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EFFECTS OF RADIOLYSIS OF B PLANT
EXTRACTION COLUMN RAFFINATES

JANUARY 1, 1971

PREPARED FOR
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EXTRACTION COLUMN RAFFINATES

By

Wallace W. Schulz*

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EFFECTS OF RADIOLYSIS OF B PLANT
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Wallace W. Schulz

INTRODUCTION

In B Plant solvent extraction operations with Purex Acid Waste (PAW) and Zirflex Acid Waste (ZAW) solutions citric acid is added to the extraction column feed to provide pH control and to suppress extraction of unwanted cations. Similarly, hydroxyacetic acid (HAcOH) and HEDTA (N-hydroxy-ethylethylenediaminetriacetic acid) are added to the scrub column to provide, respectively, pH control and additional decontamination of the extracted strontium from sodium and trace amounts of other cations. Various other organic acids, or their sodium salts; *e.g.*, acetic acid, sodium tartrate, Na₃NTA (trisodiumnitrilotriacetate), etc., have been suggested and/or tested for use as buffering and complexing agents in solvent extraction of strontium from PAW and ZAW solutions.

The organic buffering and complexing agents report to the extraction column aqueous raffinate. Normally, this waste solution is adjusted to pH >8 with 19M NaOH and routed to underground storage tanks. In some cases, however, rework of the raffinate may be necessary to recover high strontium losses. It was postulated that radiolysis of organic compounds in the raffinate could adversely affect rework operations.

Various synthetic raffinates were irradiated (⁶⁰Co) to determine the nature and magnitude of any radiolytic effects and their consequences in rework of the raffinate. Extraction tests were also performed with a raffinate solution prepared from actual plant PAW solution. Results of these studies are summarized in this report.

SUMMARY AND CONCLUSIONS

B Plant extraction column raffinate solutions prepared from reagent grade chemicals and containing various organic complexing agents were irradiated (^{60}Co) to absorbed doses in the range 110 to 840 watt hr/liter. Extraction column raffinate (citrate-complexed) was also prepared from actual plant PAW and allowed to stand three weeks until the absorbed radioactive decay energy (from the radionuclides in the PAW) amounted to about 200 watt hr/liter. Compositions of these raffinates were selected to simulate those expected if none of the strontium and rare earths were removed in extraction column operation. Physical and chemical properties of the irradiated raffinates were measured to devise feed adjustment procedures to permit satisfactory extraction column rework for recovery of strontium and rare earths.

The pH of all raffinates increased from 2 to 3.6 pH units after irradiation to about 800 watt hr/liter; pH changes were proportionately smaller at lower exposure levels. Solids did not precipitate from raffinates containing either citrate or HEDTA at any of the irradiation doses tested; however, brown solids, tentatively identified as hydrated iron(III) oxide, precipitated from NTA-complexed raffinate when irradiated to 840 watt hr/liter.

For solvent extraction rework purposes a satisfactory feed adjustment procedure is to add *both* HNO_3 and the appropriate complexing agent to the irradiated raffinate. Nitric acid serves to adjust the pH of the raffinate to approximately its original value. Buffering and complexing capacities lost by radiolysis are restored by addition of the appropriate complexing agent. Over a wide range of complexants and irradiation doses this feed adjustment procedure provides material suitable for the recovery of strontium and rare earths at

controlled pH with only minimal extraction of undesired components.

EXPERIMENTAL

MATERIALS

Raffinate Solutions

The composition of the raffinates (Solutions 1-4) irradiated in the ^{60}Co facility are listed in Table I. The gross salt composition of the NTA-complexed raffinate (Solution 3) corresponded very closely to that listed in earlier (1967) B Plant Phase III flowsheets.¹ Except for the substitution of citric acid for HEDTA, the composition of the synthetic ZAW raffinate (Solution 4) was also about the same as that listed in the 1967 flowsheets. The composition of the other raffinates irradiated in the ^{60}Co facility (Solutions 1 and 2) was essentially the same as that of the NTA-complexed solution except for appropriate changes in pH and complexant. Raffinate pH's were adjusted to conform to probable values for extraction column operations with feeds containing the different complexants. Strontium, calcium, and rare earth concentrations of Solutions 1-4 were adjusted to correspond to concentrations that would be present if little or none of these elements extracted in the extraction column.

Extraction column raffinate (Solution 5) was also prepared from current (1970) PAW by addition of appropriate amounts of citric acid, HAcOH , Na_3HEDTA , and NaOH . Concentrations of certain of the components of the resulting solution were measured and are listed in Table I. The raffinate prepared from plant PAW contained about 10-fold more aluminum than the citrate-complexed raffinate prepared from nonradioactive

¹D. E. Larson. Private Communication, Isochem Inc., Richland, Washington, 1967.

TABLE I
COMPOSITION OF EXTRACTION COLUMN RAFFINATES

Component or Property	Composition, M				
	Irradiated in ^{60}Co Facility				
	Solution 1	Solution 2	Solution 3	Solution 4	Solution 5 (a)
Na	1.53	1.55	1.66	2.4	1.7 (b)
Fe	0.16	0.16	0.16	0.10	0.13 (b)
Al	0.015	0.015	0.015	0.31	0.12 (b)
Zr	-	-	-	0.034	(c)
Cr	0.012	0.012	0.012	0.0055	(c)
Ni	0.0056	0.0056	0.0056	0.0027	(c)
La	0.002	0.002	0.002	0.00025	(c)
Ce	0.0012	0.0012	0.0012	0.00013	(c)
Ca	0.0005	0.0005	0.0005	0.0006	(c)
Sr	0.0005	0.0005	0.0005	0.001	(c)
NO_3	0.29	0.42	0.42	0.63	(c)
SO_4	0.31	0.31	0.31	0.18	(c)
F	0.0023	0.0023	0.0023	0.10	(c)
PO_4	-	-	-	0.0041	(c)
AcOH	0.28	0.28	0.28	0.25	0.19 (b)
Citrate	0.22	-	-	0.52	0.27 (b)
Tartrate	0.021	0.021	0.021	0.025	-
HEDTA	-	0.22	-	-	0.001 (b)
NTA	-	-	0.22	-	-
pH	4.7	3.9	4.45	4.4	4.8
Density, g/ml	1.09	1.09	1.09	1.14	(c)

(a) Prepared from actual plant PAW.

(b) Analyzed or estimated from make-up.

(c) Not determined.

chemicals (Solution 1); otherwise the composition of the two solutions was about the same. Radiochemical composition of Solution 5, although measured, is specifically not quoted in Table I to avoid classification problems.

Other Chemicals

The chemical forms and sources of ^{85}Sr , ^{59}Fe , and ^{144}Ce are listed elsewhere.²

Stock 11.5M HAcOH and 1.57M Na_3HEDTA solutions were obtained from the du Pont and Dow companies, respectively. A stock 1.67M Na_3NTA solution was prepared from the solid salt supplied by the Geigy Chemical Corporation. Source and purity of the HDEHP, TBP, and NPH used to prepare extractant for the distribution ratio tests have been given earlier.²

All other chemicals used were of reagent grade quality.

PROCEDURES

Irradiation Equipment and Procedures

Synthetic raffinate solutions (200 ml), contained in a stoppered and jacketed 500-ml Erlenmeyer flask, were irradiated by ^{60}Co sources positioned under water. The flask and a magnetic stirrer were contained in an aluminum canister (diving bell) fitted with a threaded top and two side arms. Twenty-foot lengths of 3/4-inch OD Tygon tubing were attached to the side arms. These pieces of tubing enclosed (a) a thermocouple wire, (b) the power supply cord to the magnetic stirrer, and (c) 1/8-inch OD Tygon tubing to carry cooling water to the jacketed flask from a circulating water bath. The temperature of the raffinate solution was controlled at $25 \pm 1^\circ\text{C}$ in these irradiation studies.

The diving bell assembly was lowered into close proximity to the ^{60}Co sources by means of a cable attached to a handle in the threaded top. A specially constructed lead and steel

²W. W. Schulz. *D2EHPA Extraction Recovery of Neptunium and Plutonium from Purex Acid Solutions*, BNWL-583, March 1968.

support bracket was carefully placed next to the ^{60}Co sources to ensure reproducible positioning of the diving bell.

Ceric sulfate dosimetry³ was used to determine periodically the radiation dose emitted by the radiation source under identical conditions used to irradiate the raffinate solutions. Two hundred-ml portions of 0.021M $\text{Ce}(\text{SO}_4)_2$ in 0.8N H_2SO_4 were irradiated 60 to 120 minutes at $25 \pm 1^\circ\text{C}$ while stirring continuously. The concentration of Ce(IV) in the dosimeter solution before and after irradiation was determined by duplicate titrations with a standardized Fe(II) solution.

Absorbed energy doses received by the raffinate solutions irradiated in the ^{60}Co facility were calculated in units of watt hr/liter by the following method. The dose rate in rads/hr was multiplied by the exposure time in hours to give the absorbed dose in rads. Multiplication by the conversion factor 2.78×10^{-9} then yielded the dose in units of watt hr/gram. The latter dose was converted to units of watt hr/liter by multiplication of the density expressed in grams/liter. Calculation methods shown in Reference 3 were used to translate changes in Ce(IV) concentration to absorbed dose rates in units of rads/hr. At the time these experiments were performed the dose rate in the ^{60}Co facility ranged from 1.5 to 1.66 megarads/hr.

Values of the *beta* decay energy, in units of watts/kCi, of various radioisotopes have been published by Van Tuyl.⁴ These values together with concentrations of corresponding radioisotopes were used to estimate the energy absorbed (watts/liter) by the raffinate prepared from actual plant PAW. Beta

³ S. D. Reeder, J. A. Dunbar, and N. P. Alloy. *Chemical Dosimetry at the MTR*, IDO-16490, December 15, 1958.

⁴ H. H. Van Tuyl. Personal Communication, General Electric Company, Richland, Washington, December 1962.

decay energy contributions for ^{90}Sr , ^{90}Y , ^{103}Ru , ^{106}Rh , ^{106}Rh , ^{95}Zr , ^{95}Nb , ^{134}Cs , ^{137}Cs , ^{144}Ce , and ^{144}Pr were summed; energy contributions of other radioisotopes were neglected. The test raffinate was contained in a 2.8-cm diameter glass vessel; the assumption that the test solution absorbed 100 percent of the beta decay energy and none of the gamma decay energy does not appear unreasonable for this geometry. Raffinate contained in plant-size tanks absorbs some gamma decay energy also.

Distribution Ratio Measurements

A standard 10-minute contact (mechanical stirring) at 25 °C was used to determine distribution ratios of iron, strontium, and cerium between raffinate solutions and an extractant of the composition 0.19M HDEHP-0.11M NaDEHP-0.2M TBP-NPH. Prior to contact with extractant, 15.6M HNO_3 was added to irradiated raffinates to adjust pH's to desired values. In some cases also, additional HAcOH and/or complexant were added to irradiated raffinates before extraction.

After addition of HNO_3 , HAcOH and/or complexant, raffinates irradiated in the ^{60}Co facility were spiked with ^{59}Fe , ^{85}Sr , or ^{144}Ce and allowed to stand 24 to 48 hours prior to distribution ratio tests. In this case conventional gamma energy pulse height analysis methods were used to determine concentration of radionuclides in aqueous and organic phases.

The organic phase obtained from the extraction contact with raffinate prepared from plant PAW was contacted with a double volume of 2M HNO_3 . Cerium-144 and $^{89+90}\text{Sr}$ concentrations of the resulting HNO_3 solution and also of the aqueous phase from the original extraction contact were measured by standard methods. Atomic absorption procedures were used to determine iron in the raffinate before and after contact with the HDEHP extractant.

RESULTS AND DISCUSSION

PROPERTIES OF IRRADIATED RAFFINATE SOLUTIONS

Tables II and III list irradiation doses, pH data, and solvent extraction results for the various irradiated raffinates. These properties and their significance to stability and solvent extraction rework of raffinate solutions are discussed in the sections that follow.

Irradiation Doses

The raffinate prepared from plant PAW solution was allowed to stand a total of three weeks; beta decay energy absorbed during this time was estimated to amount to 214 watt hr/liter (Table II). The other raffinates were irradiated in the ^{60}Co facility to exposures in the range 110 to 840 watt hr/liter corresponding roughly to the amounts of energy plant-produced raffinates would absorb after standing one to eight weeks, or even longer in the case of material derived from the ZAW solution.

It is recognized, of course, that in actual plant operation solvent extraction rework, if necessary, would likely be performed within one to two weeks of the time the extraction column raffinate was produced. The range of radiation doses used in this work was deliberately chosen wide enough to encompass those which might accrue if plant rework operations were delayed for a prolonged period.

pH Changes

The pH of all raffinate solutions increased upon irradiation. Hydroxyl radicals are a primary product of the radiolysis of water. Coupled with partial destruction of organic buffering agents, radiolytically generated hydroxyl ions increase the pH of the raffinate.

The pH of the raffinates containing NTA and HEDTA increased steadily with absorbed dose; this was also the case for the ZAW raffinate (Solution 4). Conversely, the pH of citrate- and tartrate-complexed raffinates (from PAW feed) increased sharply after only moderate exposure and then increased gradually upon further irradiation.

Precipitation of Solids

Indicative of the stability of the organic complexing and buffering agents toward radiation was the observation that all the irradiated raffinates save one were free of solids. Brown solids, presumably hydrated iron(III) oxide, precipitated from the NTA-complexed raffinate after irradiation to 836 watt hr/liter. Irradiation to this exposure without precipitation of solids was possible when additional Na_3NTA was added about midway through the irradiation period.

Solvent Extraction Rework-Feed Adjustment Procedures

Various procedures were investigated for preparing extraction column feedstock from irradiated raffinate solutions. Availability of such procedures is a prerequisite to successful rework of raffinates for removal of strontium and rare earth values.

A satisfactory feed adjustment procedure is to add *both* HNO_3 and the appropriate (*i.e.*, citrate, HEDTA, or NTA) complexing agent to the irradiated raffinate. Nitric acid serves to adjust the pH of the raffinate to approximately its original value while buffering and complexing capacities lost by radiolysis are restored by addition of complexing agent. Data in Tables II and III testify to the utility of this feed adjustment procedure. Over a wide range of complexants and irradiation doses it provides feed suitable for extraction recovery of strontium and rare earths at controlled pH with only minimal extraction of undesired components.

The optimum amount of complexant to add to the irradiated raffinate in this feed adjustment procedure to achieve best overall solvent extraction behavior was not determined. This optimum concentration varies with the type of complexant and the exposure level of the raffinate. In our work we simply added complexant to an arbitrary concentration of 0.05 to 0.1M over that present in the irradiated raffinate. At both these concentrations solvent extraction results were generally satisfactory for all the irradiated HEDTA- and NTA-complexed solutions. However, results with irradiated citrate-complexed raffinates indicate that addition of as much as 0.1M citrate to such solutions adversely affects cerium extraction. Thus, for extraction rework of such raffinates it is recommended that the added citrate concentration be limited to 0.05M.

A simpler, but less effective, feed adjustment procedure is to add *only* HNO₃ to the irradiated raffinate. Addition of HNO₃ serves to adjust the pH of the irradiated raffinate to its original value but fails to restore buffering and complexing capacity lost by radiolysis. Lack of such capacity is evidenced by subsequent solvent extraction operations in poor pH control and increased extraction of iron and other unwanted constituents (Tables II and III). In some cases also, rare earth extraction suffers when additional complexing agent is not added to the irradiated raffinate. Significantly, however, HDEHP extraction of strontium from irradiated extraction column raffinate is not greatly influenced either by 0.05 to 0.1M additional complexing agent nor by poor pH control (at least in the pH range 4.5 to about 5.5) during solvent extraction operations. This latter behavior is in accord with the known⁵ broad maximum in the pH - E_a^O curves for Sr-HDEHP systems

⁵W. W. Schulz. *Solvent Extraction of Strontium, Cerium, and Rare Earths with D2EHPA Part 1: Laboratory Studies*, HW-79762 PT 1, February 1964.

TABLE II
SOLVENT EXTRACTION RESULTS FOR IRRADIATED RAFFINATES

Irradiated Raffinate Solution		Dose Watt hr/ℓ	pH	Distribution Ratios									
				After Adjust pH ^a				After Adjust pH and Complexant ^b					
								HAcOH	Complexant ^c	Sr	Ce	Fe	pH ^d
No.			Sr	Ce	Fe	pH ^d	M	M	Sr	Ce	Fe	pH ^d	
1 (citrate)	0	4.7	17.4	14.0	0.0129	4.7	-	-	-	-	-	-	
	110.	6.2	19.6	10.0	0.0213	5.1	NA ^e	0.05	16.3	8.25	0.0118	4.5	
	239.	6.4	18.9	23.6	0.0474	5.5	NA	0.05	14.0	5.43	0.0179	4.6	
	473.	6.9	17.0	24.3	0.102	5.6	NA	0.05	14.1	9.74	0.0337	4.65	
	825.	6.8	16.3	42.7	0.195	5.7	NA	0.05	13.0	12.3	0.0706	4.85	
							NA	0.10	-	4.80	0.0307	5.0	
							0.10	NA	-	-	0.204	5.4	
							0.05	0.05	-	13.9	0.0682	5.2	
5 (citrate- plant)	0	4.7	7.0	5.85	-	4.7	-	-	-	-	-	-	
	71.4	6.3	-	-	-	-	NA	0.10	5.70	2.70	<0.1 ^f	4.4	
	143.	6.6	-	-	-	-	NA	0.10	3.48	-	0.15 ^f	4.9	
	214.	6.7	-	-	-	-	NA	0.10	4.72	1.19	0.29 ^f	4.5	
2 (HEDTA)	0	3.8	35.9	322.	0.0448	4.1	-	-	-	-	-	-	
	119.	5.0	-	-	-	-	0.05	NA	22.9	72.0	0.190	4.5	
	235.	5.3	19.1	26.5	0.360	4.5	NA	0.10	28.2	179.	0.0008	4.2	
	469.	5.9	16.2	20.6	0.404	4.5	NA	0.10	21.4	50.0	0.0086	4.4	
	823.	7.5	14.1	5.14	0.491	4.5	NA	0.10	19.3	54.7	0.120	4.5	
							NA	0.05	-	15.2	0.296	4.5	
							0.11	NA	-	19.9	0.745	4.5	
							0.05	0.05	-	28.1	0.346	4.5	
							0.10	0.05	-	50.3	0.375	4.4	

^a After add 15.6M HNO₃ to within ±0.1 pH of unirradiated raffinate.

^b After adjust pH to within ±0.1 pH of unirradiated raffinate and after addition of HAcOH and/or complexant to concentration indicated.

^c Citrate for Solutions 1 and 5, HEDTA for Solution 2.

^d After extraction contact.

^e NA = not added.

^f Estimated from analyses of aqueous phase before and after extraction contact.

TABLE III
SOLVENT EXTRACTION RESULTS FOR IRRADIATED RAFFINATES

Irradiated Raffinate Solution		Dose Watt hr/l	Distribution Ratios									
			pH	After Adjust pH ^a				After Adjust pH and Complexant ^b				
No.		Sr		Ce	Fe	pH ^d	HAcOH M	Complexant ^c M	Sr	Ce	Fe	pH ^d
3 (NTA)	0	4.5	8.86	320.	0.610	4.5	-	-	-	-	-	-
	121.	5.0	7.89	36.2	0.720	4.6	NA ^e	0.10	11.6	4.81	0.476	4.6
	239.	5.2	8.04	7.33	0.819	4.7	NA	0.05	-	22.9	0.639	4.7
							NA	0.10	11.8	58.4	0.521	4.7
	477.	5.7	8.23	12.2	0.927	4.85	NA	0.05	-	48.9	0.782	4.9
							NA	0.10	8.78	55.3	0.636	4.9
							0.05	0.05	-	46.8	0.727	4.8
							0.05	0.10	-	63.9	0.645	4.8
							0.10	NA	-	23.3	0.792	4.9
	836.7	7.1	5.65	46.4	1.0	5.0	-	-	-	-	-	-
4 (citrate- ZAW)	0	4.4	-	13.3	0.0946	4.6	-	-	-	-	-	-
	114.	4.9	1.37	2.74	0.0935	4.8	NA	0.05	3.33	10.0	0.082	4.5
	228.	5.3	2.13	4.67	0.113	5.0	NA	0.05	-	7.76	-	4.6
	456.	5.7	3.68	-	0.136	5.3	NA	0.05	3.51	6.12	0.101	4.8
	797.	6.4	2.88	2.58	0.162	5.5	NA	0.05	3.69	4.09	0.101	4.85

^a After add 15.6M HNO₃ to within 0.1 pH of unirradiated raffinate.

^b After adjust pH to within 0.1 pH of unirradiated raffinate and after addition of HAcOH and/or complexant to concentration indicated.

^c NTA for Solution 3, citrate for Solution 4.

^d After extraction contact.

^e NA = not added.

^f Solution made 0.1M Na₃NTA (in excess of that originally present) after irradiation to about 480 watt hr/liter.

and with the relatively small values of formation constants of Sr-citrate, etc., complexes.

In a few instances both HAcOH and complexant (Na_3HEDTA , Na_3NTA , or Na_3Cit) were added to the acidified (HNO_3) irradiated raffinate. In other tests only HAcOH was added to the acidified raffinate. Hydroxyacetic acid forms weaker complexes with Fe(III) , Cr(III) , etc., than do citrate, HEDTA, or NTA, and no advantage is gained by its addition in the feed adjustment procedure.

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