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DISPOSAL OF IRRADIATED WASTE "INK" SOLUTION

(PRODUCTION TEST 105-529-A)

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AEC RESEARCH AND DEVELOPMENT REPORT

DISPOSAL OF IRRADIATED WASTE "INK" SOLUTION

(Production Test 105-529-A)

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Available from Technical Information Service P. O. Box 1001, Oak Ridge, Tennessee By

H. V. Clukey

July 20, 1954

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ABSTRACT

Boron solution circulated through special poison tubes to achieve more variable control of neutron "flattening" was tested in the 100-DR Hanford reactor. About 2700 gallons of irradiated waste Ink solution from Production Test 105-529-A was discharged to an underground crib at 100-DR, after radiochemical analyses and evaluation of radiation protection aspects by the Radiological Sciences Department. In case the Ink method is considered for production use at Hanford in the future, further biological and biophysical study is recommended to determine whether irradiated waste Ink solution may be disposed of into the Columbia River, into the ground near the river, or into the ground several miles from the river.

DISPOSAL OF IRRADIATED WASTE "INK" SOLUTION

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INTRODUCTION

The Ink system provides highly flexible control of neutron fluxes at various locations in a nuclear reactor of the Hanford type while the reactor is operating. The Hanford Ink systems, both experimental and proposed for production, are fully described in documents $HW-33523^{(1)}$ and HW-33522.⁽²⁾ Briefly, a solution of boron compound which has a high neutron absorption cross-section is circulated through the reactor at various locations to prevent local overheating and to permit optimum control of the power level. During the period of unstable operation from start-up to equilibrium, the Ink solution is diluted as the need for highly flexible control decreases, until after equilibrium is reached no Ink solution may be required. The excess Ink solution from dilution as well as that from draining the system cannot be economically reused, and being radioactive from irradiation while in the reactor must be disposed of safely.

The alternate methods of radioactive waste disposal are epitomized in the phrases "dilution and dispersal" and "concentration and containment." Conditions at Hanford are especially favorable to the first method, because of the adjacent Columbia River and the large acreage of essentially desert soil. Therefore, the evaluation of radiation protection aspects of disposal of any liquid waste produced near the river, such as this Ink solution, first considers disposal into the Columbia River. If this is not advisable, consideration is given to disposal into the ground near the river, and if still unsatisfactory, into the ground several miles from the river.

Policy at Hanford limits disposal of radioactive solutions into the river, or into the ground near the river, to concentrations such that under unfavorable conditions the activity density of any radioisotope at a downstream point of public use will not exceed one-tenth of the maximum permissible concentration (MPC) given in National Bureau of Standards Handbook 52.⁽³⁾

EXPECTED CHARACTERISTICS OF IRRADIATED INK SOLUTION

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Prior to production tests of the experimental Ink system in the 100-DR Hanford reactor, calculations were made of the expected radioactive composition of two Ink solutions, one made with potassium tetraborate and the other with boric acid, based on the following criteria for the tests:

1. Eight gallons of a 10% solution of potassium tetraborate $(K_2B_4O_7 \cdot 5H_2O)$ in distilled water, containing sodium, sulfur, and chlorine ions as impurities from the salt; with radioactivity resulting from 30 days exposure in the flattened zone of a 500 megawatt reactor, in a cycle of one minute in the reactor and five minutes out.

2. Forty-five gallons of a 7.66% solution of boric acid (H_3BO_3) in distilled water, equivalent in reactor control to the 10% potassium tetraborate solution, flowing through a flux of 2.2 x 10^{13} n/cm²/sec, in a cycle of one minute in the reactor and 15 minutes out. Differences in paths inside and outside the reactor account for the larger volume and longer time outside. The boric acid is assumed to contain magnesium and sulfur impurities.

The radioisotopes expected in these two Ink solutions after irradiations are listed in Table I, together with their respective decay half-lives and MPC's in drinking water. As intended, the use of boric acid would have eliminated the high radiation levels through pipes and vessels from potassium⁴². However, potassium tetraborate was chosen for the experiments for other reasons.

RADIOISOTOPES IN IRRADIATED INK SOLUTION					
	Half-Life	MPC Water (3)	$\frac{\mu c/cc in^{(4)}}{k R O Ink}$	$\frac{\mu c/cc in^{(5)}}{\mu PO Ink}$	
Radicisotope	e Days	µc/cc	$\frac{\kappa_4}{2}$	$\frac{n_3 BO_3}{m_3}$	
К ⁴²	. 516	. 01	1280.	neglibible	
$C1^{38}$. 026	. 002*	17.9	. 0176	
Na ²⁴	. 62	. 008	1,29	.070	
s ³⁵	87.	. 005	. 033	. 054	
As ⁷⁶	1.12	. 2	. 017		
Mn ⁵⁶	. 108	. 15	. 014	. 070	
P ³²	14,1	. 0002	. 0011	. 011	
Cr^{51}	26.	. 5	. 0002	. 0005	
Zn^{65}	270.	.06	. 00008	ورزو بعده معد معد	
Ca^{45}	152.	. 0005	. 00001	. 00027	
Mg^{27}	. 0067	large		. 26	

TABLE I

Plus traces in both Ink of S³⁷, Si³¹, Al²⁸, Ni⁶³, C¹⁴ from the distilled water.

Total Activity in:

8 gal $K_2B_4O_7 = 40$ curies 45 gal $H_3BO_3 = 0.9$ curies

*Calculated from data on $C1^{36}$ in NBS Handbook 52.

CHARACTERISTICS OF IRRADIATED INK FROM 100-DR EXPERIMENTS

Four production tests of the Ink system were run at the 100-DR Hanford reactor. The waste Ink solution from each run was discharged to an underground hold-up tank, from which a sample was taken and analyzed. Recommendations were then made on the disposal of each batch. In each case, by the time the samples had been analyzed and reported, the radioisotopes had decayed to approximately the MPC for drinking, and approval was given to discharge the tank contents to an adjacent crib* (a hole filled with gravel and covered with soil several feet deep). Data on these production tests pertinent to the ground disposal are given in Table II.

*The 105-DR Pluto crib.

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By the time these samples were obtained and analyzed, the shortlived radioisotopes had decayed, so that absorption and decay analyses in addition to radiochemical analyses showed predominantly only the three radioisotopes.

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Run Number	1	2	3	4	
Last Irradiated	1-10-53	4-17-53	6-29-53	11-16-53	
Gallons	500-600	400	800	900	
Date Analyzed*	1-14-53	4-24-53	7-8-53	11-30-53	
Date to Ground	1-28-53	4-30-53	7-14-53	12-4-53	
Analysis ($\mu c/cc$)*				an a	
Total β	,0036	. 0047	. 0035	.0070	
S ³⁵	. 0014	. 0012	. 0021	. 0032	
Ca ⁴⁵	.00055	. 0012	. 0014	. 0024	
K ⁴²	.0016(diff.)	. 0023	none detect.	.0014 (diff.)	

TABLE II

DATA ON INK DISCHARGED TO GROUND

*D. L. Reid, Biophysics Section, Radiological Sciences Department.

EVALUATION OF INK DISPOSAL

On the basis of the expected radioactivity in Ink solutions (Table I), and because further information could be gained from the tests by sampling and analysis, it was recommended that provisions for the experiments should include the waste hold-up tank, with subsequent disposal of each batch after evaluation of sample analyses.

The following methods were considered for disposal of the experimental and production waste Ink solution.

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1. Discharge without hold-up into the reactor cooling water effluent system where the Ink would be diluted several hundred-fold and, in passing through the effluent retention basins, permitted to decay for 1-3 hours before entering the Columbia River.

2. Hold-up in tanks until satisfactory for discharge into the effluent system, or into the ground, either near the river or several miles away.

3. Evaporation and re-use of the concentrate.

The third method is not only expensive in equipment and operating cost (although this might be most economical where disposal to the environs is strictly limited), but also has the deleterious effect of building up the concentration of several long half-life radioisotopes.

The second method, for disposal into the Columbia River or the ground nearby, would require hold-up of production wastes for several months. The calcium⁴⁵, with an MPC of .0005 μ c/cc, a half-life of 152 days, and the activity density in Run 4 of .0024 μ c/cc, would require 342 days to decay to the maximum permissible concentration for drinking. This method was considered satisfactory for disposal of the small volumes of Ink waste from the tests into the ground near the river (but not into the effluent system), on the premise that there would be sufficient decontamination of radioisotopic contents by the soil and dilution by ground water before the solution percolated into the river. The method is not considered advisable for production volumes without further study on effects of Ink solution in the river.

However, policy at Hanford permits disposal into the plateau within the project several miles from the river of radioactive liquid wastes, so long as none of the radioisotopes of half-life greater than three years in each particular waste is detected in the ground water beneath the disposal site. $^{(5, 6)}$ Production volumes of these Ink solutions could thus be transported to the plateau and discharged immediately.

The first method, immediate discharge into the reactor effluent system, is the most convenient and economical for Hanford, but involves the most risk without study of the specific effects of irradiated Ink wastes in the river. It is apparent in Table I that the concentrations of several radioisotopes in either Ink solution are greater than MPC for drinking.

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A minimum factor of about 100 is calculated for dilution of waste Ink solution in the effluent system during reactor shut down before entering the river. This might permit disposal of the boric acid waste, but even allowing for decay time in the retention basin, several of the radioisotopes in the potassium tetraborate Ink would still exceed the MPC. Even the maximum dilution factor of about 1000 obtained during pile operation would still not be sufficient.

However, there are other factors than MPC of equal or greater importance which must be considered for disposal of radioactive wastes into the Columbia River. One is the effect on aquatic life in the river, a second is the chemical toxic effects on both humans and agricultural plants and animals using the river water, and a third is the effect of impurities taken into the cooling water systems of nuclear reactors further downstream.

During much of the annual cycle of rise and fall of Columbia River flow, the phosphorus³² in normal reactor effluent is in greater concentration than desirable. Fish in the river concentrate P^{32} in their bodies, and when eaten by humans, may contribute this radioisotope to the general human burden. In each eight gallons of irradiated potassium tetraborate Ink there was calculated to be about 33 microcuries (μ c) of P^{32} , while in 45 gallons of the boric acid Ink there was calculated to be about 190 μ c (the P^{32} content of the latter was reported to be ten times greater for some reason, perhaps due to an impurity in boric acid). It was estimated⁽¹⁾ that there may be 500 gallons per month of 10% K₄B₂O₇ Ink from each reactor if the Ink system were adopted for production at Hanford. This would be about 0.016 curies of P^{32} discharged to the river each month.

However, several years of experience at Hanford in analyzing reactor cooling water both before and after irradiation has shown that the concentration of P^{32} is always 10,000-15,000 times that which can be calculated from the known amount of phosphorus going into the reactor. Several reasons for this discrepancy are postulated, outstanding of which are hold-up in films in the reactor resulting in longer irradiation time than just the transit time of the cooling water, and a contribution of P^{32} from transmutation of sulfur in the water. (7) Since it is reported that steam condensate might be used to make up the Ink solution, and both disodium phosphate and sodium sulfite are added to Hanford steam plant feed water, (8) there could be an important contribution of P^{32} from these sources. It would be advisable not to use power plant steam condensate similarly treated to make up Ink solutions. Applying the correction factor above it would be expected that about 160 curies of phosphorus³² would be produced from the production Ink systems each month. Since this is an additional large fraction of the average P^{32} already going to the river from reactor effluent water, it is considered very undesirable to discharge production quantities of irradiated waste Ink solution to the Columbia River or to the ground nearby.

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Similar correction factors between calculated and measured activity densities of other radioisotopes in irradiated water have been determined. For these Ink solutions, note that the average in Table II of four runs for calcium⁴⁵ is 160 times the calculated activity density in Table I. It would be necessary to obtain accurate analyses on freshly irradiated Ink solutions for all of the radioisotopes in Table I before any thorough evaluation of production disposal is possible.

Consideration must be given to the effect of boron from waste Ink discharged to the river or the ground nearby on downstream reactors. Liquids, such as reactor effluent, discharged into midstream of the Columbia River or overflowing through the spillway at the shoreline are known to channel from certain reactors to the water intakes of other

reactor areas downstream.⁽⁹⁾ Liquids discharged into the ground near the reactors percolate down into the ground water, which then seeps into the river at the shoreline during most of the year (except at high river stages). In any of these three cases, there can be higher concentrations at water intakes downstream than would be calculated from gross dilution ratios of waste stream and river flow.

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CONCLUSIONS

The disposal of irradiated waste Ink solution either into the Columb River or the ground nearby in volumes resulting from production usage is not considered advisable on the basis of present knowledge. The concentrations of several radioisotopes would be greater than the maximum permissible concentrations for drinking by humans if discharged promptly after irradiation. Hold-up for decay to MPC would require several months. The amount of phosphorus³² discharged would be an additional large fraction of that already entering the river from reactor effluent, which is considered more than the desirable amount during much of the annual river cycle. Further study would have to be made of the chemical toxicity of Ink solutions on aquatic organisms, on plants, animals and humans using the river water, and of possible "poisoning" effects of the boron on downstream nuclear reactors.

It was considered acceptable to dispose of the small volumes of waste Ink solution from the experiments to an underground crib in the 100-DR Area. It would probably be acceptable to discharge production volumes into the plateau several miles from the river in the Hanford project. However, since calculated activity densities of radioisotopes in solutions irradiated in Hanford reactors are known to be greatly different from the actual measured concentrations, accurate radiochemical analyses of freshly irradiated Ink solutions are necessary before there can be further consideration of disposal.

H. V. Cluney

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