



Volume fractions calculation of a biphasic system on

cylindrical tube using gamma-ray and MCNP6 code

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ABSTRACT

This paper presents a methodology to calculate volumetric fractions on the stratified flow regime, considering a cylindrical tube. A mathematical model of a measurement system was developed using the MCNP6 code. The mathematical equation was developed to calculate the volume fractions using the pulse height distributions obtained by a radiation detector. The stratified flow regime model considers air-oil, air-gasoline and oil-gasoline biphasic flow, in order to evaluate the performance of the proposed equation in fluid combinations with different densities. A comparative study with equations developed for square-section tubes from literature was performed. The study considered geometry of a source of ¹³⁷Cs, an acrylic tubing measuring 8.0 cm in diameter and a NaI(TI) scintillator detector placed at a position diametrically opposed to a radiation source to measure the transmitted beam. The dimensions and materials to perform the simulation of the detectors were based on information obtained from the gammagraphy technique and the detector was experimentally validated. The volume fractions of each of the fluids were 0 up to 100% with variations of 25%.

Keywords: volume fraction, MCNP6 code, ray-gamma densitometry.

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1. INTRODUCTION

Due to the great demand for oil and its derivatives, extraction, refining and distribution are operations of great importance for the economy of a country. For the petroleum exploration and production industry, there is a clear need for devices that allow precise flow measurements. The most different technologies are used in the devices available on the market. However, many researchers have been investigating new methods that allow real-time and non-invasive measurement of important flow parameters, such as: flow rate, volume fraction, fluid and velocity [1 - 4]. These characteristics open a promising field for use of nuclear techniques.

Devices based on nuclear techniques are compact and efficient for flow control which is extremely useful for the development of meters that provide accurate results for all fluids present in the multiphase flow, which is very important mainly for fiscal purposes.

In this context, techniques that use gamma radiation sources have been applied to found flow characteristics rate. The great advantage of this technique, non-invasive, is the possibility of analysis the flow regime without the need to interrupt the normal operation of the installation presenting reliability in the results. In addition, the high detection sensitivity of radiation detectors allows the use of sealed radioactive sources in order to minimize the radiological risk to the health of workers. These advantages make these techniques very important in the industry and for this reason is the focus of this research [2].

Gamma densitometry has been applied in many areas, such as: chemical, mining and petroleum industry, in many applications, such as: flow measurements [5 - 7], monitoring of petroleum and petroleum by-products [8], calculations of volume fractions [4] and prediction of fluid densities [9 - 10].

The gamma–ray densitometry technique used is based on the incidence of a gamma ray beam which after interacting with the tube-fluid system is recorded by a NaI(Tl) detector diametrically positioned to a gamma ray source (¹³⁷Cs). Using this technique was possible to calculate the volume fractions of different fluid present inside a tube. Using the MCNP6 computational code [11], a measurement geometry based on the transmitted beam was developed with the aim of generating many volume fractions of a stratified flow regime under a two-phase flow. The photopeak region of

spectrum recorded in the detector is used for calculating volume fractions in a cylindrical section tube by the analytical equation. The mathematical model of a real NaI(Tl) detector was validated experimentally [12 - 18]. Therefore, this study aims to evaluate the accuracy of the analytical equation described in the literature for calculations of volume fractions in cylindrical pipelines on stratified flow regime in different fluid models (air-oil, air-gasoline and oil-gasoline) using the MCNP6 code.

2. METHODOLOGY

The methodology consists of developing a mathematical model using the Monte Carlo method through MCNP6 code to simulate a measurement geometry that consists of a tube and two different fluids in order to calculate the volumetric fractions of each fluid.

2.1 Equipment and radioactive sources

The characteristics of the spectrometric system and the radioactive sources used in this study are described:

i) **Spectrometry system:** A set of electronic modules used to obtain and process the signal generated by the detector was used. The characteristics of the spectrometric system used are:

- High voltage power source manufactured by Canberra 3106D;
- Pre-amplifier manufactured by the company Canberra model 2005;
- Amplifier manufactured by the company Tennelec model TC245;
- Mono Channel Analyzer manufactured by Ortec model 551;
- Multi-channel analyzer (AMC) manufactured by Instituto de Engenharia Nuclear;
- $1\frac{1}{4} \times \frac{3}{4}$ " NaI(Tl) detector.

ii) Radioactive Sources: The mathematical model of the detector was validated by means of experimental measurements, using radioactive standards comprising the energy range of interest of

this investigation (from 59.541 keV to 661.657 keV). The characteristics of the gamma radioactive sources that were used in calculations are shown in *Table 1*.

Source	Energy (keV)	Half-Life (days)	Probability of emission (%)	Activity (kBq)*
^{137}Cs	661.657	10976	84.99	22980.602
¹³³ Ba	356.013	3849	62.05	17800.313
²⁴¹ Am	59.541	158007	35.92	28154.197
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Table 1: Data from the standard sources of radiation.

* Activity corrected for the date of the experiment.

It is important to mention that the ¹³³Ba has other energies (276.399, 302.851 and 383.849 keV) close to the energy used in this paper (356.013 keV), and the NaI(Tl) detector has no energy resolution to completely discriminate these peaks. Therefore, a mathematical treatment was used to calculate exclusively the desired photopeak area. In this procedure it was considered the contribution of the counts for each of these peaks, since their respective energies and energy resolution are well known.

2.2 Absolute detector efficiency curve

The photopeak absolute efficiency obtained experimentally with radioactive standards, listed in *Table 1*, was used to validate the model of the NaI(Tl) detector. The measurement time was 60 minutes and the detector source distance (DSD) of 28 mm was chosen so that the associated errors due to the counting statistic remained below 5%. The simulation was done under the same conditions of experimental arrangement [19].

The emission probability of each monoenergetic photopeak associated with the corrected activity for the date of the measurements of each radioactive source was used to calculate the counting efficiency, thus determining an experimental efficiency curve of the NaI(Tl) detector. The photopeak absolute efficiency can be calculated from *Equation 1*.

$$\varepsilon(E) = \frac{C}{t.A.P} \tag{1}$$

Where:

C – number of counts recorded in the detector in the photoelectric region;

- t count time (s);
- A activity of the radioactive source (Bq);
- P emission probability for gamma rays in energy "E".

The uncertainty of the absolute efficiency is given by *Equation 2*, disregarding the uncertainties of the counting time, the emission probability and the decay correction constant, since they are very small values.

$$\sigma\varepsilon = \sqrt{\left(\frac{\partial\varepsilon}{\partial C} \cdot \sigma C\right)^2 + \left(\frac{\partial\varepsilon}{\partial A} \cdot \sigma A\right)^2} \tag{2}$$

Where:

 σC – uncertainty associated with counts;

 σA – uncertainty associated with activity.

2.3 Energy resolution curve

In order to consider the energy resolution of the NaI(Tl) detector in the mathematical simulation, using the MCNP6 code, making it more realistic, it is necessary to use a special treatment for the Pulse height distribution (PHD), in which the energy peaks behave as a Gaussian function, broadening it according to *Equation 3*.

$$f(E) = C. exp - \left(\frac{E - E_0}{FWHM/2\sqrt{\ln 2}}\right)^2$$
(3)

Where:

E – "broadening" energy (MeV);

E₀ – "non- broadening" energy (MeV);

C – normalization constant (MeV);

FWHM/ $2\sqrt{\ln 2}$ – Gauss width (MeV);

FWHM – Full Width at Half Maximum.

This treatment is performed using parameters that are supplied by the user to the input file (INP) with the Gaussian Energy Broadening (GEB) command of the FTn card, available in the MCNP6 code. These parameters were experimentally obtained from the least squares approximation of the nonlinear function of *Equation 4*.

$$FWHM = a + b\sqrt{E + cE^2} \tag{4}$$

Where:

E – Energy of incident gamma rays (MeV);

a, b, c – adjusted constants.

2.4 NaI(Tl) detector modeling

The detector must be modeled as accurately as possible because variations in crystal size influence the determination of efficiency [20 - 23]. When the technical manuals do not declare the information (shape of the crystal and the dimensions of the active volume) about the characteristics of a detector or even when the model or manufacturer of the detector is not known, such information can be obtained by performing a gammagraphy [24]. The density of the NaI(Tl) crystal used was 3.667 g.cm⁻³, the density of the reflective material (MgO) used was 3.58 g.cm⁻³ and the aluminum density was 2.6989 g.cm⁻³ [14 - 16].

Despite this, the actual dimensions of the crystal may be different, in order to determine these values accurately, two point sources, ²⁴¹Am and ¹³⁷Cs, one low and one high energy, respectively, were used. These sources were measured with the NaI(Tl) detector at a DSD of 28 mm, positioned in the direction of the longitudinal axis in relation to the crystal. Two measurements were made with the sources on the longitudinal axis of the detector in order to determine the detector response to a known radiation field. This geometry was reproduced by simulation and the results were compared with the experimental values.

The procedure for determining the real volume of the detector consists of an iterative process between the counts obtained in the simulation and the variation of the crystal thickness made in the INP. The calculations started with the value obtained by the gammagraphy which is 31.75 mm in diameter and 19.05 mm in thickness. With these values it was verified that the total recorded count was larger than that obtained experimentally. The thickness of the crystal was gradually reduced until the value of the total count obtained was in agreement with the experimental value, thus defining the actual crystal volume [22, 23]. It is worth noting that this procedure wasn't performed varying the crystal diameter in the mathematical model, therefore, crystal lateral efficiency was not considered.

The simulation response was obtained using the output command "tally card F8", available in the MCNP6 code, which presents a PHD due to the photons in the sensitive region of the crystal. The mathematical modeling considered homogeneous detectors of sodium iodide activated with thallium: NaI(Tl) [14 - 18], according to our previous papers [25-26]. Some important considerations should be mentioned: The photomultiplier, positioned behind the crystal, was simulated as a 30 mm thick aluminum disc [17]. A radioactive source of the isotropic emission volumetric type is simulated in the code. The electrons generated by gamma radiation in the NaI(Tl) crystal are completely absorbed by the crystal thickness, so they were not considered in the simulation.

2.5 Volume fractions

The method is based on the attenuation of a gamma-ray monoenergetic beam transmitted by a tube containing biphasic flow. Considering a simplified model of the stratified flow regime with liquid phases in a square cross-section tube, knowing the dimensions and characteristics of the tube, the intensity of the beam transmitted is given by the equation of Beer–Lambert's modified, according to *Equation 5*. Disregarding a tube thickness since the term is common in calculating the volume fraction, and therefore, attenuation in the tube wall is also disregarded.

$$I(E) = I_0(E). \exp\left(-\sum_{i=1}^{n} x_i \mu_i(E)\right)$$
(5)

Where:

I – transmitted intensity of gamma rays (photons. cm⁻².s⁻¹);

 I_0 – initial incident intensity of gamma rays from the source (photons.cm⁻².s⁻¹);

- x_i thickness for fluid i (cm);
- E incident radiation energy;
- μ_i linear attenuation coefficient for fluid i.

A source collimated with enough energy to penetrate and be transmitted throughout the tube– fluid system is positioned on one side and diametrically opposite the source, a NaI(Tl) scintillation detector is installed to record the incident beam counts. The signal recorded in the detector is proportional to the transmission of the gamma rays due to the tube-fluid system. High-density fluids cause a decrease in the counting rate at the detector, while the lower density fluids result in a higher counting rate. Considering a unidirectional measurement geometry and perpendicular to the fluid layers in a biphasic system, as shown in *Figure 1*.



Figure 1: Geometry used for volume fractions calculation.

The calculation of the fluid volume fraction (FVF) in heterogeneous biphasic systems under the stratified flow regime using the gamma attenuation technique can be obtained using *Equations* 6 and 7 [1].

$$\alpha_1 = \frac{\ln(I/I_2)}{\ln(I/I_1)} \tag{6}$$

$$\alpha_2 = 1 - \alpha_1 \tag{7}$$

Where:

 $\alpha_{1,2}$ – volume fraction of fluid 1 or 2 (%);

I – gamma ray intensity recorded with tube containing fluids 1 and 2 (photons. $cm^{-2}.s^{-1}$);

 $I_{1,2}$ – intensity containing only fluid 1 or 2 (photons. cm⁻².s⁻¹).

For a cylindrical straight section tube, it is quite complex to calculate, therefore, the procedure adopted was to calculate the thicknesses of each fluid that correspond to the path traveled by the radiation considering a pencil beam, which can be obtained by *Equations 8* and 9 [1].

$$x_1 = \frac{\ln(I/I_2)}{\mu_2 - \mu_1}$$
(8)

$$x_2 = \frac{\ln(I/I_1)}{\mu_1 - \mu_2} \tag{9}$$

Where:

 $\mu_{1,2}$ – coefficient of linear attenuation of fluid 1 and 2.

Knowing the thicknesses of the fluids inside the tube, it is necessary to calculate the integral of the area occupied by the fluids by *Equation 10*.

$$S(x) = 2. \int_0^h \sqrt{R^2 - (y - R)^2} dy$$
(10)

Where:

R – radius of the tube (cm);

y – mathematical parameter to calculate the integral.

By means of mathematical manipulations and using the trigonometric method, the calculation of the volume fraction is given by *Equation 11* [27].

$$\alpha(\mathbf{x}) = \frac{\pi R^2}{2} + R^2 \cdot \sin^{-1}\left(\frac{\mathbf{x} - \mathbf{R}}{\mathbf{R}}\right) + (\mathbf{x} - \mathbf{R}) \cdot \sqrt{\mathbf{R}^2 - (\mathbf{x} - \mathbf{R})^2}$$
(11)

2.6 **Proposed geometry of the detection system**

The study considered a detection geometry composed of a 137 Cs source (661.657 keV), an acrylic tube with a 3.5 cm internal diameter and a wall thickness of 0.5 cm and a NaI(Tl) scintillation detector, positioned diametrically opposite the source, to measure the transmitted beam, as shown in *Figure 1*. The interior of the tube was filled with different amounts of the fluids to investigate the attenuation of the gamma rays in the calculation of the volume fractions. The stratified flow regime model considers fluid 1 and 2 (air-oil or air-gasoline or oil-gasoline), in order to evaluate the performance of the proposed equation in fluid combinations with different densities (air – 1.205E-3, gasoline – 0.721 and oil – 0.955 g.cm⁻³). The calculation used the "Tally F8" command to estimate the simulated pulse height distribution (PHD), ranging from 40 to 800 keV. This energy range was chosen considering the 661.657 keV emission photopeak of the ¹³⁷Cs. The fluids used are air, gasoline (C₈H₁₈) and oil (C₁₀H₁₈O). The volumetric fractions were varied from 0, 25, 50, 75 and 100%. It is important to mention that only the photopeak area was used in the analytical equations to calculate the volume fractions of the fluids.

3. RESULTS AND DISCUSSIONS

3.1 Detector validation

In order to evaluate if the proposed mathematical model is in agreement with the reality, the experimental photopeak absolute efficiency was compared with the values obtained on mathematical model of the detector NaI(Tl) considering the same geometry used in the experimental procedures. Uncertainty was estimated considering the counts and activity and remained below 5%. The experimental and simulated photopeak absolute efficiencies are shown in *Table 2*.

Table 2:	Experimental	and	simulated	efficiency values.
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Radionuclide	Energy	Efficienc	Relative Error
	(keV)	Experimental	Simulation

Am-241	59.541	1.71 ± 0.05	1.71	0.18
Ba-133	356.013	0.66 ± 0.12	0.65	2.87
Cs-137	661.657	0.17 ± 0.01	0.17	0.07

The experimental data and the values obtained by the modeling showed good agreement and presented a maximum relative error of 2.87% indicating that the detector model was well developed.

The energy resolution of the detector was determined by mean of the Full Width at Half Maximum (FWHM) measurements in photopeak energy from each source. From the experimentally FWHM measured, the parameters a, b and c used to adjust the GEB function were calculated by *Equation 4* described in section 2.3, the coefficients values are presented in *Table 3* and the function represented in *Figure 2*.

With these results it became possible to perform the fit of the GEB function, through the mentioned methodology, making the mathematical model more realistic.

Coefficients	Values	Uncertainty	
а	0.009	0.002	
b	-0.24	0.01	
с	-0.35	0.02	

Table 3: Coefficient values obtained by fit Equation 4.



After the experimental validation of efficiency and the considering of the energy resolution of the real detection system, a PHD for 661.657 keV is shown in *Figure 3*.



Figure 3: Comparison of the shape of the experimental and simulated spectra in the code.

An acceptable agreement can be noted in the photopeak region, but in the low energy range (Compton region) the PHD presents some discrepancy, which is usually reported in the literature as not-so-good precision in the simulation of low energy electrons and by scattering of photons in the shield and detector support that not considered in the simulations [13].

3.2 Volume fraction results

The spectra recorded by the detector for gasoline and oil flow are shown in *Figure 4*. Important to mention that air-oil and air-gasoline flow are very similar. The percentages refer to the oil fluid. Therefore, 0% means 0% oil, or 100% gasoline. Different fluid volume fractions equal to 0, 25, 50, 75 and 100% in relation to the other fluid were simulated on the biphasic system.



Figure 4: Spectra obtained with the simulation of oil and gasoline.

The calculation of the volume fraction using the measurements by transmission was performed by means of the integral of counts referring to the photopeak area obtained by the detector, since it is the region of total absorption of energy being well characterized, as can be seen in *Figure 4*.

The results for the volumetric fractions were obtained by means of the Equations 6 and 11 to verify if both equations can be used for cylindrical tubes; these results can be visualized in *Tables 4*, *5* and *6*.

Oil – Gasoline						
Theoretical	Square	Tube	Cylindrical Tube			
Volume (%)	Simulated Volume (%)	Relative Error (%)	Simulated Volume (%)	Relative Error (%)		
25.00	29.70	18.80	24.87	0.52		
50.00	49.92	0.16	49.87	0.26		
75.00	69.85	6.86	74.56	0.59		

Table 4: Comparison of the results for the oil-gasoline system.

Oil – Air						
Theoretical	Square	Tube	Cylindrical Tube			
Volume (%)	Simulated Volume (%)	Relative Error (%)	Simulated Volume (%)	Relative Error (%)		
25.00	29.85	19.39	24.93	0.28		
50.00	49.97	0.05	49.74	0.52		
75.00	70.22	6.37	74.74	0.35		

Table 5: Comparison of the results for the oil-air system.

Table 6: Comparison of the results for the gasoline-air system.

Gasoline – Air						
Theoretical	Square	Tube	Cylindrical Tube			
Volume (%)	Simulated Volume (%)	Relative Error (%)	Simulated Volume (%)	Relative Error (%)		
25.00	29.80	19.20	24.84	0.64		
50.00	49.92	0.16	49.62	0.76		
75.00	70.08	6.56	74.50	0.67		

The relative errors for each of the procedures performed are shown in the *Tables 4*, 5 and 6, the maximum errors are around 20% for the approximation performed through *Equations 6* and 7, however for measurements with the tube filled with half of each fluid these are equations has a maximum relative error of 0.16%. While, the maximum error for the developed equation is 0.76% for all the fractions of volume.

4. CONCLUSIONS

The modeling of a detector (crystal + housing + photomultiplier tube) by means of the MCNP6 code correctly experimental validated with measurements is a very important step in the practical implementation, since it tends to approach the real case. Moreover, it avoids the need to build

practical models representative of the geometry under study (phantoms) in the initial phase of a project. The measurement geometry developed in the MCNP6 code the allowed to give reliability to the methodology of calculation of volumetric fractions in biphasic systems in the stratified flow regime with different fluids. The results obtained by *Equations 6* and 7 presented a good approximation for calculation when the volume fractions values are equal to 50% (50% oil + 50% de gasoline) of each fluid presenting a maximum relative error of 0.16%, obviously this result extends to the tube completely filled with oil or gasoline. However, for different volume fractions the maximum relative error was 19.39%. This result shows that these equations are not suitable for calculating volume fractions in cylindrical tubes. While the results for the developed analytical equations for cylindrical tubes, (*Equation 11*) presented a maximum relative error of 0.76% in all cases (25%, 50% and 75%), showing that the real results obtained match with the calculated volume fractions for all fluid combinations, even for the worst case oil-gasoline flow since the density is very close to each other when compared to the other combinations. Therefore, the authors recommend the use of *Equation 11* in cylindrical tube in applications whose precision is fundamental.

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