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MICROWAVE MEASUREMENTS OF DIELECTRIC CONSTANTS BY EXPONENTIAL AND LOGARITHMIC MIXTURE EQUATIONS

J. Sheen and Z.-W. Hong

Department of Electronic Engineering
National Formosa University
Hu-Wei, Yun-Lin 632, Taiwan

C.-W. Su and H.-C. Chen

Department of Computer and Communication Engineering
Chienkuo Technology University
Chang-Hua 500, Taiwan

Abstract—This article reports on a study of the dielectric constants of ceramic dispersions in the polyethylene matrix at microwave frequency. The exponential and logarithmic mixture rules are studied in three ceramic powders of fillers with dielectric constants 10, 20, and 36, respectively. The experimental values of the dielectric constants of the mixtures are compared to those obtained by using different mixing laws. The mixing rules are also adopted to calculate the dielectric constants of pure ceramics from the measured dielectric constants of composites with various concentrations. The theories on errors of calculations are studied. The most adequate mixture equation for measuring the dielectric constants of pure ceramics is suggested.

1. INTRODUCTION

Generalized mixture equations for dielectric properties of heterogeneous mixtures can be derived by the use of vector calculus and Maxwell's equations. Many theories were developed, and various empirical formulas were proposed. Some good overviews and comparisons on the mixture laws have also been published [1–3]. These studies generally focused on the comparisons of experimental values and the theoretical curves of various mixing laws. However, the chief development

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Corresponding author: J. Sheen (jsheen@nfu.edu.tw).

of mixture laws is often to calculate the permittivity of a substance from measurements on powder. Theoretically, the dielectric constant of a pure ceramic can be calculated from that of the ceramic-polymer composite if the mixing equation has a good accuracy. One can estimate the dielectric constant of a pure ceramic by simply measuring a composite sample which saves the high temperature sintering procedure of making a pure ceramic sample for measurements. Thus, the accuracy of estimation is an important factor to judge the practicability of a mixture rule. Recently, the theoretical analysis on the accuracy of calculating the dielectric constant from the composite samples by different mixture laws was studied for three basic particle shapes of fillers in reference [4]. The analyses of theoretical errors on dielectric constant calculations of pure ceramics were proposed in this reference. It has also shown that a mixture law with a better curve matching with experimental results does not guarantee a higher accuracy of estimating the dielectric properties of pure ceramics. However, [4] only studied six basic mixture rules. Actually, a lot of theoretical and empirical mixture rules have already been published [1–26]. Some of the rules have been studied in recent years, but they were investigated without knowledge about how high their theoretical errors could be. One of the purposes of this research is to mend this phenomenon in the development of powder mixture theories.

In the reported mixture equations, exponential mixing equations may be the most popular studied rules and are investigated in this paper. In addition, the inverse of exponential law is the logarithmic rule, and it is also studied. The goal of this article is not to submit a new law, because the existing rules are more than enough, but to analyze and compare the existing rules and choose suitable mixture laws to measure the dielectric constant of a substance in powder. In addition, from the theoretical errors of various mixture rules, we can find their potential of becoming an accurate law in the future, i.e., by improving the present formulas to obtain a better agreement with measurements, for calculating the permittivity of dielectric material in powder.

We will simply adopt the experimental results presented in [4] and apply to the eight exponential and logarithmic mixture equations in this article. There, polyethylene powder was chosen as the matrix material. Three kinds of commercial ceramic powders with dielectric constants 10, 20 and 36 at 10 GHz were adopted as the filler materials. The dielectric constants at microwave frequency of the three kinds of ceramic-polyethylene composites with various compositions will be compared with the theoretical curves of the eight mixing laws. Furthermore, the dielectric constants of composites are then converted to the dielectric constants of pure filler medium by these mixing equations. The accuracy on estimating dielectric constants of pure ceramics by various mixture rules will be compared. The most suitable mixture rule(s) to calculate the dielectric constant of pure ceramic powder will be suggested. Some rules with perspective potential for future work are suggested.

The measurement frequency is in the microwave range. However, the achievement can also be applied to low frequency case, as the dielectric constant does not have remarkable change below microwave frequency.

2. MIXTURE EQUATIONS

2.1. Mixture Equations

One of the most well known representative equations to simulated dielectric constants of composites containing the fillers are the exponential mixture rules [1–14],

$$(\varepsilon_{mix})^k = V_d(\varepsilon_d)^k + (1 - V_d)(\varepsilon_m)^k \tag{1}$$

$$k = 1 \quad (\text{parallel model}), \text{or} \tag{2}$$

$$k = -1 \ \ (\text{serial model}), \text{or} \tag{3}$$

$$k = 1/2 \ (\text{refractive model}), \text{or} \tag{4}$$

$$k = 1/3 \ (\text{random model}) \tag{5}$$

where ε_{mix}, ε_d, and ε_m are dielectric constants of a composite, pure dielectric filler, and polymer matrix material, respectively. The V_d is the volume percentage of the filler medium. Equations (2) and (3) are two extreme cases. The dielectric constant can be derived by regarding the total capacitance as the sum of two capacitances in parallel or serial, respectively. The Equation (4) is based on the optical path length of a single electromagnetic ray and will be referred to as the refractive model. Equation (5) assumes randomly dispersed filler particles which is closer to the physical condition. A differential equation related to the ε_{mix} and V_d has been developed to derive the expression of Equation (5).

Various empirical equations have also been suggested. In addition to the above four exponential rules with constant k, a modified empirical equation, Equation (1), was suggested by Wakino [5],

$$k = V_d - 0.35 \tag{6}$$

This equation was obtained using the Monte Carlo and finite element methods with the assumption of non aspect and randomly dispersed mixture of two materials. Another modified empirical equation of (1) was suggested by Stölzle et al. [6],

$$k = 1.65V_d + 0.265 \tag{7}$$

A new and interesting modified empirical equation of (1) was recently suggested by Kim et al. [7],

$$\begin{aligned}(\varepsilon_{mix})^k &= zV_d(\varepsilon_d)^k + (1 - zV_d)(\varepsilon_m)^k \\ k &= AV_d + B \\ z &= 3.23f^{0.0416} - 6.481 \\ A &= 11.0f^{0.0208} - 14.748 \\ B &= -0.22f^{0.0537} + 1.0582\end{aligned} \tag{8}$$

where f is the frequency. The spectrums at microwave frequency for complex permittivity of composites were investigated.

Finally, the eighth mixing rule is the logarithmic mixing rule, which is also a well known empirical equation and the inverse of the exponential rules [1, 6, 15],

$$\log \varepsilon_{mix} = V_d \log \varepsilon_d + (1 - V_d) \log \varepsilon_m \tag{9}$$

As mentioned, $k = 1$ and $k = -1$ are two extreme cases, and no good accuracy will be expected. The $k = 1/2$ model was first reported for the soil-water mixture. The accuracy for ceramic-polymer composite is unknown and worth studying. The random model, $k = 1/3$, is closer to the physical condition and a better accuracy than that for $k = 1$ and $k = -1$ is expected. The logarithmic rule has shown good matching situation with the experimental of aluminum-epoxy composites. All these equations along with the other empirical equations of (6) to (8) will be studied.

Some of the above rules have been studied in recent years [3, 7–10]. Unfortunately, they have been studied without knowledge about their theoretical errors. In this paper, the eight mixture rules are used to compare with the measured dielectric constant values of composite samples. In addition to the comparisons of experimental values and the function curves of various mixing laws, the theoretical and physical errors for dielectric constant estimations of pure ceramics by the above formulas are also introduced.

2.2. Error Analyses for Estimating Dielectric Constants of Pure Ceramics

To understand the adequacy of measuring the dielectric constants of pure ceramics from the measured dielectric constants of composites by each mixing rule, the computed errors on dielectric constants of pure ceramics ($\Delta\varepsilon_d$) due to the dielectric constant errors of individual rules ($\Delta\varepsilon_{mix}$) are derived. From Equation (1), we have,

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{1}{V_d}\left(\frac{\varepsilon_{mix}}{\varepsilon_d}\right)^k \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \tag{10}$$

for $k = 1$ (parallel model), $k = -1$ (serial model), $k = 1/2$ (refractive model), $k = 1/3$ (random model), $k = V_d - 0.35$ (Wakino's model), and $k = 1.65V_d + 0.265$ (Stölzle's model).

From Equation (8),

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{1}{zV_d}\left(\frac{\varepsilon_{mix}}{\varepsilon_d}\right)^k \frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \tag{11}$$

for Kim's model with $k = AV_d + B$. Finally, from Equation (9),

$$\frac{\Delta\varepsilon_d}{\varepsilon_d} = \frac{1}{V_d}\frac{\Delta\varepsilon_{mix}}{\varepsilon_{mix}} \tag{12}$$

for logarithmic mixture rule.

From the above equations, the error may change with the volume percentages and dielectric constants of both matrix and filler. As an example, Figure 1 shows the variations of the computation errors ($\Delta\varepsilon_d/\varepsilon_d$) with respect to the volume percentage of dispersed material

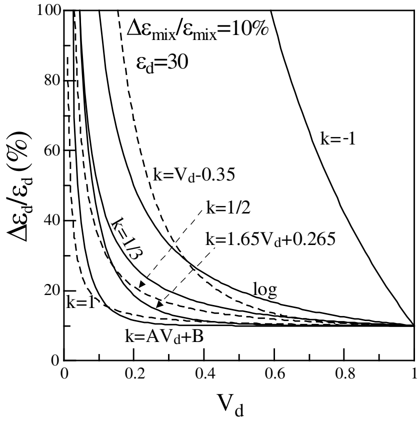

Figure 1. Theoretical analyses on the error of estimating dielectric constant of pure ceramic vs. volume concentration of filler under the condition of $\varepsilon_d = 30$ and $\Delta\varepsilon_{mix}/\varepsilon_{mix} = 10\%$.

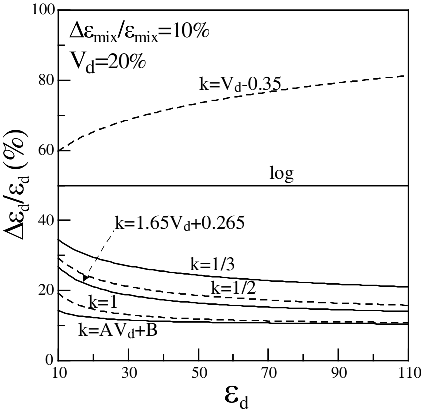

Figure 2. Theoretical analyses on the error of estimating dielectric constant of pure ceramic vs. dielectric constant of pure filler under the condition of $V_d = 20\%$ and $\Delta\varepsilon_{mix}/\varepsilon_{mix} = 10\%$.

assuming a 10% error of mixture rule ($\Delta\varepsilon_{mix}/\varepsilon_{mix}$) and $\varepsilon_d = 30$. Similar curves as Figure 1 can be observed for errors other than 10% and $\varepsilon_d \neq 30$. The dielectric constant of 2.32 for the matrix material (polyethylene) was adopted. It can be seen that the error increases with decreasing the volume concentration of dispersed material which can also be found easily in Equations (10) to (12). Therefore, to have a more accurate estimation, higher volume percentage of V_d is a better choice. Obviously, mixture rule with $k = -1$ is not a good choice for calculating the dielectric constants of pure ceramics unless the rule is very accurate ($\Delta\varepsilon_{mix}/\varepsilon_{mix}$ very small). It seems that mixing rules with $k = AV_d + B$ and $k = 1$ have lower calculation errors under the same $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value. The second choice is equations with $k = 1.65V_d + 0.265$, $k = 1/2$, and $k = 1/3$. It can also be found that the error decreases with increasing k value, which agrees with Equation (10). Figure 2 shows changes of $\Delta\varepsilon_d/\varepsilon_d$ with the dielectric constant of filler material, assuming $\Delta\varepsilon_{mix}/\varepsilon_{mix} = 10\%$ and $V_d = 0.2$. Similar curves as Figure 2 can also be observed for errors other than 10% and $V_d \neq 0.2$. The rule with $k = -1$ is not in the figure because its errors are over 100%. Again, it is found that mixing rules of $k = AV_d + B$ and $k = 1$ have lower calculation errors, and the next better rules are equations with $k = 1.65V_d + 0.265$, $k = 1/2$ and $k = 1/3$. However, the actual calculation accuracy will also depend on the $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value of the individual mixture rules. The physical $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value of each rule depends on experimental result.

3. EXPERIMENTS

The same experimental data in [4] are adopted in this paper to analyze the eight mixing equations because the existing data are enough for our analyses, and new measurements will just obtain similar results. Polyethylene powder with density 0.915 g/cm^3 and measured dielectric constant of 2.32 at 10 GHz was adopted as the matrix. Three ceramic powders, alumina with density 3.97 g/cm^3 and two commercial materials with densities 3.70 g/cm^3 and 4.50 g/cm^3, are the filler materials. Their dielectric constants are $\varepsilon_d = 10$, 20, and 36 at 10 GHz, respectively.

Suitable amounts with the desired volume concentrations of polyethylene and ceramic powders (V_d) were mixed using alcohol as a solvent and ZrO_2 balls in a plastic jar by ball milling more than 10 hours. After the well mixed suspension was stirred and evaporated until almost dry, it was baked at 80°C for 4 to 5 hours to obtain the complete dried powder mixture. The mixed powder was then pressed using a steel die at temperature 125°C (the melting point of polyethylene is 115°C) and under a pressure $\sim 7,000\,\text{lb/in}^2$ for about 0.5 hour to make disk composite samples [4].

Dielectric constants of the disk shaped composite samples are measured by the widely used Hakki and Coleman post resonance technique at 10 GHz [4, 27]. The main advantage for this method is that it can give very reliable measurement on dielectric constant value [27]. All the sample dimensions are designed to have the resonant frequency of desired $\text{TE}_{01\delta}$ mode at desired frequency for measurements [28].

4. RESULTS AND DISCUSSIONS

We applied the above eight mixing formulas to draw the theoretical curves of the dielectric constants of binary mixtures. Figures 3, 4, and 5 compare the theoretical curves to the experimental data for the ceramic-polyethylene mixtures for $\varepsilon_d = 10, 20$, and 36, respectively. From the three figures, we cannot find a mixture rule, which can match very well for all the experimental data. However, it can be seen that, in general, for low dielectric filler concentrations ($V_d < 0.25$), rules of $k = 1.65V_d + 0.265$ and $k = 1$ have better matching condition for the experimental results than other mixing rules and equations of $k = AV_d + B$, $k = 1/2$, and $k = 1/3$ are the next. For high dielectric filler concentration ($V_d > 0.25$), $k = 1/2$ and $k = 1/3$ have the best matching for the experimental data, followed by $k = V_d - 0.35$ and logarithmic rule. From Figures 1 and 2, we have already known

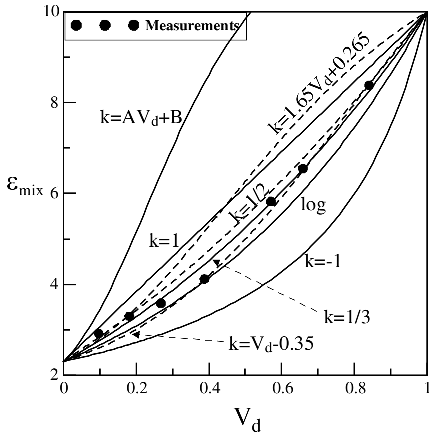

Figure 3. Comparisons of experimental data and mixture rules for $\varepsilon_d = 10$.

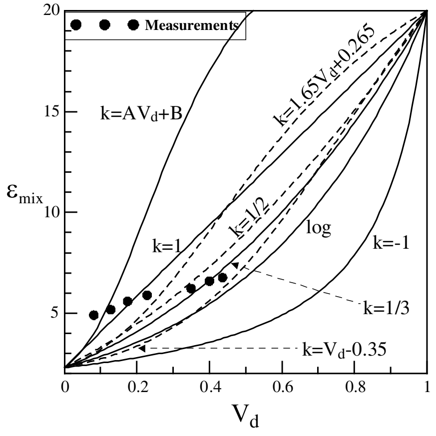

Figure 4. Comparisons of experimental data and mixture rules for $\varepsilon_d = 20$.

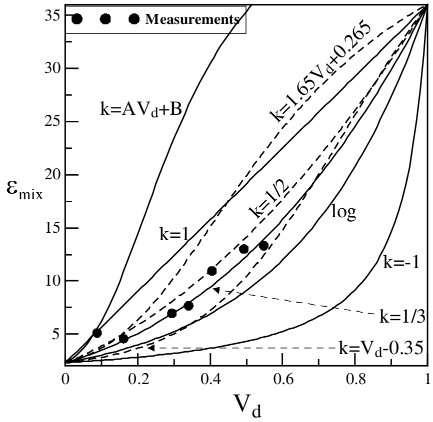

Figure 5. Comparisons of experimental data and mixture rules for $\varepsilon_d = 36$.

Table 1. Summary on the choice of mixture rules under high V_d.

	Choice of mixture rules	Summary
From analyses of errors (Figures 1 and 2)	1st choice: $k = AV_d + B, k = 1$. 2nd choice: $k = 1.65V_d + 0.265$, $k = 1/2, k = 1/3$.	$k = 1/2$ $k = 1/3$
From experimental results (Figures 3, 4, and 5)	1st choice: $k = 1/2, k = 1/3$. 2nd choice: $k = V_d - 0.35$,log rule.	

that equations of $k = AV_d + B$ and $k = 1$ have lower computation errors with the same $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value, and the next better rules are $k = 1.65V_d + 0.265$, $k = 1/2$, and $k = 1/3$. We conclude the results for high V_d in Table 1. The low V_d case is not included in the table because of its lower theoretical accuracy. From Table 1, rules of $k = 1/2$ (refractive model) and $k = 1/3$ (random model) are the best choices to estimate the dielectric constants of pure ceramics. Their expected higher accuracy are from both potential of less errors as mentioned in Figures 1 and 2 and good curve matching with the physical data in Figures 3 to 5. It can also be seen that the matching condition for the equation of $k = 1/3$ is a little better than that of the equation of

$k = 1/2$, but the equation of $k = 1/2$ has less calculation error. It is difficult to judge which one is the best choice.

On the other hand, it will be worthwhile to study and improve the curve matching situation for rules of $k = AV_d + B$, $k = 1$. If we can modify those equations to give a better curve matching result, due to their low theoretical error, they could become better choices than rules of $k = 1/2$ and $k = 1/3$. The above discussions also imply a concept, i.e., a better curve matching does not guarantee less estimation error. Of course, rule of $k = -1$ (serial model) is not adequate for predicting the dielectric constants of pure ceramics because of its high error as shown in Figure 1, and it cannot allow any very little $\Delta\varepsilon_{mix}/\varepsilon_{mix}$ value.

To give a further understanding and confirm the above discussions, Table 2 lists the estimated dielectric constants of pure ceramic materials from the measured dielectric constants of ceramic-

Table 2. Prediction of dielectric constant of pure ceramic by various mixture rules from the measured dielectric constants of ceramic-polyethylene composite samples. (V_d and ε_{mix} data are from Ref. [4]. Errors less than 25% are marked with underline.)

(a) Estimated ε_d by various mixture rules for ε_d=10

	Low V_d		High V_d				
Ceramic volume percentage, V_d (%)	9.70	18.09	26.78	38.75	57.05	65.84	84.06
Measured ε_{mix}	2.91	3.30	3.58	4.12	5.82	6.55	8.38
$k=1$	8.61	7.81	7.08	6.99	8.48	8.76	9.54
$k=-1$	−1.97	−3.42	−6.86	−16.55	−37.9	158.7	16.82
$k=1/2$	12.03	9.98	8.52	8.07	9.53	9.61	9.96
$k=1/3$	14.41	11.40	9.30	8.61	10.08	10.04	10.17
$k=V_d-0.35$	8.38	23.83	13.12	10.08	10.53	10.11	9.98
$k=1.65V_d+0.265$	12.99	9.65	7.80	7.15	8.18	8.37	9.21
$k=AV_d+B$	6.19	5.27	4.72	4.64	5.59	5.95	7.10
Log	26.19	16.84	12.00	10.33	11.72	11.27	10.71

(b) Estimated ε_d by various mixture rules for ε_d=20

	Low V_d				High V_d		
Ceramic volume percentage, V_d (%)	8.11	12.75	17.43	22.79	36.62	39.99	43.61
Measured ε_{mix}	4.90	5.17	5.58	5.89	6.44	6.59	6.76
$k=1$	34.36	24.81	21.12	18.05	13.61	13.03	12.53
$k=-1$	−41.50	−68.58	0.967	−1.37	−3.04	−3.66	−4.49
$k=1/2$	102.2	55.58	40.55	30.37	18.53	17.17	16.01
$k=1/3$	214.6	92.89	60.37	41.14	21.86	19.84	18.17
$k=V_d-0.35$	complex	complex	50452	45.72	36.78	29.03	23.84
$k=1.65V_d+0.265$	153.5	59.21	36.76	25.12	14.49	13.45	12.60
$k=AV_d+B$	32.70	17.43	13.06	10.48	7.82	7.57	7.38
Log	25816	1320	371.6	142.5	38.27	31.98	27.25

(c) Estimated ε_d by various mixture rules for ε_d=36

	Low V_d		High V_d				
Ceramic volume percentage, V_d (%)	8.97	16.06	29.41	34.01	40.51	49.31	54.78
Measured ε_{mix}	5.11	4.57	6.95	7.67	10.95	13.00	13.32
$k=1$	33.63	16.43	18.11	19.09	23.65	24.00	22.42
$k=-1$	−0.448	−1.10	−1.80	−2.17	−2.42	−3.44	−4.51
$k=1/2$	96.24	28.99	28.34	34.96	35.30	33.10	29.27
$k=1/3$	195.8	40.49	36.6	33.93	44.48	39.71	33.94
$k=V_d-0.35$	complex	4240	135.7	82.77	87.02	53.89	39.89
$k=1.65V_d+0.265$	133.7	27.6	21.74	20.17	24.57	23.22	21.18
$k=AV_d+B$	29.89	10.52	10.07	9.958	12.67	13.32	12.94
Log	16856	165.4	98.79	79.38	108.3	77.13	56.78

polyethylene composite samples. The low accuracy of rule of $k = -1$ is confirmed again in the table. For lower filler concentration ($V_d < 0.25$), the rule of $k = 1$ (parallel model) has the highest accuracy for all three kinds of composites. The rule of $k = 1/2$ (refractive model) is the second. However, no rule can accurately compute dielectric constants of ceramic powders with all the experimental data, mainly because of the high error with low V_d. For high filler concentration ($V_d > 0.25$), the rules of $k = 1/2$ and $k = 1/3$ have the best accuracy. This agrees well with our analyses in Table 1. Some other equations are only good for one material. The refractive and random models can estimate the dielectric constants of all three kinds of materials with various experimental data under high V_d condition. Both $k = 1/3$ and $k = 1/2$ equations have good performance. The random model has a better curve fitting, and the refractive model has less calculation error. Therefore, to estimate the dielectric constant of pure ceramic, the best way is to choose the refractive or random models and to be applied to the data of higher filler concentrations.

5. CONCLUSION

Dielectric constants of composite materials with various volume percentages by three kinds of ceramic fillers and polyethylene matrix have been investigated by exponential and logarithmic mixture equations at 10 GHz. Experimental results are compared for the eight mixture rules. No exact match between the experimental data with either one of the rules was found for both high and low filler concentrations. Some of the rules match better for the lower volume percentage data, and some are better for the higher volume percentage data. Errors with the estimation of the dielectric constant of 100% volume percentage ceramics by the eight mixture rules have been studied. Theoretical analyses imply that low filler concentration data is not adequate to calculate the dielectric constant of a substance from measurement on composite sample. To have a more accurate estimation, higher volume percentage of V_d is preferred. It was found that the random and refractive models are the most adequate rules for higher filler concentration. The dielectric constants of all three ceramic materials are measured accurately. Theoretically, mixture law fits better for experimental results, which does not guarantee better accuracy of estimating the dielectric properties. Finally, that rules with less theoretical error are good choices will be the research topics in the future.

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