ABSTRACT

Tc-99m as radiotracer is used to investigate the hydrodynamics of water flooding operation inside the porous media column. Tc-99m is gamma emitter isotope with a half-life of 6 hours and energy of 0.140 MeV (90%). The porous media consists of fine sandstone with a grain size of 850 µm, kerosene as trapped oil and a layer of cement on top of the rig as the bedrock. Gamma camera is arranged next to the porous media in order to capture the movement of radiotracer which has been set to 1 minute per frame. About 1.3 mCi/0.3 ml of Tc-99m is eluted from the Molybdenum generator and injected at the upstream of the porous reactor. After injection, the addition of 3 ml of water is pushed through the system to purge the line from Tc-99m. The gamma camera gives a quantitative determination of local fluid saturations over the area of observation. Besides, Au-198 that emits gamma ray with a half-life of 2.7 days and energy of 0.41 MeV (99%), it is also used to check the compatibility of other radiotracers for water flooding activities. Thus, the objectives are to check the compatibility of radiotracers as well as to provide insights into fluid hydrodynamics (flow visualization) inside the porous media using gamma camera. The study shows that water breakthrough is recorded as 5 µSv/h after 5 minutes of continuous water flooding and maintain at 20 µSv/h after 15 minutes where all tracer is collected at the outlet container. It is proven from this study that Tc-99m is the best water tracer for water flooding activities.

Keywords: Au-198, flow visualization, gamma camera, porous media, radiotracer, Tc-99m

INTRODUCTION

Crude oil development and production of oil reservoirs can include up to three distinct phases: primary, secondary, and tertiary recovery. During primary recovery, the natural pressure of the reservoir or gravity drives oil into the wellbores, combined with artificial lift techniques which bring the oil to the surface (Reddy, 2013). The expected initial extraction rate of available hydrocarbons reservoir's original oil in place is around 10-15% typically produced during primary recovery. This will leave around 85% of hydrocarbons in the reservoir. Secondary recovery techniques extend a field's productive life generally by injecting water or gas to displace oil and drive it to a production wellbore (IAEA, 2003). Pump jacks for water flooding and initial gas injection can increase the recovery up to 25-30% of the original oil in place. However, producers have attempted enhanced oil recovery (EOR) techniques, techniques that offer prospects for ultimately extracting another 10-15% of the initially available hydrocarbons or producing 30 to 60 % or more, of the reservoir's original oil in place (Tunio et al., 2011).

The petroleum potential in Malaysia has mainly fractured basement rock reservoir located offshore and characterized by a fracture system of very high heterogeneity in porosity and permeability. The
complexity of the reservoir structure introduces considerable uncertainty in the reservoir model and fluid flow simulation. The extraction of residual oil requires modification in reservoir characteristics in which water flooding is most commonly used for enhanced oil recovery. In this operation, the water is injected through the residual oil under high pressure to extract the oil (Khan et al., 2003). This situation calls for more application of tracer technique and challenges to tracer technology. In view of rising energy consumption, the use of advanced technologies to boost oil recovery from mature oil fields in Malaysia is most welcomed. In addition to it, the innovative application of the gamma-camera-imaging system as a quantitative method for investigating the physics of oil displacement within a thin slab of the porous medium has been successfully carried out using radiotracing medium (Huang and Gryte, 1988). The gamma camera can provide the behavior of the water and oil movement in the porous media during water flooding. In this paper, the preliminary result is shown on the intervention of radiotracer in porous media which mimics the reservoir with gamma camera as a tool in the quantitative determination of local fluid saturations over the area of observation.

Thus, the objectives are to investigate the compatibility of radiotracers as a radioactive tracer in porous media and to visualize the water-oil movement inside the porous media using gamma camera.

**MATERIALS AND METHODS**

**Preparation of Water-Oil Media**

The experimental set up is arranged as shown in Figure 1. In this study, a peristaltic pump is used to supply water continuously with a flow rate of 1.17 ml/s. Initially, the fine sand is compacted inside the container with a size of 20 cm in diameter; 20 cm in height and upon completion, water is saturated inside the container. The water saturated container is left overnight to ensure the water is fully saturating the sand. Once the sand is fully settled with water, about 100 ml of turpentine is injected inside the container and the water discharged is measured. The volume of water discharged is the initial oil saturation. The oil-laden container is now left overnight in ensuring the oil is considered settling in the porous media container.

![Figure 1: Schematic diagram of the experimental setup](image-url)
Flow Visualization Set Up

Gamma camera Hamamatsu model BHP6602 is used for imaging and monitoring the flow of fluid inside the porous media whereas a single NaI scintillation detector is placed at the outlet of the container. The detector is attached to the data acquisition system (DAS) which is connected to the computer to acquire the data in the form of counts per second. This data is used to determine the residence time distribution (RTD) of the tracer. RTD is a parameter to indicate the average time particle stays or resides inside the column. About 1.3 mCi/0.3 ml Tc-99m was injected inside the tracer line when everything is ready. Similarly, the gamma camera is inactive dynamic mode when the tracer starts flowing inside the column. In this study, the tracer injection is introduced at the bottom of the container. Gamma camera is set to capture an image of one frame per minute and once the gamma camera is ready for dynamic study, the experiment is set.

RESULTS AND DISCUSSION

Determination of Actual Activity

The dose rate recorded was 100 µSv/h at the surface of uncollimated liquid gold and 3 µSv/h at 1 m distance. For that matter, the actual activity is calculated using the equation as below:

\[
\text{Dose rate (mR/h)} = 0.55 \times \text{Energy (Mev)} \times \text{Activity (mCi)} \text{ for 1m distant}
\]

\[
\text{Activity (mCi)} = \frac{0.3mR}{0.55 \times 0.41} = 1.3 \text{ mCi/48.1 MBq}
\]

Thus, the actual activity for radiotracer injection is 1.3 mCi or 48.1 MBq throughout the study.

Flow Visualization Using Gamma Camera

Figure 2 shows the hotspot of the radioisotope of Tc-99m inside the vial. The bright reddish and greenish colors indicate the high intensity of highly concentrated Tc-99m which resembles to 1.8 mCi and sufficient for image detection for this experiment. It can be observed from Figure 2 also that each circle equivalent to 1 minute. Therefore, the experiment commences at t = 1 minute and ends at t = 20 minutes. At t = 2 min, Tc-99m is observed as having the highest intensity and gets into the sand column with Dirac pulse, which is due to instantaneous injection of Tc-99m at the inlet. This indicates that all tracer is fully entered the column and as time increases with a continuous supply of water due to water flooding operation, the Tc-99m has dispersed and developed significant territory inside the sand column. At t = 4 min, Tc-99m has invaded half of the column and swept almost the surface of the sand column. It can be observed clearly that at t = 5 min, Tc-99m is flowing towards the outlet which is located at the upper side of the column and continuously taking away all of the water-Tc-99m-tag out of the column and reduces in Tc-99m amount gradually. At t = 20 min, all of Tc-99m is officially leaving the column and leaves no trace of Tc-99m. It shows that Tc-99m can represent the water flooding well without adsorbing on the sand and capable to be used for water-flooding study for EOR application in the future.
Alternately, another tracer Au-198 is tested for this study to check for its compatibility with water-flooding activities. Figure 3 shows that the moment Au-198 is injected inside the porous media, the image of Au-198 as in green color is recorded and monitored. At early minutes (t = 2-3 min), it seems that the liquid radiotracer is dispersed inside the media following the water supplied with a volume of 1 L for injection. The water containing radiotracer seems to occupy the porous media containing the oil and therefore, oil recovery is expected at the outlet. This is because water reservoir is meant to push the trapped oil inside the media as water flooding mechanism. Nevertheless, after waiting for about 20 minutes, no oil is collected at the collected pot. Figure 3 also shows that the accumulation of Au-198 inside the media and the reading are around 2200-2300 counts. There is no significant tracer breakthrough although the addition of water is carried out. Besides, the dose rate is recorded at media as 2 mR/h which equivalent to 0.8 mCi or 29.6 MBq. This indicates higher intensity is trapped inside the media. These preliminary results show that Au-198 is adsorbed onto the sand and does not compatible with water flooding study and should not be used for EOR application laboratory works or related. Besides, the colloidal form of gold will agglomerate when the original concentrated solutions mixed with water. Thus, attractions occur between gold particle and sand particle that caused it gold entrapment inside the sand.
Residence Time Distribution (RTD)

Radiotracer experiment is carried out by installing NaI scintillation detector at the outlet of the column whereby the detector is attached to the DAS and PC respectively to monitor the tracer movement inside the porous column. Figure 4 shows the raw data of cps with respect to time in second. In order to measure the RTD which is the main parameters in determining the time tracer resides in the column, the raw data should be treated. Data treatment is crucial in ensuring the only radioactive signal is being measured thus, parasitic signals are removed. Kasban et al. (2010) provide detail of data treatment. Figure 5 shows the treated data of radiotracer experiment. The early spike is due to the injection point is very near to the column which contributes to the first peak. Figure 6 indicates the primary peak contains rich information of the behavior of the sand column and should start from zero and ends at zero to form RTD curve.
RTD is a fundamental parameter in reactor design which can give information on how long the substrate has been in the reactor and the RTD analysis can help to characterize the extent of their deviation from ideal behavior. The $E(t)$ function is the RTD where $E(t) \, dt$ is the fraction of the flow, measured at the exit, that is in the system between times $t$ and $(t + dt)$:

$$E(t) = \frac{C(t)}{\int_0^\infty C(t) \, dt}$$

(Eq.2)

Where, $C(t)$ is the concentration of radiotracer monitored by NaI scintillation detectors in counts per second (cps) as numerator and denominator is the area under the curve of plotted $C(t)$. The detected signal is normalised by dividing it by the area under the curve as shown in Eq. 2. The mathematical expression for the First Moment ($M_1$) in discrete form can be written as in Eq. 3:

$$M_1 = \frac{\int_0^\infty t \, C_i(t) \, dt}{\int_0^\infty C_i(t) \, dt}$$

(Eq.3)

$M_1$ is the calculation of MRT, whereas Zero Moment ($M_0$) is the measurement of area under the RTD curve. In this study, the value of the experimental MRT shows the time taken for water flooding operation is 414 s (6.9 minutes) inside the sand column reactor. The question arises as why the time taken from the gamma camera is longer. The reason is MRT measures the average of each particle travelling from the moment it gets into and leaves the reactor whereas the gamma camera shows the movement of a batch of tracer once it gets into and exits the reactor. In order to ensure there is no trace of tracer inside the reactor, the time visualization is extended using gamma camera.

International Atomic Energy Agency (IAEA) has recommended six RTD models and distributed in-house RTD software to be used for RTD study. Figure 7-9 are 3 models that fit well with the experimental curve by using the software. The models are a perfect mixer in the series model, perfect mixer in series with exchange model and perfect mixer in the parallel model.
Figure 7: Perfect mixer in series model

Figure 8: Perfect mixer in series with exchange model

Figure 9: Perfect mixer in parallel model
The best fit is chosen based upon the minimum value of the sum of the square, which is the difference between the experimental data and the model. Furthermore, the sum of the squares of the differences between the model and the data are minimized and fulfilled using the following equation (Sugiharto et al., 2009):

\[
\text{Sum of Square (SSE)} = [1N_T \int (E_{\text{exp}}(\theta) - E_m(\theta,N))^2 d\theta]^{1/2} = \text{Minimum}
\]  
(Eq.4)

Where, NT is the number of data points, \(E_{\text{exp}}(\theta)\) is the experimentally measured curve, and \(E_m(\theta, N)\) is the simulated model. Table 1 shows the results of optimized parameters for successful RTD model.

<table>
<thead>
<tr>
<th>Models</th>
<th>(\tau_1)</th>
<th>(\tau_2)</th>
<th>(J_1)</th>
<th>(J_2)</th>
<th>(T_m)</th>
<th>(K)</th>
<th>(Q_1/Q)</th>
<th>Sum of Square (SSE) (\times 10^{-7})</th>
<th>Rank</th>
</tr>
</thead>
<tbody>
<tr>
<td>Perfect mixer in series model</td>
<td>523 s</td>
<td>nil</td>
<td>1.7</td>
<td>nil</td>
<td>nil</td>
<td>1.7</td>
<td>nil</td>
<td>0.337</td>
<td>3</td>
</tr>
<tr>
<td>Perfect mixer in series with exchange model</td>
<td>98.8</td>
<td>nil</td>
<td>7.3</td>
<td>nil</td>
<td>117 s</td>
<td>4.0</td>
<td>nil</td>
<td>0.182</td>
<td>1</td>
</tr>
<tr>
<td>Perfect mixer in parallel model</td>
<td>562</td>
<td>22</td>
<td>1.4</td>
<td>3</td>
<td>nil</td>
<td>nil</td>
<td>1</td>
<td>0.189</td>
<td>2</td>
</tr>
</tbody>
</table>

From Table 1, the perfect mixer in series and perfect mixer in the parallel model can describe the experimental curve well since the value of MRT or \(\tau\) is comparable with the experimental MRT which is 414 s. The value of \(\tau\) derived from each model is greater than the experimental value can be a sign of stagnant zone inside the reactor. The stagnant zone has extended the time the tracer should reside inside the reactor.

CONCLUSIONS

Tc-99m is the best water-tracing radiotracer in porous media. It shows clearly the movement of water-bulk from the inlet to the outlet in which gamma camera has proved the clear visualization of water migration. The Au-198 is not recommended for water-oil-tagging tracer and it is definitely meant for sand tracing application. The result shows that most Au-198 tracer is trapped inside the oil-laden sand and the rest is distributed on the sand particles although water flooding is introduced continuously. Nevertheless, gamma camera can be used to study the hydrodynamics of oil inside the Laboratory Porous Media provided the selection of oil tracer is correct. This is because, for the laboratory study, the movement of water or oil can be traced accordingly (static or dynamic) and monitor respectively.

RTD model from the experiment shows that the perfect mixer in parallel model fits the RTD curve well. Moreover, the experimental MRT shows the time taken for water flooding operation is 414 s and each model provides a greater value of \(\tau\) that indicates the presence of stagnant zones inside the column. The formation of the stagnant zone has increased the time of water tracer should reside inside the reactor. The question arises as why the time taken from the gamma camera is longer. The reason is, MRT measures the average of each particle travelling from the moment it gets into and leaves the reactor whereas the gamma camera shows the movement of a batch of tracer once it gets into and exits the reactor. In order to ensure there is no trace of tracer particle inside the reactor, the time visualization is extended using gamma camera and cause the contradiction in time.
ACKNOWLEDGEMENTS

The authors would like to express their special thanks to International Atomic Energy Agency (IAEA) for the financial support from CRP-IAEA Research Contract No: 22898 and Prof Louis Brandao from Instituto de Engenharia Nuclear, Rio De Janerio and all Plant Assessment Technology (PAT) team for assisting us throughout this study.

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