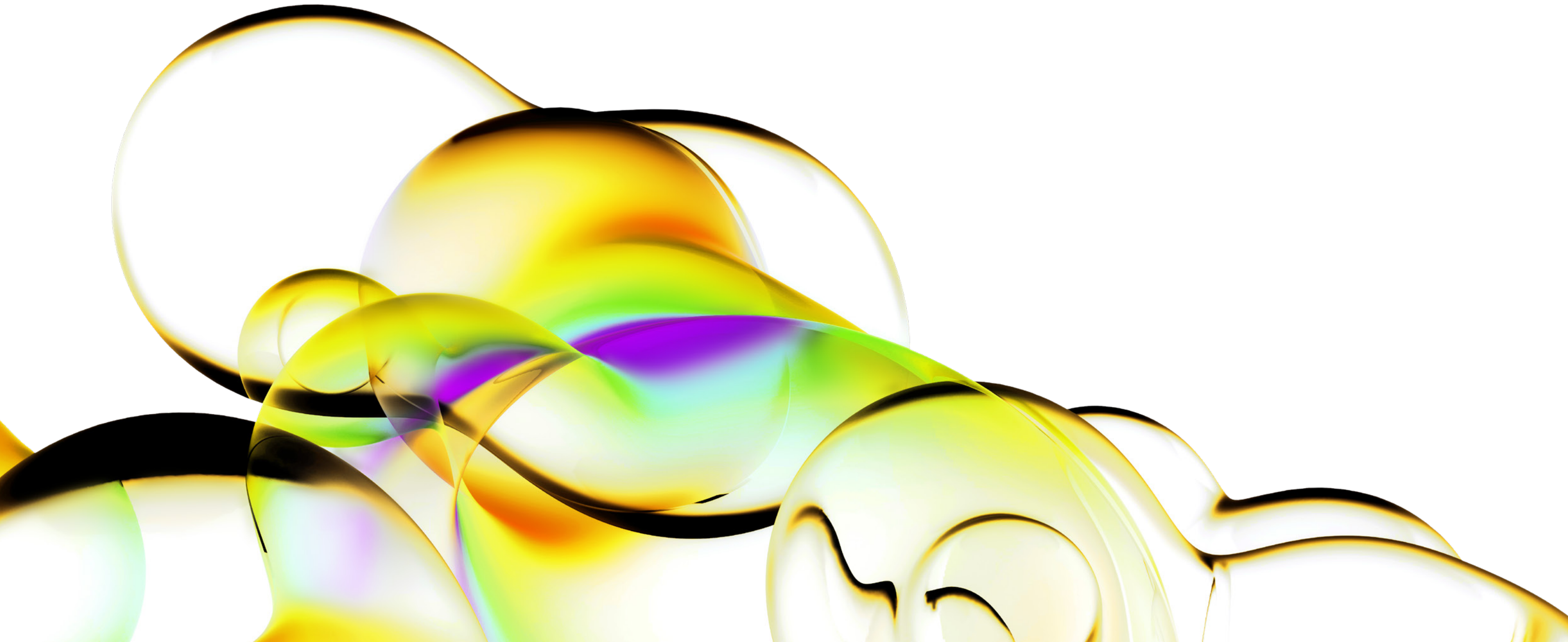
LSC counting solutions: Technical tips.

Counting solutions

The advance of the nuclear industry in all its forms coupled with growing concerns for possible environmental contamination has led to an increased interest in the quantification of radioisotopes in the environment. Radionuclides currently present in the environment originate from a variety of sources such as fallout from nuclear weapons testing, and in discharges from both nuclear and non-nuclear industries. Natural sources and fallout from nuclear devices provide the main input to terrestrial ecosystems, except for certain radionuclides emanating from nuclear installations. In the late 1950's and early 1960's, during and immediately after the period of most frequent aboveground nuclear weapons testing, numerous studies were performed to determine the distribution and movement of fallout radionuclides in air, precipitation, agricultural produce, animals and soils. Around this time the nuclear power industry was developing, and consequently, both the diversity and amount of radioactive species in the environment increased. Currently, the majority of the high level waste from the nuclear industry is stored for ultimate disposal in sites classified as stable, such as deep geological strata. However, as many nuclear facilities are situated in coastal areas, the bulk of low level radioactive waste is discharged to the sea.

Because of both nuclear fallout and discharges from the nuclear industry (including releases from the Chernobyl accident in 1986), certain radionuclides are studied more than others. This is due to either their radio-toxicity, increased presence in the environment or ease of entry into the food chain [1,2,3,4,5,6,7]; a selection of these is presented in Table 1.



The separation and isolation of these radionuclides from the complex sample matrices often encountered is presenting researchers with a myriad of problems; however many of these have been eased by the introduction of novel chromatographic separation technology (Eichrom Industries Inc. Darien, Illinois, USA). By employing this technology, previously difficult and time consuming radionuclide separations are completed more effectively and efficiently. In combination with recent advances in liquid scintillation counting (LSC) technology by Revvity, Inc., it is now possible to consider LSC as an alternative screening tool to alpha spectrometry and gas flow proportional counting.

Table 1: Environmental radionuclides of specific interest.

Radionuclide	Environmental location
^{3}H	Milk, crops, ground water and precipitation
^{14}C	Milk, crops, animals and sea water
^{35}S	Milk, crops, animals and soil/sediments
^{90}Sr	Milk, crops, animals and soil/sediments
$^{134,137}Cs$	Milk, crops, animals and soil/sediments
^{238}Pu, $^{239+240}Pu$, ^{241}Pu, ^{241}Am	Milk, crops, animals and soil/sediments

Chromatographic sample preparation

A range of products that employ the technique of extraction chromatography to efficiently pre-concentrate and separate radionuclides from a variety of different matrices were introduced by Eichrom. Extraction chromatography combines the power and selectivity of solvent extraction with the ease of use of a chromatographic column. Through careful selection of the extractant, which is bound to the resin support, a material is produced which is highly specific for a particular radionuclide or group of radionuclides. In the majority of cases, either retention or elution of the radionuclide of interest is achieved by modification of the concentration of the acidic eluent used. Employing such technology has resulted in an accurate and reproducible sample preparation method for the determination of radionuclides in environmental samples.

Eichrom have published methods for radionuclide separations using their chromatography technology and these can be conveniently explained as follows:

ACW, ACS, ACU	methods refer to Actinides in Water, Soil and Urine
SRW, SRS, SRU	methods refer to Strontium in Water, Soil and Urine
TCW, TCS	methods refer to Technetium in Water and Soil
H3W	methods refer to Tritium (^{3}H) in Water
OTW	methods refer to Other (e.g., Lead in Water)

A summary of these methods together with the recommended Revvity ULTIMA Gold™ LSC cocktail for isotopic determination by LSC is presented in Table 2. ULTIMA Gold cocktails are recommended for these applications due to the use of di-isopropyl naphthalene (DIN) as the solvent base which enhances alpha/beta resolution in LSC.[7]

The information presented in Table 2 shows both the eluent used in the final stripping of the radionuclide from the chromatographic column and the appropriate ULTIMA Gold cocktail which will accommodate either all, or at least a sizeable aliquot of the eluent. Table 2 should be used in conjunction with Table 3 which demonstrates the maximum capacity of each of the ULTIMA Gold cocktails with the each of the eluents. It should be remembered that the use of alpha/beta LSC for alpha determination will only provide a gross alpha measurement and is capable of limited alpha-alpha resolution. An example of such an alpha/beta LSC is the Revvity Tri-Carb® 2770TR/AB which uses Time-Resolved Pulse Decay Analysis (TR-PDA)[7] technology to separate the alpha spectrum from the beta spectrum.

Table 2: Compatibility of Ultima Gold cocktails with eichrom eluents.

Method no.	Analyte	Emitter	Strip volume	Eluent	Recommended ULTIMA gold cocktail
ACS06	$^{234-238}$U ^{234}Th	Alpha, gamma Beta, gamma	15 mL 20 mL	0.02 M HCl 5 M HCl	AB / XR / LLT AB*
ACW01	$^{234-238}$U ^{234}Th	Alpha, gamma Beta, gamma	20 mL 20 mL	0.02 M HCl 6 M HCl	AB / XR / LLT AB* / LLT*
ACW03	$^{234-238}$U ^{241}Pu ^{241}Am	Alpha, gamma Beta, gamma Alpha, gamma	15 mL 10 mL 3+20 mL	0.01 M HCl 0.1 M Ammonium H Oxalate 9 M+4 M HCl	AB / XR / LLT AB / XR AB* / LLT*
ACW06	$^{234-238}$U ^{234}Th	Alpha, gamma Beta, gamma	15 mL 15 mL	0.02 M HCl 5 M HCl	AB / XR / LLT AB*
OTW01	^{210}Pb	Beta, gamma	20 mL	Water	LLT
SRW01	^{89,90}Sr	Beta	10 mL	0.05 M HNO$_3$	AB / XR
SRS01	^{89,90}Sr	Beta	10 mL	0.05 M HNO$_3$	AB / XR
TCS01	^{99}Tc	Beta	0.7g (2 mL)	TEVA Resin	AB / XR / LLT
TCW01	^{99}Tc	Beta	0.7g (2 mL)	TEVA Resin	AB / XR / LLT
SRU01	^{89,90}Sr	Beta	10 mL	0.05 M HNO$_3$	AB / XR
ACW04	^{241}Am	Alpha, gamma	15 mL	2 M HCl	AB* / LLT*
ACU02	$^{234-238}$U ^{241}Pu ^{241}Am	Alpha, gamma Beta, gamma Alpha, gamma	15 mL 15 mL 3+20 mL	0.02 M HCl 3 M HCl-0.25 M Ascorbic Acid # 9 M+4 M HCl	AB / XR / LLT AB / LLT AB* / LLT*
ACW09	^{241}Pu ^{241}Am	Alpha, Beta Alpha, gamma	10 mL 3+20 mL	0.1 M Ammonium H Oxalate 9 M+4 M HCl	AB / XR AB* / LLT*
ACW07	^{241}Pu	Alpha, Beta	10 mL	0.1 M Ammonium H Oxalate	AB / XR
H3W1	Tritiu m	Beta	25 mL	Non-acidified water sample	LLT
—	^{63}Ni	Beta	15 mL	3 M HNO$_3$	AB* / LLT*

* Indicates limited sample uptake capacity (see Table 3 for further details).

Ascorbic acid causes yellowing upon storage (> 2 days)

An alternative method to acidic stripping of the radionuclide from the column is to elute with Iso-propyl alcohol (IPA). IPA effectively strips the resin coating (containing the radionuclide) and this can be counted with 4p geometry in a suitable ULTIMA Gold cocktail. The information in both these tables is presented to help researchers investigate the use of LSC as an alternative technology for gross alpha determination.

Table 3: Sample capacity of ULTIMA Gold cocktails for chromatographic eluents.

Eluent	Strip Volume	ULTIMA Gold AB mL/10 mL @ 20 °C	ULTIMA Gold LLT mL/10 mL @ 20 °C	ULTIMA Gold XR mL/10 m @ 20 °C
0.01 M Hydrochloric Acid	15 mL	10.0 mL	8.0 mL	10.0 mL
0.02 M Hydrochloric Acid	15-20 mL	9.0 mL	7.0 mL	10.0 mL
2.0 M Hydrochloric Acid	15 mL	3.5 mL	3.5 mL	1.0 mL
5.0 M Hydrochloric Acid	15 mL	2.0 mL	1.5 mL	< 0.5 mL
6.0 M Hydrochloric Acid	20 mL	1.0 mL	1.5 mL	< 0.5 mL
4.65 M Hydrochloric Acid 9M + 4M mixture20 mL	3+20 mL	1.5 mL	2.0 mL	< 0.5 mL
9.0 M Hydrochloric Acid (concentrated HCl 1.16 S.G.)	20 mL	1.0 mL	1.0 mL	< 0.25 mL
3M HCl / 0.25M Ascorbic Acid	15 mL	2.0 mL	2.0 mL	0.5 mL
0.05M Nitric Acid	10 mL	8.0 mL	7.0 mL	9.0 mL
3.0M Nitric Acid	15 mL	2.0 mL	2.25 mL	1.0 mL
0.02M HNO_3 / 0.02M HF	10 mL	8.0 mL	10.0 mL	10.0 mL
0.1M Ammonium H Oxalate	10 mL	8.0 mL	6.0 mL	9.0 mL
Water	25 mL	10.0 mL	10.0 mL	10.0 mL

Aqueous sample preparation

Many of the radioactive species of interest to low level researchers are present in an aqueous medium, usually water. Therefore, any suitable LSC cocktail must not only have a high capacity for water but also be compatible with water from a variety of different sources. These include distilled, deionized, tap, rain, river and even sea water. In addition to high sample capacity, other preferred requirements for liquid scintillation counting include a very low background contribution and high counting efficiency. Ideally the LSC cocktail should also be based on the high flash point, safer solvent DIN. ULTIMA Gold LLT is such a cocktail and is primarily designed for the low level tritium (LLT) monitoring and research sectors. Additionally, ULTIMA Gold LLT has other unique performance characteristics that set it apart from currently available cocktails. ULTIMA Gold LLT meets additional requirements for a low level counting cocktail such as long term stabilityand sub-ambient temperature stability, and it can accept the important mineral acid species normally encountered in alpha/beta counting applications. A selection of the more important properties of ULTIMA Gold LLT are illustrated in Tables 4 and 5. from either nuclear devices or the nuclear industry are summarized in Table 6.

Conclusion

Environmental sample preparation encompasses a wide variety of techniques, including extraction chromatography, acid extraction, ashing and solvent extraction, and chemical separation. Advances in liquid scintillation technology, together with new and emerging sample preparation techniques, now enable researchers to consider LSC as an alternative environmental sample radionuclide counting method, or as a potentially useful screening tool.

Element	Radionuclide	Half-life	Type of emission	Energy (MeV)	Sources
Hydrogen	^{3}H	12.3 y	Beta	0.02	Fallout, nuclear industry
Carbon	^{14}C	5730 y	Beta	0.16	Fallout, nuclear industry
Phosphorus	^{32}P	14.3 d	Beta	1.71	Fallout, nuclear industry
Sulphur	^{35}S	88 d	Beta	0.17	Nuclear industry
Argon	^{41}Ar	1.83 h	Beta Gamma	1.20, 2.49 1.29	Nuclear industry
Calcium	^{45}Ca	165 d	Beta	0.26	Nuclear industry
Chromium	^{51}Cr	27.8 d	Gamma EC	0.32 0.752	Nuclear industry
Manganese	^{54}Mn	303 d	Gamma	0.84	Fallout, nuclear industry
Iron	^{55}Fe	2.6 y	EC	0.232	Fallout, nuclear industry
	^{59}Fe	45.1 d	Beta Gamma	0.273, 0.475 0.142-1.29	Nuclear industry
Cobalt	^{58}Co	71.3 d	Beta Gamma EC	0.474 0.810, 0.864 2.31	Nuclear industry
	^{60}Co	5.3 y	Beta Gamma	0.315, 1.49 1.17, 1.33	Fallout, nuclear industry
Nickel	^{63}Ni	92 y	Beta	0.07	Nuclear industry
Zinc	^{65}Zn	244 d	Beta Gamma EC	0.325 1.11 1.11	Nuclear industry
Arsenic	^{76}As	1.1 d	Beta Gamma	0.35-2.96 0.51-2.66	Nuclear industry
Krypton	^{85}Kr	10.8 y	Beta Gamma	0.67 0.14	Fallout, nuclear industry
Strontium	^{89}Sr	52 d	Beta	1.46	Fallout, nuclear industry
	^{90}Sr	28.1 y	Beta	0.546	Fallout, nuclear industry
Yttrium	^{90}Yr	2.67 d	Beta	2.27	Nuclear industry
	^{91}Yr	58.8 d	Beta Gamma	0.33, 1.55 1.21	Nuclear industry
Zirconium	^{95}Zr	65 d	Beta Gamma	0.36-1.13 0.236, 0.723	Fallout, nuclear industry
Niobium	^{95}Nb	35.1 d	Beta Gamma	0.160 0.766	Fallout, nuclear industry
Technetium	^{99}Tc	2.12×10^{5} y	Beta	0.29	Fallout, nuclear industry
Ruthenium	^{103}Ru	39.6 d	Beta Gamma	0.203-0.90 0.04-0.61	Fallout, nuclear industry
	^{106}Ru	367 d	Beta Gamma	0.039 0.512-2.64	Nuclear industry
Silver	^{110m}Ag	253 d	Beta Gamma	0.087, 0.529 0.657, 0.818	Nuclear industry

Element	Radionuclide	Half-life	Type of emission	Energy (MeV)	Sources
Antimony	^{124}Sb	60.3 d	Beta Gamma	0.06-2.32 0.044-2.30	Nuclear industry
	^{125}Sb	2.7 y	Beta Gamma	0.10-0.619 0.036-0.671	Fallout, nuclear industry
Tellurium	^{125m}Te	58 d	Gamma	0.035, 0.110	Nuclear industry
	^{132}Te	3.25 d	Beta Gamma	0.22 0.049-0.228	
Iodine	^{129}I	1.7 x 10^{7} y	Beta Gamma	0.189 0.040	Fallout, nuclear industry
	^{131}I	8.07 d	Beta Gamma	0.257-0.806 0.080-0.723	Fallout, nuclear industry
Xenon	^{131m}Xe	11.8 d	Gamma	0.164	Nuclear industry
	^{133}Xe	5.27 d	Beta Gamma	0.267,0.347 0.080,0.382	Nuclear industry
Cesium	^{134}Cs	2.05 y	Beta Gamma	0.089, 0.410 0.475-1.40	Nuclear industry
	^{136}Cs	13 d	Beta	0.341, 0.560	Fallout
	^{137}Cs	30.2 y	Gamma Beta Gamma	0.067, 1.24 0.511, 1.18 0.662	Fallout, nuclear industry
Barium	^{140}Ba	12.8 d	Beta Gamma	0.47-1.02 0.139-0.537	Fallout
Lanthanum	^{140}La	1.67 d	Beta Gamma	1.25-2.17 0.110-2.55	Fallout, nuclear industry
Cerium	^{141}Ce	33 d	Beta Gamma	0.444, 0.582 0.145	Fallout, nuclear industry
	^{144}Ce	285 d	Beta Gamma	0.175-0.309 0.034-0.134	Fallout, nuclear industry
Neodymium	^{147}Nd	11.1 d	Beta Gamma	0.38, 0.82 0.091-0.69	Fallout
Promethium	^{147}Pm	2.5 y	Beta	0.23	Nuclear industry
Europium	^{154}Eu	16 y	Beta Gamma	0.27-1.86 0.060-1.60	Nuclear industry
	^{155}Eu	1.81 y	Beta Gamma	0.10-0.25 0.043-0.105	Nuclear industry
Polonium	^{210}Po	138.4 d	Alpha Gamma	5.30 0.803	Nuclear industry
Thorium	^{234}Th	24.1 d	Beta Gamma	0.100, 0.191 0.030-0.094	Nuclear industry
Uranium	^{234}U	2.47 x 10^{5} y	Alpha Gamma	4.60, 4.72 0.053-0.580	Nuclear industry
	^{235}U	7.1 x 10^{8} y	Alpha Gamma	4.16-4.60 0.074-0.367	Nuclear industry
	^{236}U	2.39 x 10^{7} y	Alpha Gamma	4.33, 4.44 0.050	Nuclear industry
	^{238}U	4.51 x 10^{9} y	Alpha Gamma	4.14, 4.15 0.048	Nuclear industry

Element	Radionuclide	Half-life	Type of emission	Energy (MeV)	Sources
Neptunium	^{237}Np	2.14 x 10^6 y	Alpha Gamma	4.40-4.87 0.020-0.240	Nuclear industry
Plutonium	^{238}Pu	86 y	Alpha Gamma	5.36, 5.46,5.50	Fallout, nuclear industry
	^{239}Pu	2.44 x 10^4 y	Alpha Gamma	0.044 5.01-5.16	Fallout, nuclear industry
	^{240}Pu	6580 y	Alpha Gamma	0.039-0.769 5.02, 5.12,5.	Fallout, nuclear industry
	^{241}Pu	13.2 y	Alpha Beta	17 0.045,0.104	Fallout, nuclear industry
	^{242}Pu	3.79 x 10^5 y	Alpha	4.80-5.05 0.021 4.86, 4.90	Nuclear industry
Americium	^{241}Am	458 y	Alpha Gamma	5.39-5.55 0.026-0.060	Fallout, nuclear industry
Curium	^{242}Cm	163 d	Alpha Gamma	5.97-6.11 0.044	Nuclear industry

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