Stripping of plated-out activities by various reagents - a preliminary report

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In the Sudbury water meeting of October 94, results of an activity plate-out experiment were reported (SNO-STR-94-038). Following the meeting a study of the stripping of plated out Th activities was carried out. Such stripping is to simulate both off line and on line cleaning of the D_2O system component materials.

Experimental setup:

Two sets of experiments were conducted:
1) Coupons used in the (SNO-STR-94-038) plating experiment were placed in various solutions and the residual activity measured. These included non passivated Hastelloy, PP and acrylic.
2) New coupons: Hastelloy coupons were sent for passivation at Dalpro, Stoney Creek, Ont. Other coupon materials included PP, acrylic and Filmtec membrane material (composite of Polysulfone and polyamide). Experiments are now being conducted on Filmtec water channeling spacer material. Coupons of 27x47 square mm, were cut. As in 1), PP, Hastelloy and acrylic coupons were precleaned with 1:20 Radiacwash solution. All coupons were then cleaned with 10% HNO_3 and UPW. Plating out of coupons was performed by immersing half of the coupon area (15 square cm) for 24 hours in $\approx$. 300 Bq., pH 6.8, 200 cc aqueous solution. The coupons were counted, then immersed in the various stripping solutions and residual activity measured.

Stripping solutions included: 100 ppb EDTA, 100 ppm EDTA, 1:20 Radiacwash, 10% HNO_3, 5% HCl.
-100 ppb EDTA was chosen since SNO is expected to use 1-10 ppb EDTA as an additive.
-Radiacwash 1:20 and HNO_3 10% are the standard cleaning solutions employed at CRPP/SNO and were recommended for many of the SNO cleaning procedures.
-5% HCl is recommended as part of the cleaning procedures for R/O membranes.
-Diluted NaOH (pH 12) is also a recommended cleaning agent for R/O membranes employed at CRPP and is to be tested.

Stripping was conducted by placing half of the coupon in 30 cc of stripping solution and agitating for 24 hours on an orbital shaker at 160 RPM. All experiments were carried out in duplicate. Counting was on an Ortec alpha counter at 2.3-2.4 MeV (^{228}Th) for about 1 hr., counted area of approx. 1.5 square cm.

Results:

Tables 1 and 2 include the results for the above experiments. The activities

were converted to fraction of initial activity, i.e. fraction of the initial activity plated out and before any stripping treatment is employed. Numbers are averages on two duplicate coupons and statistical variation. It should be noted that all of the above tests were done at 20 C and the effects of the cleaning reagents may be lower at 10 C.

It is evident that the 100 ppb EDTA has only a minor effect on the plated out activity. Thus while previous experiments indicate EDTA to significantly reduce plateout of Pb and Th from solution (e.g. SNO-STR-92-092,093) it does not seem to be effective at stripping pre-plated activity.

Table 1 indicates that a single Radiacwash treatment reduced plate out on PP and acrylic by 4-6. In a second experiment (table 2) Radiacwash 1:20 reduced the plateout by x8 for PP and acrylic. Further application of Radiacwash reduced the plated out activity to <10% of the initial value, proving it to be an effective initial off-line cleaning agent. However, Radiacwash is not recommended for use with membranes on line where removal of detergent residues may be impossible. A single treatment with HCl 5%, which is a recommended agent for cleanup of polyamide membranes, reduced the activity on the membrane coupons by >X15. HCl reduced the plateout on PP in the 2nd experiment to <1% of the original value, but was less effective in the 1st study (7% of the initial activity, and not significantly better than the Radiacwash treatment). A single test with Acrylic proved HCl to be highly effective, removing all detectable activity off the coupons (table 1).

The initial activity plated on the hastelloy coupons was an order of magnitude higher than on other materials while that for membranes was the lowest. Furthermore, the Hastelloy samples seemed to clean to a lesser degree than the polymer materials in both Radiacwash and acid.

In the EDTA case, repeated use of a given agent yields a smaller improvement than a one time use indicating that 24 hrs at 160 rpm was usually sufficient for cleaning the coupons and further cleaning necessitates a different cleaning compound. Conversely, a repeated use of the same cleaning agent was beneficial both in the case of the Hastelloy cleaning by HCl (table 1) and the Acrylic cleaning by Radiacwash (table 2).

Further studies:

1) Passivated Hastelloy coupons are to be further treated with HCl 5% and if necessary also with NaOH (pH 12).

2) Stripping of activity off the permeate channeling spacer is to be studied.

3) Surface areas of the various materials in the D_2O system will be estimated and the desired 'cleaning efficiency' defined and compared with the above measured values.

Table 1

Fraction of plated out activities

	0	EDTA 100 ppb		EDTA 100ppm	Radiac 1:20		HCl 5%		HNO3 10%
		1*	2		1	2	1	2	
Hast.	1	0.71 (0.02)	0.66 (0.02)	0.55 (0.03)	0.27 (0.05)	0.20 (0.04)	0.23 (0.07)	0.007 (0.001)	
PP	1	0.83 (0.03)	0.68 (0.09)	0.61 (0.05)	0.15 (0.02)	0.09 (0.01)	0.07 (0.01)	-	0.061 (0.09)
Acry.	1	0.56 (0.02)	0.57 (0.09)	0.47 (0.02)	0.26 (0.12)	0.08 (0.02)	<LOD		

1,2 refer to number of treatments with a particular reagent.

LOD: 0.0002cps

0 : Initial plated out activity before applying stripping reagents
The absolute initial activities for the various coupons (in duplicates) were:

Hast : 1.1808 cps, 1.3538 cps

PP : 0.1019 cps, 0.0764 cps

Acrylic: 0.0149 cps, 0.1065 cps

*1st EDTA treatment was 48hrs in 100 ppb , no shaking
Other treatments were 24 hours, 30 cc solutions, 160 RPM shaker

Table 2

Fraction of plated out activities

	0	EDTA 100 ppb	Radiac 1:20 1	Radiac 1:20 2	HCl 5% 1	HCl 5% 2
passivated Hast.	1	0.84 (0.02)	0.36 (0.01)			
PP	1		0.12 (0.02)		0.008 (0.005)	
Acrylic	1		0.12 (0.07)	0.03 (0.01)		
Membrane	1	1.04 (0.05)			0.06 (0.03)	

LOD: 0.0002cps

1,2 indicate repeated treatments with the same reagent

Initial plated out activities (0) for the various duplicate coupons were:

Passivated Hast :1.3841 cps, 0.7638 cps

PP :0.1268 cps, 0.0599 cps

Acrylic :0.0415 cps, 0.0444 cps

Membrane :0.0246 cps, 0.0322 cps