Measurement and Prediction of Speed of Sound in Petroleum Fluids

S. Ghafoori, A. Al-Harbi, B. Al-Ajmi, A. Al-Shaalan, A. Al-Ajmi, M. Ali Juma

Abstract**—**Seismic methods play an important role in the exploration for hydrocarbon reservoirs. However, the success of the method depends strongly on the reliability of the measured or predicted information regarding the velocity of sound in the media. Speed of sound has been used to study the thermodynamic properties of fluids. In this study, experimental data are reported and analyzed on the speed of sound in toluene and octane binary mixture. Three-factor three-level Box-Benhkam design is used to determine the significance of each factor, the synergetic effects of the factors, and the most significant factors on speed of sound. The developed mathematical model and statistical analysis provided a critical analysis of the simultaneous interactive effects of the independent variables indicating that the developed quadratic models were highly accurate and predictive.

Keywords**—**Experimental design, octane, speed of sound, toluene.

I. INTRODUCTION

KNOWLEDGE of thermodynamic and physical properties of hydrocarbon mixtures such as crude oils in petroleum production and processing, is of great importance [1]. As a reservoir is depleted, pore fluid suffers from changes in pressure, temperature and composition. Detection of fluids movements due to production gives valuable information about the depletion of a field, and can identify areas of bypassed oil and gas. Enhanced oil recovery operation often affects the reservoir properties. Changes in fluid saturation and reservoir pressure can be determined by applying 4D inversion. 4D seismic exploration method involves the acquisition, processing, and interpretation of many seismic surveys over a production field with the objective of understanding the change in the reservoir over a period of time, specifically its behavior during production [2]. The objectives of a 4D survey are concentrated on the production and development stages of a field and are focused on changes in the reservoir, usually the movement within the structures and fluid content. By tracking both of the previous points, it is possible to observe the flow of hydrocarbons within the reservoirs, gain better understanding of the reservoir behavior, and optimize its improvement. The seismic behavior is from fluid effects on the formation's seismic velocity rather than its density. The composition of reservoir fluids varies over the lifetime of the reservoir, thus affecting the acoustic properties of the rock frame also the seismic attributes. Gas invasion into liquid-filled rock and increase in temperature of hydrocarbon-filled rock both cause decrease in seismic velocity. The increase in temperature affects hydrocarbons by making them less viscous, reducing overall rigidity, therefore reducing seismic velocity. The data of speed of sound can be utilized to predict the thermodynamic properties of fluid mixtures. Several researchers have reported data on the speed of sound in some crude oils and have shown how bubble points may be determined from such data [3]-[7]. A number of researchers have measured and reported speed of sound in hydrocarbon systems at high pressures [8], [9]. These properties can be obtained by either modeling or experimental measurements. Based on the fact that the available experimental data on the speed of sound in different types of fluids such as toluene, binary mixture of toluene and octane with different concentration are limited, it is essential to generate reliable experimental data of fluids in a wide range of temperature and pressure conditions. In this report, chapter 2 delivers an overview of both experimental measurement and modeling of the speed of sound for hydrocarbons such as pure, mixtures on different concentration extracted from literature.

Experimental data of the speed of sound for pure and binary hydrocarbon mixtures in different range of temperatures, pressures, and concentrations have been presented. These results are important for the fundamental theory of liquid mixtures and also for the purpose of validating thermodynamic models [2].

The main aim of this study is to measure the speed of sound in liquids at low to high pressures and temperatures intervals for binary hydrocarbon (toluene and n-octane) at three different concentrations and to investigate the effect of each factor as well as the synergetic effects among the variables.

II. METHODOLOGY

A. Experiment Measurement

Our experimental method was from the recorded design of Peleties et al. [10]; however, the apparatus operates at a lower frequency which is 4 MHz for permitting measurements on liquids with viscosities up to approximately 21755.66 psig.

Measurement can be made on mixtures liquids that have dissolved gases or dense compress gases. The working ranges of the apparatus are pressures from 14.5 to 14503.77 psig and temperatures from 9.85 to 199.85 ºC, which was installed and commissioned by the manufacturer (Imperial College, UK). The pressure vessel was put off within a temperature-controlled oil moving cannel (Fluke model 6040) where the

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temperature was fixed with a constancy and homogeneity better than -203.14 ºC. The temperature was obtained by the means of a second-standardized platinum resistance thermometer (Fluke model 5615) which was placed into the thermos well in the wall side of vessel of pressure. This thermometer was standardized by the manufacturer with an extended uncertainty of greater than -273.13 ºC in the working array of the instrument.

A manual syringe pump with a displacement of 30 cm^3 was used to raise and alter the pressure. The equipment was label exploitation speed of sound for n-octane at air pressure and varied temperatures.

The standardization procedure was through determination of ΔL=L1–L2, wherever L1 and L2 are two totally different path lengths between electrical device and also the two reflectors. The worth of ΔL as determined from measured knowledge was 2.00370 cm compared to the particular worth of 2 cm. However, in every new spherical of experiments, the instrument was recalibrated exploitation n-octane at 25 °C and pressure 14.5 psig and the speed of sound under these conditions was taken as 1172.0 m/s one as stated by Peleties et al. [10]. Uncertainty for the measurements was regarding one $m.s^{-1}$. Measurements of speed of sound were managed at varied temperatures and pressures for toluene and octane, for binary mixtures of octane and toluene, and for many ternary mixtures of octane and toluene that was equipped by Merck with purity of Z 99%. The densities of pure liquids were measured by suggests that of a vibrating-tube measuring instrument (Anton Par model DMA 4500) antecedently label in air and pure degassed water. The measured densities at temperature 20 °C and pressure 14.5 psig were taken from Literature knowledge severally. Mixtures were ready by mix precisely-measured volumes of pure parts along at the close temperature of 24.85 °C. The number of every element was then computed from the measured densities resulting in mole fractions with associate in nursing overall enlarged uncertainty of 0.002. Measured acoustic velocities for pure hydrocarbons and mixtures are given in Tables I–IV. Table I offers the measured speeds of sound in toluene at temperatures from 30 °C, 60 °C, and 90 °C and at pressures 500 psig, 3000 psig, and 5500 psig. Literature knowledge is given in this table that takes issue with mentioned knowledge by 0.001–0.003. Experimental measurements of speed of sound for n-octane are given in Table I while for toluene, results are given in Table II. In both these cases, measurements were limited to the ambient pressure of 0.1 MPa. In addition to pure hydrocarbons, measurements were carried out for two binary mixtures of n-octane and toluene. The results are given in Table I. These data are used to estimate the proposed predictive model.

The device must be open to atmospheric pressure to empty the previous sample, with the help of nitrogen gas to flood the fluids in the system. This method is not enough to extract all fluids in the system, thus vacuum pressure instrument is used to vacuum the remaining fluid drops. It takes approximately one hour to make sure that the system pipelines are clean. System valves must be open in sequence to make sure that all system pipelines are covered in the cleaning process. Process of filling the system starts with closing the outlet valve and push the manual pump to its maximum capacity, thus it will be used to suck the experiment fluid from the prepared sample. Sample is compressed by the manual pump to check if the cell is full. If the pressure gauge indicates increase of pressure, this means the cell is saturated. Otherwise, manual pump will be pushed to its limit again to sock more from the sample.

III. RESULTS AND DISCUSSION

A. Speed of Sound in Toluene

The speed of sound of toluene (C6H5-CH3) through different pressures and temperatures is measured to find the speed of sound *u* (m/s) for the compound. The pressures that was used in this experiment was 500, 1000, 1500, 2000, 2500, 3000, 3500, 4000, 4500, and 5000 psig with three temperatures (50, 70, and 90 °C). The idea of performing this experiment is to compare the results of experiment against data from (NIST Chemistry Webbook) and check if there any error between data. The differences are generally in range between 0.015% and 0.38% from the measurement in (NIST Chemistry Webbook). The ultrasonic cell filled with fully toluene sample and the temperature rise to 50 °C and in pressures between 500 and 5000 psig are shown in Table III.

TABLE I
EXPERIMENTAL SPEED OF SOUND IN 70% TOLUENE AND 30% N-OCTANE

Temperature °C	30	60	90
Pressures (psig)	Measured speed of sound u (m/s)		
500	1259.06	1136.11	1019.64
3000	1346.7	1238.11	1134.49
5500	1423.48	1322	1227.33

B. Speed of Sound of Binary Mixture

After the efficiency of the device has been checked, the experiment run for binary mixture by 140 mL of toluene and 60 mL of n-Octane filled in device. The test has been done with three temperatures (30, 60, and 90 °C) and three pressures (500, 3000, and 5500 psig). The results are shown in Table III.

TABLE II
EXPERIMENTAL MEASUREMENT OF SPEED OF SOUND VS. NATIONAL INSTITUTE OF STANDARDS AND TECHNOLOGY (NIST) FOR TOLUENE

Temperature (°C)	50	70	90	50	70	90
Pressures (psig)	Experimental measurement			NIST results		
500	1218.12	1137.20	1058.8	1217.8	1139	1062.5
1000	1237.09	1158.19	-	1236.9	1159.8	-
1500	1254.94	1177.39	-	1255.4	1179.9	-
2000	1272.52	1197.18	1125	1273.3	1199.3	1128.1
2500	1289.49	1215.06	-	1290.7	1218	-
3000	1305.91	1233.73	-	1307.7	1236.1	-
3500	1321.42	1250.39	1183	1324.2	1253.8	1186.6
4000	1336.87	1267.52	-	1340.3	1270.9	-
4500	1352.05	1283.59	-	1356	1287.6	-
5000	1366.77	1299.37	1234.9	1371.3	1303.8	1239.7

The second experiment has been run with 100 mL of toluene and 100 mL of n-octane. The test was run as previous temperatures and pressures as well. As in previous experiment, the average of speed of sound has been taken and results are shown in Table IV.

TABLE III
EXPERIMENTAL SPEED OF SOUND IN 50% TOLUENE AND 50% N-OCTANE

Temperatures °C	30	60	90
Pressures (psig)	Measured speed of sound u (m/s)		
500	1230.056	1109.433	1001.286
3000	1322.196	1217.116	1115.296
5500	1402.54	1305.743	1214.086

This experiment was run with 60 mL toluene and 140 mL of n-octane filled in ultrasonic cell and the repeating of test for each pressure in specifying temperature was applied.

IV. RSM MODEL DEVELOPMENT

The operating conditions to measure the speed of sound were obtained using a three-factor three level Box-Behnken experimental design. The Box-Behnken design, a modified central composite design, is known as an independent, rotatable quadratic design having no fractional factorial points.

TABLE IV
EXPERIMENTAL SPEED OF SOUND IN 30% TOLUENE AND 70% N-OCTANE

Temperatures °C	30	60	90
Pressures (psig)	Measured speed of sound u (m/s)		
500	1207.55	1087.81	975.763
3000	1305.236	1198.623	1103.49
5500	1389.623	1292.676	1201.396

In this type, of design, the variable combinations are at the center and the midpoints of the edges of the variable space. Also, compared to other types of experimental design such as full factorial design, the Box-Behnken experimental design needs fewer runs. In this study, the effect of three independent variables on the speed of sound was investigated. The independent variables were the volume concentration of toluene (A), the temperature of binary mixture (B), and the pressure applied to the samples (C) that were coded as -1, 0, and 1 as presented in Table V.

TABLE V
INDEPENDENT VARIABLES AND THEIR CODED LEVELS BASED ON EXPERIMENTAL SPEED OF SOUND

Independent Variables	Symbols	Coded Levels		
		-1	0	1
Toluene Concentration (v%)	A	70	50	30
Temperature (°C)	B	30	50	70
Pressure (psig)	C	500	3000	5500

Table V illustrates the results of the three-factor three-level Box-Behnken experimental design for the speed of sound through different binary mixture.

The observed and predicted results for the speed of sound are also presented in Table VI. By applying multiple regression analysis on the design matrix and the responses given in Table VI, the following quadratic equation in terms of coded factors was determined to predict the speed of sound u (m/s):

$$\text{Speed of sound } u = 1217.12 - 18.76A - 103.9B + 97.01C + 2.62AB + 4.75AC + 10.08BC + 1.51A2 + 3.85B^2 - 8.98C^2$$

The equation in terms of coded factors can be used to make predictions about the response for given levels of each factor. By default, the high levels of the factors are coded as +1 and the low levels of the factors are coded as -1. The coded equation is useful for identifying the relative impact of the factors by comparing the factor coefficients. The equation in terms of actual factors can be used to make predictions about the response for given levels of each factor. Here, the levels should be specified in the original units for each factor.

TABLE VI
THREE-FACTOR THREE-LEVEL BOX-BEHNKEN DESIGN FOR RSM

Run	Independent Coded Variables				Speed of Sound (m/s)
	A	B	C	Observed	Predicted
1	-1	-1	0	1346.7	1356.74
2	-1	0	1	1322	1320.76
3	-1	0	-1	1136.11	1330.17
4	0	1	1	1214.09	1215.18
5	0	0	0	1217.12	1217.12
6	0	0	0	1217.12	1217.12
7	0	-1	1	1402.54	1402.82
8	1	0	1	1292.68	1292.65
9	0	1	-1	1001.29	1001
10	1	0	-1	1087.81	1089.13
11	0	0	0	1217.12	1217.12
12	0	-1	-1	1230.06	1228.96
13	-1	1	0	1134.49	1134.72
14	1	1	0	1103.49	1102.44
15	1	-1	0	1305.24	1305

This equation should not be used to determine the relative impact of each factor because the coefficients are scaled to accommodate the units of each factor, and the intercept is not at the centers of the design space.

Fig. 1 illustrates the linear graph between the observed and predicted data to show how closely the results match to each other.

V. STATISTICAL ANALYSIS

The Model F-value of 10923.18 implies that the model is significant. There is only a 0.01% chance that this large F-value could occur due to noise. Values of "Prob > F" less than 0.0500 indicate that model terms are significant. In this case, A, B, C, AB, AC, BC, B^2, C^2 are significant model terms. Values greater than 0.1000 indicate that the model terms are not significant. If there are many insignificant model terms (not counting those required to support hierarchy), model reduction may improve your model. Therefore, it can be concluded that the predictions of experimental data by the derived quadratic models for the speed of sound u are quite satisfactory. Also, a high correlation between observed and predicted data shown in Fig. 1 indicates their low discrepancies.

VI. THE EFFECT OF THE MODEL PARAMETERS AND THEIR INTERACTION

The significance of each model parameter was determined by means of Fischer's F-value and p-value. The F-value is the test for comparing the curvature variance with residual variance and probability > F (p-value) is the probability of seeing the observed F value if the null hypothesis is true. Small probability values call for rejection of the null hypothesis and the curvature is not significant.

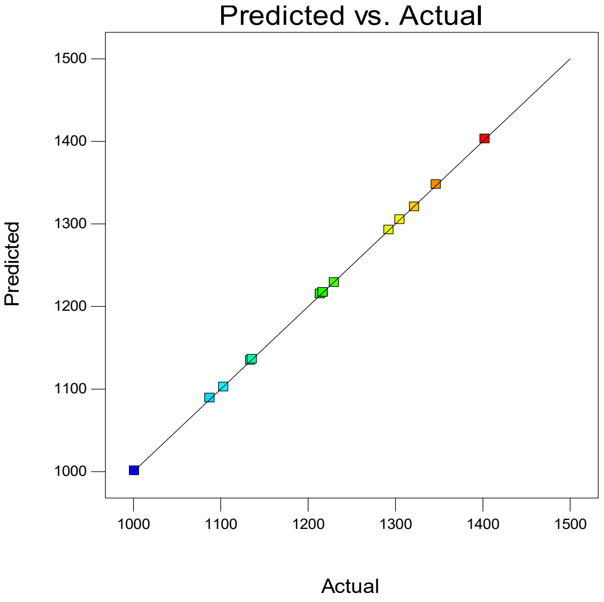

Fig. 1 Comparison of predicted speed of sound versus actual values

Therefore, the larger the value of F and the smaller the value of p, the more significant is the corresponding coefficient. As shown in Table VII, the p-value less than 0.0001 indicates that the model parameters are significant. Therefore, all three independent variables have significant effect on the speed of sound based on their p value. However, the concentration of toluene showed less significant effect compared to the temperature and pressure. Also, as it is evident in Table VI, the quadratic effect of the temperature of binary mixture has significant effect on the response function.

The interaction effects between the independent variables are illustrated in three-dimensional (3D) response surface and two-dimensional (2D) contour plots (Fig. 2). These figures are the graphical representations of the regression analysis. In such plots, the response function of two factors is presented while all the others are at the fixed levels. As shown in Fig. 2 (a), the higher pressure applied to the binary mixture results in the higher speed. However, the effect of the temperature of sample shows the reverse trend. The lower temperature results in the higher speed of sound.

VII. CONCLUSION

The application of a three-factor, three-level Box-Behnken experimental design combined with RSM and quadratic programming was investigated for the speed of sound in hydrocarbon sample effected by pressure, temperature and the concentration of toluene in binary mixture. The developed mathematical model provided a critical analysis of the simultaneous interactive effects of the independent variables. The adequacy of the proposed model was tested using various descriptive statistics.

TABLE VII
ANOVA FOR PREDICTION OF SPEED OF SOUND BY THE QUADRATIC MODEL

Source	Sum of Squares	Df	Mean Square	F Value	p-value Prob > F	
Model	1.654E+005	9	18373.77	10923.18	< 0.0001	significant
A-Tolune Conc.	2815.50	1	2815.50	1673.81	< 0.0001	significant
B-Tepmperature	86357.52	1	86357.52	51339.42	< 0.0001	Significant
C-Pressure	75279.76	1	75279.76	44753.71	< 0.0001	Significant
AB	27.35	1	27.35	16.26	0.0100	Significant
AC	90.06	1	90.06	53.54	0.0007	Significant
BC	406.43	1	406.43	241.62	< 0.0001	Significant
A2	8.39	1	8.39	4.99	0.0758	
B2	54.80	1	54.80	32.58	0.0023	Significant
C2	297.58	1	297.58	176.91	< 0.0001	Significant
Residual	8.41	5	1.68			
Lack of Fit	8.41	3	2.80			
Pure Error	0.000	2	0.000			
Cor Total	1.654E+005	14				

VIII. RESPONSE SURFACE AND 2D CONTOUR PLOTS

The statistical analysis using ANOVA indicated that the developed quadratic models were highly accurate and predictive. Good agreement between quadratic model predictions and observed values also confirmed the accuracy of the developed model. The initial concentration of toluene, the applied pressure on binary mixture, and the sample's temperature had significant effect on the velocity. The optimal operating conditions to achieve the maximum velocity of sound u (m/s) in the selected range was determined to be 5500 psig, 70% volume of toluene, and 30 °C, respectively. Finally, the model prediction for the maximum u was validated by an additional experimental run at the obtained optimum operating conditions.

The validation results clearly confirmed that a three-factor, three-level Box-Behnken experimental design combined with

RSM and quadratic programming is an effective tool for mathematical modeling of the speed of sound.

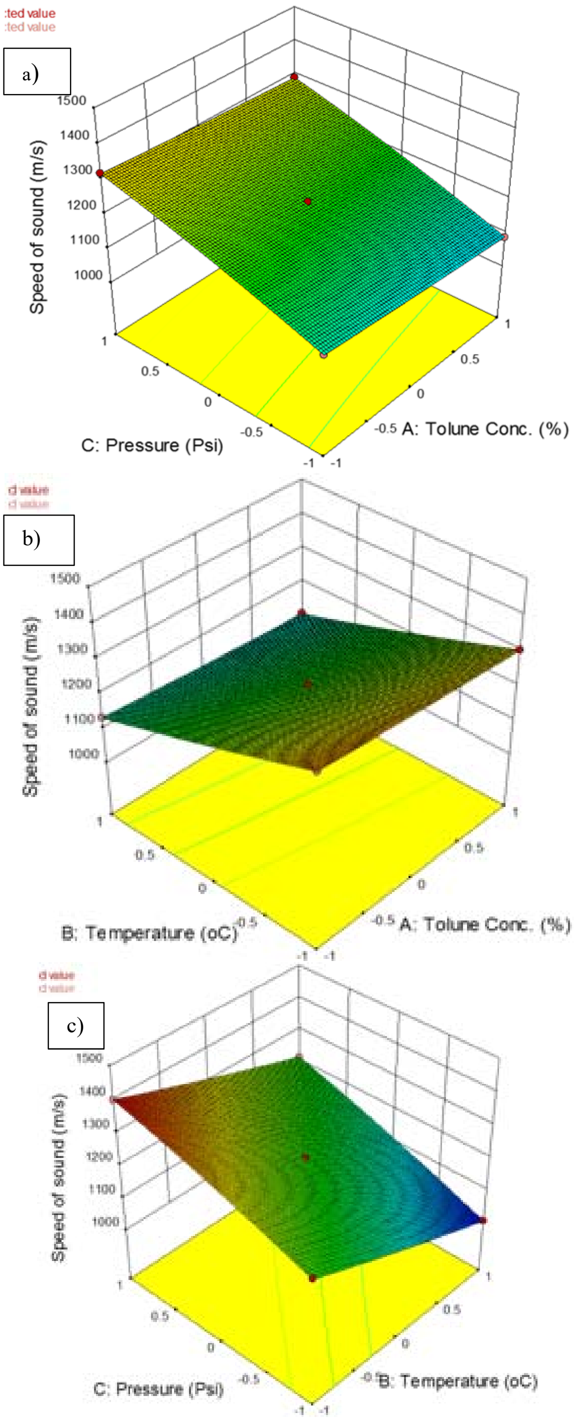

Fig. 2 3D response surface and 2D contour plots

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