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GEANT4 simulation of water volume fraction measurement in dehydrated crude oil

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Abstract Online measurement of water volume fraction (WVF) in dehydrated crude oil is a difficult task due to very little water in dehydrated crude oil and high precision requirements. We presents a method to measure water volume fraction in dehydrated crude oil with γ-ray densitometry. The Monte Carlo computer simulation packet GEANT4 was used to analyze the WVF measuring sensitivity of the γ-ray densitometry at different γ-ray energies, and effects of temperature, pressure, salinity and oil components on WVF measurement. The results show that the γ-ray densitometry has high sensitivity in γ-ray energy ranges of 16~25keV, and it can distinguish WVF changes of 0.0005. The calculated WVF decreases about 0.0002 with 1℃ of temperature increase and they have approximately linear relation with temperature when water volume fraction remains the same. Effects of pressure, salinity and oil components on water volume fraction can be neglected. Experiments were done to analyze sensitivity of the γ-ray densitometry. The results , as compared with simulations, demonstrate that simulation method is reliable and it is feasible to gauge low water volume fraction using low energy γ-rays.

Key words Water volume fraction, Dehydrated crude oil, Gamma ray densitometry, Monte Carlo method, Compensation algorithm

CLC numbers TE622.1^{+}4, O657.35

1 Introduction

It is necessary to determine water volume fraction (WVF) of crude oil in the process of petroleum dehydration and crude oil measurement. Distillation [1,2], a traditional method to determine water in crude oil, has been widely employed in petroleum and gas industries. With this method, however, crude oil is sampled and tested every two hours generally. The fact that changes in water contents in crude oil dehydration process cannot be monitored on a basis of shorter time intervals risks excessive water content in the crude oil. In addition, the distillation method demands larger expenditures on light oil solvents, electric power and labor. Therefore, developing instruments for measuring low water content in crude oil is necessitated.

Gamma ray densitometry is a technique to measure water and gas volume fraction in oil-water -gas mixtures.[3,4] But a common γ-ray density meter can be applied just for mixtures containing a lot of water, rather than dehydrated crude oil, WVF of which is generally less than 0.01. In this paper, we present a method to solve this problem by using low energy γ-rays. GEANT4, a Monte Carlo simulation software, was employed to investigate the feasibility of proposed approach. Sensitivity of the γ-ray densitometry was simulated, and the simulations were compared to experimental results. Influences of temperature, pressure, salinity and oil components on WVF measurement were analyzed, and an algorithm was given to correct the effect of temperature on measured WVF.

2 Measurement Principle

When a γ-ray penetrates a matter, its intensity is attenuated exponentially, and intensity is given by:

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$$I(E) = I_0(E)\exp(-\int_0^d \int_E^{E+\Delta E} \mu(E,x)\mathrm{d}E\mathrm{d}x) \quad (1)$$

where $I_0(E)$ is the incident intensity of γ-ray at energy E; $\mu(E,x)$ is the linear attenuation coefficient at energy E and distance x along the beam incidence within the matter; d is the distance from the γ-ray source to the detector. With single energy γ-ray and homogeneous matter, Eq.(1) can be simplified as:

$$I = I_0 \exp(-\mu d) \quad (2)$$

Assume that oil-water mixture fills the pipe (Fig.1) and that attenuation effects of the pipe wall, and of air from the γ-ray source to the pipe and from the pipe to the detector, are constant (which will not be taken into account in the following discussions). In Fig.1, the inner diameter of pipe is d, the equivalent thickness of oil and water in the pipe are d_o and d_w respectively, then the d can be expressed:

$$d_o + d_w = d \quad (3)$$

Combining Eqs.(2) and (3), one has:

$$I = I_0 \exp[-d(\mu_o\alpha_o + \mu_w\alpha_w)] \quad (4)$$

where μ_o and μ_w are the linear attenuation coefficient of oil and water respectively; α_o is the oil volume fraction and α_w is the WVF, and $\alpha_o + \alpha_w = 1$. Eq.(4) can be written as:

$$I = I_0 \exp[-d\mu_o - d(\mu_o - \mu_w)\alpha_w] \quad (5)$$

Then α_w is given by:

$$\alpha_w = \frac{\ln(I/I_{oil})}{\ln(I_{wat}/I_{oil})} = k\ln I + b \quad (6)$$

where I_{oil} and I_{wat} are intensities detected with the pipe being filled with oil or water respectively. The factors k and b are given by:

$$\begin{cases} k = 1/\ln(I_{wat}/I_{oil}) \\ b = \ln I_{oil}/\ln(I_{oil}/I_{wat}) \end{cases} \quad (7)$$

From Eqs.(6) and (7), it can be seen that a linear relationship between α_w and natural logarithm of the intensity I because I_{oil} and I_{wat} are constants for the given oil and water. The WVF is calculated using Eq.(6). The k and b can be obtained by means of the linear regression in industry applications.

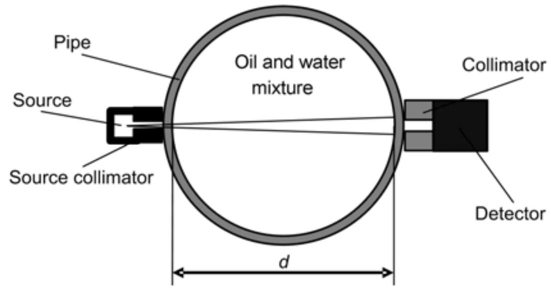

Fig.1 Schematic of a γ- ray gauge for WVF measurement.

3 Simulation Model

When γ-ray transmission method is used to measure WVF of dehydrated crude oil, it is necessary to test precision of the γ-ray densitometry by experiments or simulations. Of course, the latter was of our first choice.

There are many particle simulation softwares, such as MNCP, EGS4, GEANT4 and TART2000, to use Monte Carlo methods for instrument development.[5,6] GEANT4[7] developed by CERN is a toolkit for simulation of particles passage through the matter. Its applications include high energy, nuclear and accelerator physics, and medical and space science as well. It is an open source simulation toolkit, including a complete range of functionalities for simulation, such as managing geometry, tracking, detector response, runs, events, particle definitions, visualization and user interfaces. It was employed in this study.

A simulation geometry constructed by GEANT4 for this study is shown in Fig.2, with a pipe of Φ60mm and a NaI(Tl) detector of Φ30mm×40mm. Instead of crude oil, we used a mixture of hexadecane ($C_{16}H_{34}$, density 0.7733g/cm^3) and water (1.0g/cm^3) with salts of NaCl (2.2g/cm^3), $MgCl_2$ (2.32g/cm^3) and $NaHCO_3$ (2.2g/cm^3) in the simulations. The number of the simulated events was 1,000,000. Interactions of the particles with the oil-water mixtrue were simulated. Events reached the detector were counted as the intensities attenuated by different mixtures. The data were used to analyze sensitivity of the γ-ray densitometry and effects of temperature, pressure, salinity and crude oil composition on WVF.

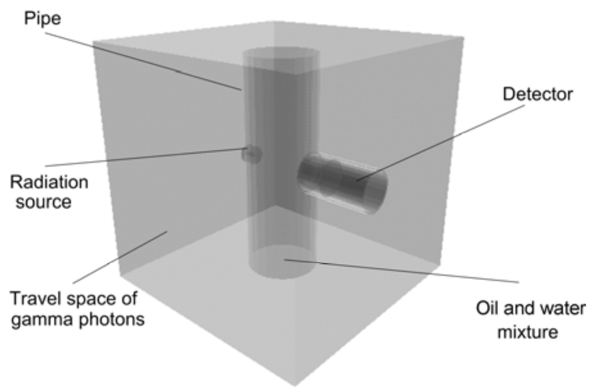

Fig.2 3D simulation geometry implemented by GEANT4.

4 Simulations analysis

4.1 Selection of γ-ray source

As mentioned above, dehydrated crude oil contains very little water, the γ-ray instrument must have high sensitivity to measure the WVF. The sensitivity S is defined as:

$$S = \left| \frac{\mathrm{d}I}{\mathrm{d}\alpha_{\mathrm{w}}} \right| \tag{8}$$

where $\mathrm{d}I$ is the change in γ-ray intensity detected, $\mathrm{d}\alpha_{\mathrm{w}}$, the change in α_{w}. According to Eq.(6), one has

$$S = |1/k| \tag{9}$$

Therefore, the sensitivity can be expressed by absolute value of $1/k$. A large S indicates that an instrument has the ability to detect slight changes of WVF.

Among the factors affecting the sensitivity, the γ-ray energy plays an important role. The oil-water mixture is composed of mainly H, C and O. The γ-ray should be of lower energy.

Fig.3 shows curves of the sensitivity versus γ-ray energy when WVF changes from 0.0 to 0.05. Fig.3(a) shows the sensitivities for the energy range of 15-40keV with a 5keV step. It has a maximum value at about 20keV. In order to know the exact position of the maximum sensitivity, range of γ-ray energy was reduced to 15-22keV with a 1keV step, as shown in Fig.3(b). The maximum sensitivity was found at about 19keV.

Combining Fig.3(a) and (b), a suitable energy range can be 16-25keV, and ^{109}Cd (22.2keV) and ^{238}Pu (16.3keV) can be used for measuring WVF in dehydrated crude oil. For the sensitivity, ^{109}Cd is better, but not for the half-life, which is just 453 days, much shorter than 87.5 years of ^{238}Pu. For online measurement, it is better to use ^{238}Pu isotope.

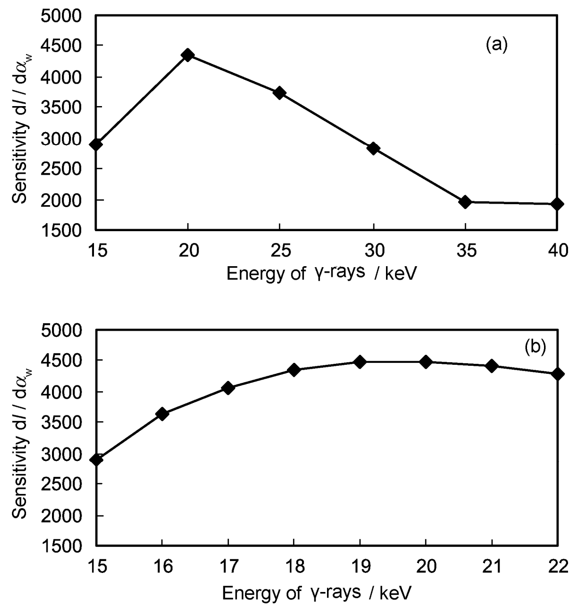

Fig.3 Sensitivity vs γ-ray energy when WVF changes from 0.0 to 0.05. (a) 15-40keV with a 5keV step, (b)15-22keV with a 1keV step.

The ^{238}Pu was judged on whether it meets the requirements of measuring WVF in dehydration crude oil. Oil-water mixtures in different ratios were tested. The proportion of water volume in a mixture was regarded as the true WVF. Simulations were done by changing the WVF from 0.0 to 0.01 with a 0.001 step.

As shown in Fig.4, the detected radiation intensity changes with WVF obviously. A linear regression was carried out using simulation data. Based on Eq.(6), the regression parameters k and b were obtained. The WVF was calculated with the linear regression parameters. The maximum absolute error of WVF is 0.00026 between the calculated WVF and true WVF. The results are shown in Fig.5. The linear regression equation is $\alpha_{\mathrm{w}} = -0.201314684 \times \ln I + 2.301022993$.

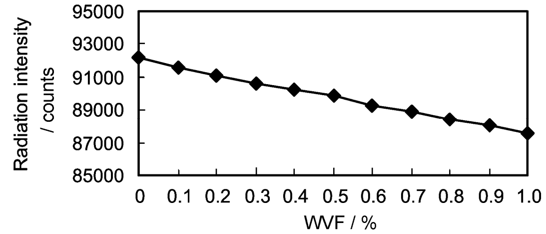

Fig.4 Variations of radiation intensity (counts) with WVF.

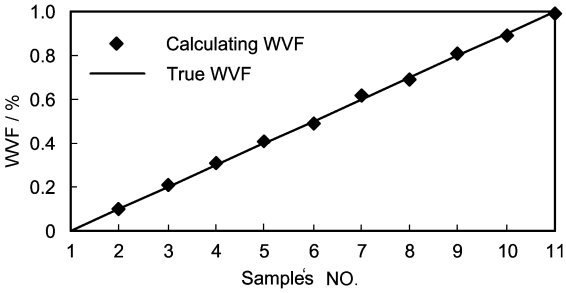

Fig.5 Comparison between calculated WVF and true WVF.

4.2 Effects of temperature and pressure

The above simulations were done under standard temperature and pressure. But in on-line applications, the γ-ray densitometry measures WVF of the mixture with changing temperature and pressure, which cause density changes of the water and oil, and finally affect the measurement result of WVF.

Fig.6 shows the calculated WVF at different temperatures of the mixture. The constants of k and b were calculated at 50℃, 1.0MPa and 0.004 WVF. The WVF varies almost linearly with temperature, decreasing about 0.0002 with 1℃ of temperature increase. Thus, the temperature compensation algorithm can be given by:

$$\alpha_{\rm w} = k\ln I + b + c(T_{\rm s} - T) \qquad (10)$$

where c=0.0002 is the temperature compensation coefficient, $T_{\rm s}$ is the calibration temperature and T is temperature of the mixture.

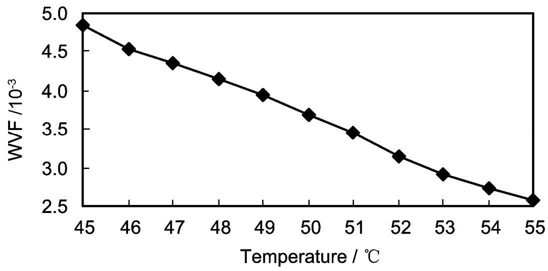

Fig.6 Calculated WVF at different temperatures. The simulations were done with WVF of 0.004. The linear regression was done at 50℃ and pressure 1.0MPa. The linear equation is $\alpha_{\rm w}$=-0.215468872×ln$I$+2.470111724.

The calculated WVFs at different pressures are shown in Fig.7. The WVF changes very little when pressure increases from 1.0MPa to 1.5MPa. It is most likely that the coefficients of compressibility of oil and water change very little with pressure. Thus, the effect of the pressure on WVF can be neglected.

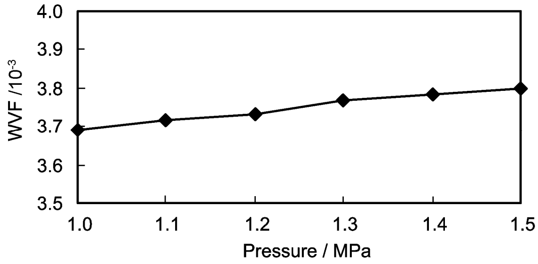

Fig.7 Calculated WVF at different pressures. The simulations were carried out with WVF of 0.004. The linear regression was done at 50℃ and 1.0MPa. WVF was calculated by linear equation $\alpha_{\rm w}$=-0.215468872 ×ln$I$+2.470111724.

4.3 Effect of salinity

The water in crude oil contains different salts, which contain typically positive ions of Na^{+}, Mg^{2+} and Ca^{2+} and negative ions of Cl^{-} and HCO_3^{-}. The water in crude oil has different origins, such as formation water, injected water or water to clean oil well, hence different compositions. Fig.8 shows the simulation results of the salinity effect on calculated WVF, with WVF of 0.004 and defining mass ratio of salt to water as unit of salinity. The calculated WVF changes little with the salts in different salinity. Therefore, for the dehydrated crude oil containing very small amount of water, the salinity effect on WVF is negligible, too. For crude oil having a large amount of water, however, the salinity effect cannot be neglected, and the single energy γ-ray transmission method cannot be used to correct the variations of salinity. Refs. [8] and [9] gave an approach to produce water characterization by the dual mode densitometry.

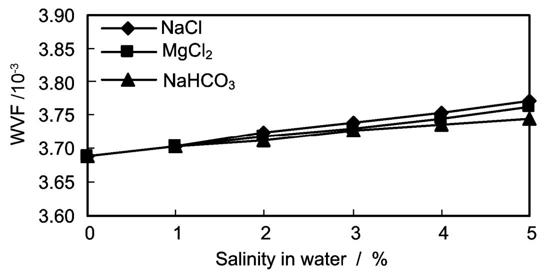

Fig.8 Effects of salinity on calculated WVF for different salts. The simulations were carried out with WVF of 0.004. The linear regression was done at 50℃ and 1.0MPa. WVF was calculated by linear equation $\alpha_{\rm w}$=-0.215468872×ln$I$+2.470111724.

4.4 Effect of composition of crude oil

Crude oil is hydrocarbon mixtures of alkanes, cycloalkanes and aromatic hydrocarbons, etc. However, as all of them consist of hydrogen and carbon, it is expected that composition change of crude oil has little effects on calculated WVF, as can be clearly seen in Fig.9. Heptadecane was used as the adding component. The mass ratio of heptadecane to hexadecane is as abscissa. The calculated WVF changes very little up to a heptadecane content of 5%.

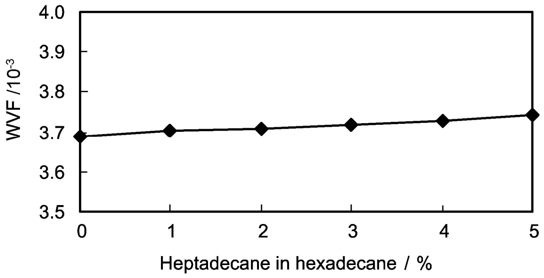

Fig.9 Effects of composition in crude oil on calculated WVF. The simulations were done with WVF of 0.004. The linear regression was done at 50℃ and 1.0MPa. The WVF was calculated by linear equation α_w= -0.215468872×ln$I$+2.470111724.

5 Experimental setup and results

In order to evaluate validity of the simulation results, an experimental setup was established, with a ^{238}Pu source of 3.7GBq and a Φ60mm pipe with oil-water mixture flowing in it. The 16.3keV γ-ray was detected by a NaI(Tl) scintillator coupled to a photomultiplier tube. The signals were processed by an ORTEC 575A amplifier. Fig.10 is a schematic illustration of the experimental setup.

Measurements were carried out using diesel oil (0.826g/cm^3) mixed with different volume proportions of water (used as the true WVF). Each counting of the γ-rays lasted 3 min at 40℃ of the mixture. The results are shown in Fig.11, which is similar to Fig.4.

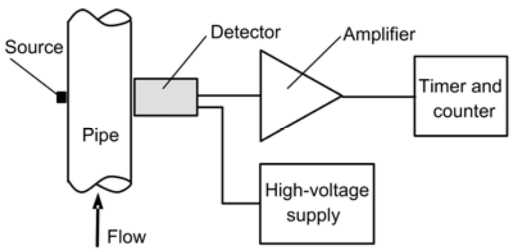

Fig.10 Schematic of the experimental setup.

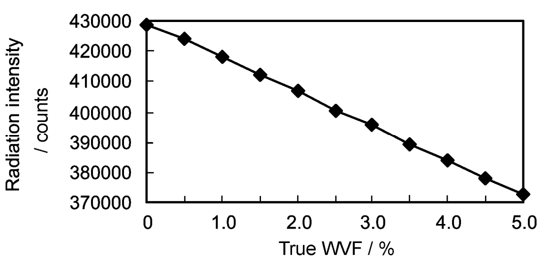

Fig.11 Change of radiation intensity detected with WVF.

Experiments were done also for testing sensitivity of the setup. The results were shown in Fig.12. The calculated WVFs were computed using the regression parameters obtained from the data of Fig.11. The maximum absolute error between the true WVF and the calculated WVF is 0.19%, greater than that of simulation. This is reasonable, as the simulation was done at ideal conditions, but the experiment was affected by many factors such as statistical error of source and time error.

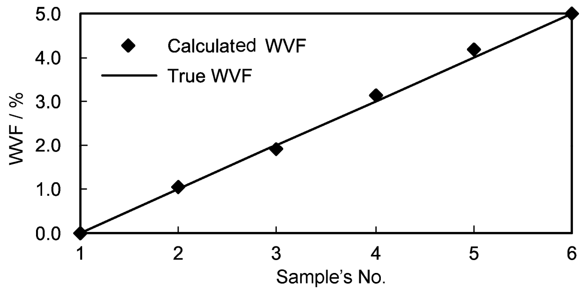

Fig.12 Comparison between measured WVF and true WVF. The WVF is magnified 100 times. The linear regression parameters come from the data in Fig. 11. The equation is α_w=- 0.35218355×ln$I$+ 4.5683593.

6 Conclusions

The simulations show that the system has high sensitivity at the energy range of 16~22keV. The maximum absolute error of WVF is about 0.00026 between the calculated WVF and true WVF at 16.3keV. This indicates the low energy γ-ray method can satisfy the requirements for high precision measurement of WVF in dehydrated crude oil, and ^{238}Pu is acceptable. Temperature changes of oil-water mixture will introduce large deviations to the measurement of WVF. But the error can be corrected by a temperature compensation algorithm according to the simulations. The effects of pressure, salinity and compositions of crude oil on WVF can be neglected. The experimental results are similar to the simulations. Our work demonstrates that the Monte Carlo computer simulations can be used to design and optimize the radiation measurement system.

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