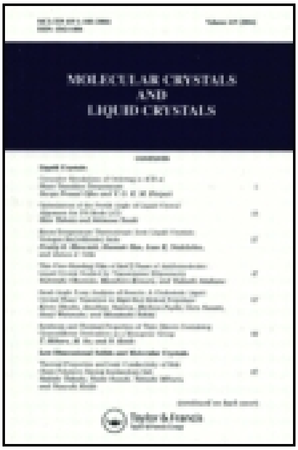

Molecular Crystals

Sensitivity enhancement for low level activities by complete syntheses of liquid scintillation solvents

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Sensitivity enhancement for low level activities by complete syntheses of liquid scintillation solvents

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Abstract—In order to take full advantage of liquid scintillation measurements for low level radioactivity, a large part of the solvent should be synthesized from the material of the sample to be counted. This procedure has been applied to three natural radioactivity dating methods. For carbon-14, the most sensitive and practical of the various liquids investigated is benzene. The compound, in addition to being a good liquid scintillation solvent, contains 92% carbon. The work on the synthesis of benzene for carbon-14 dating is now complete with yields approaching 100% and less than eight hours total chemical preparation time. Furthermore, the isotope effect for carbon has been proven to be small and reproducible to better than $^1/_2$%.

A synthesis of benzene for low level tritium measurements has also been developed. Benzene contains 8% hydrogen, but the chemical technique is considerably simpler than that used for radiocarbon dating. Due to the faster preparation, large quantities of benzene for tritium dating can be produced and the sensitivity increased in this way. An isotope effect of −34% has been observed, but it seems to be sufficiently reproducible not to pose a problem to the use of the liquid scintillation counter for tritium dating.

The possibility of age determinations with naturally occurring chlorine-36 is attractive due to the long half-life of the isotope. Dating of samples with ages of more than a million years is possible. The procedure now being developed consists of the synthesis of an inorganic liquid, silicon tetrachloride, from the chlorine of the sample to be dated. The chemical preparation is rapid and has an average yield of 75%, but the compound is not suitable for use without the presence of an organic solvent in which the scintillator must be dissolved. Despite this, with silicon tetrachloride much larger quantities than were previously possible can be counted and the method offers promise of eventually making chlorine-36 dating practical.

1. Introduction

Since its development more than ten years ago, the liquid scintillation counter has undergone constant improvement in sensitivity, convenience, and reliability. Important advances were the introduction of automatized sample changing and transisterized electronics. These changes were of interest principally to workers employing high levels of radioactivity, but there were also ameliorations in sensitivity. Most important here was the application of low noise quartz-face photomultipliers and the use of very fast coincidence circuits.

At the same time as the performance of the counter was being improved, the composition of the counting solution was examined for possible optimization. Information was obtained on quenching and ways in which it could be controlled as well as the introduction of new scintillators for better kinetic energy to light conversion efficiency. For researchers working with low level activities, the principal problem was how to put maximum amounts of their samples into the counting chamber and detect the activities with good efficiencies. The obvious solution was complete syntheses of liquid scintillation solvents from the materials to be counted. Furthermore, the best solvents would have as many of the following characteristics as possible:

1. The synthesis should be relatively facile and capable of being carried out with not more than eight man-hours of work.
2. The compound should have good scintillation properties, i.e., not be a quencher.
3. The compound should be stable and easily purified.
4. The compound should have a high percentage of the element containing the radioactivity being measured.
5. High density liquids would be preferable.

A number of different chemical methods were examined for the detection of low level activities of a variety of radioisotopes. In this work, three isotopes will be discussed. Carbon-14 has been the most thoroughly studied for the application of liquid scintillation techniques, followed by tritium and, recently, chlorine-36. These isotopes all exist naturally, but with low specific activities, and are of principal interest in the determination of ages. With their widely differing half-lives they are useful for distinct portions of the time scale. Furthermore, the limits of pairs of the three isotopes overlap to some extent and they can, potentially, be used as checks on each other. In the following sections the chemical syntheses that were developed in order to count the maximum amounts of the samples will be discussed separately for each isotope.

2. Low Level Carbon-14

Of all the radioisotopes occurring naturally, carbon-14 is the most widely used at the present time. Its half-life of 5730 years permits age determinations of materials from less than a hundred to about 60,000 years old. This period covers the entire span of modern man and, therefore, carbon-14 dating is capable of being applied to all archeological investigations where carbon containing artifacts are encountered. The isotope has also been used in geological studies, but here it is limited to the recent period and the end of the Pleistocene.

The carbon-14 specific activity before nuclear weapon contamination was 13.56 dpm/gm C,[1] a value that has been almost doubled at the present time.[2] This quantity of radioactivity is not difficult to measure, but great sensitivity is needed for the older samples or when reduced errors are necessary. The liquid scintillation counter is in extensive use for carbon-14 dating and has a sensitivity as high as that of the best of the gas counter methods. Table 1 shows the various solutions that have been investigated. Acetylene[3] and carbon dioxide[4] are gases that are dissolved in organic liquids at low temperatures. Practical difficulties have prevented either of these systems from being used for routine dating. The aliphatic hydrocarbons method,[5] which, along with ethanol, was the first attempt made for liquid counting sample carbon-14 dating, suffers from the disadvantages of a difficult chemical preparation and the fact that a carbon analysis had to be made after each synthesis. A few dates were obtained,[6] but the method is now abandoned. Ethanol[5] and methanol[7, 8] are poor scintillation solvents and can be used only in dilute solutions. Despite this, one list of dates was published with methanol.[9] Ethyl benzene[10] and toluene[7] are only partially synthesized from the sample carbon and also have difficult chemical preparations. Methyl borate was studied[11] as an extension of the synthesis of methanol. It shows improved scintillation properties, but still requires relatively dilute solutions. Paraldehyde was promising,[12, 13, 14] but the work has not led to a routine dating method because of the difficulty of the synthesis.

It is seen that benzene offers the most advantages for carbon-14 dating using liquid scintillation techniques. The only disadvantage is the relatively low density, but this is more than compensated for by the practical synthesis, good scintillation properties, and the possibility of

TABLE 1

Chemical characteristics of some organic compounds investigated for carbon-14 dating using the liquid scintillation counter

Liquid	% carbon	% carbon coming from sample	% of total counting sol'n coming from sample	Density	Scintillation properties	Synthesis	Reference
Acetylene	92	92	11	0.62	fair	facile	3
Aliphatic hydrocarbons	84	84	64	1.4	fair	difficult	5
Carbon dioxide	27	27	8	1.98	fair	facile	4
Ethanol	52	52	20	1.36	poor	facile	5
Ethyl benzene	91	23	23	1.50	good	difficult	10
Methanol	37	37	~10	1.33	poor	facile	7
Methyl borate	35	35	~10	0.92	fair	facile	11
Paraldehyde	54	54	38	1.40	fair	difficult	14
Toluene	78	13	13	1.50	good	difficult	7
Benzene	92	92	92	0.88	good	facile	27

having 92% of the counting solution carbon from the sample being dated. The following equations describe the chemical procedure for the synthesis of benzene:

$$\text{Sample} + \begin{matrix} O_2 \\ \text{or} \\ \text{acid} \end{matrix} \rightarrow CO_2 + \cdots$$

$$\left[\begin{array}{rcl} CO_2 + 2NH_4OH & \rightarrow & (NH_4)_2CO_3 + H_2O \\ (NH_4)_2CO_3 + SrCl_2 & \rightarrow & SrCO_3 + 2NH_4Cl \\ 2SrCO_3 + 5Mg & \rightarrow & SrC_2 + SrO + 5MgO \\ SrC_2 + 2H_2O & \rightarrow & C_2H_2 + Sr(OH)_2 \end{array}\right]$$

or

$$\left[\begin{array}{rcl} 2CO_2 + 10Li & \rightarrow & Li_2C_2 + 4Li_2O \\ Li_2C_2 + 2H_2O & \rightarrow & C_2H_2 + 2LiOH \end{array}\right]$$

or

$$\left[\begin{array}{rcl} 2C + 2L & \rightarrow & Li_2C_2 \\ Li_2C_2 + 2H_2O & \rightarrow & C_2H_2 + 2LiOH \end{array}\right]$$

$$3C_2H_2 \rightarrow C_6H_6$$

The samples are usually first converted to carbon dioxide, which is accomplished by either a combustion or an acid attack. Acetylene must then be produced. Originally procedures based on modifications of the Suess method[15] were used[16, 17] as illustrated in the first series of equations for acetylene production written above. This has now largely been replaced by the molten lithium procedure developed by the radiocarbon dating laboratory of the British Museum[18] and shown in the second group of equations for acetylene. The direct use of charcoal with molten lithium was introduced by the Leningrad radiocarbon dating laboratory[19] and is the simplest of all the reactions. It is the presently preferred method of the Instituto Venezolano de Investigaciones Científicas radiocarbon dating laboratory for most samples and has been seen to give high yields if the temperature is brought up to more than 650°C for a half-hour after the initial reaction. However, the British museum procedure is used when it is desirable to know the exact amount of carbon in the sample, which is the case for studies of ground water and bones.

The conversion of acetylene to benzene was first carried out by a pyrolysis.[16, 17] The use of Reppe,[19] Zeigler,[20] and diborane activated silica-alumina [22] catalysts have produced improved yields. More recently, vanadium and chromium activated silica-alumina catalysts were found to be much more convenient and give yields of almost 100%.[23, 24] This development has brought the complete synthesis of benzene to a high degree of perfection. With total yields approaching 100%, the chemical preparation can be carried out by a laboratory technician in the space of one working day.

The counting procedure for benzene solutions has also been studied. It was shown[25] using a single photomultiplier counter, that the sensitivities increase with sample size only up to a certain point where larger amounts of benzene cause losses in detection efficiency. A 60 cc. counting bottle was capable of producing carbon-14 dates on samples as old as 61,000 years, based on 2σ statistics and 48 hours counting time. Another study of the reproducibility of carbon-14 measurements using synthesized benzene, which is even more important than sensitivity, showed that errors due to causes other than the random nature of the radioactive disintegration process are certainly less than 1% and probably less than $^1/_2$%.[26] This means that isotope effects in the syntheses do not introduce appreciable errors in the carbon-14 dates. Two other potential problems, radon contamination and impurity quenching, were also examined.[27] Radon has never been seen to be present in a synthesized benzene solution. It should be effectively removed by the chemical procedure and, in addition, discriminated against by the counter. Quenching in liquid scintillation counters can be accurately monitored by several procedures now well known. It is absent in the benzene method if tihe acetylene is purified by passing over a phosphoric acid absorber which removes any ammonia present. The use of synthesized benzene as a routine procedure for carbon-14 dating has been reviewed in detail recently.[27] A number of lists of dates obtained by this method have been published.[28-35]

3. Low Level Tritium†

When discussing low levels of tritium, results are often presented in terms of "tritium units" or "T.U.". One T.U. corresponds to a ratio of tritium atoms to hydrogen atoms of 10^{-18}. A convenient factor to re-

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† An expanded version of this section had been privately distributed previously—Packard Technical Bulletin No. 12: "Liquid Scintillation Counting of Low Level Tritium", by M. A. Tamers.

member is that 100 T.U. = 6.5 dpm T/gm hydrogen. The tritium unit is useful because it represents the order of magnitude of tritium in natural waters before contamination by the termonuclear weapon tests that have taken place since 1952 in the northern hemisphere. The T.U. is also approximately the limit of detectible activity using present day counting systems. Some natural tritium concentrations are shown in Table 2.

TABLE 2 Approximate tritium concentrations in northern hemisphere waters, taken or estimated from information contained in references[36–40]. Tritium concentrations in the southern hemisphere before 1952 were the same as in the north. After this date contamination has appeared in the south, but it is considerably less than that present in the northern hemisphere.[41]

	1951	1963
Rain	6 T.U.	100–1000 T.U.
Rivers and shallow lakes	1–6 T.U.	10–1000 T.U.
Underground waters	0–6 T.U.	0–1000 T.U.

The most sensitive arrangement for tritium detection in a liquid scintillation spectrometer involves the viewing of the counting solution by two co-axial photomultiplier tubes. Photomultipliers not exactly in line will result in unnecessarily high backgrounds,[42] presumably caused by Cerenkov radiation in the walls of the tubes. It was also found that a small opaque screen on the edges of the photomultiplier windows reduced the background due to Cerenkov radiation. In the best scintillation solutions, about two-thirds of the tritium beta particles are sufficiently energetic to produce at least two electrons in the photomultiplier cathodes.[43] But with coincidence-type liquid scintillation counters, in order for a tritium decay to be detected a minimum of one electron must be produced in each photomultiplier; two electrons in one photomultiplier and none in the other does not result in a count. About 45% of the tritium disintegrations are capable of this. Therefore, the upper limit for the tritium detection efficiency in a two-photomultiplier liquid scintillation spectrometer will not be more than this. Efficienty will only be improved when better scintillators and photocathodes are developed.

Three separate efforts can be made to lower the count rate of accidental coincidences. First of all, the photomultipliers should be carefully selected from the available stock to have low thermal noise levels. These spurious counts can be reduced further by cooling;[42–44] however, this is not always feasible since the temperature must be high enough to prevent the solidification of the scintillator liquid or the precipitation of one of its components. The thermal noise level of the photomultipliers decreases exponentially with the temperature, but only down to a few degrees below zero centigrade. At lower temperatures, the background decreases very slowly due to residual ion feedback pulses. Consequently, no important background reductions would be obtained by excessive cooling of the photomultipliers. The point of diminishing return with cooling was observed by Bibron to be —6° to —8°C. The water containing scintillator solutions are counted at —7°C, which is just above their freezing point, and, therefore, they can take advantage of noise reduction by photomultiplier cooling.[45] With the use of synthesized benzene, which freezes at 6°C, 15% toluene must be added as an antifreeze so that the photomultipliers and solution can be cooled down to —10°C for tritium counting.[46]

The use of fast electronics resulting in a low value of the resolving time of the coincidence selector will directly affect the coincidence count rate. Having a coincidence unit with a very small resolving time is perhaps more important than finding ultra-low noise photomultipliers. Presently, the best all around photomultiplier for low level tritium work is the 13 stage E.M.I. 6255 S, which was used in four of the system[44-50] listed in Table 3. These tubes, with quartz windows, have such less potassium-40 contamination than the ordinary pyrex glass face tubes.

Water counting in a liquid scintillation spectrometer first become practical for low level tritium when it was shown that the addition of naphthalene to 1,4-dioxane significantly increased the scintillation efficiency.[51] Various improvements have been made on this original solution, the highest scintillation efficiency for tritium being reported with a mixture of naphthalene, dioxane, alcohol, and toluene.[52] However, the most sensitive water counting solutions contain simply dioxane and naphthalene,[48] besides the scintillators. Even though this system has a lower scintillation efficiency, a larger volume of water can be added and the net effect is a greater sensitivity than that of the solution also containing alcohol and toluene. The optimum conditions found by Kaufman *et al.*[48] were naphthalene concentrations of 100 gm/liter dioxane and 18.6% by volume water in the dioxane-PPO-POPOP solution. Boyce and Cameron[49] used the same dioxane-naphthalene concentration, but increased the amount of water to 20.5%. Purification of the dioxane is very important since the addition of water to impure dioxane shows strong luminescence that decays with a half-life of almost a day, thus making the system impractical.

The most sensitive liquid scintillation method for tritium counting involves the synthesis of benzene.[46, 47] This liquid is produced by the following reactions:

$$CaC_2 + 2H_2O \rightarrow C_2H_2 + Ca(OH)_2$$

$$3C_2H_2 \rightarrow C_6H_6$$

The water sample to be measured is added to calcium carbide and acetylene gas obtained. Certain commercially available calcium carbides are made from ancient carbon containing virtually no carbon-14. This type of material must be used here. The acetylene is polymerized to benzene. Originally, this was carried out at 600°C[46] and the final liquid required a purification with bromine and a distillation.[47] However, more recently the diborane and chromium activated silica-alumina catalysts[21, 23, 24] have been tested and it was found that if the acetylene had been previously purified, the resulting benzene could be used directly without further treatment. Large quantities of benzene can be rapidly produced by means of these catalysts.

Half of the water used for the attack of calcium carbide goes into the production of calcium hydroxide, which automatically reduces the chemical yield by 50%. This loss could be avoided by heating the calcium hydroxide formed to 500°C in order to drive off water, but the quantities involved in tritium studies are almost always very large and yields are not as important as in carbon-14 dating. The total yields in the production of benzene for low level tritium measurements are 10–20% in the case of the pyrolysis conversion of the acetylene and almost 50% when the chromium activated silica-alumina catalyst is used. An isotope effect of −34% is present, presumably coming from the acetylene generation step, but it has been shown to be reproducible under varying conditions[47] and will not introduce a large error in the measurements.

Fifteen per cent toluene is added to the benzene solution, as well as the PPO and POPOP scintillators. Using this system, measurements of tritium in northern hemisphere rain were made without the necessity for the troublesome isotope enrichment by electrolysis.[47] The limit of detection of the method, based on 2σ statistics and 48 hours counting, was 6 T.U.

An alternative to the use of water containing or benzene solutions is the isotope exchange method developed by Dostrovsky *et al.*[53] In this procedure the water sample to be measured is reacted with SO_3 gas to produce 50% sulfuric acid. The acid is shaken with toluene for several days and about 25% of the tritium content of the water sample can be transferred to the toluene. This liquid, having good scintillation properties, is then used as the solvent of the counting solution. However, an isotope effect certainly takes place in the exchange reaction between the water and toluene and whether or not this is large and reproducible must be ascertained.

A similar procedure is that introduced by Anbar *et al.*[54] where the hydrogens of cyclohexene are exchanged with a water sample, which has been acidified by trifluoroacetic anhydride. This method represents an improvement over the toluene-water exchange in that it is more rapid and has a smaller loss. However, cyclohexene is a poor scintillator and must be mixed with an equal volume of toluene in order to show approximately the same scintillation efficiency as the water containing scintillator, which is about half that of the benzene scintillation solution. The cyclohexene method is potentially more sensitive than those using water containing solutions, but still less than the benzene method.

The Table 3 summarizes the characteristics of the different liquid scintillation systems studied for low level tritium measurements. The figure of merit used was grams hydrogen counted times counting efficiency divided by the square root of the background. The hydrogen counted includes a factor for isotope effects and incomplete chemical reactions, where applicable.

4. Low Level Chlorine-36

Chlorine-36 is formed on the earth by the action of cosmic ray secondary neutrons with chlorine-35. The cross section for the reaction is 32 barns, a value so high that is has been calculated that in a common igneous rock containing only 0.35% chlorine, 30% of the neutrons are absorbed in the production of chlorine-36.[55] To a smaller extent the isotope is also produced by neutrons liberated from naturally occurring uranium and thorium. The fluxes of cosmic ray neutrons on the surface of the earth increase with altitude and decrease very rapidly with depth in the ground. The concentrations that can be expected have been cal-

Table 3 Low level liquid scintillation counting systems

	Chemical Yield (%)	Hydrogen Counted (gm)	Counting Efficiency (%)	Back-ground (cpm)	Figure merit	Detection limit (T.U.)	Refer-ence
Chemical Synthesis Method							
68 cc benzene in an 80 cc cell	10–50	4.7 × 0.66 = 3.1	29	52	12	6	47
17 cc benzene in a 20 cc cell	10–50	1.2 × 0.66 = 0.79	34	15	6.9	10	47
Water Solution Method							
31.6 cc water in a 170 cc cell	100	3.5	6.9	56	3.4	21	48
16 cc water in a 78 cc cell	100	1.8	14	61	3.2	22	49
1.4 cc water in a 20 cc cell	100	0.16	26	19	1.0	72	50
Exchange Method							
75 cc toluene in a 75 cc cell	—	5.65 × 0.25 = 1.4	25	120	3.2	22	53
Half cyclohexene, half toluene solution	100	—	5.2	—	—	—	54

culated as 0.3 dpm/gm Cl at sea level and 2.3 dpm/gm Cl on a mountain 10,000 ft high.[55] The half-life of chlorine-36 is 308,000 years, which means that it would be potentially useful for dating the entire Pleistocene period, and would overlap several tens of thousands of years with the period of time that can be reached by carbon-14 dating. Therefore, chlorine-36 could be used to verify certain older carbon-14 dates.

Chlorine-36 has been measured in natural systems,[56, 57] but in most materials investigated the counting arrangement was not sensitive enough to detect the low level activities. There appears to be considerable contamination from nuclear weapons testing, which will probably eliminate from study, by this method, surface materials that are water soluble. However, there are still numerous types of samples for which contamination can either be effectively removed or be sure not to have contacted. Examples would be consolidated rocks and some ground water aquifers.

The previous method for low level chlorine-36 detection consisted of counting solid ammonium chloride.[55] The counter was protected by a massive amount of bulk shielding as well as an anti-coincidence guard ring of counting tubes that resulted in a very low background. The sensitivity limiting factor was self-absorption in the ammonium chloride. With the maximum amount of counting material used, 41 grams, the detection efficiency was only about 10%. There seems to be little hope of significantly improving this counter's performance due to the fact that the background is already a minimum value and any additional ammonium chloride would be almost useless because of self-absorption of the beta particles.

The original difficulty with the use of the liquid scintillation counter for measurements of low level chlorine-36 was to find a suitable compound that contained a large amount of chlorine, could be synthesized easily, and was not a quencher. Carbon tetrachloride, for example, fulfilled the first two conditions, but was seen to be a poweful quenching agent and could be used in only the most dilute solutions. The counting of NaCl and HCl dissolved in gel systems[58, 59] permitted only small amounts to be used because of solubility problems. The same disadvantage would be the case if inorganic salts of chlorine were to be counted in water containing scintillation solutions.

After a survey of the various possibilities, it was discovered that the compound silicon tetrachloride offered promise.[60] This liquid contains 83% chlorine, is relatively easily synthesized, has a density of 1.47, is stable, and is not a quencher. The following reactions illustrate the synthetic method employed in the laboratory:

$$2NaCl + K_2S_2O_8 \xrightarrow{70^\circ\,C} Na_2SO_4 + K_2SO_4 + Cl_2$$

$$2Cl_2 + Si \xrightarrow{450^\circ\,C} SiCl_4 \text{ (impure)}$$

$$SiCl_4 \xrightarrow{\text{distillation}} SiCl_4 \text{ (pure)}$$

The compound is used with toluene, in which the PBD scintillator can be dissolved. Other common scintillators present problems with silicon tetrachloride. Also, the solutions must be counted at room temperature since the PBD precipitates out with even a moderate amount of cooling. Various proportions of silicon tetrachloride and toluene were investigated, all with 20 cc total volume and 10 gm PBD/liter solution. The optimum sensitivity was obtained using 11 cc silicon tetrachloride and 9 cc toluene, which permitted a counting efficiency of 55% and a background of 44 cpm. This solution contained 13.4 grams of chlorine.

The liquid scintillation counting method using silicon tetrachloride still has a figure of merit lower than that of the best of the other methods for low level chlorine-36. However, it is capable of being considerably improved. The shielding here consisted of only 5 cm of lead. This can be increased by ten times and an anti-coincidence guard also added in order to reduce the background. Furthermore, the 20 cc counting bottle used is unnecessarily small since the amount of sample material available is often very large and the laboratory synthesis is capable of producing 100 cc or more of silicon tetrachloride in one day. Counting bottles of several hundred cc or even a liter should be feasible. With these minimum improvements, the detection of low level chlorine-36 by the use of silicon tetrachloride solutions in a liquid scintillation counter should greatly surpass in sensitivity the older ammonium chloride procedure.

5. Discussion

The sencitivities of low level counting systems are directly proportional to the amount of material in the counter, but inversely proportional to only the square root of the background. Therefore, it is generally more interesting to make improvements that will increase the counting sample size than to spend an equal effort on means of reducing the background. In liquid scintillation counting high sensitivity is obtained because large quantities of material can be counted without the self-absorption losses that limit solid sample counters or excessive sizes that restrict the gas sample detectors. The most effective means of counting large amounts of material is to synthesize the liquid scintillation solvent from the substance to be measured.

For carbon-14 dating and other low level activities of this isotope, the best liquid scintillation technique consists of the synthesis of benzene, which is then used as the solvent of the counting system. This laboratory technique has been brought to a high state of perfection, both with respect to ease of execution and chemical yield. As for the choice of benzene as the counting solvent, there seems little hope that another liquid will be capable of offering more advantages for carbon-14 dating using the liquid scintillation counter.

The best liquid scintillation technique for low level tritium is, at present, the synthesis of benzene using carbon-14-free calcium carbide. Although the chemical and scintillation properties of benzene are very good, there are still possibilities for improvement. Benzene contains only 8% hydrogen and any saturated hydrocarbon would have more than twice that percentage. Methane, which can be easily liquified, has 25% hydrogen, three times that of benzene. There seems every reason to believe that another compound will be discovered that contains a larger percentage of hydrogen, but still can compete with benzene in so far as the ease of synthesis and scintillation properties are concerned.

Research on the liquid scintillation counting of silicon tetrachloride solutions for low level chlorine-36 has begun only recently. Silicon tetrachloride offers several advantages, especially the rapidity and good yield of its synthesis and its high chlorine content. However, it would be interesting to find another compound that contained as high a percentage of chlorine, but that had scintillation properties comparable to that of toluene and in which it would be possible to directly dissolve the scintillator. In that case, a counting efficiency of more than 90% could be obtained besides larger amounts of chlorine being counted if a second solvent was not required. Until the time when a liquid better than silicon tetrachloride is discovered, considerable work remains to be done using this compound and on the counter in order to reduce the background and increase the counting sample size.

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