

# DENSITY MEASUREMENTS DURING SEDIMENTATION OF MINE WATER TREATMENT SLUDGE AND MINE TAILINGS USING DUAL-SOURCE GAMMA-RAY TRANSMISSION

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## Abstract

The technique of gamma-ray transmission has been used to measure the variation of the density of acidic mine water treatment sludge as it settled in a 15 cm diameter, 180 cm high Plexiglas column. The previous system, using one radioactive source, was upgraded to two sources, <sup>153</sup>Sm with gamma-ray energy 103 keV and <sup>198</sup>Au 412 keV, to determine simultaneously the solids density and the water concentration during sedimentation. The sources and Sodium-Iodide detector scan the column vertically in 20 minutes to measure density profiles. Accuracy of 0.04 g/cm<sup>3</sup> was achieved for both solids density and water concentration in one type of sludge, but the technique failed to distinguish between water and solids in sludge with low heavy metal content. Changing one of the sources to reduce the energy of the gamma-rays from 103 keV to 60 keV should improve the accuracy to 0.02 g/cm<sup>3</sup> for any sludge.

## 1. Introduction

Mining operations which produce acid mine drainage need to treat their effluent before discharge. These acid waters typically contain high amounts of solubilized elements such as sulphates and heavy metals which are then precipitated with lime and recovered in the treatment sludge [1]. Lined ponds are often used to store the large volume of sludge produced. The design of these ponds remains largely empirical as little is known about the hydro-geotechnical behaviour of treatment sludge, which typically contains only 2 to 20% solids by weight. There is thus a need to develop measurement techniques to assess the behaviour of the sludge after discharge.

A key parameter needed for comparison with sedimentation-consolidation theories is the variation of the density of the sludge as the solids settle. A laboratory testing system, employing a 15 cm diameter, 180 cm high Plexiglas column, was constructed at Ecole Polytechnique de Montreal and used to study the sedimentation and consolidation of various types of sludge [2,3]. Density, as a function of position along the column, was measured by gamma-ray transmission.

The gamma ray source, <sup>153</sup>Sm, with half-life 47.5 h and gamma-ray energy 103 keV, is placed in a lead casing near the side of the column. The gamma-rays are not collimated, in order to avoid alignment

problems and to reduce the necessary source activity.

Depending on the density of the sludge, a certain fraction of the emitted gamma-rays are transmitted through the column along its diameter and are detected by a NaI detector, with a 38 mm diameter, 25 mm thick crystal, placed on the other side at the same elevation, as shown in Fig. 1.

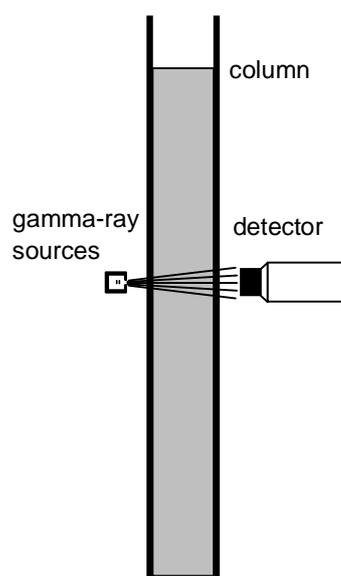


Figure 1: Schematic diagram of the test column with the density measurement system.

Both source and detector are fixed to a platform that can be moved along the column with a motorized screw mechanism. A smaller column of the same diameter can be placed below the main column for calibration measurements. These details are shown in the Fig. 2.

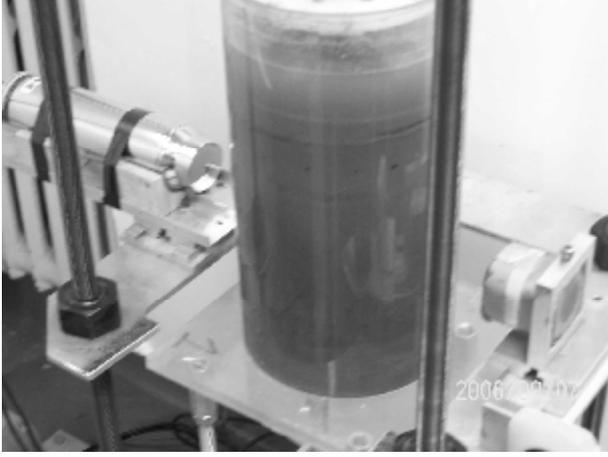


Figure 2: Photo of a small test column placed in between the source and the detector.

The number of gamma-rays,  $I$ , detected in a given interval of time is given by the following equation:

$$I = I_0 \exp(-\mu r x) \quad (1)$$

where  $I_0$  is the number which would be detected if the column were empty,  $x$  the thickness of sludge along the path of the gamma-rays, which is the same as the inside diameter (15.25 cm) of the column,  $\mu$  the mass attenuation coefficient ( $\text{cm}^2/\text{g}$ ) for 103 keV gamma-rays in the sludge in the column,  $\rho$  the density ( $\text{g}/\text{cm}^3$ ) of the sludge.  $I_0$  and  $\mu$  are determined from calibration measurements using columns filled with pure water and sludge of known density. The density is determined by solving Eq. 1 for  $\rho$  for measurements made along the column at various times. For each measurement,  $I_0$  is corrected for decay of the  $^{153}\text{Sm}$  source.

Previous experience with this one-source technique revealed two weaknesses: First, as sedimentation occurs, the relative amounts of water and solids at a given position change, leading to a change in the mass attenuation coefficient, which depends on the chemical composition of the material. Thus there was always some uncertainty in the value of this coefficient. Second, the one-source technique gives only the total bulk density; it would be preferable to have both the dry (solids) density and the

volumetric water concentration (as defined below). We have therefore developed a dual-source technique.

## 2. Dual-Source Methodology

With the dual-source technique, the two sources have different gamma-ray energies and therefore different mass attenuation coefficients in solids and water. To maximize the difference in the coefficients for solids and water, one of the sources should emit gamma-rays with energy as low as possible. For soil studies, several authors have used  $^{241}\text{Am}$ , 60 keV [4,5]; we continue to use  $^{153}\text{Sm}$ , 103 keV, because the expected heavy metal content of mine sludge and tailings and the thickness of our column may result in an insufficient number of gamma-rays transmitted at 60 keV. The second source used is  $^{198}\text{Au}$ , with half-life 64.8 h and gamma-ray energy 412 keV. It was placed in the lead casing just behind the  $^{153}\text{Sm}$  source. The sources were produced by irradiating 30 mg of  $\text{Sm}_2\text{O}_3$  and 10 mg of gold wire in the neutron flux of the SLOWPOKE nuclear reactor [6] at Ecole Polytechnique. The activities used were 40 MBq  $^{153}\text{Sm}$  and 20 MBq  $^{198}\text{Au}$ . Because of the short half-lives, the sources were re-activated in the reactor each day.

With the dual-source technique, we have two simultaneous attenuation equations [4,7]:

$$I_1 = I_{01} \exp(-m_{s1} r_s x - m_{w1} r_w x) \quad (2)$$

$$I_2 = I_{02} \exp(-m_{s2} r_s x - m_{w2} r_w x) \quad (3)$$

where  $I_1$  is the number of gamma-rays detected at 103 keV and  $I_2$  is the number detected at 412 keV.  $\mu_{w1}$ ,  $\mu_{s1}$ ,  $\mu_{w2}$  and  $\mu_{s2}$  are the mass attenuation coefficients for water and for the solids at the two gamma-ray energies. These coefficients do not change as the solids settle, as long as the solids do not change chemical composition. The solids (dry) density  $\rho_s$  ( $\text{g}/\text{cm}^3$ ) and the volumetric water concentration  $\rho_w$  ( $\text{g}/\text{cm}^3$ ) are found by solving the above simultaneous equations for these two variables, which gives:

$$r_s = \frac{m_{w1} \ln(I_2/I_{02}) - m_{w2} \ln(I_1/I_{01})}{(m_{s1} m_{w2} - m_{s2} m_{w1}) x} \quad (4)$$

$$r_w = \frac{m_{s2} \ln(I_1/I_{01}) - m_{s1} \ln(I_2/I_{02})}{(m_{s1} m_{w2} - m_{s2} m_{w1}) x} \quad (5)$$

Our term for the volumetric water concentration,  $\rho_w$  ( $\text{g}/\text{cm}^3$ ), is essentially equivalent to the commonly

used [1,4] volumetric water content,  $\theta$  ( $\text{m}^3/\text{m}^3$ ), since the density of water is  $1 \text{ g}/\text{cm}^3$ .

The dual-source technique was tested for two different types of sludge using the set-up described above. Sludge 1 is a reddish treatment sludge containing mainly fine particles. Sludge 2 contains mine tailings (i.e. finely crushed rock) with coarser particles. Calibration measurements were performed to determine  $I_{01}$ ,  $I_{02}$ ,  $\mu_{s1}$  and  $\mu_{s2}$  for each sludge.  $I_{01}$  and  $I_{02}$  were measured with the calibration column filled with pure water.  $\mu_{s1}$  and  $\mu_{s2}$  were measured with the column filled with homogenized sludge of known solids density and volumetric water content. Sludge was then placed in the main column to determine the accuracy of the density measurements. After each measurement a sample of the sludge was removed from the column, weighed, its volume measured, dried and weighed again. Thus, the accuracy of the dual-source density measurements could be determined by comparison with weighing. The volumetric water content of the sludge was increased each time by adding pure water and homogenizing.

The gamma-ray spectra, accumulated for 80 s, were stored on computer disk for calculation of  $I_1$  and  $I_2$  off-line. A typical spectrum is shown in Fig. 3.

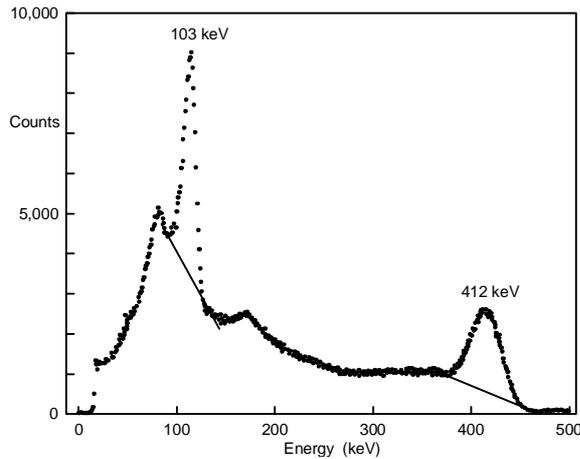


Figure 3: Dual-source gamma-ray spectrum.

For the 103 keV and 412 keV peaks, the peak area was calculated by subtracting a straight-line background from beneath the peaks as shown in the figure. This background is caused by Compton scattering of gamma-rays in the column and in the detector. From measurements with the  $^{198}\text{Au}$  source only, it was found that the background at 103 keV due to Compton scattered 412 keV gamma-rays was non-linear and a straight line assumption led to a peak area at 103 keV equal to 0.69% of the area

of the 412 keV peak. Thus, for each measurement, the peak area at 103 keV was corrected by subtracting 0.69% of the peak area at 412 keV. The solids densities and water concentrations were calculated with Eq. 4 and 5.

The system was used to scan the column vertically to determine density variations. The measurement time was reduced to 20 s to enable the entire column to be scanned quickly before significant variations could occur in order to obtain (quasi)static profiles. The precision of counting measurements is known to vary as the square-root of the counting interval. To determine the precision of 20 s measurements, a number of them were performed on the two types of sludge.

### 3. Results and Discussion

The mass attenuation coefficients determined for the two types of sludge are shown in Table 1. The coefficients for water were calculated by combining the values for H and O interpolated from standard tables [8]. To differentiate between solids and water, the dual-source technique uses Eq. 4 and 5 where the denominator contains the quantity  $k = \mu_{s1} \mu_{w2} - \mu_{w1} \mu_{s2}$ . From Table 1 it can be calculated that  $k = 0.00284$  for sludge 1 and  $k = 0.00005$  for sludge 2. With  $k$  so close to zero, especially for sludge 2, the uncertainty in  $k$ , relative to the value of  $k$ , is significant and the dual-source technique cannot easily differentiate between solids and water.

Table 1: Gamma-ray mass attenuation coefficients ( $\text{cm}^2/\text{g}$ )

	103 keV	412 keV
Water	0.1656	0.1044
Solids Sludge 1	$0.162 \pm 0.003$	$0.085 \pm 0.002$
Solids Sludge 2	$0.148 \pm 0.002$	$0.093 \pm 0.002$

In Table 2, we compare the solids densities and volumetric water concentrations determined by the dual-source technique with those determined gravimetrically for sludge 1. From the observed differences, the uncertainty in the measurements by the dual-source technique is estimated to be  $0.04 \text{ g}/\text{cm}^3$ , for both the solids densities and water concentrations.

These results are for 80 s measurements. When the measurement time was reduced to 20 s, the uncertainties increased from  $0.04 \text{ g}/\text{cm}^3$  to  $0.07 \text{ g}/\text{cm}^3$ .

Table 2: Comparison of solids density and volumetric water concentration measured gravimetrically and by dual-source gamma-ray transmission

Solids density (g/cm <sup>3</sup> )		Water concentration (g/cm <sup>3</sup> )	
gravimetric	dual-source	gravimetric	dual-source
0.157	0.106	0.950	0.980
0.136	0.123	0.957	0.966
0.120	0.074	0.962	1.001
0.107	0.139	0.966	0.938
0.094	0.067	0.970	0.995

If the solids densities and water contents of Table 2 are added to obtain total bulk densities, then the comparison with the gravimetric results suggests an uncertainty in the dual-source total bulk densities of 0.01 g/cm<sup>3</sup>. This reduction in uncertainty from 0.04 g/cm<sup>3</sup> to 0.01 g/cm<sup>3</sup> is due to the fact that the errors in the values of solids densities and water contents are negatively correlated and they cancel when they are added.

The results for sludge 2 are not shown; they are much less accurate than those for sludge 1 because of the very low value of  $k = 0.00005$ , as was mentioned above. It is concluded that when the quantity  $k = \mu_{s1} \mu_{w2} - \mu_{w1} \mu_{s2}$  is near zero the dual-source technique fails to distinguish between solids and water.

For a given sludge, the value of  $k$  depends on the relative amounts of hydrogen (high mass attenuation coefficient), light elements (low coefficient) and heavy metals (high coefficient). These are usually not well known in advance. It is thought that the coefficient for the solids of sludge 1 at 103 keV is higher than that for sludge 2 because of a higher iron concentration in sludge 1.

The relatively poor accuracy of the present measurements, especially those on sludge 2 with tailings, is thought to be due to the choice of the low-energy gamma-ray source, <sup>153</sup>Sm at 103 keV. Several previous studies measuring water content in soils used <sup>241</sup>Am at 60 keV [4,5]. Our calculations show that, with a 60 keV low-energy gamma-ray, the value of  $k$  will exceed 0.009 for sludge with any expected concentration of heavy metals. This is more than three times the value we found for sludge 1 and it should lead to an improvement of a factor of two in the accuracy of the measured solids densities and water concentrations. The lower gamma-ray energy means that fewer gamma-rays

are transmitted through the column, which may reduce the precision for measurements with large diameter columns and high solids densities. Better accuracy may also be achieved using 80 keV gamma-rays; a possible source is <sup>166</sup>Ho, 80.8 keV, half-life 27 hours.

#### 4. Conclusions

The technique of dual-source gamma-ray transmission, using <sup>153</sup>Sm with gamma-ray energy 103 keV and <sup>198</sup>Au 412 keV, enabled the simultaneous measurement of solids density and the water content with an accuracy of 0.04 g/cm<sup>3</sup> for sludge containing a relatively high amount of heavy metals. For tailings containing a lower amount of heavy metals, the technique failed to distinguish efficiently between water and solids. Calculations show that replacing the <sup>153</sup>Sm source with <sup>241</sup>Am (60 keV) would enable the measurement of solids density and water content with an accuracy of 0.02 g/cm<sup>3</sup> for most types of sludge and tailings.

#### 5. References

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