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TO: File

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DISTRIBUTION RESTRICTED TO U.S. ONLY ^{ED}PARTICLE SIZE OF PARTICULATE MATTER IN THE DISSOLVER OFF-GAS STREAMIntroduction

During a visit on August 2, 1951, W. Bain of the Vitro Corporation (Keller) and J. Hayner of the AEC - Washington, D. C. discussed with Hanford personnel the desirability of obtaining a characterization of the particle size of the particulate matter in the dissolver off-gas stream at one of the Hanford plants. Prior to the installation of Fiberglass filters in the dissolver off-gas lines, this particle size measurement would have been useful in evaluating the requirements of the proposed Keller method of handling this air stream, viz.

- a. Injection of propylene or ethylene glycol into the air stream to produce large particles from the small ones,
- b. Injection of steam into the air stream to increase the particle loading in the air stream,

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- c. Passage of the air stream through sonic towers to agglomerate the particles,
- d. Removal of the agglomerated particles whose size would now be larger than five microns by conventional cyclones.

The much simpler solution of this problem by the use of high efficiency, fixed bed filters in the off-gas line obviated the necessity of considering the Kollax method of handling this gas stream. Since the fixed bed filters were designed to remove particles whose size was from 0.4 - 0.6 microns, the size range of particles most difficult to remove from air streams by filtration, it had never been considered particularly worth while at Hanford to make a particle size measurement of the exceedingly radioactive dissolver off-gas stream. Considerable previous experience in connection with the iodine problem and with the recontamination of the ventilation air stream after the installation of the sand filters had shown that the dissolver off-gas stream contained very large quantities of radioactive particulate matter, and would be a difficult air stream on which to make particle size determinations. Work along these lines was therefore not undertaken at Hanford.

During the discussion with Kaysor and Pahn it became apparent that Hanford would be requested to make particle size determinations on the dissolver off-gas line, or to assist Kollax personnel in obtaining this information. The Kollax method of making particle size measurements by the use of a cyclone, a Greenburg-Smidt impinger and filter paper in series posed serious problems for use in the Canyon Building, particularly if handled by men with no experience at working at the high activity levels that would be involved. In the interest of having the minimum expenditure of time and effort on the part of Hanford personnel, it was agreed that characterization of the particle size of the particulate matter in the dissolver off-gases would be obtained by Hanford personnel by methods previously used at Hanford on the ventilation air stream. In this way the measurements would be obtained with less effort than that required to train and assist off-site personnel in getting less meaningful information.

Summary and Conclusions

Particle size measurements with a modified cascade impactor of the particulate matter in the dissolver off-gases during the period of maximum gas evolution, and during the air sparge of the dissolver solution after the completion of the dissolving cycle indicate that the mass-size median of the particles was less than 0.1 micron in both cases.

The measurements were made, unfortunately, at a time when the efficiency of the silver reactor upstream to the sampling point was low enough to permit appreciable amounts of iodine through to the cascade impactor. The samples were therefore permitted to decay for thirty-five days before the relative amounts of radioactivity on the stages of the impactor and the follow-up filter paper were evaluated.

The size of the particles obtained, 0.1 micron, should be considered primarily as an order of magnitude rather than an absolute value, because the cascade impactor (as well as any other impaction device) can not give an accurate value for the particle size in this size range.

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Experimental

A modified cascade impactor with a follow-up filter as a fifth stage was installed in the line used to measure the pressure upstream to the Fiberglass filter which was installed in the dissolver off-gas line in 4-5L cell in the B Plant. The sampling line was therefore downstream to the silver reactor. An orifice in the line permitted the samples to be taken at a rate of one cubic foot per minute.

The first sample was taken during the dissolution of the second cut on September 11, 1951 at a time after the start of the dissolving cycle during which maximum gas evolution occurs. The sampling time was limited to two minutes because of the very high activity that could be expected on the filter paper. A second sample was then taken for a five minute interval as soon as the slides in the impactor and the filter paper could be changed; only eighteen minutes elapsed between the finish of the first sample and the start of the second.

The third and fourth samples were taken after the finish of the dissolving cycle during the period that the solution is being air sparged. Water is not circulated in the reflux condenser during this interval. The sampling time for the third and fourth samples was four and three minutes respectively.

Cutie pie readings at two inches were taken of the slides and filter paper immediately after they were removed from the sampling equipment. The filter paper and slides were then given to the Analytical Services Unit laboratory at B Plant to handle according to established practices.

About the time these samples of the dissolver off-gases were taken, it became apparent that the efficiency of the silver reactor in 4-5L cell had dropped from 99.9 per cent to only 85-90 per cent. It was therefore likely that some of the activity obtained on the slides and filter paper was because of the presence of I^{131} . The slides were therefore permitted to decay for thirty-five days before an evaluation was made on the basis of the activity on the slides. The method of preparing filter paper samples in the laboratory prior to making counting plates removes iodine very completely. For this reason the counting plates were not retained, although it would have been helpful to have had them for a final count after the thirty-five day decay period.

Data and Discussion

The dissolver off-gases were sampled without any radiation incident during the peak of the gas evolution and also during the sparging period after dissolution. That the off-gas air stream contains a large amount of radioactive particulate matter can be readily seen from the compilation in Table I. The filter paper, which acts as the fifth stage of the impactor, was found to have cutie pie readings (beta plus gamma) as high as 1750 mrep per hour per cubic foot of air collected.

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Table I -- Cutie Pie Readings of Dissolver Off-gas Samples**Sampler: Modified Cascade Impactor - Filter Paper for Fifth Stage****Sampling Rate: One Cubic Foot per Minute**

Sample No.	Sample Taken During	Sampling Time (mins)	Cutie Pie Readings - mrem/hr				
			Stage No.				
			1	2	3	4	5
1	Reaction peak	2	20	10	10	100	3500
2	Reaction peak	5	30	20	15	15	5000
3	Air sparging	4	12	12	30	60	160
4	Air sparging	3	60	75	100	150	780

The cascade impactor slides and plates containing the radioactive material collected by the filter paper were counted in the laboratory on the day after the samples were taken. Counts were made subsequently at frequent intervals in order to observe the decay trend of the individual stages of the impactor. Although there was considerable variation in the results for the four runs, the fraction of the activity due to I^{131} appeared to decrease in going from the first slide, which collects the coarsest cut of the particles, to the fifth stage which, of course, collects all the particles that go through the impactor. The counting data are summarized in Table II.

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Table II - Counting Data for Dissolver Off-gas Sampler

Refer to Table I for sample designation. All values are disintegrations per minute.

Sample	9-12	9-28	10-1	10-4	10-10	10-20	10-22
Run 1 -							
Stage 1	96,800	39,300	38,500	31,700	26,980	24,370	21,170
Stage 2	72,600	36,800	35,000	29,600	27,380	23,110	21,550
Stage 3	238,000	176,000	166,000	162,500	151,300	139,020	132,100
Stage 4	914,000	778,000	750,000	701,000	651,700	648,100	608,000
Stage 5	5,871,400	--	--	--	--	--	3,050,000*
Run 2 -							
Stage 1	357,000	107,000	85,700	64,700	43,590	22,820	20,780
Stage 2	346,000	92,900	77,400	59,500	37,180	20,580	15,240
Stage 3	519,000	154,700	127,500	102,500	71,650	46,880	39,120
Stage 4	690,000	281,900	249,000	222,500	183,500	150,500	142,100
Stage 5	12,312,000	--	--	--	--	--	3,850,000*
Run 3 -							
Stage 1	78,300	38,500	34,900	27,900	20,500	14,360	14,750
Stage 2	97,500	46,300	39,200	33,600	24,170	14,850	14,580
Stage 3	236,000	152,000	131,400	117,900	102,100	85,530	81,840
Stage 4	695,000	508,000	470,000	418,000	365,900	342,800	324,400
Stage 5	10,826,800	--	--	--	--	--	7,815,840*
Run 4 -							
Stage 1	218,000	99,000	82,300	63,900	46,790	28,640	24,560
Stage 2	429,000	189,500	151,000	117,000	76,590	42,230	35,150
Stage 3	988,000	478,000	396,000	319,000	240,200	167,600	152,100
Stage 4	--	--	892,000	731,000	604,200	528,900	493,200
Stage 5	3,530,000	--	--	--	--	--	3,035,000*

* Calculated value

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The calculated value for the fifth stage was obtained on the basis of the following two assumptions. First, that the original count for the fifth stage included little or no I^{131} ; second, that the decay rate for the fifth stage was the same as for the fourth after the I^{131} had decayed to a low value on this stage. Although the samples had only decayed for thirty-five days, hardly long enough to have all the I^{131} decay and establish a decay rate for the I^{131} free material, the last portion of the decay curve was extrapolated to zero time to give an initial count value of the I^{131} free material. The same fractional decrease of the I^{131} free material on the fourth stage was assumed to occur on the fifth stage during the decay interval. The calculated value of the fifth stage is then readily obtained by reducing the initial fifth stage value by the same fraction that occurs in the fourth stage during the same interval.

After the fifth stage value has been established, the mass-size median particle size can be obtained by methods described in HW-20847, "Filtration of Radioactive Aerosols by Glass Fibers". The data in the last column in Table 2 are handled as shown below in Table III. The cumulative per cent of activity less than the stage median value given in this table is plotted against the stage median diameter on a logarithmic-probability grid. The median particle size is obtained from the point where the best fitting line through the experimental points crosses the fifty per cent line of the cumulative per cent of activity less than the stage median axis. In fitting the best line to the experimental data emphasis is placed on the three middle values.

Table III -- Cascade Impactor Calculations

Sample Flow Rate - cfm

Density of Particles - 2.3 gm/cc (assumed)

Refer to Table I for Sample Designation

	Stage Median (microns)	Cumulative Per Cent of Activity Less Than Stage Medium			
		Run 1	Run 2	Run 3	Run 4
Stage 1	5.48	99.7	99.7	99.9	99.6
Stage 2	2.30	99.2	99.3	99.7	98.6
Stage 3	1.21	99.1	98.6	99.1	93.0
Stage 4	0.81	87.5	96.4	96.5	84.9
Stage 5	0.48	39.8	47.4	32.3	38.6

A plot of this data on the logarithmic-probability grid indicates that the median particle size is approximately 0.1 micron. This is lower than the value one would surmise from an inspection of Table III. The large uncertainty of the value of the stage median for the fifth stage (filter paper), however, must be recognized, and therefore less weight should be given to this value in obtaining the best fitting line for the experimental.

Regardless of the type of impaction device used, the accuracy of the particle size measurement is always less when the median size gets much below a micron. For the purposes to which this work would be used, the characterization of the particle size with the modified cascade impactor is believed to be adequate. Further work with more refined methods of making this measurement, such as the thermal precipitator would not appear to be justified.

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