of each of the sampling units, representative sampling, and the absorption of alpha and beta activity in the filter media.

As illustrated in Figure KH-9A2403, the sampling system consists of a filter unit to collect efficiently the entrained particulate material followed by a caustic scrubber to collect such radioactive components as ruthenium and iodine which may be readily volatilized. This sampling system was considered adequate since it collects or detects a portion of the radioactivity discharged from the stack with a known efficiency. The efficiencies of the Hollingsworth and Vose, H-70 filter paper were determined for a particle density of 2.7 gm/cm^3 over a wide range of particle sizes and linear face velocities. At an operating face velocity of approximately 5 cm/sec the H-70 filter paper was 97.7 per cent for 0.2 micron particles. The efficiency of the caustic scrubber shown in Figure KH-1104414 was determined for a variety of flow rates, quantities of berl saddles and of caustic solutions. In the range of operating flow rates the efficiency was 95 per cent for the collection of volatile iodine.

Isokinetic sampling was considered since it is not only necessary that the monitoring system be adequate but that the sample taken be representative. Although the system was designed for isokinetic sampling, particle size analyses

made the need for the balancing of the sampling and stack linear flow rates less stringent.

Absorption studies of the alpha and beta fission product activity of the entrained particulate material collected in the H-70 filter papers revealed average absorptions of 55 and 25 per cent for the alpha and beta activities, respectively.

PARTICLE SIZE DISTRIBUTION OF STACK EFFLUENT

Since the stack effluent due to separations operations is composed of a heterogeneous mixture of entrained radioactive and non-radioactive particles, the particle size distribution was studied in relation to the physical size of the heterogeneous mixture of the particles, and in relation to the radioactive distribution.

The molecular filter paper was used as a filtering medium to collect a representative sample of the stack effluent. This type of filter paper was chosen for its efficiency in the collection of submicronic particles and ease in detecting particles in the same medium under the microscope. The particle size distribution during various chemical operations of the Separations Process are illustrated in Figure KH-9A2354. The data reveal the abundance of submicronic particles and the similarity of distributions during various phases of the chemical process. A geometric mean of 0.2 micron in each case is readily observed when the data are plotted on log-probit paper as shown in Figure KH-9A2354. An average of 10 analyses during all phases of the process as shown in Table 1, indicated a geometric mean of 0.2 micron and a standard deviation of 2.7. Since the limit of detection with the light microscope is 0.1 micron, it was felt at the time that the true geometric mean was less than 0.2 micron. This feeling was later substantiated by electron microscopic analyses of the filter samples and by autoradiographic studies of the radioactive particle size distribution.

TABLE 1

SIZE DISTRIBUTIONS OF PARTICULATE MATERIAL

<u>Repetitive Run</u>	<u>Separations Operation</u>	<u>Geometric Mean, microns</u>	<u>Standard Deviation</u>
1	Dissolving	0.2	2.5
1	Dissolving	.2	3.1
2	Dissolving	.2	2.9
2	Dissolving	.2	2.7
3	Dissolving	.2	2.6
3	Dissolving	.2	2.6
4	Head-End	.2	2.5
4	Head-End	.2	2.3
4	Extraction	.2	2.9
5	Extraction	.2	2.4

The modified cascade impactor with a molecular filter paper in the fifth stage was used to determine a relationship between particle size and activity.

Even with flow rates of 34 l/min through the impactor, nearly all of the activity was deposited on the molecular filter paper, indicating that most of the activity was composed of or deposited on sub-micronic particles.

Autoradiographic techniques were investigated to determine further the relationship between particle size and radioactivity. A stripping film technique similar to the methods employed by La Riviere* and Boyd** indicated the presence of many sub-microscopic particles and the need for electron microscope studies.

Samples were analyzed under the electron microscope at the General Electric Research Laboratory. Silicon dioxide was evaporated on a small section of the Millipore filter, under a vacuum of 0.1 micron of mercury. This section of the filter paper was then dissolved in acetone. Upon hardening, the silicon retained an impression of the surface structure of the filter and served to hold the sample particles in position. The electron micro-graph of an unexposed filter paper used for control purposes is shown in Figure 1121212. The surface of the Millipore filter paper under a magnification of 15000 is seen in this electron micrograph. The electron micrograph of a portion of an exposed filter paper which had a geometric mean and standard deviation under light microscope studies similar to those previously indicated, is illustrated in Figure 1121213. The number of particles in the range of 0.01 to 0.05 micron are far in excess of those greater than 0.05 micron. The true geometric mean, then, is closer to 0.05 micron than 0.2 micron as determined under the light microscope.

ISOTOPIC DETERMINATION OF FISSION PRODUCTS DISCHARGED FROM PILOT PLANT STACK

Knowledge of the isotopes contributing to the discharged radioactivity was an important requisite in this investigation. The biological effects and consequently the maximum permissible concentrations depend not only on the level or radioactivity, but also upon the body metabolism of the elements that comprise the activity. To determine the maximum permissible concentration that may be discharged from the Pilot Plant stack, the activity was isotopically analyzed during all phases of the separations process for several repetitive runs.

ISOTOPIC DETERMINATION OF FISSION PRODUCTS COLLECTED IN THE PARTICULATE FORM

The isotopic composition of the radioactive particulate components present in the effluent was determined by radiochemical analyses of the Hollingsworth and Vose, type H-70, filter papers. These filter papers are used as the particle collecting media in the health physics stack monitoring system.

Radiochemical analyses of the filter paper samples collected during all phases of the separation process revealed that the radioactivity emitted from the Pilot Plant stack in the particulate form was composed of the rare earths, ruthenium, zirconium, niobium, barium, strontium, and iodine.

*USNRDL-342, "An Autoradiographic Method of Detecting and Identifying Beta-Active Particles in a Heterogeneous Mixture," by Philip D. LaRiviere and Stephen K. Ichiki, April 1952.

**UR-209, "Stripping Film Techniques for Histological Autoradiographs," by George A. Boyd and Agnes Williams, May 1948.

During the initial analyses of the stack effluent, separations process operations without variation in procedures were repeated. These processes were called repetitive runs. The relative proportions of beta-gamma emitting radioisotopes discharged in the particulate form during each of the chemical operations for seven repetitive runs and several non-repetitive runs were analyzed. An analysis of the third repetitive run is shown in Figure MH-9A8127. The rare earths predominated throughout nearly all of the operations, representing from approximately 50 to 80 per cent of the particulate activity during the dissolving and extraction phases. Ruthenium-106 contributed the greatest portion of the particulate activity during the head-end operations and, in most instances, exceeded the rare earths* during the first part of the dissolving and latter part of the cake dissolution operations (the third repetitive run shown here was an exception). The cake dissolution operation usually takes place following the extraction cycle but it is physically a part of the head-end. Niobium was emitted in varying amounts during all operations, representing from less than 1 per cent to approximately 50 per cent of the particulate activity. Zirconium represented less than 10 per cent of the activity during all operations except the cake dissolution. During the cake dissolution zirconium reached a maximum of 20 to 25 per cent of the particulate activity. Strontium contributed from approximately 5 to 20 per cent of the activity during nearly all of the dissolving, extraction, and cake dissolution operations of the third and fourth repetitive runs. The strontium component was as high as 40 per cent during the extraction cycle of the fifth repetitive run.

The relative proportions of the particulate fission products discharged during 5 repetitive and 2 non-repetitive runs are listed in Table 2.

The rare earths and Ru-106 composed the largest portions of the total particulate activity during repetitive and non-repetitive runs. The per cent rare earth particulate activity in the stack effluent appears to be reduced significantly when the cooling time of the slugs are reduced from 95 to 85 days. Some variations in the isotopic percentage of activity discharged, however, are attributable to the variation in the decontamination factors obtained during different runs. During the dissolving, head-end and extraction operations, on the average, 5, 85 and 10 per cent, respectively, of the total particulate activity was discharged.

ISOTOPIC IDENTIFICATION OF VOLATILE MATERIALS IN STACK EFFLUENT

The total volatile activity, excluding the radioactive noble gases, collected in the caustic scrubber comprised less than 1 per cent of the total activity discharged from the stack during all repetitive and non-repetitive runs. Analyses of the contaminated caustic solution revealed varying percentages of I-131 and Ru-106 collected in the scrubber during operating phases. The volatile components Kr-85, Xe-133 and Xe-135 detected by the constant air monitor comprised the majority of the activity discharged from the stack. Detail analyses of the percentage composition of these volatile materials during phases of the separations process are given in KAPL-814 and KAPL-863.

*KAPL-814, Semi-Annual Progress Report of Radiological Development Activities in the Health and Safety Unit, Jan.-June 1952.

TABLE 4

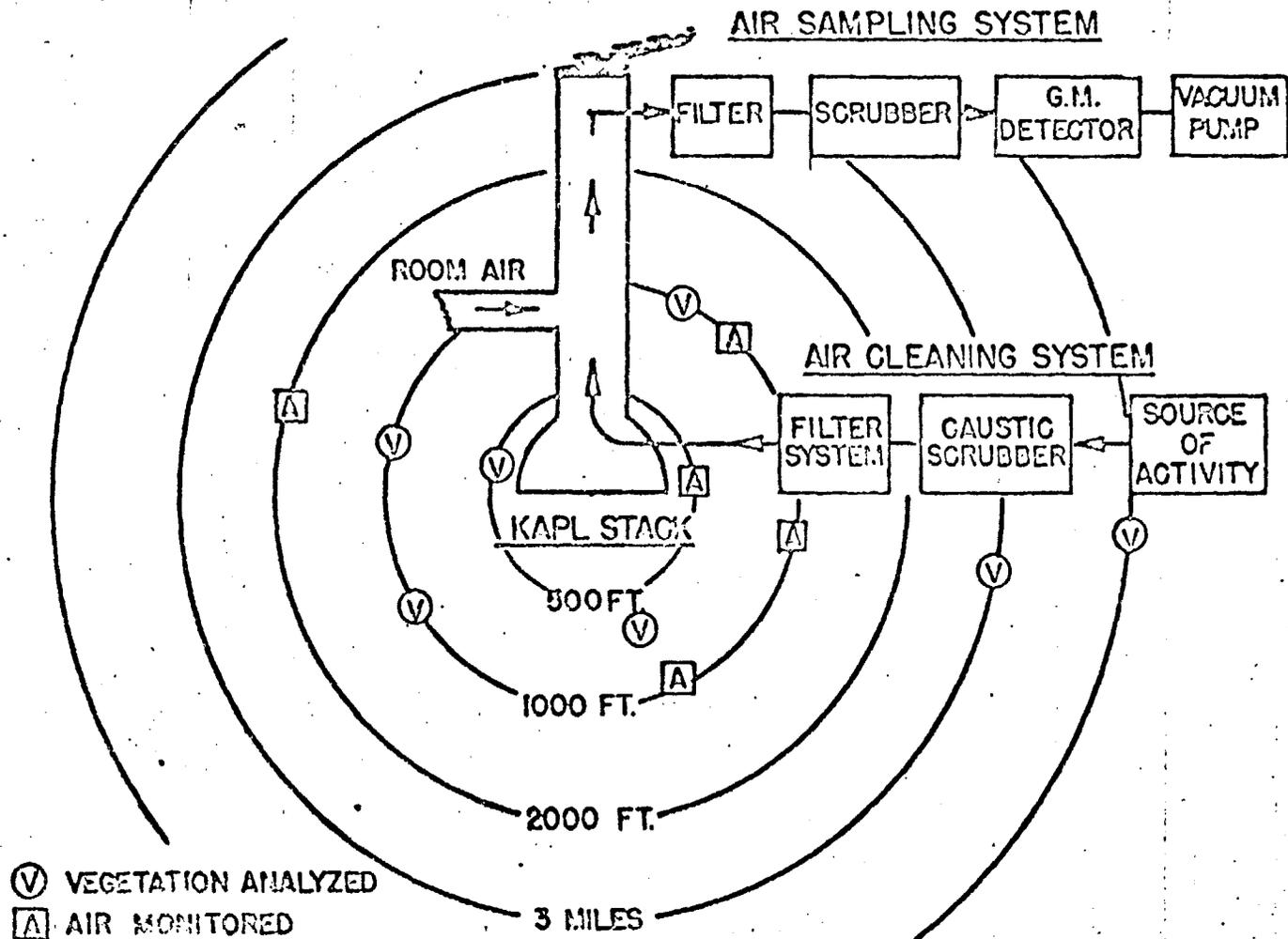
MAXIMUM PERMISSIBLE CONCENTRATIONS IN AIR AND STACK EFFLUENT

<u>Isotope</u>	<u>Max. Percentage in Stack Effluent</u>	<u>MPC in Air, $\mu\text{c}/\text{cc}$</u>	<u>MPC in Stack, $\mu\text{c}/\text{cc}$</u>
<u>Beta-Gamma</u>			
Kr-85 + Xe-133 + Xe-135	99	4×10^{-6} ** (body)	4×10^{-4}
Rare Earths & Y-91	14	7×10^{-9} * (bone)	5×10^{-6}
Ru-106	20	3×10^{-8} * (kidney)	2×10^{-5}
Ru-103	50	2×10^{-7} *** (kidney)	4×10^{-5}
Sr-89	1	2×10^{-8} * (bone)	2×10^{-4}
Zr-95	1	1×10^{-8} *** (lung)	1×10^{-4}
Nb-95	4	4×10^{-7} * (bone)	1×10^{-3}
Ba-140	1	6×10^{-8} * (bone)	$<6 \times 10^{-4}$
I-131	1	3×10^{-9} * (thyroid)	3×10^{-5}
<u>Alpha</u>			
Pu-239	100	2×10^{-12} * (bone)	2×10^{-10}

*MPC listed in National Bureau of Standards Handbook 52.

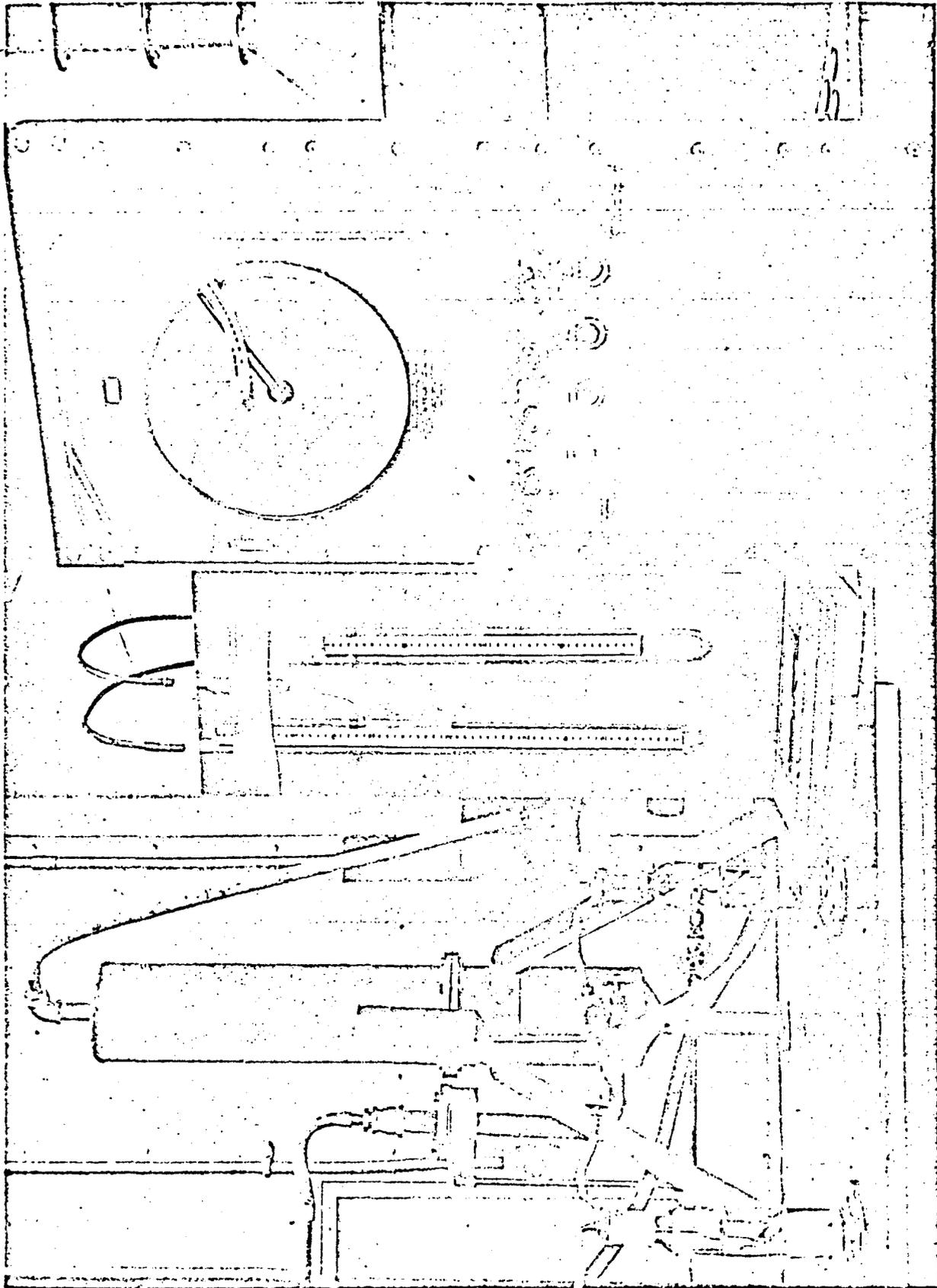
**MPC listed in Handbook 52 or calculated using formula in Handbook 52.

***MPC calculated using formula listed in Handbook 52.

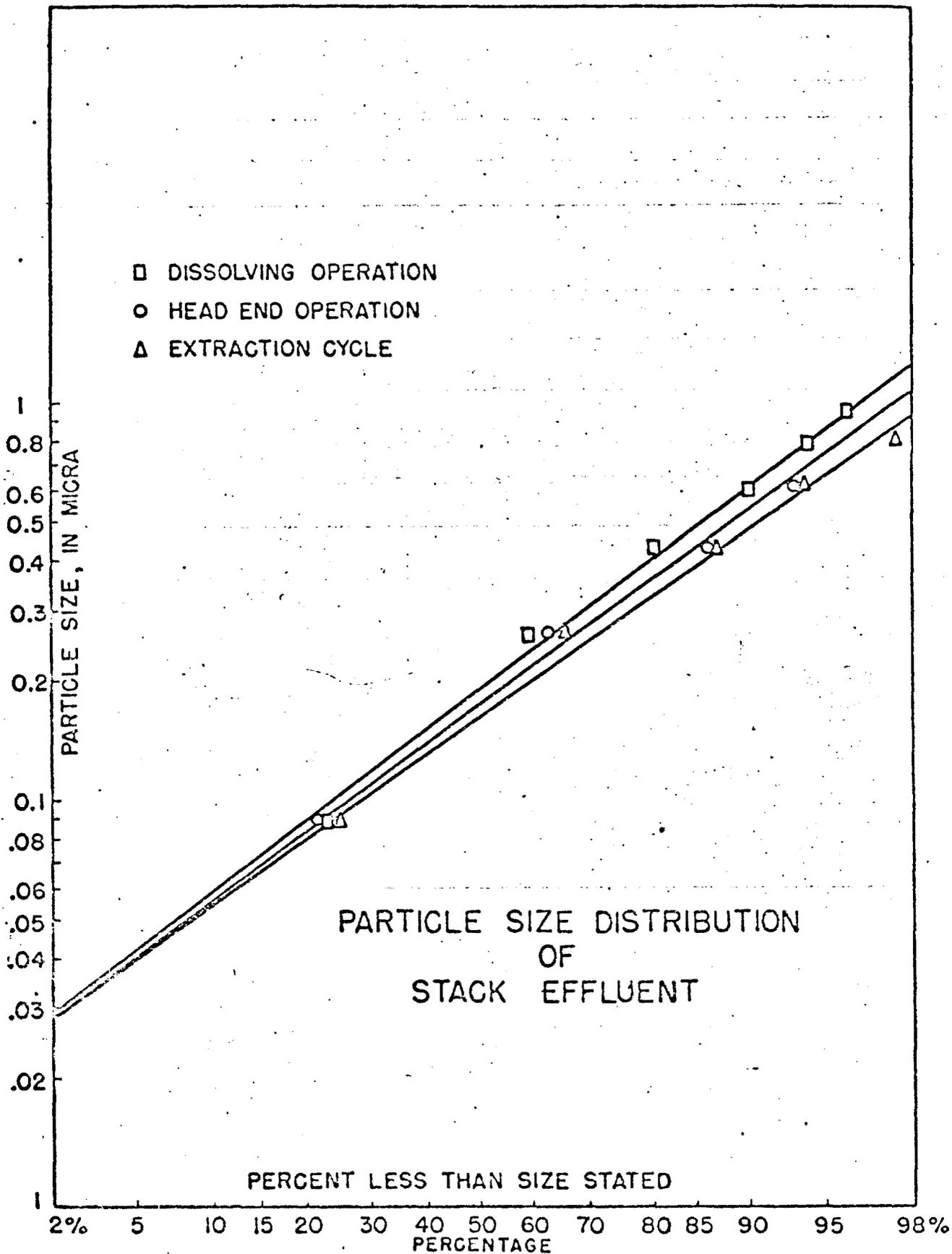


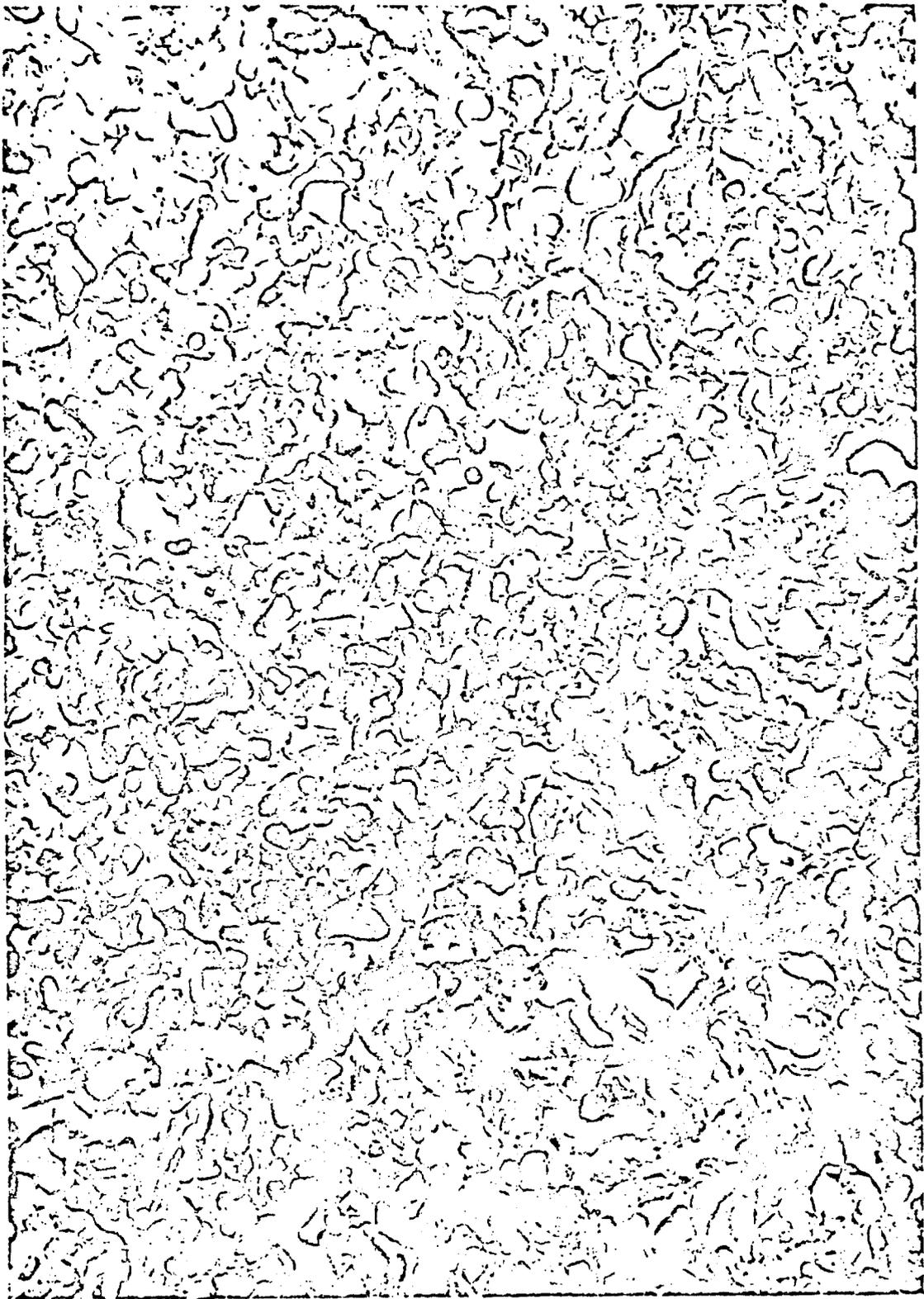
KAPL - AIR MONITORING AND CLEANING SYSTEMS

KH-9A2403



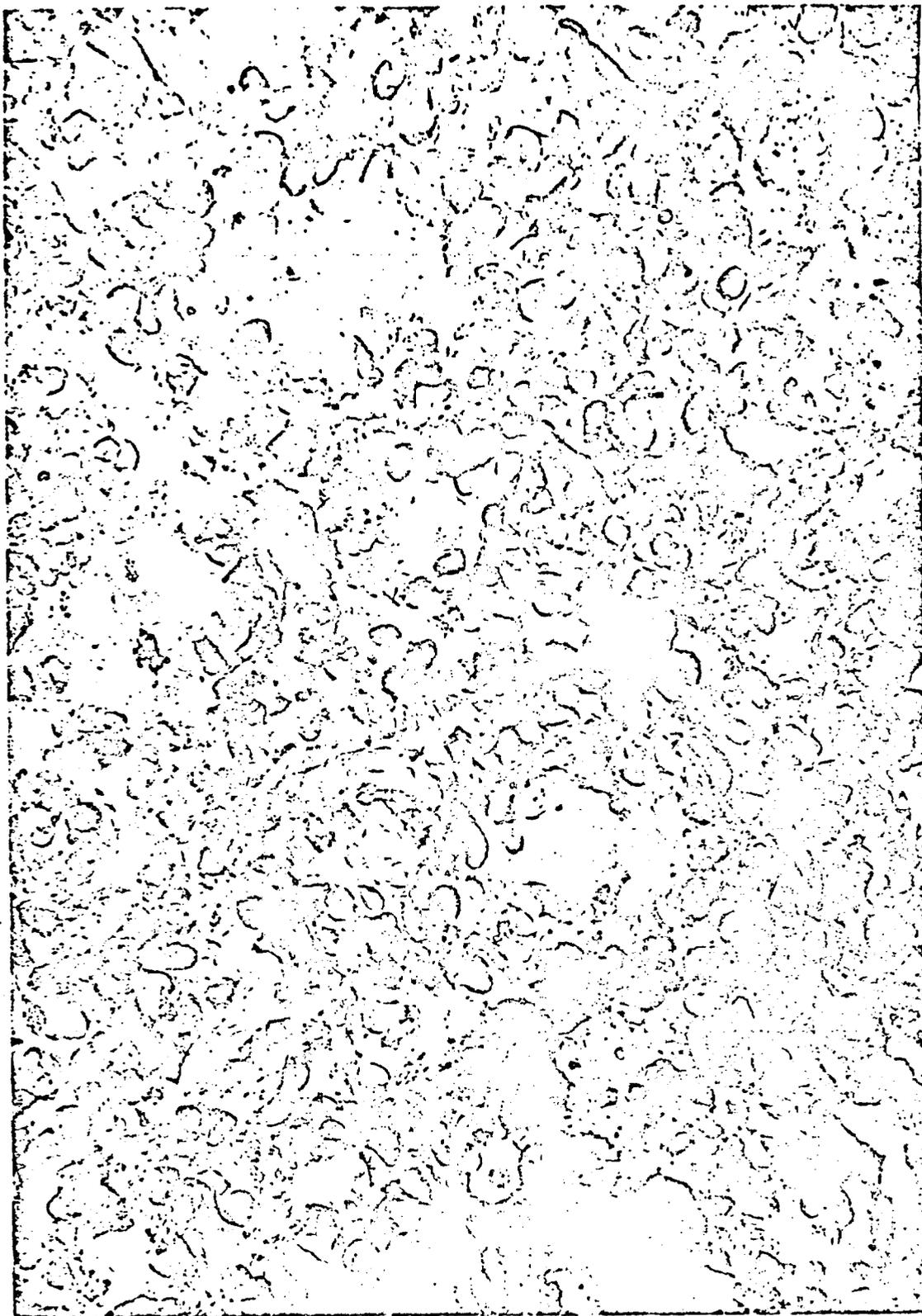
41414014-EN





MILLIPORE FILTER UNEXPOSED-CONTROL FILTER
Magnification - 15000 — Scale 0.5 micron

KH-1121212



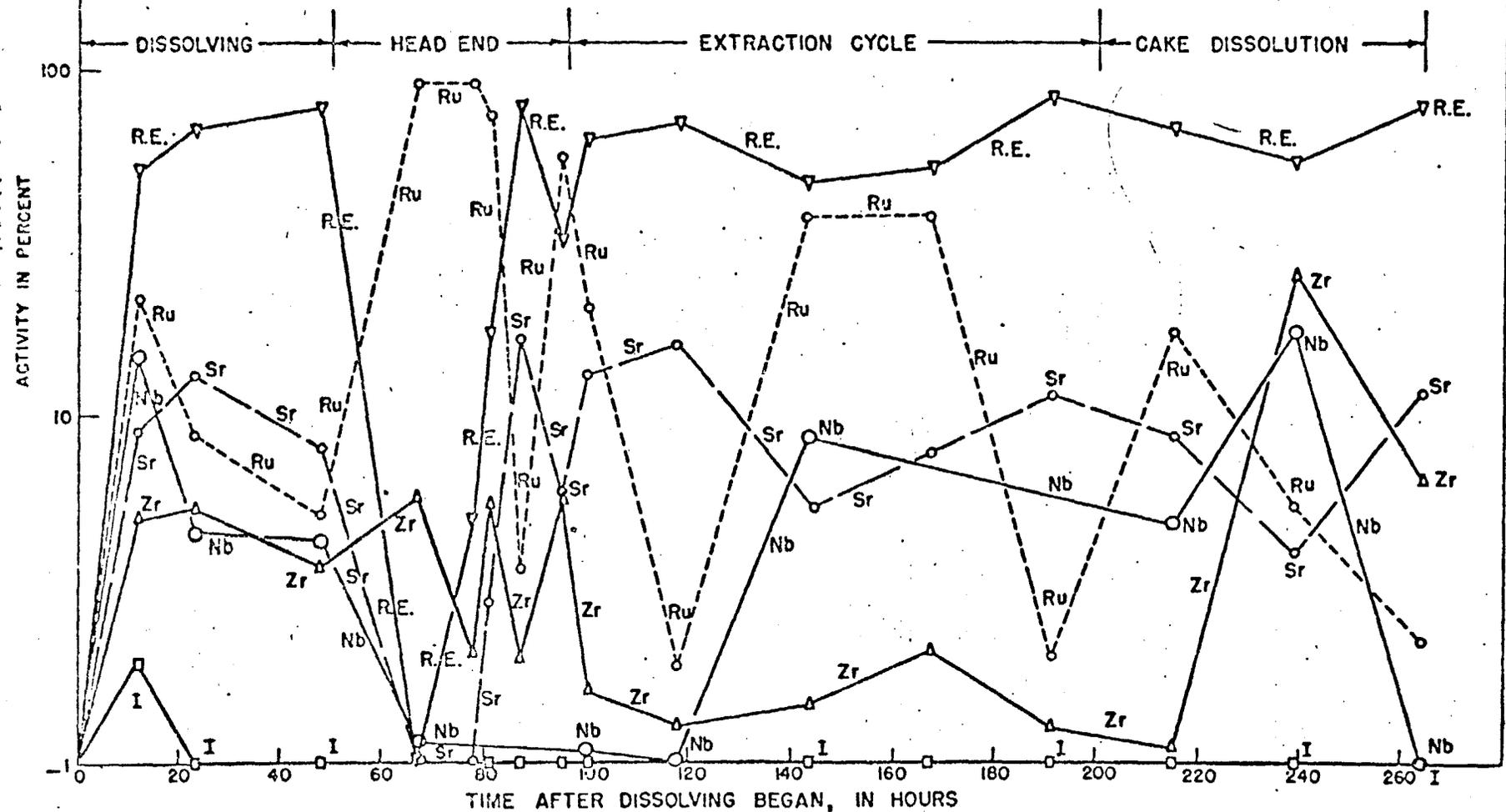
MILLIPORE FILTER EXPOSED TO KAPL STACK EFFLUENT
Magnification - 15000 — Scale 0.5 micron

KH-1121213

RELATIVE PROPORTIONS OF PARTICULATE FISSION PRODUCTS RELEASED FROM STACK DURING THIRD REPETITIVE RUN

[EACH PLOTTED POINT REPRESENTS THE AVERAGE RELATIVE ACTIVITY DURING THE
INTERVAL PRECEEDING IT.]

CONFIDENTIAL
SECURITY INFORMATION



MI-9A8127