DISCRIMINATOR SETTING AND COCKTAIL PREPARATION FOR ANALYSIS OF ALPHA AND BETA EMITTERS IN AQUEOUS SOLUTION USING LIQUID SCINTILLATION COUNTER

(Penetapan Diskriminator dan Penyediaan Koktil Untuk Analisis Pemancar Alfa dan Beta Didalam Larutan Berakueus Menggunakan Pembilang Sintilasi Cecair)

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Abstract

Liquid scintillation counting (LSC) is not only being used to measure pure beta emitters, but it can be used to measure both alpha and beta emitters simultaneously. Measurement of alpha and beta emitters in aqueous solution is done using a single sample. For the sample preparation, colorless detergent or emulsifier was used to incorporate the water into an organic based scintillator to produce a clear homogeneous solution, since this is the best form to give the highest count rate and detection efficiency. The instrument also need some attention, where after calibration, the LSC was set for the discriminator level which is suitable for measurement of both alpha and beta radiations. In this study, the focus is on the development of the best scintillation cocktail and establishes the best discriminator setting. From this study the best proportion of scintillation cocktail is 2:4:4 for water, toluene, and Triton-N101 (emulsifier) respectively and the best discriminator setting for alpha and beta counting are 120.
Abstract in English

Keywords: alpha emitter, beta emitter, cocktail, LSC, scintillant.

Abstrak

Pembilang sintilasi cecair (PSC) bukan hanya digunakan untuk mengukur pemancar beta tulen, tetapi boleh juga digunakan untuk mengukur kedua-dua pemancar alfa and beta serentak. Pengukuran pemancar alfa dan beta di dalam larutan berair boleh digunakan menggunakan sampel yang sama. Untuk penyediaan sampel, emulsifier tanpa warna telah digunakan bagi mengikat air ke dalam sintilator berasaskan organik untuk menghasilkan larutan homogeneous, kerana ini adalah merupakan bentuk terbaik yang memberikan kadar bilangan dan efisiensi pengesanan yang tertinggi. Peralatan juga memerlukan perhatian, dimana selepas proses kalibrasi, PSC perlu penentuan tahap diskrimanator yang sesuai untuk pengukuran kedua-dua radiasi alfa dan beta. Ini adalah untuk memastikan kadar limpahan dari kedua-dua radiasi rendah. Di dalam kajian ini, fokus ditumpukan kepada membangunkan koktil sintilasi dan menentukan tahap diskriminator yang sesuai. Dari kajian ini didapati kadar terbaik bagi koktil sintilasi adalah 2:4:4 bagi air, toluen dan Triton N-101 (emulsifier) masing-masing dan tahap diskriminator terbaik adalah pada 120 untuk pengukuran zarah alfa dan beta serentak.

Kata kunci: pemancar alfa, pemancar beta, koktil, PSC, sintilan.

Introduction

Measurement of alpha and beta emitters in environment especially water samples are normally carried out using separate method, since both radiations having different properties. Recently, with the advancement of liquid scintillation counting (LSC), the measurement of radionuclides in the natural environment where radionuclide concentrations are low, becoming more feasible since the instrument background is very low. The contribution of instrument background to the precision of the measurement is often important. The “low level” instruments have their background reduction features that are the efficiency to background (E^2/B) factor is increased or minimum detectable activity (MDA) is decreased [1].

This method is based on the recently available liquid scintillation counting (LSC) systems with the ability of discrimination of alpha and beta emitters by the time distribution of the light emission each generates from scintillators, in the term of pulse shape discrimination (PSD) or pulse decay analysis (PDA). Pulse Shape Analysis (PSA) is a pulse shape discrimination technique that is based on a method that integrates the charge of 'tail' of scintillation pulse and compares it with the total charge in the same pulse. Different settings of the PSA level assign the pulse into either a long (alpha-like) or short (beta-like) category. Thus, different PSA settings allow pulses to be categorized according to their length (shape). Typically, increasing the PSA setting will direct more pulses toward the long or alpha category. However, depending on how the technique is implemented, the reverse can also be true, i.e., increasing the setting may direct more alpha counts into the beta category. There are several methods of accomplishing pulse shape analysis including slow crossover timing, fast crossover timing, and constant fraction of pulse-height trigger [1].

Water quality is an important aspect of environmental studies. Natural waters contain both alpha (such as ^{238}U) and beta (such as ^{40}K) emitters in widely varying concentrations which are responsible for a generally small fraction of the total dose received from natural and artificial radioactivity [2]. For practical purposes, the recommended guideline for activity concentrations are 0.1 BqL^{-1} for gross alpha and 1 BqL^{-1} for gross beta in the drinking water [3]. The recommendations do not differentiate between natural and man-made radionuclides. Below these reference levels of gross activity, drinking water is acceptable for human consumption and any action to reduce radioactivity is not necessary. In the last decades, nationwide studies have been performed in practically every developed country, where drinking water is regularly sampled and analyzed. In developing nations, data on the natural radioactivity in water are not always available. According to a UNSCEAR report [4], drinking water is considered to be an important factor in increasing the natural radiation exposure in humans.

The occurrence of natural radionuclides in drinking water poses a problem of health hazard, when these radionuclides are taken into the body by ingestion. Several naturally occurring alpha or beta emitting radionuclides such as ^{238}U, ^{226}Ra, ^{222}Rn, ^{210}Pb, ^{228}Ra and others are frequently dissolved in domestic water supplies and their concentrations vary over an extremely wide range, mainly depending upon the amount of radio elements present in bedrock and soil with which the water comes in contact [5].

Terlikowska *et. al.* [6], has done some work on the application of alpha/beta discrimination in liquid scintillation counting for the purity control of ^{99m}Tc medical solutions. Shing-Fa Fang *et al.* [7], has worked on the comparison of Alpha/Beta separation performance of Commercially Available Scintillation Cocktails and counting by Quantulus™ 1220 Liquid Scintillation Counter. Maurizio Forte *et. al.* [8], has done some natural radionuclides measurements in drinking water using Ultra-low Level Liquid Scintillation Counting. Cantaloub *et.al.* [9] studied the interaction of sample, cocktail and headspace volume when measuring aqueous radon in small volume sample. The study focused on measurement of radon using commercial cocktail, Ultima Gold F.

In this study, the focus is on the development of the best scintillation cocktail and establishes the best discriminator setting for LSC TRICAB 2700 from Packard that available in Malaysian Nuclear Agency (NM) in Bangi, Malaysia. The scintillation cocktail was developed using emulsifier (Triton N-101) mixed with solvent (toluene) and scintillator (2,5-diphenyloxazole, PPO and 1,4-bis(5-diphenyloxazol-2-yl)-benzene, POPOP).

Experimental

Instrument Calibration

Liquid Scintillation Counter TRICAB 2700 is a Packard instrument which is capable for measuring both alpha and beta radiations in water. Before any samples are counted, the instrument must be calibrated. This is performed automatically with the use of the calibration (SNC) protocol plug and the unquenched ^{14}C calibration standard.

Alpha/ beta Calibration and Discriminator Setting

Using Packard TRICAB models with alpha/beta discrimination, the optimum setting is the setting where there is equal and minimum spillover of alpha pulses into the beta Multi Channel Analyzer (MCA) and beta pulses into the alpha Multi Channel Analyzer (MCA). The determination of an optimum PDD requires two standards: one of the pure alpha emitter of interest and one of the pure beta emitter of interest. ^{241}Am and ^{36}Cl samples in Packard Ultima Gold AB, a cocktail specifically designed for alpha/beta separation were used for setting the percent spillover or percent misclassification of alpha and beta emitters. To achieve this, one need to vary the discriminator setting on the instrument (LSC). The instrument determines it optimum setting that results in the minimum misclassification of alpha and beta activity, and will generate the percent misclassification plot on demand. Several observations can be made by comparing these two curves. The first observation is the obvious shift in the instrument determined optimum PDD to a slightly higher value with TR-PDA than without TR-PDA. The next observation is the flattening of the alpha curve.

Cocktail development

Cocktail development is the method where we attempt to incorporate the water in an organic solvent. A triangular method was used to get the best proportion of water, organic solvent and Triton N-100 with 4.0 g/L PPO and 0.4 g/L POPOP (Figure1).

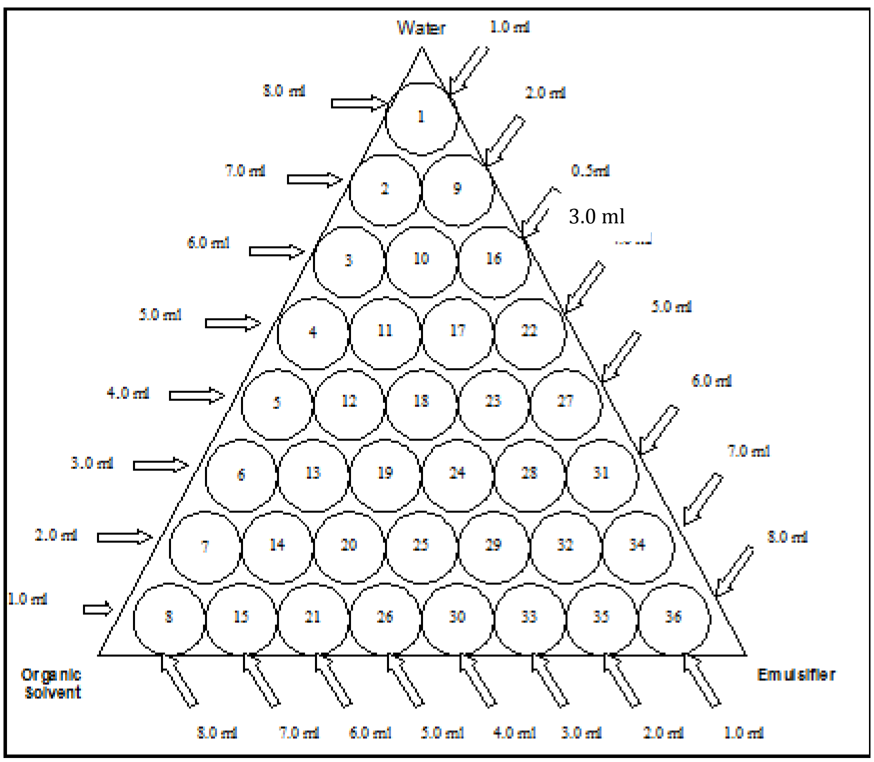


Figure 1: The position of vials for different composition of water, toluene and Triton N-100

Results and Discussion

Instrument Calibration

During the calibration procedure, the high voltage applied to photomultiplier tube (PMT) is adjusted individually until the tubes are normalized (synchronized) in their response to the ^{14}C calibration standard. Then, the high voltage to both tubes is adjusted simultaneously until the end point of ^{14}C spectrum fall in the appropriate position on the spectra analyzer (on-board 400-channel) multi-channel analyzer.

It is highly recommended that the SNS cassette (containing the ^{14}C calibration standard) be left in the instrument at all time. If the instrument is not IDLE, the calibration procedure is performed once every a 23-hour timer is checked. If 23 hour has elapsed since the previous calibration, the instrument will automatically perform the SNC procedure. If the time has NOT elapsed, the SNC cassette is bypassed. When the calibration is complete, the message SYSTEM NORMALIZED appears on the printout. This indicates that the instrument is ready for counting.

Discriminator Setting

Separate optimum PDD setting should also derived for unique alpha and beta standard pairs. For gross alpha and gross beta measurements where the particular radionuclides may not be known, an alpha and beta standard of similar energy to the alpha and beta in the samples is desirable. To arrive at the optimum setting, each standard is counted individually at range of PDD settings and the percent misclassification of alphas into the beta MCA and vice versa are plotted against PDD on the same graph. When only the beta emitter is of interest, a PDD value below the instrument determined optimum may be used, which minimizes the misclassification of alpha activity into the beta MCA at the expense of reducing the beta efficiency. Similarly, when only the alpha emitter is of interest, a PDD value greater than the optimum, can be used. This minimizes the mis-classification of beta events into the alpha MCA at the expense of reduced alpha counting efficiency.

The effect of Time-Resolved Pulse Decay Analysis (TR-PDA) on the misclassification curve is slightly higher value with TR-PDA and the flattening of alpha curve is the cause of the shift in the intersection of the curves and results in a higher optimum PDD value. The flattened shape of the alpha curve is due to TR-LSC discrimination of misclassified alpha events in the beta MCA. At this higher value, beta misclassification is also reduced. Manual adjustment the discriminator to a higher or lower value than the optimum will further reduce either beta or alpha misclassification at the expense of some loss of alpha or beta counting efficiency, respectively.

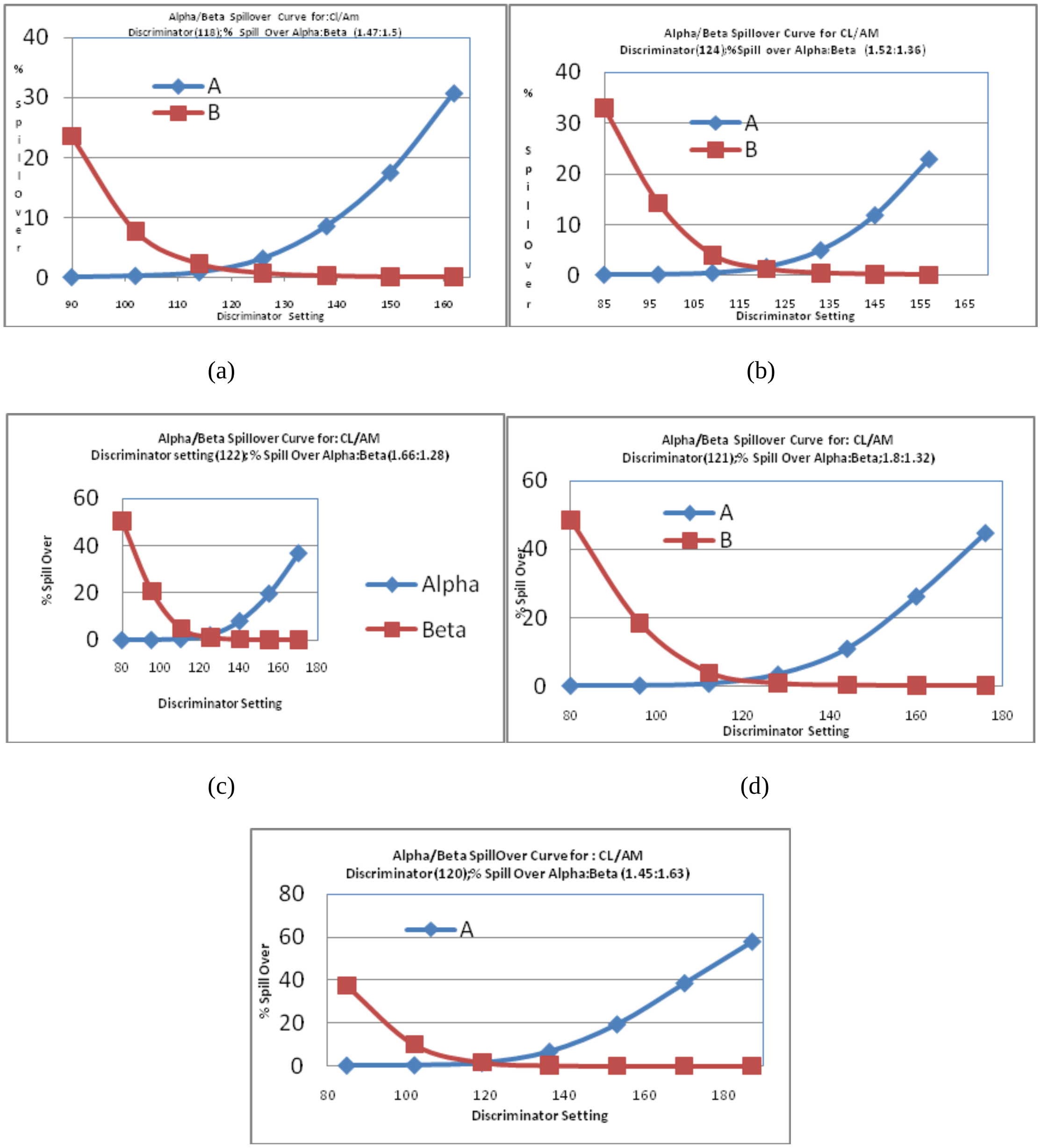
Figures 2(a) to 2(e) show the alpha/beta spillover curve for $^{36}Cl/^{241}Am$. From these curves we have chosen the discriminator 120 with the percent spillover alpha:beta (1.45:1.63).

Cocktail development

In this study, the emulsifier used was Triton N-101 and the solvent was toluene scintillation grade. The amount of primary and secondary scintillator was 4.0 g/L and 0.4 g/L of PPO and POPOP respectively. Using the triangular method [10], 36 vials were used to be filled with different proportions of water, toluene and emulsifier (Triton N-100).

Tables 1(a) and 1(b) show the proportion of water, toluene, and triton N-101 in different vials. The appearance of liquid in each vial was recorded as cloudy, clear and opaque. The aim of this cocktail is to get a clear solution and that solution is giving the best count rate. The count rate of ^{3}H (pure beta emitter) and ^{226}Ra (alpha emitter) are listed in the column 2 of Table 1.

Both ^{3}H and ^{226}Ra show the same characteristics when mixed with cocktail. The aim of this cocktail preparation is to get the clear stable solution, with the best count rate as well as giving the best merit value. From Tables 1 (a) and (b), the best proportion for the mixture of water:toluene:Triton N-101 is 2:4:4. So, for 20 mL mixture, the amount of water is 4 mL.



(a) (b) (c) (d) (e)

Figures 2(a) – 2(e): Discriminator Setting 118, 124, 121,122 and 120

Table 1(a): The proportion of water, toluene, and triton-N100, appearance and the activities for beta emitter (^{3}H)

Vial no.	Count (CPM)	Efficiency	Water	Toulene	Triton	Appearance	Homogenity	Viscosity	% of water in solution	Merit value	% Triton
MA-38	51	0.5	8	1	1	milky	heterogenous	viscous	80	42.8	10
MA-39	140	1.5	7	2	1	milky	heterogenous	viscous	70	103.1	10
MA-40	471	5.0	6	3	1	milky	heterogenous	not viscous	60	297.3	10
MA-41	728	7.7	5	4	1	milky	heterogenous	not viscous	50	383.0	10
MA-42	1044	11.0	4	5	1	milky	heterogenous	not viscous	40	439.2	10
MA-43	780	8.2	3	6	1	milky	heterogenous	not viscous	30	246.1	10
MA-46	968	10.2	2	7	1	milky	heterogenous	not viscous	20	203.7	10
MA-47	953	10.0	1	8	1	milky	heterogenous	not viscous	10	100.3	10
MA-51	3096	32.6	1	7	2	milky	heterogenous	viscous	10	325.6	20
MA-52	1748	18.4	2	6	2	opaque	heterogenous	viscous	20	367.7	20
MA-53	1420	14.9	3	5	2	opaque	heterogenous	viscous	30	448.0	20
MA-54	853	9.0	4	4	2	opaque	heterogenous	viscous	40	359.0	20
MA-55	513	5.4	5	3	2	opaque	heterogenous	viscous	50	269.9	20
MA-56	290	3.1	6	2	2	opaque	heterogenous	viscous	60	183.3	20
MA-57	131	1.4	7	1	2	cloudy	heterogenous	viscous	70	96.1	20
MA-58	176	1.8	6	1	3	cloudy	heterogenous	viscous	60	111.0	30
MA-59	435	4.6	5	2	3	clear	homogeneous	not viscous	50	228.6	30
MA-60	694	7.3	4	3	3	clear	homogeneous	not viscous	40	291.8	30
MA-61	1085	11.4	3	4	3	cloudy	heterogenous	viscous	30	342.5	30
MA-62	1739	18.3	2	5	3	cloudy	heterogenous	viscous	20	365.8	30
MA-63	2634	27.7	1	6	3	clear	homogeneous	not viscous	10	277.0	30
MA-64	2298	24.2	1	5	4	clear	homogeneous	not viscous	10	241.7	40
MA-65	***1436***	***15.1***	***2***	***4***	***4***	***clear***	***homogeneous***	***not viscous***	***20***	***302.0***	***40***
MA-66	891	9.4	3	3	4	cloudy	heterogenous	viscous	30	281.3	40
MA-67	529	5.6	4	2	4	clear	homogeneous	viscous	40	222.7	40
MA-68	235	2.5	5	1	4	clear	homogeneous	viscous	50	123.7	40
MA-69	310	3.3	4	1	5	clear	homogeneous	viscous	40	130.3	50
MA-70	651	6.8	3	2	5	clear	homogeneous	viscous	30	205.4	50
MA-71	1191	12.5	2	3	5	clear	homogeneous	not viscous	20	250.6	50
MA-72	1847	19.4	1	4	5	clear	homogeneous	not viscous	10	194.3	50
MA-73	1588	16.7	1	3	6	clear	homogeneous	not viscous	10	167.0	60
MA-74	899	9.5	2	2	6	clear	homogeneous	not viscous	20	189.0	60
MA-75	411	4.3	3	1	6	cloudy	homogeneous	not viscous	30	129.8	60
MA-76	541	5.7	2	1	7	clear	homogeneous	viscous	20	113.9	70
MA-77	1126	11.8	1	2	7	c;ear	homogeneous	viscous	10	118.5	70
MA-78	681	7.2	1	1	8	clear	homogeneous	not viscous	10	71.6	80

Note : The best portion of cocktail mixture

Table 1(b): The proportion of water, toluene, and triton-N100, appearance and the activities for alpha emitter (^{226}Ra)

Vial ID	Count (CPM)	Efficiency	Water	Toulene	Triton	Appearance	Homogenity	Viscosity	water in solu	Merit value	% Triton
MA-79	203	4.9	8	1	1	milky	heterogenous	viscous	80	393.6	10
MA-80	254	6.2	7	2	1	milky	heterogenous	viscous	70	432.2	10
MA-81	471	11.5	6	3	1	milky	heterogenous	not viscous	60	689.8	10
MA-82	334	8.1	5	4	1	milky	heterogenous	not viscous	50	406.8	10
MA-83	478	11.7	4	5	1	milky	heterogenous	not viscous	40	467.2	10
MA-84	503	12.3	3	6	1	milky	heterogenous	not viscous	30	368.4	10
MA-85	529	12.9	2	7	1	milky	heterogenous	not viscous	20	258.2	10
MA-86	575	14.0	1	8	1	milky	heterogenous	not viscous	10	140.4	10
MA-87	719	17.6	1	7	2	milky	heterogenous	viscous	10	175.7	20
MA-88	640	15.6	2	6	2	opaque	heterogenous	viscous	20	312.8	20
MA-89	581	14.2	3	5	2	opaque	heterogenous	viscous	30	426.1	20
MA-90	405	9.9	4	4	2	opaque	heterogenous	viscous	40	395.5	20
MA-91	307	7.5	5	3	2	opaque	heterogenous	viscous	50	373.9	20
MA-92	313	7.6	6	2	2	opaque	heterogenous	viscous	60	457.9	20
MA-93	405	9.9	7	1	2	cloudy	heterogenous	viscous	70	691.7	20
MA-94	326	7.9	6	1	3	cloudy	heterogenous	viscous	60	476.0	30
MA-95	336	8.2	5	2	3	clear	homogeneous	not viscous	50	408.7	30
MA-96	**357**	**8.7**	**4**	**3**	**3**	**clear**	**homogeneous**	**not viscous**	**40**	**348.4**	**30**
MA-97	415	10.1	3	4	3	cloudy	heterogenous	viscous	30	304.0	30
MA-98	494	12.1	2	5	3	cloudy	heterogenous	viscous	20	241.1	30
MA-99	637	15.6	1	6	3	clear	homogeneous	not viscous	10	155.7	30
MA-100	574	14.0	1	5	4	clear	homogeneous	not viscous	10	140.1	40
MA-101	***379***	***9.2***	***2***	***4***	***4***	***clear***	***homogeneous***	***not viscous***	***20***	***184.6***	***40***
MA-102	351	8.5	3	3	4	cloudy	heterogenous	viscous	30	256.4	40
MA-103	378	9.2	4	2	4	clear	homogeneous	viscous	40	369.0	40
MA-104	359	8.7	5	1	4	clear	homogeneous	viscous	50	437.1	40
MA-105	327	8.0	4	1	5	clear	homogeneous	viscous	40	318.8	50
MA-106	397	9.7	3	2	5	clear	homogeneous	viscous	30	290.5	50
MA-107	400	9.7	2	3	5	clear	homogeneous	not viscous	20	194.9	50
MA-108	578	14.1	1	4	5	clear	homogeneous	not viscous	10	141.2	50
MA-109	502	12.3	1	3	6	clear	homogeneous	not viscous	10	122.6	60
MA-110	359	8.8	2	2	6	clear	homogeneous	not viscous	20	175.1	60
MA-111	322	7.9	3	1	6	cloudy	homogeneous	not viscous	30	235.6	60
MA-112	321	7.8	2	1	7	clear	homogeneous	viscous	20	156.6	70
MA-113	614	15.0	1	2	7	c;ear	homogeneous	viscous	10	150.1	70
MA-114	441	10.8	1	1	8	clear	homogeneous	not viscous	10	107.5	80

Note : The best portion of cocktail mixture

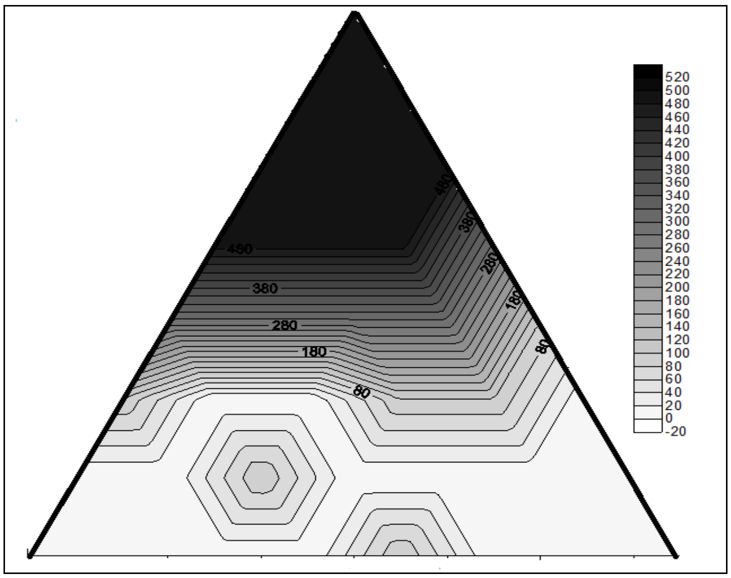


Figure 2(a): Appearance

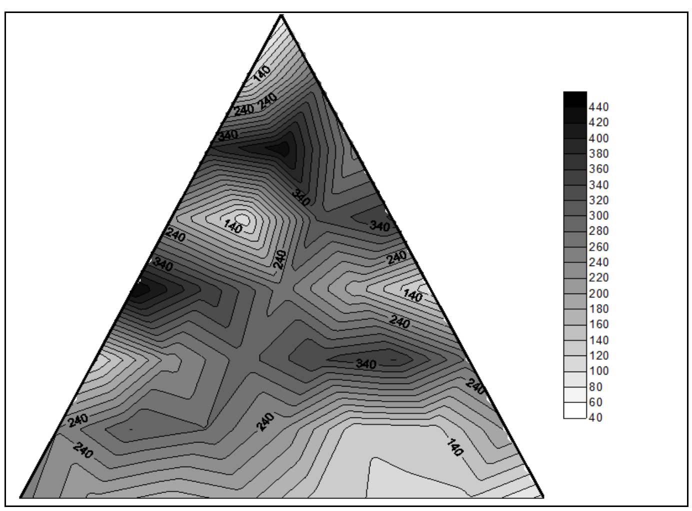


Figure 2(b): Merit Value

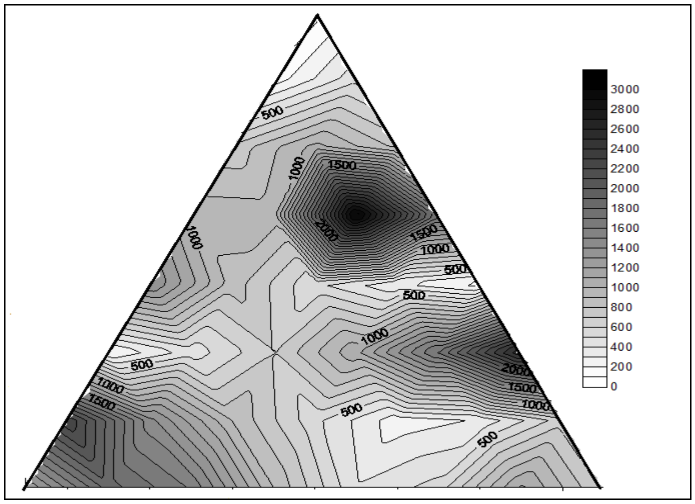


Figure 2(c): Count Rates of ^{3}H

From Figures 2(a), (b) and (c) are the contour for appearance, merit value and count rate of ^{3}H for 36 vials. It gives clear indication of the best appearance, merit value and count rate.

Conclusion

This instrument, LSC Packard TRICAB 2700 used ^{14}C for its calibration. The best discriminator setting for alpha/beta counting obtained in this study is 120 with 1.45:1.63 percentage ratio of alpha/beta spillover. The best proportion for the scintillation cocktail is found to be 2:4:4 for water, toluene, and triton N-100 with 4 g/L PPO and 0.4 g/L POPOP.

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