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EUROCHEMIC ASSISTANCE PROGRAM: COMPILATION OF HAPO-ORIGINATED INFORMATION SENT TO EUROCHEMIC VIA TRANSMITTAL LETTERS (THROUGH TRANSMITTAL LETTER No. 87)

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ABSTRACT

HAPO-originated information sent to Eurochemic via transmittal letters is combined in this report to facilitate general distribution. The subjects covered include instrumentation, de-entrainment, waste, pulsers, fuel shipment, and criticality.

NOTICE

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1.0 INTRODUCTION

As part of the Eurochemic Assistance Program, information was sent and identified in transmittal letters. Specifically prepared information was generally issued with an identifying number and sent via the Division of International Affairs. Information needed quickly or small in quantity was included in the transmittal letters. The HAPO-originated information, which was not given the general distribution prescribed in the procedures for obtaining and transmitting information, is combined in this report.

2.0 DISCUSSION

2.1 Instrumentation (via transmittal letter 28)

The "contact alpha monitor," an alpha scintillation phosphor, protected from exposure to process solutions by a 0.00025-in.-thick plastic covering, has been under development at HAPO for sometime. Prototypes of this instrument have given good performance under some conditions but have been less satisfactory under conditions where it is believed that abrasive solids in the solution may have degrading effects upon the integrity of the protective covering. The development of this instrument is continuing in an attempt to find alternative phosphors with chemical resistance to process solutions.

Eurochemic reports on in-line instrumentation and TR-37 have been reviewed by HAPO. The following comments are offered:

(1) We feel that the bold approach for complete in-line instrumentation is a commendable one. We do wonder, however, whether it is necessary to provide as complete instrumentation as it appears they are planning. Again, we recommend that the Eurochemic group discuss this general subject with United States experts at the various AEC sites.

(2) Uranium analysis by X-ray fluorescence is a good laboratory analytical method. However, considerable development work is required to adapt this procedure to in-line analysis. Scattered background limits the detection and precision in an in-line application. We believe that the use of dual detector crystals, or some other method for measuring scattered background intensity, would be required for successful in-line application at low uranium concentrations.

(3) The HAPO scintillation counter measures activity down to about 10^{-5} c/l; 10^{-10} c/l is a very low level and difficult to reach by

any in-line instrument. The background of the crystal in the laboratory is about 200 counts/min. With only 10^{-10} c/l, Cs-137 gives about 6 counts/min. The upper limit of 10^{-2} c/l should be all right.

(4) The use of Bremsstrahlung for gamma absorption techniques should be possible but is of questionable value. The main problem would be source intensity. Self-absorption of the beta source limits the source size and intensity.

2.2 De-entrainment (via transmittal letter 50)

Your inquiry in letter No. 18 on de-entrainment from submerged combustible evaporation was referred to HAPO. HAPO has commented as follows:

> It is their experience that the vertical vapor velocity is of little consequence in the designing for high decontamination factors when a de-entrainer is used with a concentrator. Literature data and HAPO's experience indicate that about one part per million parts of vapor (DF = 10^6) that is entrained is one micron or less and would not settle out, even at very low vapor velocities (less than 0.2 fps). Baffles or the vapor-space deflector you mentioned would be of little use for this small particle-size entrainment. A recent paper by C. S. Schlea and J. P. Walsh, SRP, entitled "De-entrainment in Evaporators," substantiates HAPO's views and includes information on the latest development of de-entrainment devices. (Copies of this paper have been requested for Eurochemic from du Pont.) The submerged combustion evaporator differs from evaporators using steam coils or tubes in that the amount of submicron entrainment produced is about 1,000 times greater. This can be reduced substantially (to about 2 ppm of vapor, DF = 5×10^5) by lowering the sparge gas temperature. Mr. Rey's thesis (being obtained) covers this last peculiarity of the submerged combustion evaporator.

2.3 Waste (via transmittal letter 54 and 67)

The questions in your letter No. 21 relative to the data in HW-20847 and to deep bed 115K glass fiber ventilation filters were referred to HAPO. Most of the questions are answered in HW-30142, "Comparative Study of Alternative Fibrous Glass and Sand Exhaust Ventilation Air Filter Installation for Purex". This document does not discuss the allowable pressure drop increase for Purex ventilation or the effect of accidental air flow stoppage on filter life and performance. Additional comments are given below.

At the Purex Plant, a modulating damper is installed downstream of the fans to control the total air flow through the ventilation system. At the present time, the partially opened damper has a differential pressure of 3 in. of water at an air flow of 125,000 cfm. Thus the differential pressure across the filter could increase these 3 in. of water with the damper open prior to affecting the total air flow. Inspection of the fan performance curves indicates that pressure could drop an additional 1 in. of water before the total air flow would drop to the minimum acceptable, ~ 110,000 cfm.

To date, the Purex Plant has never experienced a complete air flow stoppage of the large ventilation filter. On the other hand, the air flow through the dissolver off-gas system has been interrupted on numerous occasions without any noticeable effect on the small glass fiber filters. At the Redox Plant, where a sand filter is installed (rather than the large glass fiber ventilation filter), the air was completely stopped during a thorough stack rinse. No alteration in filtration or permeability characteristics was observed on the sand filter.

HAPO has provided the following information on wiremesh demister pads:

As a part of an investigation performed for HAPO Chemical Processing Department, samples of A-55 titanium, zirconium, and commercially pure tantalum were exposed as simulated demister pads in the vapor phase over boiling synthetic 6 *M* HNO_3 - Purex IWW. These same materials were also exposed to other solutions which approximated the composition of Purex IWW. The titanium samples were 6, 11, and 60 mil wire, and the tantalum and zirconium samples were 10 mil wire.

The titanium samples exhibited corrosion rates ranging from 0.1 to 0.5 mil penetration per month; considerably higher than one would expect. This relatively high rate was believed to have been due to contamination of the wire during drawing. Titanium was excluded as a material of construction because of the pyrophoric behavior of the samples. The 6 mil wire could be ignited with a paper match and would sustain burning. The 11 mil wire could be ignited with an oxygen-propane flame, but sustained burning was erratic. This behavior constituted a serious hazard in fabrication and installation of the demister pad.

Commercially pure zirconium wire exhibited corrosion rates of .01 mil penetration per month, or less. However, the pyrophoric behavior of zirconium was similar to that of titanium, and zirconium was excluded as a material of construction.

The tantalum wire exhibited corrosion rates of .01 mil penetration per month, or less, in all exposures. The wire would melt and oxidize severely in an oxygen-propane flame, but would not burn. In view of this, tantalum was selected as the material of construction for the demister pad.

The Purex demister pad is six inches thick, of 5 mil wire, and packed to 330 ft^2/ft^3 surface area. It gives a de-entrainment factor of about ten, to raise the evaporator overhead de-entrainment factor to total of about 2×10^6. It is our judgment that sprays may give an additional de-entrainment factor of two, not enough to warrant their use.

2.4 Pulsers (via transmittal letter 67)

HW-51884, "Plug Piston Pulse Generator," was sent via transmittal letter 19. Results of recent work with this type of pulser is summarized as follows:

A six-inch piston pulser has been operated pulsing recuplex CAF at a 1/2 inch amplitude and 60 cyc/min for 10 million cycles. There has been no change in the amplitude ratios indicating no significant piston wear. A 3-inch diameter piston pulser has been operating at 1-3/8 in. amplitude at 100 cyc/min pulsing water for about 3 million cycles with no change in the amplitude ratio.

2.5 Fuel Shipment (via transmittal letter 70)

If U.S. owned low-enrichment fuels are processed by Eurochemic and are returned to the U.S., the uranium would go to the Y-12 plant and the plutonium to the Rocky Flats plant. It has not been decided whether this material will be returned directly to the U.S. or to the reactor operator. Shipping container specifications for uranyl nitrate solutions are currently being developed. HAPO has made a preliminary survey on methods and costs of shipping low-enriched uranyl nitrate solution within the continental U.S. Tentative conclusions indicate the following:

(a) It appears feasible to ship low-enriched UNH solutions in bulk containers by employing rigid administrative controls, on a double contingency basis, of limiting uranium concentration and preventing freezing of the solution.

(b) Shipment in nuclear safe containers appears too costly.

(c) Soluble poisons do not offer sufficient assurance and advantages in nuclear safety control to justify their use.

(d) About equal capital investment is required for tank car or drum shipment. The tank car shipment is favored because of its greater operability, an anticipated greater service life, and a lower potential for human error.

2.6 Criticality (issued as HW-62845 and HW-63061)

HW-62845, by E. D. Clayton

I do have a further comment relative to the Eurochemic criticality study, Technical Report No. 68. This concerns item: 6.23, Cadmium or Boron Lined Vessels, p. 31-32. As you may know, there are plans to revise TID-7016, "Nuclear Safety Guide". In connection with this revision, it now appears that the "safe" slab thickness for tanks containing aqueous Pu solutions up to 1000 gm/L for a nominal reflector ($\leq$ 1 inch of water) may be revised downward slightly to 2.3 inches, or 5.84 cm (the current value is 6.6 cm). There is no new experimental data to effect this change; this is simply the result of further analysis and interpretation of the existing data.

There is a further point to consider. I believe a thin Cd lining will result in the slab tank being safe for the case of a nominal reflector. However, to be certain in the above case I would limit the slab thickness to no more than 2 inches for a concentration of 1000 gm/L. In the Hanford Pu Critical Mass Facility, which is now under construction, we have slab tanks which are 2.5 inches thick, Cd lined, and separated by 30 cm of polyethylene. In view of the possible revision of TID-7016, and the lack of experimental data, we now plan to check these slab tanks by critical experiments to be sure they are safe for concentrations up to 1000 gm/L; if this concentration is not safe we will determine the safe concentration. Since the Critical Mass Facility will not be completed until about June, 1960, we will not be able to conduct these experiments until after that time.

HW-63061, Appendix A, by G. W. Stuart

REACTIVITY CHANGES

A considerable number of exponential experiments[1] have been performed on light water - slightly enriched uranium systems. The results of these experiments may be used to set column size limits on chemical plant dissolvers used to process enriched fuel elements. There is an additional

twist to this procedure, however, since in the dissolver our fuel elements are set not in water, but instead are in a solution containing uranium of the same enrichment as is contained in the fuel rods. The purpose of this note is to relate, by extremely pedestrian means, the infinite multiplication constant, k'_∞, of the fuel rod-uranium bearing solution system to the measured k_∞ for the fuel rod-water system.

We may write

(1) $$k_\infty = \epsilon P_r f \eta_r$$

and

(2) $$k'_\infty = \epsilon P_r P_s \left[f' \eta_r + (1 - f') \eta_s \right]$$

where the subscripts r refer to fuel rods and s to solution. The quantity ϵ is assumed not to change between the cases. The resonance escape probability of the solution is

(3) $$F_s = 1 - \frac{V_s}{V_s + V_r} (1 - P_{s\,\infty})$$

where V is volume and $P_{s\,\infty}$ is calculated for an infinite homogeneous medium.[2] The η's are respectively;

(4) $$\eta_r = \frac{\nu_{25} \Sigma^f_{25}}{\Sigma^a_r} \quad ; \quad \eta_s = \frac{\eta_r}{1 + \frac{\Sigma^a_s (H_2O)}{\Sigma^a_s (U)}}$$

Furthermore, one can show[3]

(5) $$f = \frac{1}{1 + C \Sigma^a_s}$$

where C is a constant determined by lattice spacing, rod size and enrichment, while

(6) $$\Sigma^a_s = \Sigma^a_s (H_2O) + \Sigma^a_s (U)$$

It should be noted that we neglect, in the interests of conservatism, additional poisoning effect due to the presence of nitrogen.

Combining equations (1) and (2), substituting into this equations (4) and (5) and expanding, we find

(7) $$k'_\infty = \mu k_\infty$$

where

$$(8) \qquad \mu = P_s \left[1 + C\, \Sigma_s^a{}_{(U)}\, (1 - f) - (C\, \Sigma_s^a{}_{(U)})^2\, f\, (1 - f)\, t \ldots \right]$$

Let us apply equation (8) to three sample lattices constructed of 0.750 inch diameter, 1.01% enriched uranium rods.[4]

Case	$\frac{V_s}{V_r}$	f	C
1	1.3	0.91	4.2
2	2.0	0.87	6.6
3	3.5	0.79	12.0

In the accompanying graph we plot the μ's for these cases versus the H/U atomic ratio of the solution.

In conclusion we may state that for any reasonable lattice, i.e., any lattice for which the initial thermal utilization is not rediculously small, addition of uranium of the same enrichment as the fuel rods to the water moderator will reduce k_∞.

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(1) Kouts, et al, Geneva paper, p/600.

(2) By using Eq. 9.11.5 and the equation on p. 256, Glasstone and Edlund.

(3) Feinberg, Geneva paper, p/669, see Eq. 16.2.

(4) The values of f are only approximate.

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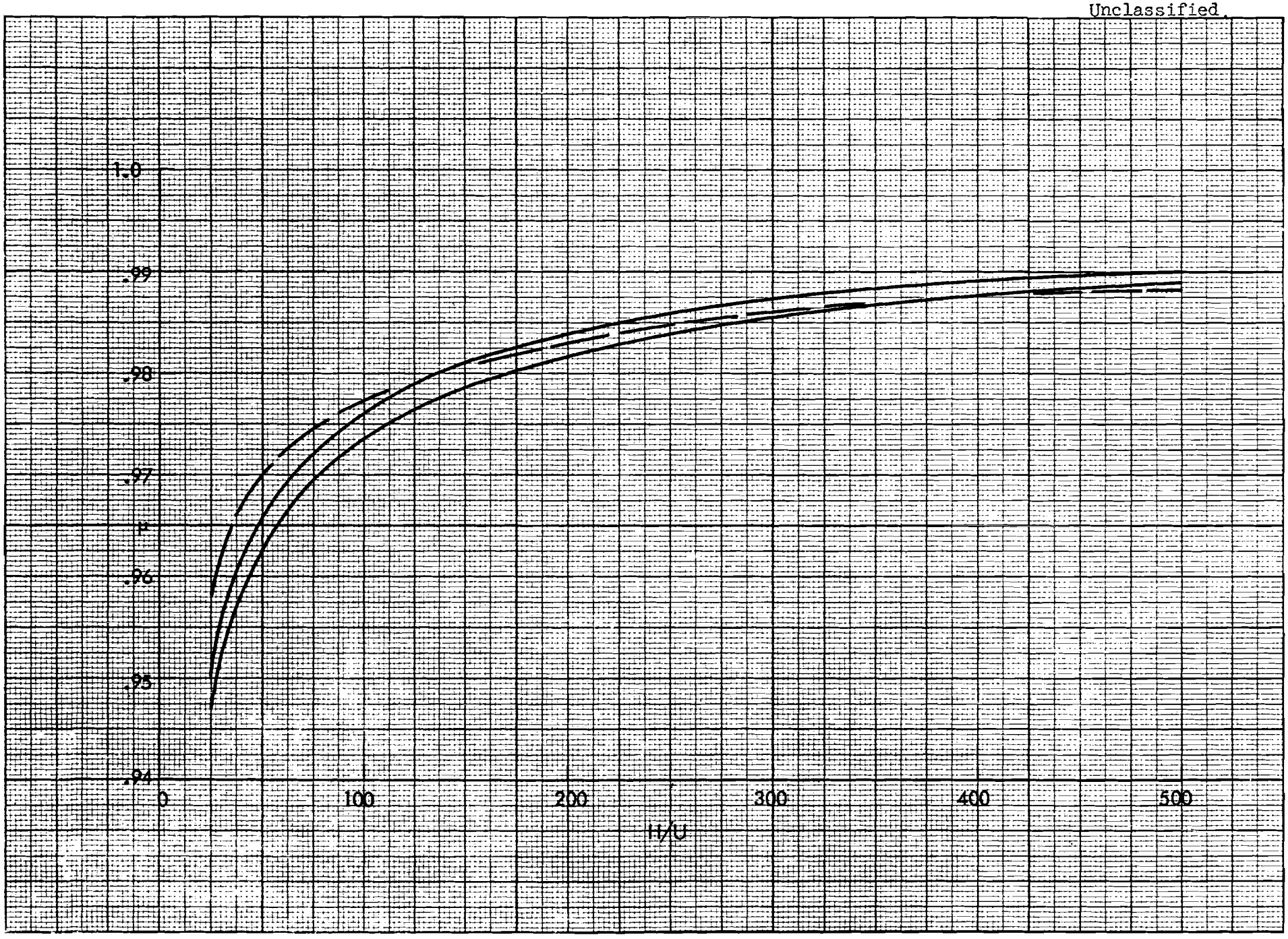

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