Mercury Recovery Results of Microwave Digested Tritium Facility Pump Oil

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DISCLAIMER

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INTRODUCTION

This report is a follow up of work done earlier this year and recorded in document WSRC-RP-97-322. The scope of this document is to demonstrated the viability of digesting two non-radioactive Tritium facility pump oils, Welch Duoseal and Spindura, neat and spiked with low-level mercury to determine completeness of digestion and recoverability of mercury. As noted in document WSRC-RP-97-322 a microwave digestion methodology was developed with CEM's ultimate digestion vessel system (UDV) and is the technique used for the follow up task of digesting the above mention pump oils for the preparatory step of cold-vapor mercury analysis.

All analytical development for this project was performed at TNX. The determination of the mercury concentration in each digested sample was by cold vapor atomic absorption. The instrument used was a Varian SpectrAA 800 with a vapor generation attachment. This flameless AA procedure is a physical method based on the absorption of radiation at 253.7 nm of mercury vapor. Organo-mercury compounds will not respond to the cold vapor atomic absorption technique, therefore, to acquire a total mercury value it is necessary for a complete digestion to oxidize and convert the organo-mercury species to the mercuric ion.

Method Development

CEM Ultimate Digestion Vessels (UDV) (200 °C, 1000 PSI)

The digestion of the Tritium facility pump oils was performed with a MDS 2000 (650 Watt) microwave digestion system and the UDV. A 0.5 gram oil sample was weighed into the UDV and 20 mL of nitric acid subsequently added. The vessel was sealed and placed in the microwave oven. The UDV allows the reaction to react at a maximum temperature of 200 °C and a maximum pressure of 1000 PSI. It was determined in the earlier work that a maximum pressure of 580 PSI was sufficient for complete digestion of oil matrix material. These same parameters were used in this follow up work. The temperature was not recorded due to a probe malfuction, however, the digestion process is controlled by the pressure limits rather than the temperature.

After completion of the microwave digestion process, a minimum of two hours cooling period is required prior to opening the vessels to allow the pressure and temperature to drop to ambient conditions. Upon opening the vessel the dark green digestion solution was diluted to a final volume of 100 mL. The dissolution remained clear with no signs of precipitation, which is evidence of complete digestion of the organic material. A summary of the microwave oven power, pressure, and timing sequencing are shown in table 1 below.

Stage	1	2	3	4	5
Power	70%	70%	70%	70%	70%
Pressure (PSI)	35	100	200	340	580
Ramp Time	5:00	5:00	4:00	4:00	4:00
Hold Time	1:00	1:00	1:00	5:00	7:00

Table 1 Summary of the Ultimate Digestion Vessel program

A range of mercury oil standards (10 μg/g to 1000 μg/g) were spiked into the different oil matrix materials. After the digestion process they were analyzed and demonstrated acceptable mercury recoveries. A summary of the results are shown in table 2 and table 3. In test number 1 and 2, the low recoveries are believe to be the result of incomplete digestion. These digestion solutions were a pale yellow color prior to diluting up to 100 mL rather than the dark green color. To insure complete digestion of the oil matrix material an adjustment in the timing sequence was made by increasing the hold time at the higher pressures. In test number 3, after the timing sequence adjustment, the digestion is complete and the mercury recoveries improved considerably. 8 mL of 1000 μg/L Hg standard was added to vessel #11 and 20 mL of nitric acid. 8 mL of 1000 μg/L Hg standard was added to vessel #12, 0.4772 g of Spindura pump oil, and 20 mL of nitric acid. 8 mL of 1000 μg/L Hg standard was added to vessel #13, 0.0362 g of 1000 μg/g Spex Hg oil, 0.4245 g of Spindura pump oil, and 20 mL of nitric acid. 0.4859 of the 1000 μg/g Spex Hg in oil standard was added to vessel #15 and 20 mL of nitric acid.

Vessel	Approx. µg/g Hg in Oil	Oil Type	Theoretical µg/L	Actual µg/L	% Recovery
TEST#1					
Vessel#1	18.73	Spindura	46.5	46.77	100.6
Vessel#2	78.72	Spindura	36.0	23.70	65.8
Vessel#3	17.50	Welch	8.2	6.22	75.9
Vessel#4	62.20	Welch	57.8	47.83	82.8
Vessel#5	60.64	Conostan 20	57.0	43.21	75.8
TEST#2					
Vessel#6	48.77	Welch	51.6	47.01	91.1
Vessel#7	37.59	Welch	35.0	30.03	85.8
Vessel#8	64.53	Spindura	58.8	54.43	92.6
Vessel#9	64.22	Spindura	64.4	50.30	78.1
Vessel#10	58.03	Conostan 20	53.8	43.41	80.7
TEST#3					
Vessel#11	-	No Oil	40.0	41.41	103.5
Vessel#12	-	Spindura	16.0	16.13	100.8
Vessel#13	78.58	Spindura	44.2	43.32	98.0
Vessel#14	41.94	.0235	47.0	46.76	99.4
Vessel#15	1000	Spex Oil Std.	38.9	37.5	96.5
TEST#4					
Vessel#16	10.72	Spindura	10.4	10.17	97.8
Vessel#17	10.88	Spindura	10.8	10.47	96.9
Vessel#18	52.59	Spindura	51.8	48.94	94.5
Vessel#19	46.42	Spindura	44.2	41.94	94.9
Vessel#20	58.81	Spindura	54.8	52.10	95.1

Table 2 Summary of mercury oil standards using ultimate digestion vessels.

Vessel	Approx. µg/g Hg in Oil	Oil Type	Theoretical µg/L	Actual µg/L	% Recovery
TEST#5					
Vessel#21	44.30	Welch	45.2	44.08	97.5
Vessel#22	13.13	Welch	12.8	12.87	100.5
Vessel#23	18.14	Welch	17.0	16.81	98.9
Vessel#24	15.38	Welch	14.8	14.37	97.1
Vessel#25	14.71	Welch	14.2	13.71	96.5
TEST#6	**Digestion**	**Blanks**			
Vessel#26	0	20 mL $HN0_3$	0	-0.44	
Vessel#27	0	20 mL $HN0_3$	0	-0.51	
Vessel#28	0	20 mL $HN0_3$	0	-0.56	
Vessel#29	0	20 mL $HN0_3$	0	1.43	
Vessel#30	0	20 mL $HN0_3$	0	-0.59	
TEST#7					
Vessel#31	418.1	Welch	39.7	39.87	100.3
Vessel#32	437.0	Spindura	40.3	42.00	104.2
Vessel#33	291.1	Welch	26.3	25.71	97.7
Vessel#34	274.1	Spindura	25.9	25.84	99.6
Vessel#35	170.2	Welch	16.0	15.28	95.5
TEST#8					
Vessel#36	72.44	Welch	35.9	35.41	98.6
Vessel#37	76.17	Spindura	37.1	36.46	98.3
Vessel#38	40.72	Welch	19.8	19.33	97.6
Vessel#39	38.66	Spindura	18.7	18.79	100.5
Vessel#40	24.17	Welch	11.5	11.29	98.2

Table 3 Summary of mercury oil standards using ultimate digestion vessels.

There was no evidence of cross contamination or absorption of mercury by the UDV Teflon liners demonstrated by the results of the nitric acid digestion blanks run between oil digestions and analyzed in the test #6 phase. The UDV results suggest that the microwave digestion methodology reported in this document for the digestion of Tritium facility pump oils is a viable preparatory step for cold-vapor mercury analysis.