



ELSEVIER

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Micelle size effect on Fe-55 liquid scintillation efficiency



Denis E. Bergeron*, Lizbeth Laureano-Pérez

Physical Measurements Laboratory, National Institute of Standards and Technology, 100 Bureau Dr., Gaithersburg, MD 20899-8462, USA

HIGHLIGHTS

- No efficiency reductions attributable to the micelle size effect were observed.
- Calculated micelle corrections are small compared to other uncertainties.
- Metal ions did not affect micelle size or fluorescence quenching.
- Tracing ^{55}Fe efficiencies with ^{54}Mn reduced model dependence.

ARTICLE INFO

Available online 1 December 2013

Keywords:

Reverse micelle

Liquid scintillation counting

Cocktail

Mn-54

H-3

ABSTRACT

We used efficiency tracing techniques to study the micelle size effect on liquid scintillation counting of the electron capture nuclide, ^{55}Fe . We determined micelle hydrodynamic diameters for specific LS cocktails via dynamic light scattering, and sought trends in efficiencies as a function of micelle size. The presence of Fe^{3+} or Mn^{2+} ions in the cocktails did not significantly affect micelle sizes or fluorescence quenching. We did not detect any reductions in counting efficiencies due to the micelle size effect.

Published by Elsevier Ltd.

1. Introduction

We recently undertook a series of dynamic light scattering (DLS) measurements to determine the size of micelles in several commercial liquid scintillation (LS) cocktails (Bergeron, 2012). An electron emitted from a radionuclide residing within a reverse micelle will lose energy to the aqueous medium before interacting with the scintillant residing in the organic phase, resulting in a reduction in counting efficiency typically referred to as “the micelle effect” or “micelle size effect” (Grau Carles, 2007; Kossert and Grau Carles, 2008, 2010). Our measurements revealed that micelles in commercial LS cocktails are smaller than previously assumed (based on measurements by Rodríguez et al., 1998), so that the micelle size effect is smaller than was thought. We showed that even for the electron capture nuclide ^{55}Fe , the effect would reduce counting efficiencies by $< 0.2\%$ under typical counting conditions (Bergeron, 2012).

In this work, we set out to determine whether it might be possible to observe the micelle size effect under ordinary experimental conditions. We selected ^{55}Fe because, as has been described previously (Grau Carles, 2006, 2007; Kossert and Grau Carles, 2010; Bergeron, 2012), its low energy Auger electron emissions make it particularly sensitive to the effect. Through careful cocktail selection, we sought to maximize the achievable range of micelle sizes. We were also careful to select cocktails with ionic and nonionic surfactants since the

location of the nuclide within the micelle, which may be affected by the surfactant type (Pileni et al., 1985; Pant et al., 1998; Andrade and Costa, 2002; Faeder and Ladanyi, 2000, 2001, 2005; Stahla et al., 2008), should affect the magnitude of the efficiency reduction. Since the variation of micelle size is achieved by varying the total aqueous fraction in the cocktail, and since the addition of water changes the general quenching properties of the cocktail, the experimental scheme included CIEMAT/NIST efficiency tracing (CNET) with both ^3H and ^{54}Mn . It was hoped that careful efficiency tracing would help to minimize contributions from quenching mechanisms that might obscure the detection of the micelle size effect.

2. Methods

2.1. Sample preparation

Two experiments were performed. All of the selected scintillants were diisopropyl naphthalene (DIN) based. Table 1 provides details on the scintillation cocktails used in both experiments. In Experiment 1, cocktails were prepared from HiSafe II and HiSafe III (PerkinElmer, Waltham, MA, USA).¹ The scintillants were

¹ Certain commercial equipment, instruments, or materials are identified in this paper to foster understanding. Such identification does not imply recommendation by the National Institute of Standards and Technology, nor does it imply that the materials or equipment identified are necessarily the best available for the purpose.

* Corresponding author. Tel.: +1 301 975 2282; fax: +1 301 926 7416.
E-mail addresses: bergeron@nist.gov, denis.bergeron@nist.gov (D.E. Bergeron).

Table 1

Summary of the LS cocktails used in the experiments. Each series includes N_s separate samples, each with a different total aqueous fraction, f , covering the indicated range. C_{Fe} and C_{Mn} indicate the total concentration of the metal ions; the metal ions are introduced as carrier solution, or (indicated by italicization) standard solutions of the appropriate radionuclide. For more details on the surfactants and other components of the scintillants, refer to the manufacturer's specifications and material safety data sheets.

Radionuclide solution	Scintillant	Surfactant information	N_s	$C_{Fe}/(\mu\text{g g}^{-1})$	$C_{Mn}/(\mu\text{g g}^{-1})$	f
<i>Experiment 1</i>						
^{55}Fe	HiSafe II	In addition to nonionic surfactants, includes 10–20% sodium dioctyl sulphosuccinate (anionic surfactant)	6	0.065		0.02–0.17
^3H			6	0.065		0.02–0.17
Blank	HiSafe III	20% to 40% ethoxylate polymers (nonionic surfactants)	8	0 or 0.065		0.02–0.17
^{55}Fe			5	0.065		0.09–0.17
^3H			5	0.065		0.10–0.17
Blank			7	0 or 0.065		0.09–0.17
<i>Experiment 2</i>						
^{55}Fe	Ultima Gold	In addition to nonionic surfactants, includes $\leq 2.5\%$ sodium dioctyl sulphosuccinate (anionic surfactant)	6	0.67	0.50	0.03–0.16
^{54}Mn			6	0.67	0.54	0.05–0.17
^3H	HiSafe III	20–40% Ethoxylate polymers (nonionic surfactants)	8	0 or 0.67	0 or 0.50	0.05–0.17
Blank			7	0 or 0.67	0 or 0.50	0.05–0.17
^{55}Fe			5	0.67	0.50	0.04–0.18
^{54}Mn			5	0.67	0.54	0.05–0.17
^3H			7	0 or 0.67	0 or 0.50	0.05–0.17
Blank			6	0 or 0.67	0 or 0.50	0.05–0.17

selected to include one formulation that contains an ionic surfactant, and one that includes only non-ionic surfactants (Table 1). Matched ^{55}Fe , ^3H , and blank cocktails were prepared with a range of aqueous fractions ($f = v_w/v_{\text{tot}}$; where v_w is the total aqueous sample volume and v_{tot} is the total cocktail volume, including added aqueous sample) selected to maximize the range of micelle sizes as measured by DLS in terms of hydrodynamic diameter (Bergeron, 2012). The ^{55}Fe cocktails were prepared gravimetrically from a 14.2 kBq g^{-1} ^{55}Fe solution with $19 \mu\text{g g}^{-1}$ of Fe^{3+} (as FeCl_3) in 1 mol L^{-1} HCl (NIST, 2006). The matched ^3H (NIST, 2008) cocktails and blanks contained Fe^{3+} carrier solution. While the Fe^{3+} content for all cocktails was the same, the desired range of f was achieved via addition of different amounts of distilled water, so that the cocktails with highest f had the lowest Fe^{3+} concentration. In Experiment 2, HiSafe II was replaced by Ultima Gold (PerkinElmer, Waltham, MA, USA). All cocktails contained the same total amount of Fe^{3+} and Mn^{2+} , but different amounts of distilled water to achieve the desired range of values for f . In addition to the matched series, ^3H sources for each scintillant were prepared without any Fe^{3+} or Mn^{2+} in order to quantify quench effects owing to the presence of the metal ions.

In both experiments, scintillant was dispensed via dispensette while distilled water was added by micropipette. In Experiment 1, carrier solution was added via pycnometer, while in Experiment 2, carrier solutions were added by micropipette. All other additions were performed gravimetrically via pycnometer. 20 mL high performance glass LS vials with poly-cone lined urea screw caps (PerkinElmer) were used in all experiments.

2.2. Dynamic light scattering

DLS measurements were made using a Zetasizer Nano ZS (Malvern Instruments, Inc., Westborough, MA, USA), using the same protocols as in our previous work (Bergeron, 2012). Briefly, samples in quartz cuvettes were allowed to equilibrate to 20°C for 60 s prior to three measurement cycles of ten 10 s scans (for a total measurement time of 300 s). Input values for viscosities and refractive indices (of the pure scintillants) were provided by PerkinElmer, and average hydrodynamic diameters (defined as the diameter of a hard sphere that diffuses at the same rate as the particle being measured) generated by the instrument software were recorded and uncertainties handled as described previously (Bergeron, 2012). All blank cocktails were measured.

2.3. Liquid scintillation counting efficiencies

LS sources were counted on three commercial instruments: a Beckman LS6500, a Packard Tri-Carb A2500 TR (PerkinElmer, USA), and a Wallac 1414 Winspectral (PerkinElmer, USA) instrument. The different operating conditions of the three counters helps to eliminate instrumental idiosyncrasies as a source of bias in the comparison of counting efficiencies (Laureano-Pérez et al., 2007).

After preparation, sources were dark adapted for at least 1 h before counting. After three counting cycles, the blanks were removed for DLS measurements (vide supra). In Experiment 1, the initial cycles revealed an overall standard deviation on the count rate from all blanks of < 0.05 counts per second ($\approx 5\%$), and no systematic relationship between the blank count rate and the aqueous fraction, so subsequent measurement cycles included only a subset (2 for each cocktail) of the original 15 blanks. In Experiment 2, the standard deviation on the count rate in the initial cycles was > 0.2 counts per second ($\approx 13\%$), and so all blanks were counted in subsequent cycles so that the appropriate matched blank could provide the background subtraction data for its complementary ^3H , ^{54}Mn , and ^{55}Fe cocktails.

2.4. MICELLE2 calculations

Historically, difficulties in calculating LS counting efficiencies for electron capture nuclides, especially those emitting low-energy Auger electrons, arose from reliance on overly simple models for atomic rearrangement processes (Günther, 1998; Kossert and Grau Carles, 2006, 2010). Recently, the stochastic approach to atomic rearrangement as implemented in the MICELLE2 code has produced better agreement with experimental results, reducing typical discrepancies with experiment to $\leq 2\%$ (Kossert and Grau Carles, 2010).

In this study, MICELLE2 was run for each specific cocktail formulation. In Experiment 1, calculations were run using the two models for source distribution within the micelles (central and random) and ignoring the micelle effect. The actual difference between the corrections calculated by the central model and the random model is very small ($\leq 3 \times 10^{-4}$ counts per decay in this study), and so in Experiment 2, the central distribution model was dropped. For each sample, simulations (each with 5×10^4 events) were run with the appropriate micelle radius and percent added water entered into the CTL.DAT and EFFCOMP.DAT files, respectively. The ^3H efficiencies from the H3X.TAB output file are based on an analytical method and were not used in this study; instead, separate simulations were run for each specific ^3H

Download English Version:

<https://daneshyari.com/en/article/1878668>

Download Persian Version:

<https://daneshyari.com/article/1878668>

[Daneshyari.com](https://daneshyari.com)