

CERENKOV COUNTING AND LIQUID SCINTILLATION COUNTING
FOR THE DETERMINATION OF ^{18}F FLUORINE

D.N. Abrams, S.A. McQuarrie, C. Ediss, and L.I. Wiebe
Division of Bionucleonics and Radiopharmacy
University of Alberta
Edmonton, Alberta

Abstract

The positron emitted (E_{max} 635 KeV) upon radioactive decay of ^{18}F has been used to measure ^{18}F in liquid scintillator solutions which have low or high water capacity, and in hydrophilic and hydrophobic liquids which have different refractive indices. The influence of nitromethane, a chemical quenching agent, on counting efficiency in each of these liquids has been measured, and is discussed on the basis of observed shifts in the pulse height spectra. The contribution of coincident Compton events arising from the annihilation gamma rays, to the overall counting efficiency, has been estimated using methyl salicylate as the counting medium.

Introduction

Fluorine-18 is a short lived ($T_{1/2} = 110$ min) radionuclide which decays primarily (97%) by the emission of positrons (β^+ ; E_{max} 635 KeV), and some (3%) electron capture.¹ Annihilation γ rays (511 KeV, 194%) and x-rays are associated with these respective decay modes. It is the coincident γ -rays that are most frequently used in the radioassay of this and other positron emitting radionuclides.

Although most radioisotopes analysed by liquid scintillation counting are either α or β^- emitters², and those assayed by Cerenkov counting in liquid solution are β^- emitters³, positrons can be counted equally well by these methods.

We now report preliminary observations from the radioassay of ^{18}F using liquid scintillation fluors (toluene/PPO/POPOP, and Aquasol^{T.M.}), water and methyl salicylate as photon generating media for counting with commercial liquid scintillation spectrometers.

Experimental

Fluorine-18 was produced by irradiation of ^{20}Ne gas with 6.5 Mev neutrons using the 7 MV Van de Graaff electrostatic generator at the University of Alberta. Fluorine-18 was recovered from the glass target vessel with a methanol flush. Carrier NaF (1 mg ml^{-1}) was added to that solution prior to liquid scintillation radioassay. The ^{18}F content of an aliquot of the final solution was standardized by γ - γ coincidence counting of the 511 Kev annihilation photons, using a $1\frac{3}{4} \times 2$ " NaI well crystal detector. After cross calibration with ^{137}Cs (11% efficiency), a final counting efficiency of 35 ± 5 percent was calculated. This value was used to estimate the counting efficiency of the liquid scintillation (LS) and Cerenkov (C) procedures.

Aliquots (10 μl) of the Na^{18}F -methanol solution were accurately pipetted into glass scintillation vials (Kimble, Toledo) containing 15 ml of either distilled water, methyl salicylate (MS) (redistilled, Reagent Grade, Fisher Scientific, Edmonton), Aquasol^{T.M.} or toluene containing PPO (4 g l^{-1}) and POPOP (50 mg l^{-1}) (Scintillation grades, Fisher Scientific, Edmonton). Samples were counted in the LS spectrometers, and then all but the water samples were quenched with 200 μl of nitromethane (reagent grade, Fisher Scientific, Edmonton) and counted again. Water (200 μl) was added to one Aquasol sample as an alternate quenching agent.

To determine the contribution of pulses arising as a result of the interaction of the 511 Kev coincident annihilation γ 's, a liquid scintillation vial was fitted with a stainless steel cylindrical tube which was open at the upper end but sealed at the bottom (Figure 1). An aliquot (10 μl) of the Na^{18}F /methanol solution was introduced into the tube, and the tube was then inserted in a LS vial containing 15 ml of MS. The sample was counted, the tube removed, and then recounted to ensure that the generating liquid was free of ^{18}F contamination. That vial was then recounted containing 10 μl of the Na^{18}F methanol intimately mixed with the MS in the vial.

All samples were counted using both the Liquimat 220 (Picker Nuclear, New York) and the Mark III (Searle Instrumentation, Chicago) LS spectrometers. Pulse height spectra (PHS) were stored in a Northern Scientific NS636 multi-channel analyzer.

All count rates were decay corrected to the time of standardization of the Na^{18}F solution.

COUNTING FOR ^{18}F FLOURINE

Table 1. Counting efficiency and figure of merit (E^2/B for ^{18}F in toluene/POP/POPOP (TPP), Aquasol^{T.M.}, methyl salicylate (MS) and water, using the Picker Liquimat 220 and the Searle Mark III LS spectrometers. NM indicates the addition of 200 μl of nitromethane, H_2O the addition of 200 μl of water. Counting precision is estimated to be ± 10 percent.

Generating System	Liquimat 220 % E	Mark III (^{32}P window) % E	E^2/B
TPP	80.7	78.1	135.6
TPP + NM	53.1	56.7	
Aquasol	98.2	99.3	200.8
Aquasol + H_2O	98.7	98.5	
Aquasol + NM	87.0	90.3	
MS	78.8	85.9	282.7
MS + NM	70.2	76.6	
H_2O	2.3	3.7	0.41

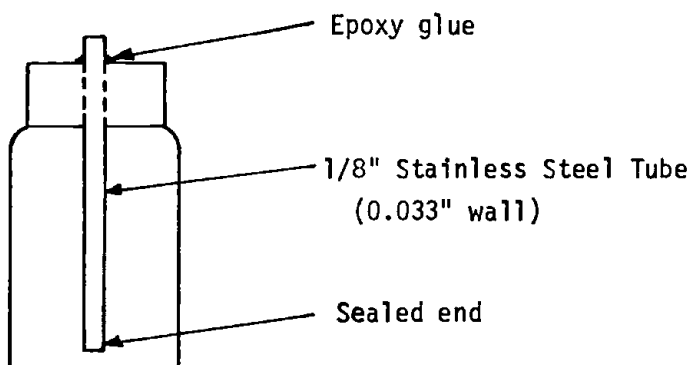


Figure 1. Modified liquid scintillation vial for determination of Compton electron contribution to ^{18}F Cerenkov pulse height spectrum.

Results and Discussion

Counting efficiencies observed for ^{18}F in the various media both with and without the addition of the chemical quenching agent (nitromethane) are presented in Table 1, and Pulse height spectra for ^{18}F in TPP, Aquasol and MS are depicted in Figure 2. Of particular interest are the high counting efficiencies (essentially 100%) observed using Aquasol and the relatively high counting efficiencies (78.8%) using MS, compared with TPP (80.7%). MS has been shown to be an excellent Cerenkov counting medium, with its high refractive index (1.522) and wave-shifting characteristics.⁴ Counting efficiencies for ^{36}Cl (β^- E_{max} 714 Kev) in MS were 82.2 and 91.6 percent of TPP values respectively for the Liquimat 220 and Mark III LS spectrometers. Furthermore, Aquasol has been found to be less efficient than TPP when counting weak β^- emitters⁵, and TPP has been widely used to obtain efficiencies approaching 100 percent for most energetic β^- emitting nuclides. Dissolution of Na^{18}F is a possible explanation of the phenomenon. The low MS background, even in the wide window necessary for LS counting, gives the MS system a distinct advantage over the LS systems in terms of figures of merit (E^2/B) (Table 1).

The annihilation gamma photons were found to produce a count rate of 9.6 percent of that observed when the ^{18}F (hence the β^+ s) was dissolved directly in the methyl salicylate. Although pulses derived via the gamma interactions would not likely contribute to the count rate, they could well decrease the sensitivity of the system to chemical or to color quench.

The counting efficiency for ^{18}F in water was found to be too low to be of any practical interest. The PHS observed was similar to that reported for ^{32}P in water, using the same LS spectrometer.⁶

Figures 3 and 4 depict the pulse height spectra obtained for ^{18}F positrons and ^{137}Cs Compton electrons (External standard) in MS, using the Liquimat 220 LS spectrometer. It is apparent that the pulse height ranges are virtually identical, although there are qualitative differences in the nature of the spectral shifts when the systems are quenched. It has been shown that a sufficiently high count rate can be obtained from the ^{137}Cs Liquimat 220 external standard source to allow precise standardization for ^{36}Cl samples by the ESCR method.⁴ Similarities between the ^{18}F and ^{36}Cl PHS in MS would lead to the conclusion that ESCR quench correction would be ideal for ^{18}F in MS.

COUNTING FOR ^{18}F FLOURINE

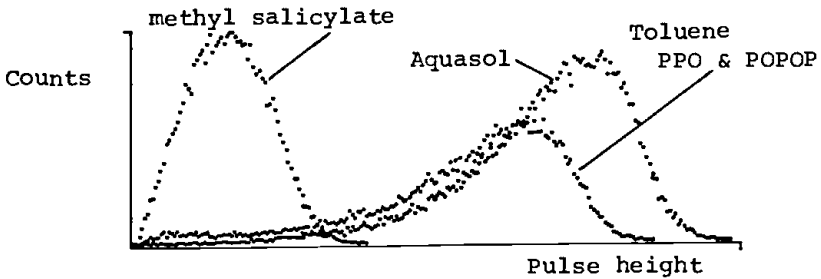


Figure 2. Pulse height spectrum of ^{18}F positrons in methyl salicylate, aquasol & toluene PPO & POPOP.

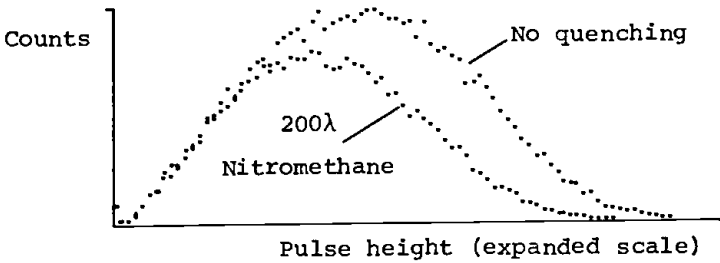


Figure 3. Pulse height spectrum of ^{18}F positrons in methyl salicylate with and without quenching.

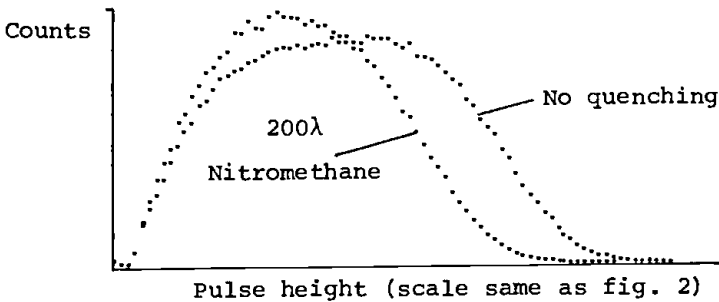


Figure 4. Pulse height spectrum of the Liquimat 220 ^{137}Cs external standard in methyl salicylate with and without quenching.

References

1. C.M. Lederer, J.M. Hollander, and I. Perlman, *Table of Isotopes*, 6th Ed., J. Wiley and Sons, N.Y. (1968) p. 7.
2. A.A. Moghissi, *in* *The Current Status of Liquid Scintillation Counting*, E.D. Bransome, Jr. (Ed.), Grune and Stratton, N.Y. (1970) p. 86.
3. H.H. Ross and G.T. Rasmussen, *in* *Liquid Scintillation Counting: Recent Developments*, P.E. Stanley and B.A. Scoggins (Eds.), Academic Press, N.Y. (1974) p. 363.
4. L.I. Wiebe and C. Ediss, *Int. J. Appl. Radiat. Isotopes* (in press) (abstract).
5. L.I. Wiebe, A. Stevens, A.A. Noujaim and C. Ediss, *Int. J. Appl. Radiat. Isotopes*, 22, 663 (1971).
6. L.I. Wiebe, A.A. Noujaim and C. Ediss, *Int. J. Appl. Radiat. Isotopes*, 22, 463 (1971).