

CALIBRATION OF DUAL-ENERGY GAMMA SYSTEMS FOR DETERMINING LIQUID SATURATIONS DURING MULTIPHASE FLOW IN SOIL

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Abstract. The purpose of this study was to calibrate a dual-energy gamma system for simultaneous determination of aqueous and non-aqueous phase liquid (NAPL) saturations in soil columns. A dual-energy gamma system containing ^{241}Am and ^{137}Cs was used to study the infiltration and displacement of aqueous liquid by NAPL and vis versa. Distilled water, NaI solution, and Soltrol 130 were used as fluids. The system was calibrated to minimize errors in liquid saturations due to the random nature of gamma photons emission and drift in the photon detection equipments.

The measurement accuracy in liquid saturations was substantially improved by increasing counting time and fluid attenuation coefficient. NaI salt was used to increase the attenuation coefficient of water. Measured and predicted liquid saturations during immiscible displacement in soil were consistent with mass balance measurements and anticipated deviations in saturations from probable error calculations. The calibration procedure resulted in a significant improvement in the prediction of liquid saturations using dual-energy gamma system.

Key words: radiation attenuation, infiltration, multiphase flow, immiscible displacement

INTRODUCTION

Dual-energy gamma systems have been used for simultaneous determination of soil bulk density and moisture content and recently been used for simultaneous determination of volumetric contents of oil and water in soil columns. Errors associated with these measurements, due to the random nature of gamma ray emission and the

sorption characteristics of these rays in soil [5], are likely to be problematic for measurement of liquid saturations in soil columns. Bali and Grismer [2] have evaluated the various errors associated with the simultaneous determination of volumetric contents of non-aqueous phase liquid (NAPL) and water in soil columns. Herein, we focus on the calibration of dual-energy gamma systems for simultaneous determination of volumetric liquid contents or saturations of NAPL and water in soil columns as well as for determination of soil bulk density.

THEORY

The general attenuation equation for a soil cell which contains NAPL and water is given by:

$$I = I_e \exp(-\beta_o S_o \Phi X - \beta_w S_w \Phi X - \mu_{sp} X) \quad (1)$$

where I is the total gamma count for a cell which contains soil, NAPL, and water, I_e is the total gamma count through an empty cell, β_o is the attenuation coefficient for NAPL (cm^{-1}), β_w is the attenuation coefficient for water (cm^{-1}), Φ is porosity (cm^3/cm^3), S_w is water saturation, S_o is oil saturation,

μ_s is the mass absorption coefficient of a given soil (cm^2/g), X is the radiation path length (cm), and ρ_b is the bulk density of the soil (g/cm^3). The attenuation coefficient, β equals $\mu\rho$ where μ is the mass absorption coefficient of the material and ρ is the material density.

Equation (1) may be written for count rates using two different energy sources [5,7]; Americium (^{241}Am) and caesium (^{137}Cs) are commonly used, however, any two different gamma energy sources could be used if their mass absorption coefficients are different [3]. The count rate obtained from an americium source is given by:

$$I_a = I_{ea} \exp(-\beta_{oa} S_o \Phi X - \beta_{wa} S_w \Phi X - \mu_{sa} \rho_b X) \quad (2)$$

and similarly for a caesium source:

$$I_c = I_{ec} \exp(-\beta_{oc} S_o \Phi X - \beta_{wc} S_w \Phi X - \mu_{sc} \rho_b X) \quad (3)$$

where the subscript a and c refers to americium and caesium, respectively.

Equations (2) and (3) can be solved simultaneously for the saturations, S_w and S_o yielding:

$$S_w = \frac{\beta_{oc} \ln[I_a/I_{ea}] - \beta_{oa} \ln[I_c/I_{ec}]}{\Phi X (\beta_{oa} \beta_{wc} - \beta_{oc} \beta_{wa})} + \frac{(\beta_{oc} \mu_{sa} - \beta_{oa} \mu_{sc}) \rho_b}{\Phi (\beta_{oa} \beta_{wc} - \beta_{oc} \beta_{wa})} \quad (4)$$

and

$$S_o = \frac{\beta_{wc} \ln[I_a/I_{ea}] - \beta_{wa} \ln[I_c/I_{ec}]}{\Phi X (\beta_{wa} \beta_{oc} - \beta_{wc} \beta_{oa})} + \frac{(\beta_{wc} \mu_{sa} - \beta_{wa} \mu_{sc}) \rho_b}{\Phi (\beta_{wa} \beta_{oc} - \beta_{wc} \beta_{oa})} \quad (5)$$

and when S_w and S_o have been calculated, the volume fraction of gas-filled pores, S_g , is

given by $S_g = 1 - S_w - S_o$.

The probable errors in S_w and S_o due to the random emissions from the radioactive sources are inversely proportional to the counting rate or counting time, and are implicitly functions of water and NAPL saturations, path length, soil attenuation coefficient, and porosity.

The probable error for any measured value of S_w and S_o can be calculated from (1):

$$\sigma_{S_w}^2 = \frac{1}{[\Phi X (\beta_{oa} \beta_{wc} - \beta_{oc} \beta_{wa})]^2} \left(\frac{\beta_{oc}^2}{I_a} + \frac{\beta_{oc}^2}{I_{ea}} + \frac{\beta_{oa}^2}{I_c} + \frac{\beta_{oa}^2}{I_{ca}} \right) \quad (6)$$

and

$$\sigma_{S_o}^2 = \frac{1}{[\Phi X (\beta_{wa} \beta_{oc} - \beta_{wc} \beta_{oa})]^2} \left(\frac{\beta_{wc}^2}{I_a} + \frac{\beta_{wc}^2}{I_{ea}} + \frac{\beta_{wa}^2}{I_c} + \frac{\beta_{wa}^2}{I_{ca}} \right) \quad (7)$$

where I_a and I_c are determined from Eqs (2) and (3). The probable errors in S_w and S_o can be significantly reduced by increasing counting time and fluid attenuation coefficients. NaI salt was used to increase the attenuation coefficient of water. A counting time of 360 s was used for all measurements. Complete analysis of the effect of NaI on the attenuation coefficients of water and the effect of counting time on the probable errors are given by Bali [1].

METHODS

A dual-energy gamma system containing line sources of 100 mCi of ^{241}Am (59.6 Kev) and 40 mCi of ^{137}Cs (662 Kev) was used in this study. The system produced a collimated beam, 2.54 cm in height and 0.31 cm in width. The counting system included a photomultiplier base (EG&G ORTEC Model 266) with a NaI(Tl) scintillation detector

(EG&G ORTEC Model 905-3), two amplifiers and high voltage power supply (EG&G ORTEC Model 925 SCINT), and a multichannel analyzer (EG&G ORTEC Model 7100). The porous media used in this experiment was Yolo clay loam. The attenuation coefficients of soil, water, and Soltrol 130 (a light oil, thereafter Soltrol) were determined using a cubical plexiglass cell which had five compartments of constant width of 1.185 cm. Gamma count rates for the Am source were obtained first on an empty cell (I_{ea}) and then sequentially after filling each cell one at a time. The same procedure was employed for the other materials and repeated using the ^{137}Cs radiation source. Attenuation coefficients of water, NaI solution (10 g/l), Soltrol, and soil were also determined by taking count rates through an empty experimental flow cell and through cells filled with soil, 10 g/l NaI solution, Soltrol, and soil.

The effective path length in the cylindrical flow cell is less than the diameter of the flow cell because the radiation emanates from line sources rather than point sources. The effective path length for the flow cell was calculated using the mass attenuation coefficient of water obtained from count rates through a cubical plexiglass cell and a cylindrical flow cell filled with water. The effective path length can also be calculated from the configuration of the cylindrical flow cell and beam height.

All counts were corrected for resolving time using the method described by Bali [1]. The resolving time method accounts for the fact that the measured count rate does not always reflect the actual number of gamma-ray photons encountered by the scintillation crystal. The resolving time or the system dead time of a gamma-attenuation counting system is defined as the minimum time separating two consecutively recorded gamma photons, or the minimum time necessary to process a pulse [8]. Any photon that arrives at the scintillation crystal before the minimum time has elapsed is not counted.

Correction for the electronic drift in the measuring system during measurements was applied to all observed counts prior to any other correction. Systematic drift in count rates occurs due to micro-fluctuations in voltage and temperature of the counting system and over stimulation of the photon detection crystal particularly from the higher energy ^{137}Cs source. This drift was minimized by maintaining the system in a constant temperature room ($25\text{ }^\circ\text{C} \pm 0.2\text{ }^\circ\text{C}$) and minimizing detector exposure to ^{137}Cs radiation. To account for electronic drift, counts were taken on a standard absorber before and after each set of measurements.

Correction was based on a linear interpolation with time [6] using the following equation:

$$R_{tc} = \left(\frac{R_s(0)}{R_s(t)} \right) R_t \quad (8)$$

where R_{tc} is the corrected count rate at time t . $R_s(0)$ and $R_s(t)$ are the standard absorber count rates at time 0 and t , respectively, and R_t is the count rate at time t . $R_s(t)$ was found from the linear interpolation between the standard absorber count before and after each set of measurements.

The dual-energy gamma system was used to study the displacement of water by Soltrol. A column 5.08 cm in diameter and 16.5 cm long was packed with soil in lifts so as to minimize the variability in bulk density within the column. The initially water wet soil column had a uniform saturation of $0.08675\text{ cm}^3/\text{cm}^3$ of 10 g/l NaI solution. As mentioned earlier, the NaI was added to the water to decrease the probable errors in liquid saturation measurements. The average bulk density was 1.3629 g/cm^3 . Count rates for the standard absorber, empty column, column filled with NaI solution, and column filled with Soltrol were taken before each set of measurements. A standard absorber count rate was also taken after each set of measurements. The standard absorber count rates were used to correct the observed

counts for any electronic drift in the system.

Soltrol was introduced to the column at a constant rate of 0.2 ml/h using a syringe pump. The column was scanned at 3 different times during the displacement process (22.00, 38.75, and 64.25 h), at a 0.5 cm interval for the first 5 cm and at 1 cm intervals thereafter. The syringe pump was disconnected from the columns during scanning, and the infiltration rate was such that there was negligible change in saturations during the scanning process.

RESULTS AND DISCUSSION

Soil bulk density and effective radiation path length were needed in order to make measurements of liquid saturations during immiscible displacement (Eqs 2 and 3). The effective path length, x , was determined from columns filled with 10 g/l NaI solution and Soltrol. Effective path length for each source was calculated from:

$$x_a = \frac{1}{\beta_{ia}} \ln \frac{I_{ea}}{I_a} \quad (9)$$

$$x_c = \frac{1}{\beta_{ic}} \ln \frac{I_{ec}}{I_c} \quad (10)$$

where I_a , I_{ea} , I_c , I_{ec} , β_{ia} , and β_{ic} are as defined earlier, and the subscript i refers to the fluid of interest (NaI solution or Soltrol).

The effective path length, which is slightly less than the diameter of the column due to the curvature of the column, was calculated from the Am and Cs count rates for NaI solution and Soltrol using the independently determined attenuation coefficients from the cubical plexiglass cell. The calculated effective path length (Table 1) based on liquid attenuation, unfortunately, depended on the fluid and source of radiation used. The data from the Cs source yielded similar path lengths for both fluids that were greater than those obtained from the Am data.

Conceptually, path length of a beam 2.54 cm wide passing through the center

of soil column 5.08 cm in diameter is 4.7397 cm which is 1.8 % greater than the average value obtained from the liquid attenuation measurements. The dependence of effective path length on radiation source and this discrepancy between conceptual and measured values, though small, causes problems in correctly calculating S_o and S_w from count data during multi-phase flow experiments.

Solving Eqs (2) and (3) simultaneously to determine liquid saturation and soil bulk density in a single-phase wetted soil column yields:

$$\theta_i = \frac{\mu_{sc} \ln \left(\frac{I_a}{I_{ea}} \right) - \mu_{sa} \ln \left(\frac{I_c}{I_{ec}} \right)}{x(\mu_{sd} \beta_{ic} - \mu_{sd} \beta_{ia})} \quad (11)$$

and

$$\rho = \frac{\beta_{ia} \ln \left(\frac{I_c}{I_{ec}} \right) - \beta_{ic} \ln \left(\frac{I_a}{I_{ea}} \right)}{x(\mu_{sa} \beta_{ic} - \mu_{sd} \beta_{ia})} \quad (12)$$

where $\theta_i = \Phi S_i$.

When Eqs (11) and (12) were used to calculate soil bulk density and liquid saturation along initially NaI solution (10 g/l) packed to a known average bulk density and saturation, errors in saturations and bulk densities as compared to the known values at packing were extremely high when either the conceptual or any one of the measured path lengths were used. This was apparently due to the variability in path lengths and errors in the independent determinations of the attenuation coefficients for the NaI solution and Soltrol. Therefore, a correction procedure was developed to eliminate the effect of path length and the liquid attenuation coefficients on the measurements of bulk density using the dual-energy gamma system and Eqs (11) and (12). Assuming that: 1) the average bulk density and initial saturation of the soil packed column are known, 2) the fluid of interest is uniformly distributed in the soil, and 3) that the empty

Table 1. Calculated effective path length (cm)

Path length (cm)					
Am NaI sol.	Cs NaI sol.	Am Soltrol	Cs Soltrol	Average	Actual*
4.4601	4.8053	4.5525	4.7942	4.6530	4.7397

* The conceptual path length of a 2.54 cm wide beam passing through the center of 5.08 cm in diameter soil column is 4.7397 cm.

column and liquid-filled column count rates are known, then the following equations can be written for each radiation source:

$$\ln\left(\frac{I_o}{I_f}\right) = \beta_f x = A_f \tag{13}$$

and

$$\ln\left(\frac{I_o}{I}\right) = \beta_f x \Phi S + \mu_s x \rho_b \tag{14}$$

where I_o is the count rate through the empty column, I_f is the count rate through the liquid-filled column, I is the count rate through the soil column at uniform saturation, β_f is the attenuation coefficient of the fluid, S is the fluid saturation, and x is the radiation path length. Rewriting Eq. 14:

$$\ln\left(\frac{I_o}{I}\right) = \ln\left(\frac{I_o}{I_f}\right) \Phi S + \mu_s x \rho_b \tag{15}$$

enables $\mu_s x$ to be determined from:

$$\mu_s x = B = \frac{1}{\rho_b} \left[\ln\left(\frac{I_o}{I}\right) - \ln\left(\frac{I_o}{I_f}\right) \Phi S \right] \tag{16}$$

where I can be replaced by the average count rate over the entire column (I_{avg}) and S , Φ and the bulk density are the average liquid saturation, porosity and bulk density obtained when packing the soil column. When the value of B is determined, the bulk density at any given location along the column is determined from:

$$\rho_b = \frac{1}{B} \left[\ln\left(\frac{I_o}{I}\right) - A_f \Phi S \right] \tag{17}$$

using either radiation source.

We applied Eqs (13)-(17) to the attenuation data obtained from scanning the column packed with an initially NaI solution wet soil ($\Theta_w = 0.08675 \text{ cm}^3/\text{cm}^3$, $S_w = 17.861 \%$) to determine the soil bulk density along the length of the column after we found that Eqs (11) and (12) with independently determined parameters yielded uninterpretable results. Scanning with both sources at 20 locations along the column yielded an average bulk density of 1.3633 g/cm^3 when the Am source was used and 1.3630 g/cm^3 when the Cs source was used (Table 2). Both values are not significantly different from each other or the overall bulk density of 1.3629 g/cm^3 determined from the mass and volume of the soil measured at packing. The average of the absolute difference between the bulk densities calculated from the Am and Cs sources is 0.0120 g/cm^3 or less than a 1% from the predetermined bulk density. Bulk density distributions along the column were also practically the same for data from both sources with a maximum variability of no more than 2.5%. Those low values indicate that the method is reliable in determining bulk density distribution along soil columns of known over all bulk density and uniform saturation. The accuracy of this method is improved by increasing the counting time and by thoroughly mixing the soil before packing.

After completing the correction procedures discussed above, a similar procedure described by Bali [1] was used to calculate Soltrol and NaI solution saturations in the initially NaI solution-wet soil

Table 2. Bulk density distribution in the NaI solution-wet column

Distance (cm)	Am bulk density (gm/cm ³)	Cs bulk density (gm/cm ³)	Average bulk density (gm/cm ³)
0.5	1.367046	1.337431	1.352239
1	1.374366	1.388341	1.381354
1.5	1.371363	1.369173	1.370268
2	1.391583	1.398867	1.395225
2.5	1.327659	1.340679	1.334169
3	1.356049	1.366152	1.361101
3.5	1.383123	1.394114	1.388619
4	1.369011	1.369396	1.369204
4.5	1.383417	1.392757	1.388087
5	1.377424	1.385356	1.381390
6	1.387629	1.376425	1.382027
7	1.338940	1.334535	1.336738
8	1.387387	1.362404	1.374896
9	1.392171	1.386145	1.389158
10	1.373075	1.372464	1.372770
11	1.330116	1.325162	1.327639
12	1.329459	1.320886	1.325173
13	1.355447	1.354172	1.354810
14	1.306204	1.341389	1.323797
15	1.364112	1.344228	1.354170
Average	1.363279	1.363004	1.363142

measurements to the next due to photon saturation of the crystal in time. This correction was simple ratio analogous to the standard absorber correction (Eq. 8), and may, or may not be necessary for particular gamma systems.

The correction procedure was used to improve the accuracy of the dual gamma system in predicting fluid saturations. The correction procedure was based on the $\ln(I_{oa}/I_a)$ for the NaI solution column at the time of measurements. The correction was defined as:

$$\text{Correction 1} = \frac{\ln\left(\frac{I_{oa}}{I_a}\right)_i}{\ln\left(\frac{I_{oa}}{I_a}\right)_t} \quad (18)$$

where the subscripts i and t represent the initial time of infiltration and the time at which the measurements were taken, respectively. I_{oa} is the empty count rate and I_a is the count rate of a NaI solution column. All Am count rates were corrected by multi-

plying each value by the correction presented in Table 3. A similar correction based on the same principle was used to correct the Cs counts (Table 3).

NaI solution and Soltrol contents in the soil column for corrected and uncorrected data for the first time step are presented in Table 4. This correction resulted in a significant improvement in the calculated volumetric contents of NaI solution and Soltrol 130. For the first time step ($t = 22.00$ h), the total volumetric content prediction based on the infiltration mass balance was $0.13017 \text{ cm}^3/\text{cm}^3$, the calculated total volumetric content was $0.10924 \text{ cm}^3/\text{cm}^3$ for corrected counts and $0.05985 \text{ cm}^3/\text{cm}^3$ for uncorrected counts, similar results were also found for the second and the third time steps. Most measured saturations were within the predictable saturations based on the theoretical probable errors. There were few measurements that were not within the predictable error due to sudden changes in voltage during the experiment.

Table 3. Am and Cs count correction. Initial NaI solution content $0.08675 \text{ cm}^3/\text{cm}^3$

Parameter		Count correction			
		Am			
Time (h)	0.00	22.00	38.75	64.25	
NaI solution infiltrated (cm^3)	0.00	4.40	7.75	12.85	
I_{0a} (counting time = 3 min)	3499618	3478069	3488075	3469845	
$\ln(I_{0a}/I_a)$ (NaI solution column)	1.10974466	1.10473553	1.10634290	1.11613808	
Correction	1.00000000	1.00453423	1.00307478	0.99427184	
		Cs			
Time (h)	0.00	22.00	38.75	64.25	
NaI solution infiltrated (cm^3)	0.00	4.40	7.75	12.85	
I_{0a} (counting time = 3 min)	979781	988990	985928	984845	
$\ln(I_{0a}/I_a)$ (NaI solution column)	0.40752534	0.41163533	0.40885039	0.40951913	
Correction	1.00000000	0.99001546	0.99675909	0.99513139	

Table 4. Liquid contents (cm^3/cm^3) during Soltrol displacement of NaI solution. Time = 22.00 h, infiltration rate 2 ml/h

Distance (cm)	Liquid contents (cm^3/cm^3)			
	Uncorrected		Corrected	
	NaI solution	Soltrol	NaI solution	Soltrol
0.5	0.222	-0.130	0.166	-0.024
1.0	0.138	-0.026	0.081	0.079
1.5	0.161	-0.066	0.104	0.040
2.0	0.138	-0.067	0.081	0.039
2.5	0.132	-0.072	0.075	0.034
3.0	0.146	-0.097	0.089	0.009
3.5	0.157	-0.132	0.100	-0.017
4.0	0.149	-0.180	0.137	-0.074
4.5	0.168	-0.131	0.111	-0.025
5.0	0.169	-0.132	0.113	-0.026
Average	0.162	-0.102	0.106	0.003

error due to sudden changes in voltage during the experiment.

CONCLUSIONS

A correction procedure was established and tested to reduce the errors in the calculated bulk density and fluid saturation in a single-phase wetted soil column. The procedure minimized the errors introduced by the path length and bulk density of the porous media.

Another correction procedure was introduced to account for system electronic drift from one set of measurements to the

next due to photon saturation of the crystal in time. Most measured saturations were within the predictable saturations based on the theoretical probable errors. The calibration procedure resulted in a significant improvement in the prediction of liquid saturations using dual-energy gamma system.

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