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Optimization of Cocktail AquaLight LLT Volume for Estimation of ^{14}C Activity in Aqueous Sample using LSC Hidex 300 SL

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Abstract

Optimization of the comparison between the AquaLight LLT cocktail and an aqueous sample had been carried out. AquaLight LLT cocktail was mixed with aqueous sample. The variation ratio between the aquaLight cocktail and the sample were 19: 1; 18: 2; 17: 3; 16: 4; 15: 5; 14: 6; 13: 7; 12: 8; 11: 9; 10:10; 9:11; 8:12; and 7:13. Then, each mixture was analyzed by LSC Hidex 300 SL. Optimization is seen by comparing the highest efficiency which is stated by the TDCR value. The results indicate that the best ratio between AquaLight cocktail and an aqueous sample was 12: 8. The CPM, DPM, and TDCR values from a mixture of 12 mL cocktail and 8 mL sample were 306,400 ± 3,578; 418,000 ± 7,348; and 0,733 ± 0,013.

Keywords: ^{14}C; Liquid Scintillation Counter; Optimization Performance; AquaLight LLT

1. Introduction

Liquid Scintillation Counting (LSC) is widely used in the radionuclide field because it has advantages over conventional techniques, high efficiency, simple, fast preparation, and analyzes 2 particles (alfa and beta particles) at once [1-5]. The new generation Hidex 300 SL liquid scintillation counter is equipped with a TDCR (Triple to Double Coincidence Ratio) system, capable of correcting simultaneous blackouts [6-8]. The pre-treatment of the sample for radiocarbon analysis using LSC depends on the sample type, the sample by burning for organic samples or dissolving the carbonate sample with HCl for inorganic samples. Then, both pre-treatments produce releasing CO_2. The resulted CO_2 gas can be absorbed through 2 methods, direct CO_2 absorption (LSC A) or benzene synthesis (LSC B) [9-11]. Direct CO_2 absorption is applied by mixing the specified sample volume with a cocktail. AquaLight is used for aqueous samples, while mineral oil scintillator (a scintillator made from toluene), MaxiLight, and Ultima Gold F are used for organic samples [12].

Efficient use of possible cocktail volume is required since there is a necessity to reduce radioactive wastes and taking into account high expenditures of liquid scintillation cocktails [13]. In addition, performance indicators need to be balanced against the specific laboratory needs which include waste treatment regulations [14]. Another identified factor called quenching insufficient separation from contaminants, forming matrix components and leading to lowering of scintillation efficiency is influenced by cocktail chemical composition (whose precise composition is usually unknown), sample

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chemical composition, and sample v. cocktail ratio [13]. The quantity of quenches in a sample is mostly driven by the cocktail quantity [15]. So how the measurement predisposed by the ratio of cocktail and water added into the samples is essential, which is the significant factor of optimal counting condition [13,15].

Research showed the comparison cocktail and sample volume, such as ^{3}H analysis in water sample using 10 mL cocktail and 10 mL water [16], ^{3}H analysis with the optimum ratio between cocktail and sample, namely 10:10 [17]. Mixture of 2 g sample, 6 g ultrapure water, and 12 mL scintillator [18], 1:2 ratio used for maximizing the counts [15], ^{3}H analysis using 8:12 ratio (sample water: AquaLight cocktail) in LSC Hidex 300 SL [19]. Analysis ^{222}Rn in water used a ratio of 1:1 between sample and cocktails [20]. Analyze ^{222}Rn by mixing 10 mL of cocktail and 10 mL of water sample [21]. Analysis of ^{222}Rn by mixing 14 mL of scintillation liquid and 6 mL of water sample [22]. ^{14}C analysis by mixing 8 mL sample and 12 mL aquaLight LLT in vial 20 mL [23]. Analysis of ^{14}C sample from coral *Porites Lobata* with the ratio between sample and scintillator is 8:12 [24]. Analysis ^{14}C used 8-12 mL of Carbo-Sorp E and mixture with Permaflour E+ scintillation cocktail and then analysis with LSC [8]. Several researches previously used specific mixture cocktails and samples volume, but no research that describes in detail the optimization comparison of AquaLight cocktails and aqueous samples. The data obtained from the analysis are expressed by the values of CPM, DPM, and TDCR. The analysis efficiency is expressed in the TDCR or Triple to Double Coincidence Ratio [7,8]. Therefore, this research was conducted to determine the ratio of appropriate AquaLight cocktail for ^{14}C analysis in the aqueous sample with LSC Hidex 300 SL.

2. Experimental

2.1. Equipment

The equipment used in this research were glass tools commonly used in laboratories, mortar, scales, preparation tools in the form of a series of CO_2 absorption devices, ^{14}C activity analysis using LSC Hidex 300 SL.

A low-level background Liquid Scintillation Counter Hidex 300 SL uses three PMTs aligned at 120^{o} (Optimal detection geometry) from each other, resulting in a better detection geometry. LSC Hidex 300 SL software is operated using an external PC with MikroWin 300 SL software [25].

2.2. Materials

Materials used in this research were scintillation cocktail AquaLight LLT (Hidex), coral sample, HCl 10%, $AgNO_3$ (99,8 % purity), silica gel, N_2 gas (>99,999%, High Purity), KOH (≥ 85,0 % purity), filter paper, silica gel, distilled water.

2.3.1. The Absorption of CO_2 Gas

The coral sample was weighed 5 grams with a mass variation of 5 to 55 grams and put into a round bottom flask. Furthermore, the sample was added with HCl 10% to produce CO_2 gas. Nitrogen gas released from nitrogen cylinder was used to carry CO_2 gas [26] through $AgNO_3$ to absorb HCl gas [10,27] and silica gel to absorb water [28] and CO_2 absorber as KOH [27,29]. The reaction was as follows [26]:

$$CaCO_3 + 2HCl \longrightarrow CO_2 + CaCl_2 + H_2O$$

The series of CO_2 absorption methods for measuring ^{14}C was shown in the figure below [26, 27]:

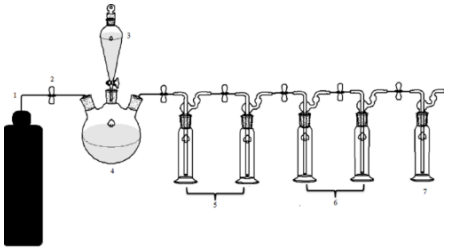

Figure 1 Design tool for the CO_2 absorption method. 1). Nitrogen Cylinder and Flow Meter, 2). Control Valve, 3). HCl 10 %, 4). Three necks 1000 mL flask, 5). Acid absorber ($AgNO_3$), 6). Water Absorber (silica gel), 6). CO_2 gas Absorber (KOH)

2.3.2. Sample Preparation

AquaLight Cocktails and aqueous samples were mixed in ratios 19: 1, 18: 2, 17: 3, 16: 4, 15: 5, 14: 6, 13: 7, 12: 8, 11: 9, 10:10, 9:11, 8:12, 7:13 and 20: 0 as control. Sample Load determination based on the following formula [14]:

$$SL = \frac{V_s}{V_s+V_{LSC}} \text{ x } 100 \qquad (1)$$

Where:
SL = Sample Load (%)
V_s = Sample Volume (mL)
V_{LSC} = Liquid Scintillation Cocktail Volume (mL)

2.3.3. Analysis with LSC Hidex 300 SL

The ^{14}C activity in the sample was expressed in units of activity, which was the minute decay (DPM) of ^{14}C. The counting of the sample with the LSC Hidex 300 SL counters produce data in units of CPM and TDCR or the efficiency of counting [30]:

$$E = \frac{CPM}{DPM} \times 100\ \% \qquad (2)$$

The statistical calculation of radioactive sample count using LSC was a very natural decay calculation of radioactive elements emitting pure β particles every time (random decay).

Samples and cocktails were mixture with a specific ratio, then analyzed with the LSC Hidex 300 SL device [31]. The count was carried out for 30 minutes with five repetitions. The standard deviation was determined for each value CPM, DPM and TDCR.

3. Result and Discussions

3.1. Composition of The Cocktail and Sample Volume

Sample load is the number of samples to the total number of the final mixture of samples and cocktails used [14]. The sample load data was stated as follows:

Table 1. Sample Load

No	Cocktail Volume (mL)	Sample Volume (mL)	Sample Load (%)
1	19	1	5
2	18	2	10
3	17	3	15
4	16	4	20
5	15	5	25
6	14	6	30
7	13	7	35
8	12	8	40
9	11	9	45
10	10	10	50
11	9	11	55
12	8	12	60
13	7	13	65

Even though the more the sample volume is mixed, the higher the percentage of sample load is needed, further analysis is needed to determine the appropriate cocktail and sample ratio.

3.2. CPM Value

CPM is obtained from the accumulated count multiplied by the time of death. Figure 2 shows the relationship between sample volume (mL) and CPM. Based on Figure 2, it can be seen that the increasing sample volume (mL), the higher the CPM value. When calculating 100 % efficiency, CPM will be identical in value to DPM. With the application of quench correction, it changes the value CPM to DPM [32].

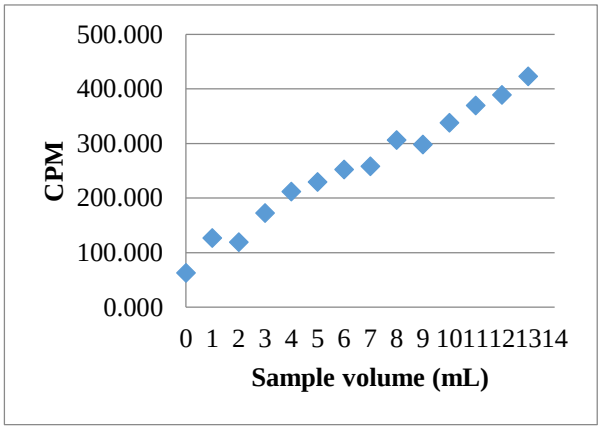

Figure 2. Relationship between Sample Volume (mL) and CPM

3.3. DPM Value

DPM is obtained from the CPM rate divided by the TDCR. DPM is the number of atoms in some radioactive materials that decay in one minute. 1 Bq is equal to 60 DPM [32]. Based on Figure 3, it is observed that an increase in the DPM value occurred with an increase in the volume of the sample added. The increase and decrease in the DPM value during the counting process were influenced by the phase instability between the carbonate solution and the scintillator at the beginning of the counting process and the quenching effect. There are 3 types of quenching effects, chemical quenching, ionization, and color quenching. Ionization quenching relates to the density of the solvent molecules excited in the cocktail. Color quenching occurs when the analyzed sample is colored. The color quenching phenomenon causes the absorption of photons of light in the vial before being detected and measured by PMT.

Chemical quenching (Chemiluminescence) is a disturbance that often occurs in the analysis of radioactive samples in light production in cocktails due to chemical reactions. Chemical reactions caused by chemiluminescence often occur when the cocktail is mixed with the sample solution in the vial. Several samples can produce chemiluminescence, for example, when a cocktail is added to the sample solution (pH 8-14) or when a chemical such as a hydrogen peroxide is present in the sample. The effect of pH and chemical interactions with some of the scintillation can cause molecular excitation and light emission. Several types of samples that can produce chemiluminescence are tissues or cells with inorganic bases (such as NaOH and KOH) or organic bases [33].

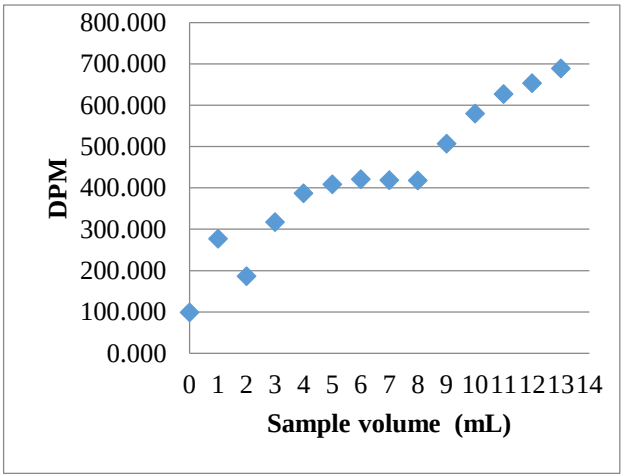

Figure 3. Relationship between Sample Volume (mL) and DPM

3.4. TDCR

TDCR shows the number of counting efficiency or as a quench index. Measurements with TDCR allow the determination of ^{14}C activity for pure beta-emitting radionuclides with a high degree of accuracy. TDCR does not require an external source and any known activity bottles. Therefore, the automatic TDCR absolute measurement is used in the LSC Hidex 300 SL [7]. The TDCR system is the main advantage of LSC. TDCR allows accurate estimates of the intrinsic efficiency based on the likelihood that increased quenches in the sample will follow the photon signal in the PMT. The output of the LSC includes total count, counts per minute (CPM), TDCR, and estimates of nuclear disintegration per minute in samples (DPM) which are CPM and TDCR [34]. A comparison between LSC cocktails and the correct sample volume optimizes performance calculations [2]. The following is Table 2. The relationship between the time of counting (m) and the TDCR of each AquaLight LLT cocktail ratio and the aqueous sample:

Table 2. Relationship between Ratio Cocktail: Aqueous Sample and TDCR

No	Ratio AquaLight LLT: Sample	TDCR
1	Control (20:0)	0,635 ± 0,004
2	(19:1)	0,494 ± 0,113
3	(18:2)	0,646 ± 0,057
4	(17:3)	0,548 ± 0,028
5	(16:4)	0,536 ± 0,047
6	(15:5)	0,568 ± 0,047
7	(14:6)	0,610 ± 0,063
8	(13:7)	0,621 ± 0,038
9	(12:8)	0,733 ± 0,013
10	(11:9)	0,579 ± 0,014
11	(10:10)	0,583 ± 0,003
12	(9:11)	0,589 ± 0,001
13	(8:12)	0,594 ± 0,008
14	(7:13)	0,613 ± 0,016

Table 2 shows changes in the ratio of cocktails and samples affect counting efficiency. The highest value of TDCR for comparison of AquaLight LLT and sample is 12 mL of aquaLight LLT and 8 mL of sample. TDCR value for 12:8 is 0,733 ± 0,013. The efficiency value in the enumeration with LSC Hidex 300 SL can be seen with the TDCR value. Based on the Table 2, the fairest comparison between the AquaLight LLT cocktail and the sample is 12: 8.

Regarding research related to tritium activity in air samples, optimization of the comparison between AquaLight and aqueous samples, the obtained results show the optimal ratio at a ratio of 12: 8 mL [35]. The ratio 12 mL cocktail and 8 mL sample maximize the sample water amount in 20 mL vial [34].

4. Conclusions

To conclude, the best comparison between AquaLight LLT cocktail and aqueous sample (^{14}C) was 12: 8. Furthermore, the CPM, DPM and TDCR values from the mixture of 12 mL cocktail and 8 mL sample were 306,400 ± 3,578; 418,000 ± 7,348; and 0,733 ± 0,013.

5. Conflicts of interest

The authors declare that there are no conflicts to declare.

6. Funding

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