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# Measurement of wavelength-dependent refractive indices of liquid scintillation cocktails

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## HIGHLIGHTS

- Refractive indices of several liquid scintillation cocktails were measured.
- The wavelengths cover a range from 404.7 nm to 706.5 nm.
- Measurements were carried out at 16 °C, 18 °C, 20 °C and 22 °C.
- For some cocktails, mixtures with water or nitromethane were studied.

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## ABSTRACT

Refractive indices of several commercial liquid scintillation cocktails were measured by means of an automatic critical-angle dispersion refractometer in the wavelength range from 404.7 nm to 706.5 nm. The results are needed for various applications. In particular, detailed Monte Carlo simulations of liquid scintillation counters that include the computation of optical light require these data. In addition, the refractive index is an important parameter for studies of micelle sizes by means of dynamic light scattering. In this work, the refractive indices were determined for Ultima Gold™, Ultima Gold™ F, Ultima Gold™ LLT, Ultima Gold™ AB, Hionic Fluor™, Permafluor® *E*+, Mineral Oil Scintillator, Insta-Gel Plus, OptiPhase HiSafe 2, OptiPhase HiSafe 3, Ultima Gold™ XR, Insta-Gel Plus, AquaLight, MaxiLight and Ultima Gold™ MV at 16 °C, 18 °C, 20 °C and 22 °C. The carbon dioxide absorber Carbo-Sorb® *E* was also analyzed.

For some scintillators, various batches were compared and mixtures with water or nitromethane were studied.



## 1. Introduction

Liquid scintillation (LS) counting is widely used for activity determination and can be considered as an active research field (Broda et al., 2007). Some researchers are developing Monte Carlo simulations which comprise the modelling of the scintillation photons (Hurtado et al., 2009; Bobin et al., 2012; Thiam et al., 2012). Such simulations require excellent knowledge of material parameters since the transmission, reflection and refraction of light is emulated using software packages, like the GEANT4 tool (Agostinelli et al., 2003). The refractive index of scintillation cocktails belongs to these parameters. For accurate simulations, it must be taken into account that the refractive index depends on the wavelength of light. When the emitted light is not monochromatic, as in the case of Čerenkov light, dispersion plays an important role.

In many cases, LS counting is used to measure aqueous radioactive solutions. Thus, the cocktails usually contain a surfactant which ensures the miscibility of the organic and the aqueous phases. The radioactivity usually remains within the aqueous phase which is embedded in the organic phases in the form of (reverse) micelles. Electrons which are ejected as a result of a radioactive decay may lose a part of their kinetic energy and, consequently, only a reduced energy remains to create scintillation light (Grau Carles, 2007). The effects are very small in LS samples with low water content (Kossert and Grau Carles, 2010; Bergeron, 2012), but they might increase when using gel samples with high water content. Tarancón Sanz and Kossert (2011) have also shown that the general idea of subtracting the energy loss in the aqueous phase works well when using plastic scintillation beads in an aqueous radioactive medium. The energy loss within the micelles and the corresponding reduction of the counting efficiency are referred to as the micelle size effect. For accurate studies of this effect, it is crucial to know the dimensions of micelles. Rodríguez et al. (1998) and Bergeron (2012) have shown that the size of

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micelles can be evaluated by applying the Stokes–Einstein equation. To this end, the viscosity and the diffusion coefficient are required. The latter can be measured by means of dynamic light scattering (DLS). This measurement technique, however, also requires information about the refractive index of the samples under study. Thus, mixtures of LS cocktails and water as well as mixtures of cocktails and the quenching agent nitromethane ($CH_3NO_2$) were included in this study. These measurements are of course also important since most activity measurements are carried out with corresponding mixtures.

## 2. Description of measurements

The measurements were carried out with an automated multi-wavelength refractometer DSR-λ (Schmidt+Haensch, 2010) between 17 January and 13 June 2013.

According to the manufacturer's manual, the instrument can be used to measure the "refractive index of liquid media independent of opacity, viscosity and colour". The refractive index is determined via the measurement of the critical angle for total reflection. To this end, the position of the dark line is determined by means of a high resolution CCD linear array with 2048 elements. Seven LEDs are part of the system, allowing the sequential measurements at different wavelengths in the range from 404.7 nm to 706.5 nm. The system comprises a heater as well as a Peltier cooler and can be used for measurements in a temperature range from 10 °C to 80 °C. The system used in this work allows measurements of the refractive index in a range from 1.33 to 1.70 at 589 nm. According to the manufacturer's description, the accuracy is ±0.0001 at 20 °C.

In this work, small volumes of about 1 mL of the liquid to be measured were placed on the surface of the sapphire prism. All measurements were carried out according to the following sequence:

– Cleaning of the measurement chamber and the sapphire surface
– Measurement with distilled water to ensure sufficient cleaning
– Cleaning and drying of the prism surface
– Placing the sample to be measured on the surface
– After waiting period of several minutes to ensure stable and homogeneous temperature distribution, the automated measurement is started with 5 repetitions at each of the 9 wavelengths. A waiting period of two minutes between the measurements was adjusted using the instrument software.

In this way, several commercial LS cocktails as well as the carbon dioxide absorber CarboSorb E were measured at 16 °C, 18 °C, 20 °C and 22 °C. Information on these liquids is summarized in Table 1. In addition, some measurements were carried out using mixtures of scintillation cocktail and water. We define the ratio $V_r = V_{water}/V_{scintillator}$, where $V_{water}$ and $V_{scintillator}$ are the volumes of water and the scintillator, respectively. LS samples with $V_r=0.067$, $V_r=0.033$ and $V_r=0.010$ were prepared using the scintillators Ultima Gold and AquaLight, respectively. The ratio $V_r=0.067$ is also obtained when mixing 15 mL of a scintillator and 1 mL of water. Such a sample composition is frequently used at PTB. In addition, the influence of the chemical quenching agent nitromethane was studied. To this end, a mixture of nitromethane and pseudocumene (1:1 by volume) was prepared and small amounts of this quenching agent were added to pure Ultima Gold as well as to a mixture of Ultima Gold and water with $V_r=0.067$. All liquids were stored at a room temperature of about 20 °C in a dark place prior to the measurements.

The measurements with distilled water were also used as a validation of the experiment in a manner similar to that conducted

**Table 2**
Measured refractive index of distilled water at 20 °C as measured in this work compared to reference values, $n_{ref}(\lambda)$, which were calculated according to Thormählen et al. (1985) for 19.993 °C.

| $\lambda$ in nm | $n(\lambda)$ this work | $n_{ref}(\lambda)$ | $n(\lambda)-n_{ref}(\lambda)$ |
|---|---|---|---|
| 404.7 | 1.3428 | 1.3432 | −0.0004 |
| 435.8 | 1.3403 | 1.3408 | −0.0005 |
| 486.1 | 1.3372 | 1.3377 | −0.0005 |
| 546.1 | 1.3345 | 1.3350 | −0.0005 |
| 587.6 | 1.3331 | 1.3335 | −0.0004 |
| 589.3 | 1.3330 | 1.3334 | −0.0004 |
| 632.8 | 1.3318 | 1.3321 | −0.0003 |
| 656.3 | 1.3312 | 1.3314 | −0.0002 |
| 706.5 | 1.3301 | 1.3301 | −0.0001 |

**Table 1**
LS cocktails and CarboSorb *E* used in this study.

| No. | Name of LS cocktail | Solvent | Expiry date | LOT no. |
|---|---|---|---|---|
| 1 | Ultima Gold | DIN | January 2011 | 77-090601 |
| 2 | Ultima Gold F | DIN | July 2010 | 78-090601 |
| 3 | Ultima Gold LLT | DIN | June 2014 | 97-1121 |
| 4 | Ultima Gold AB | DIN | May 2006 | 91-050501 |
| 5 | Hionic Fluor | Pseudocumene | April 2007 | 55-040901 |
| 6 | CarboSorb E | – | June 2010 | 99-071102 |
| 7 | Permafluor E+ | Pseudocumene | July 2012 | 90-091201 |
| 8 | Permafluor E+ | Pseudocumene | March 2014 | 90-11311 |
| 9 | High Efficiency Mineral Oil Scintillator | White mineral oil | March 2013 | 137-100801 |
| 10 | Insta-Gel Plus | Pseudocumene | December 2008 | 95-060501 |
| 11 | OptiPhase 'HiSafe' 2 | DIN | Unknown | 1222836 |
| 12 | OptiPhase 'HiSafe' 3 | DIN | 2003 | 0416733 |
| 13 | Ultima Gold | DIN | July 2014 | 77-12501 |
| 14 | Ultima Gold AB | DIN | January 2015 | 91-12241 |
| 15 | Ultima Gold F | DIN | January 2014 | 78-12491 |
| 16 | Ultima Gold LLT | DIN | June 2014 | 97-12471 |
| 17 | Ultima Gold XR | DIN | May 2014 | 79-12441 |
| 18 | Insta-Gel Plus | Pseudocumene | June 2015 | 95-12451 |
| 19 | Hionic Fluor | Pseudocumene | April 2015 | 55-12361 |
| 20 | AquaLight | DIN | 2014 | Unknown |
| 21 | MaxiLight | DIN | 2014 | Unknown |
| 22 | Ultima Gold MV | DIN | August 2015 | 80–13051 |