

ROUND-TABLE ON 'SUSPENSION COUNTING'

SAMUEL HELF

Picatinny Arsenal, Dover, New Jersey

SUSPENSION counting is used when a labeled compound cannot be dissolved in either a simple or a mixed liquid scintillator. Here, one merely suspends a quantity of the radioactive solid in a simple liquid scintillator, with or without a suspending agent. For these systems the same instrumentation is used as for homogeneous solution counting.

Suspension counting of low-energy beta-emitters has been reported by three different groups: (1) by HAYES, ROGERS and LANGHAM¹ of the Los Alamos Scientific Laboratory, (2) by FUNT and HEATHERINGTON² of the University of Manitoba and (3) by WHITE and HELF³ of Picatinny Arsenal.

The Los Alamos group added their radioactive particles directly into a simple liquid scintillator (PPO-POPOP-toluene) without the use of a suspending agent. Funt and Heatherington prepared their samples in the form of rigid gels using aluminum stearate as a gelling agent. The suspending medium consisted of *p*-terphenyl (4 g/l.)-POPOP (100 mg/l.)-toluene solution containing 70 g/l. of aluminum stearate. Samples were prepared for counting by shaking a quantity of the radioactive precipitate in a 10 ml scintillator volume and heating the glass sample vial to 80°C in order to form the rigid gel. Funt and Heatherington have reported that the resulting gels are completely transparent in the absence of any suspended material.

We used a somewhat different system for counting toluene-insoluble compounds. Our scintillator consists of PPO-toluene solution combined with a thickening agent commercially known as Thixcin. Thixcin is a castor oil derivative, completely organic in nature which when combined with aromatic solvents yields a thixotropic gel possessing excellent supporting strength for solid particles. The gel is prepared by adding 25 g of powdered Thixcin to a liter of liquid scintillator and homogenizing in a blender for about 3 min. The resulting gel is pourable and capable of supporting 1 g of suspended material in a 22 ml volume. No heat is required to effect gelation and this is obviously advantageous for materials that are not stable above room temperature. The addition of the thixotropic agent reduces the optical transmission of the simple solution scintillator by only 10% for scintillations produced by C¹⁴ betas. The addition of POPOP has very little or no effect on the efficiency of suspension counting. Samples are prepared for counting by adding 22 ml of gel scintillator to a glass vial containing a pre-weighed

amount of tagged compound. Vigorous hand-shaking of the closed vial for a few seconds is sufficient to assure the uniform dispersion of the solid in the scintillator.

There are three important factors which tend to reduce the efficiency of suspension counting as compared to homogeneous solution counting. These are:

1. Settling of the suspension which causes a departure from 4π geometry,
2. Opacity of the resulting medium which increases light scattering.
3. Self-absorption of the low-energy betas within the individual solid particles.

With the Los Alamos method, where a suspending agent was not used, settling of the particles was a definite problem. Two methods were used for obtaining the count of a particular sample. In one, the sample was shaken between each of a series of 1 min counts and the results were averaged. In the other, a series of 1 min counts was obtained and the results plotted and extrapolated back to zero time. The former method was preferred for routine assay. However, where good reproducibility and short counting times are important, this settling of the suspension is a decided disadvantage.

Funt and Heatherington report that their aluminum stearate gels showed some settling with C^{14} - $BaCO_3$ particles during the first day after preparation. After the first day there was no additional settling and the counting rate remained stable after three weeks standing. The published data shows that up to 700 mg of $BaCO_3$ can be suspended in a 10 ml scintillator volume.

With the Picatinny gel scintillator, no settling effects at all are observed and experience has shown that the counting rate of a sample remains stable after many weeks of standing without additional shaking required. The counting rates of 10 replicate samples using the same weight of C^{14} -labeled compound in the same volume of gel scintillator agreed to better than $\pm 1\%$.

With regard to opacity effects, the same type of results are obtained with or without the use of a suspending agent. With increasing concentration of solid material, there is a gradual decrease in counting efficiency as the opacity of the suspension increases. For white solids, this opacity effect is a function of the nature of material, being less pronounced with the more transparent crystalline compounds. For colored solids, the same effects are observed as for homogeneous solution counting, i.e. if the color of the resulting medium is strong, light transmission will be poor or non-existent.

The phenomenon of self-absorption is perhaps the most interesting factor in suspension scintillation counting. The Los Alamos group studied self-absorption by determining the ratio of suspension counting efficiency to homogeneous internal-standard counting efficiency and denoted the calculated ratio as the f -value. This ratio is determined by adding an internal standard of the same isotope after absolute counting of the radioactive suspension. The f -value should be independent of scattering or absorption

of light and any deviation of this value from one depends only on the self-absorption of the beta particles. For various concentrations of finely pulverized C^{14} -labeled $BaCO_3$, phenylalanine, liver and bacteria, f -values close to one were obtained, indicating essentially no self-absorption for these suspensions. For H^3 -labeled sodium acetate an f -value of 0.28 was obtained indicating a large degree of self-absorption of the relatively lower energy beta-particles for essentially the same particle-size material.

At Picatinny Arsenal, in our initial work with C^{14} -compounds using the gel scintillator, no special attempts were made to obtain our radioactive materials in the finely divided state necessary to eliminate self-absorption. We wanted to evaluate our suspension counting technique with precipitates as they are normally obtained from our tracer experiments. However, by passing some of our C^{14} precipitates through a series of U.S. standard sieves, we were able to obtain some information on the effects of particle size on suspension counting efficiency. We observed that within a series of powders ranging in particle size from 40 to 250μ , a constant f -value of approximately 0.75 was obtained, thus indicating a constant degree of self-absorption for this range of particles studied. More recent work has demonstrated that by grinding our precipitates down to between 1–10 μ , f -values of 1 can be obtained. Thus, it appears that for C^{14} suspensions, an 'infinite thickness' phenomenon occurs such that above a finite small particle size no additional self-absorption takes place. From a theoretical standpoint, this is difficult to understand and we are doing further work on the influence of particle size on self-absorption.

The following illustrations will demonstrate the type of results which have been obtained by the three different groups on suspension counting:

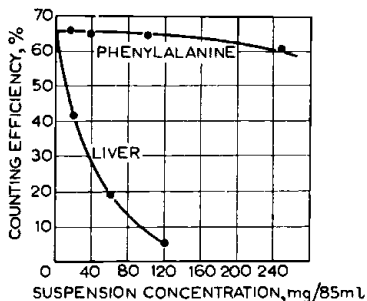


Fig. 1.

Figure 1. HAYES, ROGERS and LANGHAM—Los Alamos, compares suspension counting data between a white crystalline C^{14} compound (phenylalanine) and a highly colored material (liver tissue). There is relatively very little loss in counting efficiency with increasing concentration with the phenylalanine as compared to the liver.

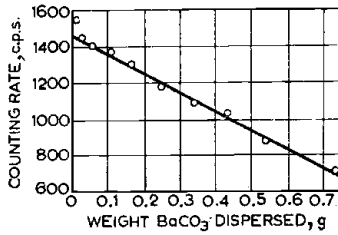


Fig. 2. Variation of counting rate with weight of suspended material at constant activity ($0.05 \mu\text{c}$).

Figure 2. FUNT and HEATHERINGTON—University of Manitoba, shows variation of counting rate with increasing quantities of C^{14} -tagged BaCO_3 of the same total activity suspended in aluminum stearate gels. Since BaCO_3 is a white amorphous material, light-scattering effects due to opacity are fairly pronounced.

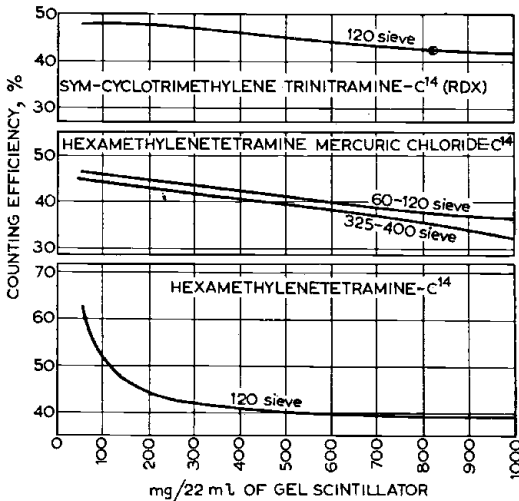


Fig. 3. Counting efficiency vs. concentration for C^{14} -suspensions.

Figure 3. WHITE and HELF—Picatinny Arsenal, shows similar type of data for three different C^{14} -tagged compounds suspended in 'Thixcin' gel scintillator. Suspended material ranges from 50 to 1000 mg in a 22 ml scintillator volume. The upper curve for *sym-cyclotrimethylene trinitramine* (commonly known as the high-explosive, RDX) is analogous to the curve obtained by the Los Alamos group for phenylalanine, i.e. a very small light-scattering effect with increasing quantity of suspended material. The bottom curve for hexamethylenetetramine appears to be an anomalous behavior in that there is an initial sharp decline in counting efficiency at low concentrations and a

gradual leveling off to a relatively constant efficiency with increasing concentrations. This is due to the slight solubility of this material in the scintillator (ca. 2 g/l.). Increasing the amount of solid material thus changes the behavior from that of a non-quenching homogeneous system to that characteristic of a suspension.

The two center curves gives results for the complex mercuric chloride salt of hexamethylenetetramine. Here the effect of particle size was observed by passing a sample through No. 60, 120, 325 and 400 U.S. Standard Sieves, equivalent to 250, 125, 44 and 37 μ respectively. There was no noticeable difference in the counts obtained from equal quantities of 60- and 120-sieve material (the upper curve); surprisingly the 325- and 400-sieve materials gave slightly lower counting efficiencies despite their smaller size. As mentioned previously, the f -values (ratio of suspension counting efficiency to homogeneous internal-standard counting efficiency) for this entire range of particle sizes were the same, indicating a constant degree of self-absorption. Therefore, the lower counting efficiencies exhibited by the smaller particles must be due to a greater degree of light scattering by the finer suspensions.

Table 1. WHITE and HELF—Picatinny Arsenal, illustrates the effect of bottle size and scintillator volume on suspension counting rates. Because a suspension medium is optically more dense than a pure solution scintillator, it was

TABLE I
*Effect of Bottle Size and Scintillator Volume**

Small bottle†		Large bottle‡	
Scintillator volume (ml)	Counting rate (counts/min)	Scintillator volume (ml)	Counting rate (counts/min)
7.5	6050	22.0	4640
15.0	6513	44.0	5173
22.0	6743	66.0	5267

* There were 200 mg of hexamine $.2\text{HgCl}_2$ in each sample.

† Kimble Opticlear, 28 ml capacity, 27 mm o.d.

‡ Kimble weighing bottle, 80 ml capacity, 54 mm o.d.

expected that sample-bottle dimensions and scintillator volumes would be more critical. The table gives the counting rates obtained with 200 mg samples of hexamine- C^{14} . 2HgCl_2 in various volumes of gel scintillator contained in both small and large bottles. With both bottle sizes an increase in gel scintillator volume resulted in increased counting rates. However, the smaller bottle with its narrower absorbing path gives higher counting rates.

REFERENCES

- 1 F. N. HAYES, B. S. ROGERS and W. H. LANGHAM. *Nucleonics* 14 No. 3, 48 (1956).
- 2 B. L. FUNT and A. HETHERINGTON. *Science* 125, 986 (1957).
- 3 C. G. WHITE and S. HELF. *Nucleonics* 14 No. 10, 46 (1956).