MODELS OF THE VOLTAGE INDUCED BY CAVITATION IN HYDROCARBONS

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When an ultrasonic wave passes through a liquid medium, containing microscopic gas inclusion, it can produce cavitation. At the boundary of a cavitating zone, a voltage appears as a effect of the collapse/rebound cycle of the cavitation bubbles. In this paper we establish a mathematical model of the voltage induced at the boundary of an acoustic cavitation zone, when the liquid was the crude oil. It is compared to that obtained for diesel, in order to prove that the ARIMA process can appropriately describe the fluctuations of generated voltage in different liquids. We also discuss the hypothesis of the relation between the ARIMA parameters and the liquid nature.

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1. Introduction

Cavitation is the process of non-equilibrium rise of cavities (bubbles, vortices *etc.*) filled with vapour or vapour–gas mixture due to local pressure drop below certain critical value (usually close to that of the saturated vapour) and followed by sudden collapse due to subsequent pressure rise [1].

An ultrasonic field that goes over a liquid can produce or move cavitation bubbles. Their collapse/rebound cycle results in physical and chemical effects relieved in many scientific papers [2–8]. The apparition of a variable voltage induced between different points at the boundaries of an acoustic cavitation zone in water have been studied [7, 9–12]. In [11] we gave some possible explanation of its generation mechanism.

In this paper we want to prove that the Box–Jenkins methods can be successfully used to describe the electrical voltage induced in cavitating sour oil and diesel. In fact, we shall prove that the autoregressive integrated moving average process can describe with sufficient accuracy the voltage

occurring between two points of the cavitation zone boundaries in the two liquids and we shall compare to those obtained in distilled water.

It is only an intermediate stage of a study that has the aim to determine and to explain the relation between the cavitation phenomenon and its links with the physical and chemical properties of the liquids.

We formulate the hypothesis that the autoregressive parameter, p, of an ARIMA(p, d, q) model that describes the voltage induced by cavitation in a group of liquids with the same basic chemical compound is a characteristic of this group. Till now, this hypothesis has been verified and we discuss the known cases. But, a lot of aspects related to the features of the liquids with a common p remains to be studied and clarified.

2. Experimental set-up

In order to determine the voltage appeared at the boundary of an acoustic cavitation zone in sour petroleum, the experimental set-up drawn in Fig. 1 was used and we worked in the same conditions as in the case of the experiment for distilled water [11].

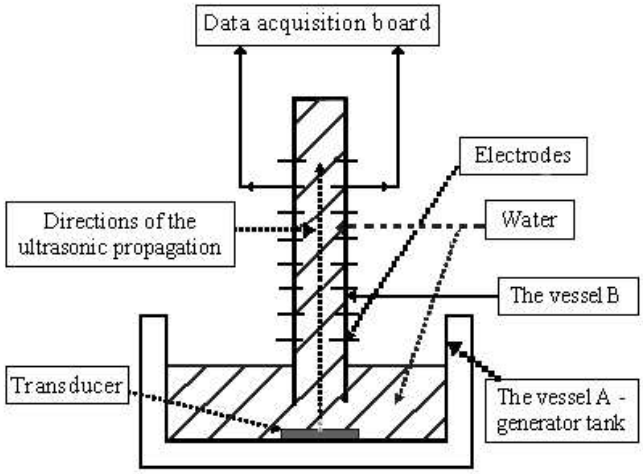


Fig. 1. The experimental set-up.

It consists of:

— The vessel A (the ultrasonic generator tank) that contains a transducer, connected to a high frequency generator that excites the transducer to produce the ultrasound. The ultrasonic generator operated at the frequency of 20 kHz and at the power of 120 W.

— The vessel B which contains pairs of electrodes, that measure the potential difference produced between different points at the cavitation zone boundaries.

The vessels A and B contained:

— crude oil of Siberian Light type, with the density 833 kg/m^3, with a low sulphur content (0.55%), a low water content (0.1% vol.) and salts (0,0005%), light acid (1,5 mg KOH/100 g sour oil);

— diesel, with the density 863 kg/m^3, sulphur: 0.011%, diesel index: 55.

The data acquisition board is connected to the electrodes and collects the signal, which is not amplified.

3. Results

The values of the voltage induced by the cavitation are represented in Figs. 2 and 3.

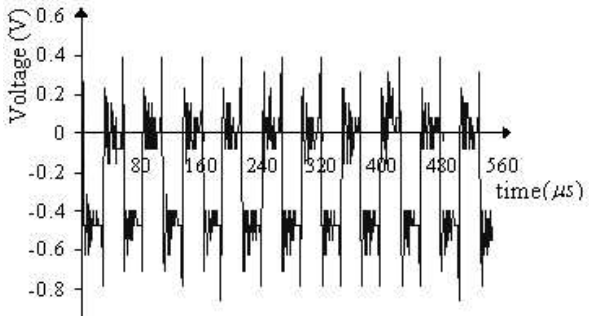


Fig. 2. The values of the voltage induced in crude petroleum.

The study was made in a period of 5326 μs. For clarity, in the Figs 2 and 3 the voltage graph is plotted only for 560 μs. In both cases it can be seen that there is a decreasing trend of the voltage and the pulses are not interchangeably shifted to positive and negative side.

We could give the following explanation of the apparition of a potential difference between two points when an ultrasound propagates through a liquid [11, 16].

At the contact area between the cavitation bubble and the liquid, an electrical double layer appears, as the effect of the electric loading accumulation at the internal and the external surfaces of the bubble. At the moment, some bubbles collapse, this process is followed by the reappearance of the bubbles, the electrical charging and the increase of their size, resulting in a new collapse.

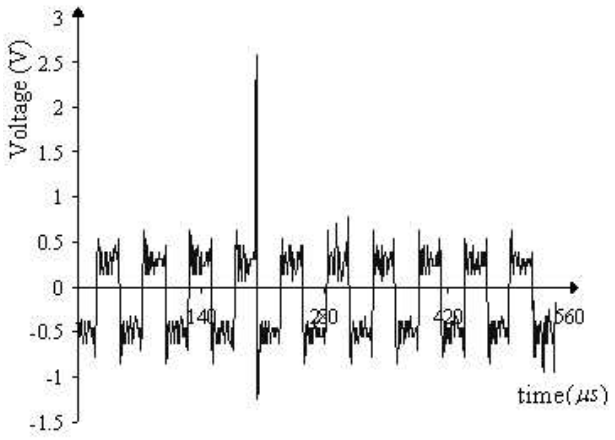


Fig. 3. The values of the voltage induced in diesel.

Since the bubbles do not collapse all at the same moment, an alternating current appears between the measurement electrodes put in the studied liquids. The frequency and the amplitude of the induced voltage are not constant because the pressure and the value of the surface tension are not constant.

Our experiments have proved the dependence of the voltage induced by the ultrasonic field on the ultrasonic frequency, the distance between the electrodes, the power of the ultrasonic generator and the liquid nature.

4. Mathematical modelling

It has been attempted to describe the process of voltage variations between two points of an acoustic cavitation zone using simple models, as polynomial, hyperbolic, sinusoidal, Gaussian *etc.* The best model determined, using CURVEExpert program was of exponential type. It was not satisfactory because the high standard deviation and small correlation coefficient associate to it.

The Box–Jenkins methods were used to describe the voltage induced in a cavitation zone in distilled water [8–11], so it was considered that an autoregressive integrated moving average model could be satisfactory also in the new cases.

The following definitions are known [11, 14, 15].

Definition 1. *A discrete time process* $X_t, t \in \boldsymbol{Z}$ *is called stationary if*:

$$\forall t \in \boldsymbol{Z}, M(X_t^2) < \infty\,,$$

$$\exists \mu \in \boldsymbol{R}, \forall t \in \boldsymbol{Z}, M(X_t) = \mu\,,$$

$$\exists \gamma : \boldsymbol{R}^+ \to \boldsymbol{R}, \forall t \in \boldsymbol{Z}, \forall h \in \boldsymbol{Z},\ \mathrm{Corr}(X_t,\, X_{t+h}) = \gamma(h)\,,$$

where $M(X)$ is the expected value of the random variable X, μ is a constant, $Corr(X, Y)$ is the correlation of the random variables X and Y and γ is a real function.

Definition 2. *A stationary process $\xi_t, t \in \boldsymbol{Z}$ is called a white noise if $\gamma(h) = 0$, for $h \neq 0$, $M(\xi_t) = 0$ and $D^2(\xi_t) = \sigma^2 = \gamma(0)$, $\forall t \in \boldsymbol{Z}$.*

Definition 3. *Let p, d, q be natural numbers, $(\varphi_1, \ldots, \varphi_p)$ and $(\theta_1, \ldots, \theta_p)$ finite sequences of real coefficients, I — identity function and $X_t, t \in \boldsymbol{Z}_+$ a time process. Let us consider*

$$\begin{aligned} BX_t &= X_{t-1}, \\ \boldsymbol{\Phi}(B) &= I - \varphi_1 B - \cdots - \varphi_p B^p,\ \varphi_p \neq 0\,, \\ \boldsymbol{\Theta}(B) &= I - \theta_1 B - \theta_2 B^2 - \cdots - \theta_q B^q,\ \theta_p \neq 0\,, \\ \Delta^d X_t &= (1 - B)^d X_t\,. \end{aligned}$$

The time process is called an autoregressive integrated moving average process of p, d, q orders and is denoted by ARIMA(p, d, q) *if*:

$$\boldsymbol{\Phi}(B)\Delta^d X_t = \boldsymbol{\Theta}(B)\xi_t\,,$$

where ξ_t, $t \in \boldsymbol{Z}_+$ is a white noise. ξ_t, $t \in \boldsymbol{Z}_+$ is called the residual in the ARIMA *process. An autoregressive moving average process of p and q orders, denoted by* ARMA(p, q) *is an* ARIMA *process of p, 0, q orders. p is called the autoregressive order, q — the differencing degree and q — the order of moving average.*

The software SPSS 10 for Windows was used to process the signal captured by the acquisition card. It was created in the sixties to assure the management and the analysis of the statistical data in the social sciences and psychology. Afterwards, its utilisation was extended to other scientific branches, since 1995, with the version 7, being a Windows product.

The TRENDS module assures the analysis and the graphical representation of the time processes. The trend of a time series can be estimated by: smoothing procedures, regression methods, Box–Jenkins methods, decompositions of series that present seasonality or the analysis of random compounds.

The software also permits to chose the algorithm for the coefficients estimation for a model proposed by user and returns the values of t — ratio necessary in the validation of the coefficients.

The autocorrelation function (ACF) and the partial autocorrelation function (PACF) are implemented in SPSS. (For the definitions of ACF and PACF, see [11]).

If $X_t, t \in \boldsymbol{Z}$ is a time process, ACF measures the dependence between X_t and PACF expresses the linear dependence between X_t and $X_{t-h}, t \in \boldsymbol{Z}$, striking off the intermediate terms, $X_{t-1}, \ldots, X_{t-k+1}$.

Since we work with samples of thousands data, it is helpful to use a software to calculate these functions that play a major role in the model selection.

First, the obtained data were subjected to the Fourier analysis and the obtained graphs were plotted in Fig. 4. The graphs of the Fourier transforms are alike, but the amplitudes for the diesel are almost 2.5 times bigger at frequencies less than 200 kHz. After 400 kHz, we remark the stationary trend of the process in diesel, marked by the absence or small amplitude of harmonics and subharmonics. Therefore we expect that the two models for the liquids under consideration do not differ too much and the second be a stationary one. Another argument could be that the two liquids have the same chemical basic compounds.

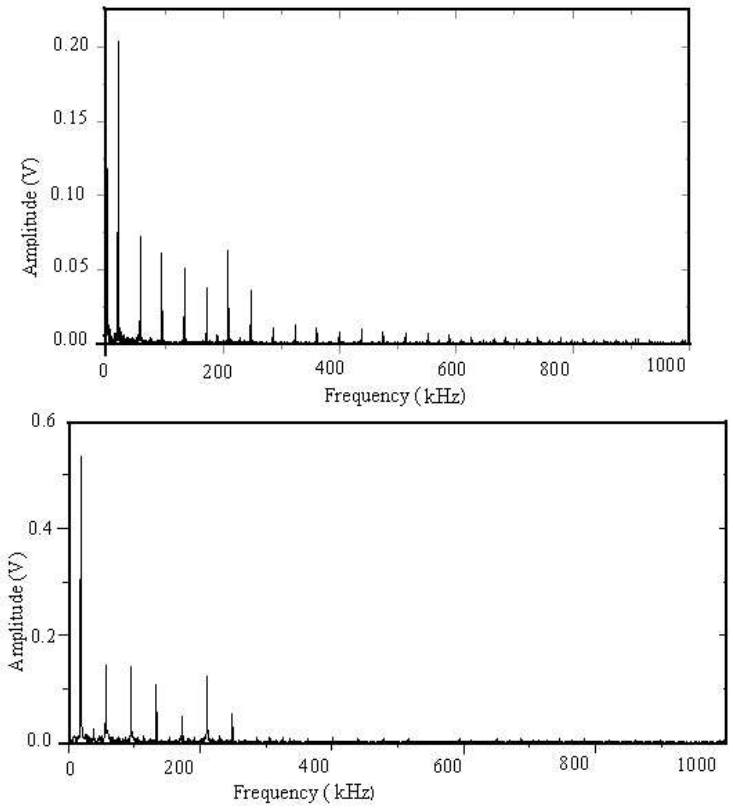


Fig. 4. The Fourier transform of the induced voltage.

In what follows we shall discuss the results obtained for the crude petroleum. In Fig. 2 it can be seen that there are some aberrant values, which must be removed. After this process, the remained values will be studied. The autocorrelation function of the voltage, at the lags between 1 and 16, was calculated and the confidence interval, at the confidence level 95%, was determined. The values of ACF are outside the confidence interval. The form of ACF was of an exponential decreasing and that of PACF was of damped sine wave oscillation. These remarks enable us to think that the process is not stationary and it can be of ARIMA type.

Twenty models were analysed. To chose between them, the Schwarz (SBC) and Akaike (AIC) criteria were used. The preferred value was that of Schwarz' criterion. The selected model — ARIMA(3,1,4), without a constant term — had the least SBC value.

Fixing the significance level $\alpha = 5\%$, the next step was to test the hypothesis: H_0: the coefficients of the model are zero.

The values of the model coefficients and of the Student test (t-ratio) are given in Table I. The values of the t-ratios (Table I, Variables in model — the column 4) are bigger than the values of the quantile of the Student function with 15 degrees of freedom, at the significance level α. The probability of hypothesis H_0 validity is practically zero (Table I, Variables in model — the last column). Thus, H_0 is rejected, *i.e.* the assumption that the coefficients of the model are zero is not justified.

To prove that the model is a good one, point of view of statistics, the ACF (Fig. 5, Table II) and PACF (Fig. 6) of residuals were calculated.

The following data are provided by Table II:

— In the column 1 — the lags, between 1 and 16;

— In the column 2 — the ACF values for errors;

— In the column 3 — the standard errors;

— In the column 4 — the values of Box–Ljung statistics, which are in the interval [0.023, 19.946];

— In the last column — the probability to accept the hypothesis that the residuals form a white noise, which is in the interval [0.720; 0.984].

The values of the probabilities to accept the hypotheses that the residuals form a white noise are big enough and the values of the Box–Ljung statistics are less than $\chi^2(15)$.

Also, the values of ACF and PACF of the residuals are inside the confidence intervals and the standard errors are very small, so the model is well selected. For the better understanding of the voltage fluctuation simulation,

TABLE I

The coefficients of the ARIMA(3,1,4) model.

Variable: U
Non-seasonal differencing: 1
No seasonal component in model.
Parameters: AR1, AR2, AR3, MA1, MA2, MA3, MA4
95% confidence intervals will be generated.
Split group number: 1 Series length: 5326
Melard's algorithm will be used for estimation.
FINAL PARAMETERS:
Number of residuals 5325
AIC −10701.729
SBC −10655.668

Analysis of variance

	DF	Adj. sum of squares	Residual variance
Residual	5318	41.676893	0.00783582

Variables in the model

	B	SEB	t-ratio	Approx. prob.
AR1	0.8400813	0.20794523	8.8488749	0.0000000
AR2	−1.4524325	0.030479608	−4.7652597	0.00000194
AR3	0.3521657	0.18436609	1.9101437	0.05616845
MA1	1.6665377	0.20851582	7.9923799	0.0000000
MA2	−1.2339601	0.27363218	−4.5095578	0.00000664
MA3	0.2888236	0.16778443	2.7213971	0.08523695
MA4	0.0397538	0.02174703	2.8280102	0.06760402

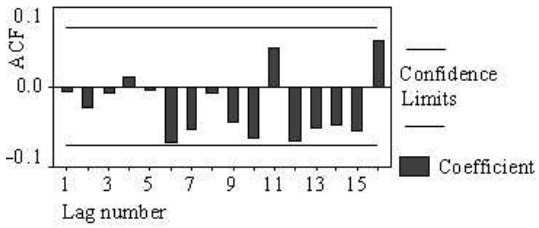


Fig. 5. The ACF of residuals in the ARIMA(3,1,4) model.

the softwares MATLAB or P&R ([17], with the installation kit) can be used to generate an white noise. Providing the mean, the variance, the sampling rates (Hz) and the time points, as input data in P&R, the output will be

TABLE II

The ACF values of residuals in the ARIMA(3,1,4) model.

Lag	Auto-Corr.	Stand. Error	−0.5	−0.25	0	0.25	0.5	Box–Ljung	Prob.
1	−0.002	0.014			.*.			0.023	0.881
2	−0.010	0.014			.*.			0.512	0.774
3	−0.002	0.014			.*.			0.545	0.909
4	+0.005	0.014			.*.			0.664	0.956
5	−0.001	0.014			.*.			0.673	0.984
6	−0.034	0.014			*I.			3.832	0.937
7	−0.020	0.014			.*.			4.932	0.858
8	−0.003	0.014			.*.			5.967	0.945
9	−0.016	0.014			.*.			6.337	0.824
10	−0.045	0.014			*I.			8.143	0.720
11	+0.018	0.014			.*.			10.858	0.819
12	−0.040	0.014			*I.			11.476	0.902
13	−0.019	0.014			.*.			12.476	0.901
14	−0.018	0.014			.*.			13.257	0.901
15	−0.021	0.014			.*.			15.575	0.911
16	+0.021	0.014			.*.			19.942	0.931

Plot Symbols: Autocorrelations* Two Standard Error Limits .
Total cases: 5326 Computable first lags: 5324

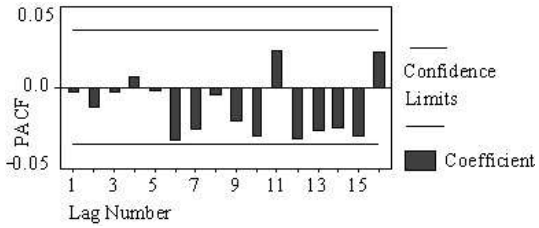


Fig. 6. The PACF of residuals in ARIMA(3,1,4) model.

the white noise values and its graph. In our case, the residual values being known, the mean and the variance of residuals are easily computed; they are respectively 0 and 0.008, so the white noise graph is drawn in Fig. 7.

To compare the collected data and those calculated for a single period, using the ARIMA(3,1,4) model, one can see the Fig. 8. In its upper part the values of the signal are plotted and in its lower part, those calculated. It is obvious that the two curves are alike.

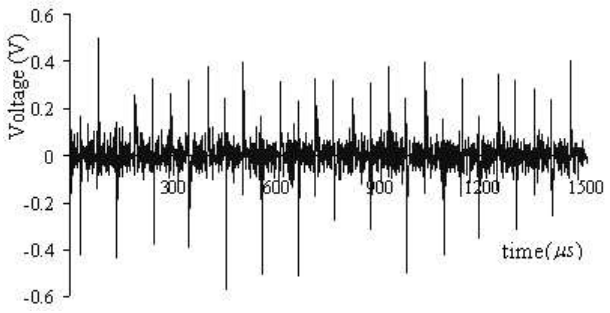


Fig. 7. The white noise.

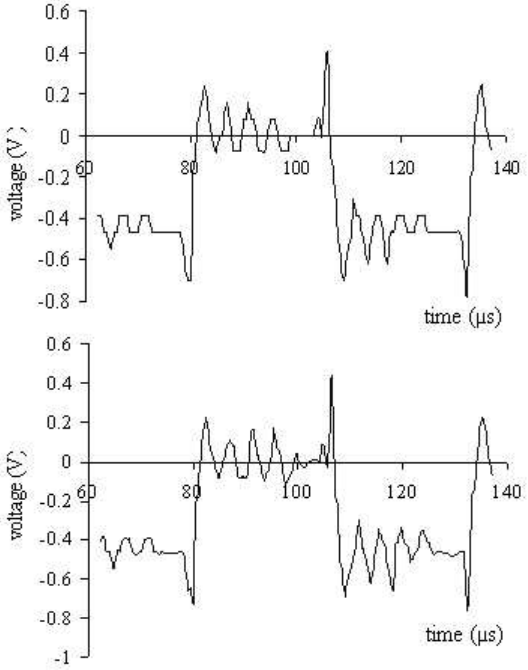


Fig. 8. The voltage experimentally determined and that calculated in crude petroleum.

The equation of the voltage induced in crude petroleum is:

$$(1 - 0.84B + 1.4524B^2 - 0.352B^3)(1 - B)U_t = \xi_t - 1.666\xi_{t-1} \\ +1.2339\xi_{t-2} - 0.2888\xi_{t-3} - 0.0397\xi_{t-4}\,, \tag{1}$$

where $t \in \boldsymbol{N}, t \geq 4$, U_t is the voltage at the moment t and ξ_t, $t \in \boldsymbol{N}$ is the residual.

To make the comparison, the same experiment was reproduced for diesel. The model for the voltage induced in this case was determined. It is of ARMA(3,6) type, without constant and it has the equation:

$$U_t = 2.3798U_{t-1} - 2.2495U_{t-2} + 0.8465U_{t-3} + \xi_t - 1.279\xi_{t-1} \\ +0.8222\xi_{t-2} + 0.03134\xi_{t-3} - 0.1189\xi_{t-4} + 0.0998\xi_{t-5} - 0.0416\xi_{t-6}\,, \quad (2)$$

where $t \in \boldsymbol{N}, t \geq 4$, U_t is the voltage at the moment t and ξ_t, $t \in \boldsymbol{N}$ is the residual.

Bearing in mind the fact that $BU_t = U_{t-1}$, $B^kU_t = U_{t-k}$, $k \in \boldsymbol{N}$ (Definition 3), the equation (2) is equivalent to:

$$(1 - 2.3798B + 2.2495B^2 - 0.8465B^3)U_t = (1 - 1.279B + 0.8222B^2)\xi_t \\ +(0.03134B^3 - 0.1189B^4 + 0.0998B^5 - 0.0416B^6)\xi_t\,. \quad (3)$$

An important remark is that a fixed number of iteration of the computational algorithm (100), at a precision of 0.001, the coefficients in the equations (1) and (3) remains constant, at each repetition of the procedure, for any period analyzed. Varying the iterations number or the precision (0.0001), the coefficients are insignificantly affected (only at the third decimal). This means that these formulas are well reproducible.

5. Conclusions

Our experiments proved the apparition of voltage in cavitating liquids. This alternative potential difference is distinct from the metal–liquid boundary one (which is continuous) and the components of the induced voltage frequency are the ultrasound frequency and some harmonics and subharmonics.

Analysing the formulas (1) and (3) and the relation (7) from [11] we make the following remarks:

1. Working in the same experimental conditions, when the generator power was 120 W, the model of the voltage induced by cavitation in diesel was an ARMA(3,6), *i.e.* it was a stationary process. It is remarkable that a stationary model — ARMA(2,1) — was also obtained in the case of distilled water [11]. It would seem that the models of the induced voltage are stationary in all the cases, but the analysis performed on the sour oil proved that this assertion is not true. A first conclusion is that the stationarity/nonstationarity of the processes discussed in this paper is not directly correlated with the generator power. The question that arises is which chemical compound or physical propriety of the liquid is responsible of this situation. At this stage of our research, it is difficult to give a well-founded answer.

2. Another remark to be proved by our following experiments is that the autoregressive parameter, p, of ARIMA(p, d, q) is a characteristic of the liquid/group of liquids. It was proved that for water (distilled, sea or drinking water), $p = 2$, *i.e.* a value of the voltage induced in a cavitating liquid is affected by the preceding 2 values (independently of one another). For oil products (sour oil, diesel, gasolines of different types), $p = 3$. Bearing in mind these remarks, the aim of our forthcoming studies is to classify the liquids function of p and to determine exactly which are their common properties, responsible for the presence of a liquid in a certain group.

3. The formulas (2) and (7) from [11] are analogous, describing stationary autoregressive moving average processes. They differ from (1), that describes a nonstationary process. If $V_t, t \geq 2$ is the series with the general term $V_t = U_{t+1} - U_t, t \geq 1$ and U_t is of ARIMA(3,1,6) type, then V_t is of ARMA(3,6) type. In other words, the series obtained by the first degree differentiation is stationary. An analogous situation was found for the distilled water, at 120 W, when the process was of ARIMA(2,1,0) type.

Since the voltage variation between two points of an acoustic cavitation zone is a phenomenon produced in time, the Box–Jenkins method was appropriate to describe it, when the studied liquids were sour oil, diesel or water [9–11]. But it is only a stage in the determination of relation between the cavitation intensity and the liquids properties.

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