

## THE IMPORTANCE OF CALCULATING RESULTING PERMITTIVITY AND CONDUCTIVITY IN MIXTURES OF TWO LIQUIDS

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### Summary

Calculation of resulting permittivity and conductivity in mixtures of two liquid components is important for reliable use of capacitance and micro-wave sensors for concentration measurement in mixtures. Especially, in three component flows (e.g. crude/water/gas), a reliable model is necessary for accurate determination of the different component concentrations. There do exist many formulae worked out by i.e. Maxwell (1873), Fricke (1924), Bruggeman (1935), Sillars (1937) and Van Beek (1967), Ramo and Rau (1973) and Hammer (1983) and many other but non of them have shown to be reliable when the continuous component is electrically conducting. A homogeneous mixture of saline water (5 S/m) and crude oil from the Gullfax field has been measured at different frequencies in a homogeneous electric field (Specially designed test cell) with water fractions of 0 - 100% and compared with the results calculated by the different existing formulae. A numerical model has also been developed and data simulations show that the droplet size does not influence on the permittivity of the mixture.

This work shows that Bruggeman's formula for permittivity in two component mixtures agrees best with the eksperimental results on a mixture of crude oil and saline water.

### 1. INTRODUCTION

Capacitance and microwave transducers are commonly used in multi-component flowmeters to determine the fraction of process water in crude oil/water mixtures. Reliable results are dependent on accurate mathematical models of permittivities in multi-component mixtures. Many models have been developed but they give different results and modification based on empirical data has ben necessary to obtain the demand accuracy.

In this work we have developed a numerical model based on the finite element method and Laplace equation which we have used as the reference in our analyses of the existing formulae together with permittivity measurement on a mixture of Gullfax crude oil and saline water.

A special test cell for measuring the permittivity in mixtures of oil and water has ben developed for obtaining reliable experimental data.

### 2. SOME EXISTING FORMULAE FOR CALCULATING PERMITTIVITY AND CONDUCTIVITY IN TWO COMPONENT MIXTURES

In 1873 Maxwel [1] published a theory of calculating the permittivity of a mixture of two dielectric materials. He assumed small spheres of one material uniformly distributed in the other. He also assumed a homogeneous electrostatic field, constant diameter of the spheres and that this diameter was small compared with the distance between the spheres. Bruggeman [2] developed, in 1935, a model based on Maxwells theory in which the discontinuous component consisted of spheres of random diameter and distribution.

The Bruggeman's formula is:

$$\frac{\epsilon_2 - \epsilon_m \left( \frac{\epsilon_1}{\epsilon_m} \right)^{\frac{1}{3}}}{\epsilon_2 - \epsilon_1} = 1 - \beta \quad \{1\}$$

Where

$\epsilon_1$  = relative permittivity of the continuous component

$\epsilon_2$  = relative permittivity of the discontinuous component

$\epsilon_m$  = relative permittivity of the mixture

$\beta$  = fraction of component 2

This model has shown to fit very well with experimental results up to 40% water cut and a modified version of this model has therefore been used in most of multi-component meters. This model can be used for mixtures of crude oil and process water. However, since the process water has a conductivity of approximately 5 S/m the water droplet acts as electrical short cuts, equivalent to put the permittivity of the water component ( $\epsilon_2$ ) equal to infinity, if the measurement frequency  $f \ll f_d$ , where  $f_d = 1,3$  GHz is the dispersion frequency of process water.

By introducing  $\epsilon_2 = \infty$ , the Bruggeman formula then reduces to:

$$\epsilon_m = \frac{\epsilon_{oil}}{(1 - \beta)^3} \quad \{2\}$$

Where  $\epsilon_{oil}$  is the relative permittivity of the oil.

Many others including Rayleigh [3], Wiener [4], Fricke [5], Sillars [6], van Beek [7] and Hammer [8] have developed formulae for calculating the permittivity and conductivity of mixtures of two different dielectrics.

Ramo and Rao [9] claim, however, that these formulae are inaccurate if one component of the mixture has a high conductivity as i.e. process water in crude oil. Based on a model developed by van Beek, Ramu and Rao have derived formulae which are also valid under such conditions. These formulae have been derived under the assumption that a number of small spheres (material 2) of radius  $r_2$ , permittivity  $\epsilon_2$  and conductivity  $\sigma_2$  are uniformly dispersed in a spherical medium (material 1) of radius  $r_1$ , permittivity  $\epsilon_1$  and conductivity  $\sigma_1$  which in turn is surrounded by continuum of material 1. It is further assumed that  $r_1 \gg r_2$ . The authors have found that these hold well in the case of low-loss liquid mixtures even if the volume fraction of the dispersed component is as high as 0.4 - 0.45. In the case of high-loss additives, they can only be used with reasonable accuracy when the volume fraction of the dispersed component is less than 0.2 - 0.25. For higher concentrations Ramu and Rao found a considerable difference between the predicted and experimental values. They assumed that this discrepancy was due to higher order interaction between the dispersed particles themselves and between the dispersed particles and the continuum.

These formulae are the only ones developed which can be used for any of the components as the continuous component and can therefore be used for calculation of the permittivity and conductivity of a mixture of oil and water with the water as the continuous component even at saline water with conductivity of 5 S/m (used as substitute for North Sea process water in the laboratory experiment).

For mixtures of oil and process water of  $\sigma = 5 \text{ S/m}$ , the Ramo and Rao's formulae can be written:

Oil continuous phase:

$$\epsilon_m^o = \epsilon_{oil} \frac{1 + 2\beta}{1 - \beta} \quad (3)$$

Water continuous phase:

$$\epsilon_m^w = \epsilon_w \frac{2\beta}{3 - \beta} \quad (4)$$

where  $\epsilon_w$  is the relative permittivity of the process water.

### 3. THE NUMERICAL MODEL FOR CALCULATION OF PERMITTIVITIES AND CONDUCTIVITIES IN TWO COMPONENT MIXTURES

Our numerical model consists of a unit size parallel plate capacitance sensor equipped with sensor screen and guard electrode (Figure 1). The regime between the parallel plate electrodes is a two component mixture where the discontinuous component, i.e., the spheres, is uniformly distributed in the continuous component. The potential, electrical field, and capacitance are calculated by solving the Laplace equation using the Finite Element method (FEM).

Since FEM solves Laplace equation exactly, and thus takes the polarization effect into account, the calculated potential, electrical field distribution, capacitance and thus the permittivity of the homogeneous mixture should be reliable for the droplet distribution simulated. By using guard electrodes in the FEM-model the electrical field in the detector volume will not fringe and, hence, increase the accuracy of the calculations.

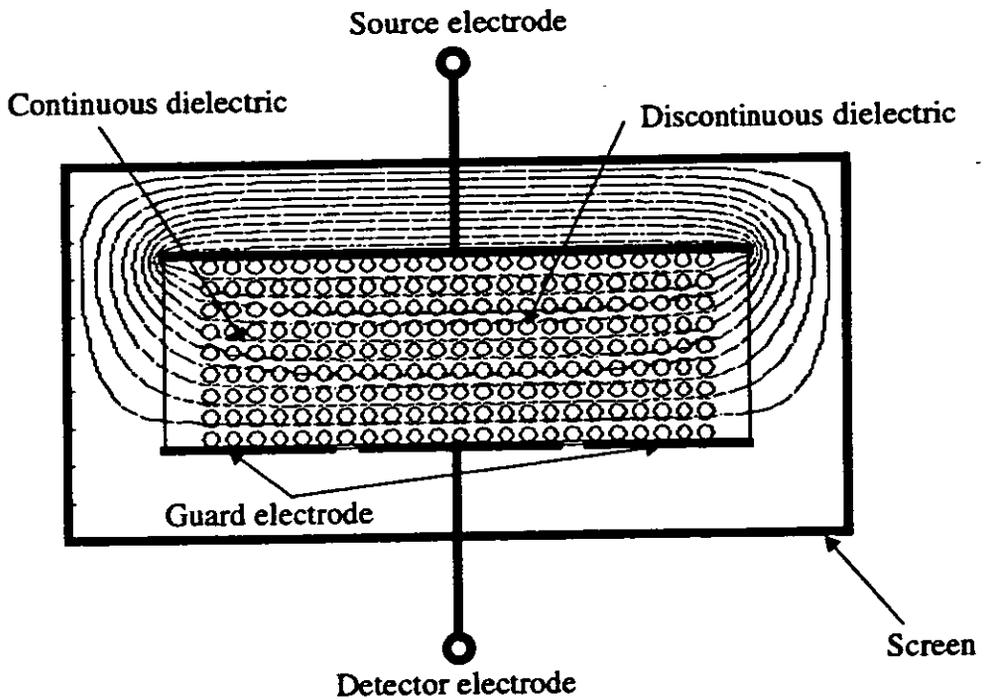


Figure 1. Sketch of the model used for the numerical calculation

#### 4. THE TEST CELL

The test cell arrangement is shown in Figure 2. Two propellers keep the water and oil homogeneously mixed in the test cell.

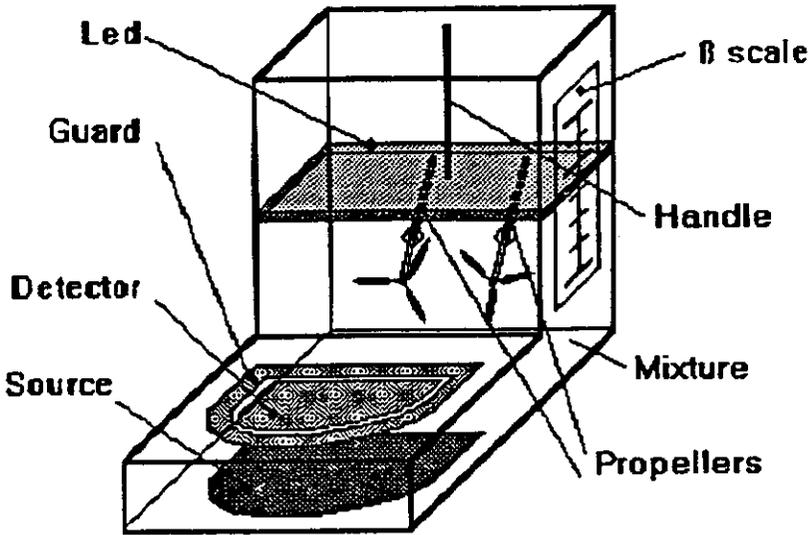


Figure 2. The test cell arrangement

The cell is equipped with a guard electrode surrounding the detector electrode in such a way that the electrostatic field in the measurement volume is kept homogeneous if the mixture is homogeneous. The capacitance and conductance between the electrodes are measured by HP - Impedance Analyzer

The equivalent diagram of this cell is shown in Figure 3.

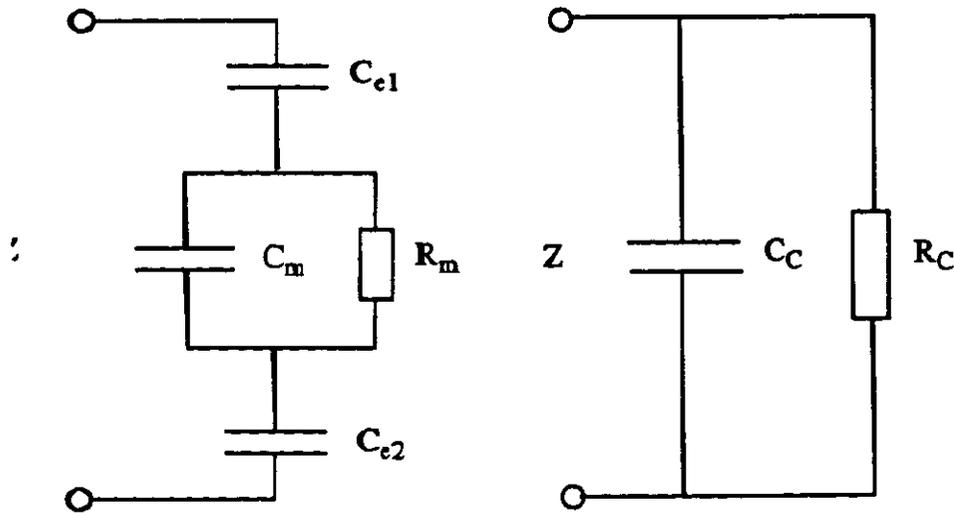


Figure 3. The equivalent diagram of the test cell.

Here  $C_c$  and  $R_c$  are the measured cell capacitance and resistance,  $C_{e1}$  and  $C_{e2}$  is the electrode capacitance, e. i. the capacitance between each electrode and the mixture with the electrode

insulation layer as dielectricum.  $C_m$  is the capacitance of the mixture and  $R_m$  is the resistance of the mixture between the detector electrode and source electrode.

## 5. CALCULATION OF THE MIXTURE PERMITTIVITY AND CONDUCTIVITY

To calculate the permittivity and conductivity of the mixture,  $C_m$  and  $R_m$  must be determined from the measurement of  $C_c$  and  $R_c$  according to the following formulae:

$$C_c = \frac{C_e \{1 + \omega^2 R_m^2 C_m (C_e + C_m)\}}{1 + \omega^2 R_m^2 (C_e + C_m)^2} \quad \{5\}$$

$$R_c = \frac{1 + \omega^2 R_m^2 (C_e + C_m)^2}{\omega^2 R_m C_e^2} \quad \{6\}$$

Where  $\omega = 2\pi f$  is the angular frequency of the impedance analyzer and  $C_e = 1/2C_{e1} = 1/2C_{e2}$

It is important to be aware of that at oil-continuous phase,  $R_m$  is high due to low conductivity in crude oils and if the detector frequency is low the polarization loss in the mixture is negligible. Thus  $\omega^2 R_m^2 C_m^2 \gg 1$  and equations {5} and {6} will be reduced to:

$$C_c = \frac{C_e C_m}{C_e + C_m} \quad \{7\}$$

$$R_c = \frac{R_m (C_e + C_m)^2}{C_e^2} \quad \{8\}$$

This means that if the measurement frequency is low so the dielectric loss is low, the permittivity in the mixture will be frequency independent and also independent of the conductivity of the process water when the oil is the continuous phase in the mixture.

The permittivity and conductivity can then be found according to:

$$\epsilon_m = \frac{C_m d}{\epsilon_0 A} \quad \{9\}$$

$$\sigma_m = \frac{d}{R_m A} \quad \{10\}$$

Where  $d$  is the distance between the sell electrodes and  $A$  is the area of the detector electrode.

When water is the continuous phase the given inequality ( $\omega^2 R_m^2 C_m^2 \gg 1$ ) is not valid and it can be seen that the measurement results will be dependent both on the frequency and the water conductivity.

## 6. THE CALCULATED AND EXPERIMENTAL RESULTS

### 6.1 The experimental results

The experimental results are given in Figure 4.

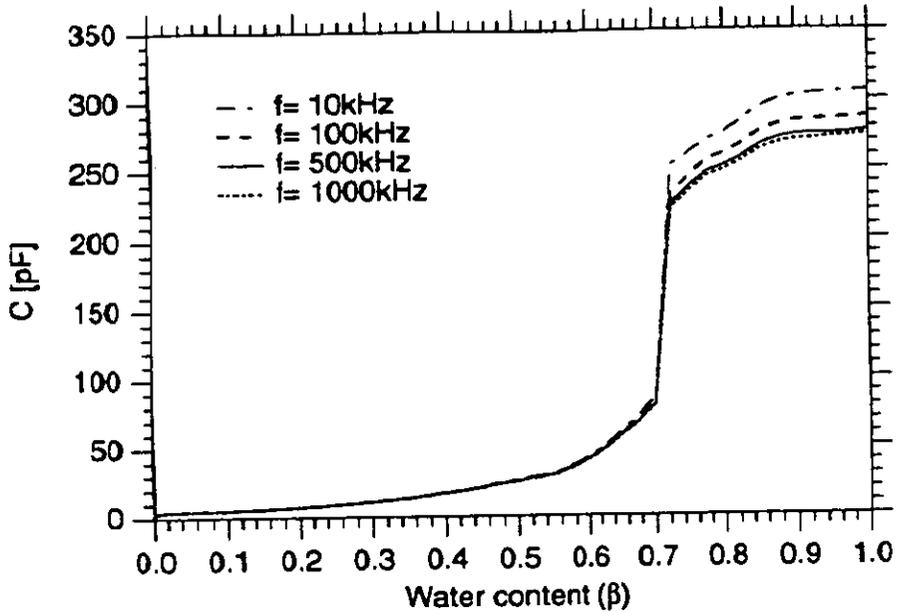


Figure 4. The capacitance of the test cell - homogeneous mixture of Gullfax crude oil and saline water (5 S/m)

The simulation results are shown in Figure 5. together with the result from Bruggeman's, Ramo & Raos', and Hammer's model. The permittivity  $\epsilon_{oil}$  is 2.2 and  $\epsilon_w$  is 71. The conductivity of the water  $\sigma_w$  is 5 S/m.

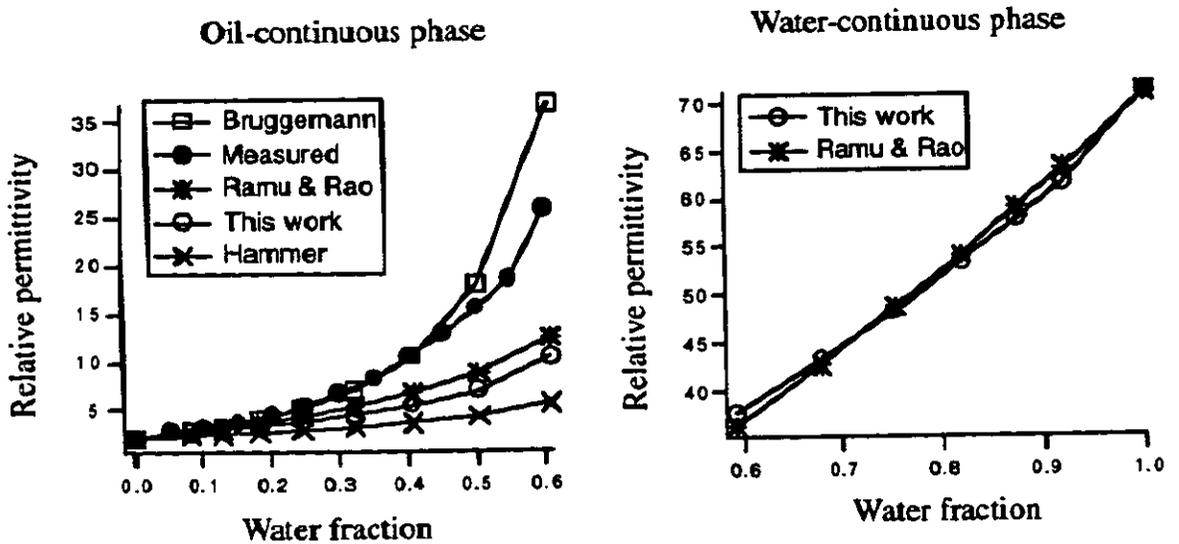


Figure 5. Permittivity of a homogenous mixture of crude oil and water (5 S/m) versus water concentration according to Bruggeman, Ramo & Rao, Hammer, measured capacitance of gull-fax crude and simulation results derived in this work.

Our numerical model also shows that the mixture capacitance is independent of the droplet diameter if the diameter is less than 1/5 of the width of the detector electrode.

## 7. CONCLUSIONS

From Figure 5 we can see that Bruggeman's formula gives the best result compared with the test cell measurements but even this formula must be modified for water fractions greater than 40%. All the other formulae under-estimate the mixture permittivity and so does our numerical model. The explanation of this is probably that both Ramo and Rao, Hammer and our numerical model are calculating the permittivity in mixtures where the discontinuous phase is arranged in columns between the sensor plates as shown in Figure 1. In a crude oil/process water mixtures the droplets will be randomly distributed resulting in an higher measured capacitance. Bruggeman's formula is based on random distributed diameters and position of the droplets in the mixture and gives therefore the most accurate result among all the existing formulae. Our numerical model must therefore be altered to generate random distributed droplets in the measurement volume.

Figure 4. shows that the capacitance of a mixture of crude oil and saline water is independent of the detector frequency when the mixture is oil continuous. It is also independent of the conductivity of the water component if the frequency is low ( $f \ll f_d$ ) so that polarisation losses are negligible. Above the transition point we can see the frequency dependency and the result will also be dependent on the water salinity.

Figure 4. also shows that the measured capacitance is dependent on the water fraction  $\beta$  even at water-continuous phase. This dependency might be utilized to measure the water content in the mixture even at water-continuous phase but more experiments and analysis have to be done before anything can be said about the reliability.

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