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We have developed methods to measure the Th and U concentration in the picogram/gram sensitivity range for samples of acrylic. The objective of the program is to assure adequate radiopurity of the structural acrylic which will be used in the SNO vessel. Our technique is based on using the Los Alamos Omega West Reactor to neutron activate the acrylic samples. Pneumatic sample delivery line limitations restrict us to irradiating samples which are around 3 gm of acrylic..

Neutron Irradiation of the Acrylic Samples.

Samples of acrylic are machined, using either tungsten carbide or high speed steel cutting bits, so that they will fit into plastic pneumatic carriers. The finished samples are $\approx$ 4 mm diameter by 40 mm long and weigh about 3.5 g. The samples are ultrasonically washed with HCl, HF, and water prior to insertion into the plastic carriers. Irradiations were typically performed for $\sim$ 7 hours using the INC-5 Omega West reactor which has a neutron flux of about 1×10^{13} n/cm^2/s. The total irradiation fluence is typically 2.4×10^{17} n/cm^2.

The Th and U concentrations were obtained by gamma counting for the ^{233}Pa and the ^{239}Np induced activities. The samples were either directly counted or vacuum volatilized and chemically processed (see below for a description of this procedure). For the direct counting (no chemistry) cases, a suitable cooling time of one to several days was necessary depending on trace element activation contaminants in the sample. The samples were placed in a calibrated Ge well detector and counted for one to several days. The sensitivities were limited by activation backgrounds associated with the other trace elements present in the sample. For low background samples, levels at, or below, the pg/g concentration were measurable. Acrylic samples were obtained from four sources: Reynolds Polymer, Polycast, Swedlow and a sample prepared locally (technique described below). Also we did one set of measurements on Suprasil quartz. The results are summarized in the following table:

Neutron Activation Results

Supplier	ID	Thorium (pg/g) No Chem	Thorium (pg/g) Chem (CY)	Uranium (pg/g) No Chem	Uranium (pg/g) Chem (CY)	Note
Reynolds	1301	26.2 ± 5.5	14.8 ± 3.6 (49%)			
	2149	21.3 ± 2.8				
		4.5 ± 2.4				(1)
	1302	3.4 ± 2.7				
Polycast	2001		0.8 ± 0.9 (70%)		0.1 ± 0.2 (77%)	
	2002	20.2 ± 1.3				(2)
		13.2 ± 1.1				(3)
			1.6 ± 4.9 (12%)			(4)
Swedlow A	1503		19.2 ± 3.3 (40%)			
B	1603		16.8 ± 2.1 (64%)		1.7 ± 0.8 (52%)	
C	1703		26.2 ± 7.3 (10%)		2.2 ± 0.6 (60%)	
D	1802		93.0 ± 8.7 (51%)			
E	1901		10.1 ± 4.8 (28%)			
	1902		12.1 ± 2.8 (57%)			
Fowler	3001	106.0 ± 10.0				(5)
Suprasil	2301	6.8 ± 0.9		3.2 ± 0.9		

Notes:

(1) The second value for Reynolds 2149 represents a count following acid cleaning of the acrylic rod.

(2) The sample Polycast 2002 was counted in two different orientations. (2) was in the "up" direction.

(3) Polycast 2002 counted in the "down" orientation.

(4) The Th concentration following chemical processing of Polycast sample 2002.

(4) Analysis of an acrylic sample produced at Los Alamos by slush casting the inhibited monomer with powdered acrylic resin.

Chemical Processing of Acrylic Samples for U and Th Determination.

Volatilization of the Acrylic.

After irradiation, the samples are taken out of the plastic carrier and placed in a volatilization tube along with ^{231}Pa and ^{235}Np for chemical yield determination. The volatilization tube, 25 mm diam. by 75 mm long has a 12 mm diam. connection tube 100 mm long terminating in a 14/35 standard taper joint. The tube is connected, through a glass trap cooled in liquid nitrogen, to a vacuum pump. The system is evacuated to less than 0.01 torr and the tube is heated in a small furnace. Initially the temperature is held at 200-220 °C. until the sample melts and quits degassing. During this period the pressure will often increase to greater than 1 torr. When the pressure again drops to less than 0.05 torr, the temperature is the gradually raised to 400-420 °C. as the acrylic distills away. This process takes about 3-4 hrs and depends somewhat on the type of acrylic sample. At the end of the volatilization the tube is cooled to room temperature while still under vacuum. In most cases, there is only a small amount of carbonaceous material left in the tube. The tube is removed from the trap then the joint and upper end of the connection tube are cleaned to remove vacuum grease and any soluble high boiling material that may have condensed near the joint.

Digestion of the Residue.

Next 0.5 mℓ of conc. HNO_3 and 0.5 mℓ of conc. $HClO_4$ are added to the tube. The tube is then heated to boil the acid mixture. More nitric acid is added as it distills from the tube. After most of the carbon is washed from the walls the last of the nitric acid is distilled off and the perchloric acid destroys the carbon yielding a pale green clear solution. The perchloric acid is the distilled down to near dryness, 0.5 mℓ nitric acid is added and distilled off to near dryness. Next 1 mℓ of conc. HCl is added and distilled off. This is repeated until no more NO_2/N_2O_4 fumes are evolved. The solution is then transferred along with 1 mg of Fe^{+3} to a centrifuge tube and $Fe(OH)_3$ is precipitated with conc. NH_4OH. The $Fe(OH)_3$, carrying the Np and Pa activities, is centrifuged down and the supernant is discarded. The precipitate is then dissolved in conc. HCl, reprecipitated with conc. NH_4OH, and centrifuged. Again the supernant is discarded and the precipitate is dissolved in 0.5 mℓ conc. HCl.

Ion Exchange Separation of Np and Pa.

^{239}Np and ^{233}Pa are separated on a small column with AG 1X8 anion exchange resin (200-400 mesh). The column is a plastic tube 3.5 mm diam. and 100 mm long that holds 1 cm^3 of resin. The column is pretreated with water then with 3 mℓ of conc. HCl. The solution from the digestion step is saturated with HCl gas the transferred to the column with 4 washes of 0.5 mℓ of conc. HCl. The Pa activity is eluted from the column with 3 mℓ of 9M HCl that contains 0.05M HF. After the Pa is collected, the Np activity is eluted with 2 mℓ of 4M HCl containing 1M HF. Both the Pa and Np are collected directly in small counting vials. The chemical yield has been improving with time and is now about 70% for both Np and Pa.

Counting of the Np and Pa Activities.

The activities are measured with a well type intrinsic Ge detector. The detector is made with low background components and has a NaI(Tl) compton suppression shield. The background of the detector is about 3 C/hr/keV at 300 keV. With our sample size and irradiation conditions, 1 ppt of U in the acrylic gives a counting rate of 10.7 C/h while 1 ppt of Th gives a counting rate of 1.7 C/h assuming unity chemical yields.

In our irradiation conditions, with a poor cadmium ratio, the formation cross section for ^{233}Pa is about 9 barns while that for ^{239}Np is about 13 barns. Both of these effective cross sections are quite a bit larger than for the (n,γ) reactions with thermal neutrons.

The Th is estimated from the intensity of the 312 keV transition associated with the decay of ^{233}Pa and the U concentration comes from the intensity of the 228 keV transition in the decay of ^{239}Np.

Preparation of Acrylic Traced with ^{229}Th.

We have investigated several methods for the preparation of acrylic samples that contain known amounts of Th tracer. We would use these samples to verify that various chemical processing steps involved in the estimation of Th and U content of acrylic give reproducible results. We need to be able to prepare samples of acrylic that weigh a few hundred grams and have cross sections of a few inches. These relatively thick pieces are difficult to prepare due to the large exothermic reaction during polymerization. For normal casting from the monomer, it would take many days at very carefully controlled thermal conditions to achieve satisfactory results. We have elected to use a variety of "slush casting" to avoid some of the thermal problems. We are currently using a mixture of equal parts of inhibited monomer and Dupont molding powder (polymer). When the mix is made one has about 45 min. of "pot life" before the mix becomes a stiff gel. During that time the mix is put into appropriate containers, vacuum degassed, and then the containers are placed in an autoclave. The autoclave is pressurized to 150 psig with nitrogen and then allowed to warm slowly to 40 °C. After about 5 hrs at 40 °C, the temperature is raised slowly to 80 °C and maintained there for 15 hrs. The autoclave is then cooled to room temp, depressurized, and the samples are removed. We have found that the molding powder has sufficient initiator still in it to promote the polymerization of the inhibited monomer in the slush. This allows us to use the inhibited monomer without treatment to remove the hydroquinone inhibitor. This has several other benefits; first the gel has a long lifetime at room temperature so that the polymerization step can be done when convenient. Secondly, the monomer makes up only half of the mix so the exothermic heat is reduced by a factor of 2 while the heat capacity is doubled. The heat capacity and the inhibitor in the monomer make control of the polymerization much easier than when one uses the pure monomer. We have prepared several samples up to 300 g with good success with this method. The finished acrylic has good clarity and has a density of 1.18 indicating complete polymerization. We are planning to prepare the traced acrylic samples soon.