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# **Solubility of metallic mercury in octane, dodecane and toluene at temperatures between 100°C and 200°C**

M. M. MIEDANER,\* A. A. MIGDISOV, and A. E. WILLIAMS-JONES Department of Earth and Planetary Sciences, McGill University, Montréal, Québec, Canada H3A 2A7

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**Abstract**—The solubility of metallic mercury in dodecane, octane and toluene has been investigated experimentally at temperatures up to  $200^{\circ}$ C and pressures up to 6 bars (toluene). The equilibrium Hg concentrations are very similar in octane and dodecane, reaching values of 821 ppm and 647 ppm, respectively at 200 $^{\circ}$ C, whereas they are significantly lower in toluene (e.g., 280 ppm at 200 $^{\circ}$ C). The behavior of Hg in toluene is nevertheless similar to that in the alkanes. There is a strong prograde dependence of Hg concentration on temperature in both types of solvent, which can be described by the following experimentally determined relationships:

> Dodecane:  $Log(X_{Hg}) = -1943.1/T[K] + 0.8884$ Octane:  $Log(X_{Hg}) = -2346.3/T[K] + 1.6017$ Toluene:  $Log(X_{Hg}) = -2768.3/T[K] + 1.9726$

The solubility of metallic mercury can be modeled satisfactorily using a modification of the scaled particle theory designed to predict the behavior of nonpolar solutes in polar solvents. This modification involves an adjustment of the Lennard Jones Potential and the hard-sphere diameters of solute and solvent to account for the effects of temperature.

The results of this study suggest that hydrocarbons could play a significant role in transporting Hg in natural systems, which may explain the high concentrations of metallic mercury in some crude oils, as well as the common occurrence of petroleum in epithermal mercury deposits. *Copyright © 2005 Elsevier Ltd*

### **1. INTRODUCTION**

An important feature of many mercury deposits is their close spatial association with hydrocarbons. For example, cinnabar occurs together with small droplets of oil in the cinnabar deposits of the Huancavelica district, Peru (Concha et al., 1952), and the Culver Baer mercury deposit, California (Bailey, 1959; Peabody and Einaudi, 1992), and with more bituminous hydrocarbons in the Don Dnieper Depression, Russia (Gavrish et al., 1987). Despite examples such as these and evidence of appreciable concentrations of Hg in crude oils (up to several 10's of ppm, Wilhelm and Bloom, 2000), little consideration seems to have been given to the possibility that petroleum could be an important medium of mercury transport (cf. Fein and Williams-Jones, 1997). On the contrary, most studies have attributed the formation of mercury deposits to hydrothermal processes, even where a spatial association with hydrocarbons is clearly evident, e.g., the paper of Peabody and Einaudi (1992) for the Culver Baer deposit. In large part, the failure of researchers to propose or apparently consider models involving transport of mercury by hydrocarbons is probably due to the paucity of data on the solubility of Hg in hydrocarbons at elevated temperature.

Previous experimental studies of the solubility of mercury in hydrocarbons have been conducted at temperatures  $\leq 60^{\circ}$ C,

and have focussed on *n*-alkanes, the principal constituents of liquid petroleum. These studies, as summarized in Table 1, showed that the solubility of Hg in  $C_{5-8}$  alkanes ranges from 1.3 ppm at 25°C to 4.4 ppm at 45°C (Spencer and Voigt, 1968; Okouchi and Sasaka, 1981), and that the solubility increases with increasing carbon number. The solubility of mercury in aromatic hydrocarbons, which may represent more than 30% by weight of a crude oil (Killops and Killops, 1997), has not been investigated experimentally at temperatures above 25°C. Studies of the speciation of mercury in hydrocarbons have considered Hg°, HgCl<sub>2</sub>, CH<sub>3</sub>Hg<sup>+</sup> and (CH<sub>3</sub>)<sub>2</sub>Hg, and concluded that the solubility of these species decreases in the order presented here (e.g., Snell et al., 1998). Moreover, most researchers have concluded that Hg<sup>o</sup> is the overwhelmingly dominant form of mercury in crude oil (Snell et al., 1998; Wilhelm and Bloom, 2000).

The objectives of the present study were to investigate the behavior of mercury in hydrocarbons at temperatures similar to those reported for the formation of mercury deposits (Peabody and Einaudi, 1992), i.e., 100 to 200°C, and to find a tool with which to predict mercury solubility at elevated temperatures. In this paper, we report the results of experiments designed to determine the solubility of metallic mercury in octane, dodecane and toluene, and compare the experimentally determined solubilities with those predicted using scaled particle theory (SPT) (Pierotti, 1976).

### **2. EXPERIMENTAL METHOD**

Previous experiments designed to determine the solubility and speciation of mercury in liquid hydrocarbons have been conducted at

<sup>\*</sup> Author to whom correspondence should be addressed, at Earth Sciences Department, Mineralogy–Environmental Geochemistry Group, Johannes Gutenberg University, Becher Weg 21, 55128 Mainz, Germany (Miedaner@uni-mainz.de).

Table 1. Previously published experimental data on the solubility (ppm) of liquid mercury in organic solvents.

Reference	Octane	Dodecane	Toluene	Temp (°C)
De Ligney and Van der				
Veen (1972)	1.6 <sup>a</sup>		$1.8^{\rm a}$	25
Clever and Iwamoto (1987)	1.0 <sup>b</sup>	$1.5^{b}$		25
	$1.5^{b}$	$2.1^{\rm b}$		45
Spencer and Voigt (1968)	1.1		13	25
Okouchi and Sasaka (1981)	0.8			25
	25			45

<sup>a</sup> Recalculated from  $log(P/X_{\text{Hg}})$ .<br><sup>b</sup> Recalculated from mol/L.

sufficiently low temperature  $(<60^{\circ}C$ ) that the solvent could be sampled at the experimental conditions, and transported to the analytical device without fear of precipitation of a mercury phase (Clever and Iwamoto, 1987; Snell et al., 1998). However, exploratory experiments conducted during the initial stages of our study showed that the equilibrium concentrations of Hg in alkanes (octane and dodecane) at elevated temperatures (100 to 200°C) are not preserved after quenching the experimental vessel to room temperature. We therefore surmise that small amounts of mercury precipitated on the walls of the reactor during quenching. Subsequent experiments were therefore conducted using a method that separates solvent from solute at the end of an experiment, i.e., before quenching.

The experimental approach used in this study involved a combination of mass loss and solubility methods. The experiments were performed at temperatures up to 200°C and a maximum pressure of 6 bars, in autoclaves constructed from titanium alloy (grade 2 ASTM B348) to ensure the inertness of the reactor (Fig. 1). These autoclaves were preconditioned with nitric acid to produce an inert layer of  $TiO<sub>2</sub>$  on the internal surface, and so further reduce the possibility of reaction be-



Fig. 1. Drawing illustrating type of thin-walled titanium alloy V8T autoclave used in the experiments, partly filled with organic solvent, and quartz ampoule containing liquid mercury.

tween the experimental fluid and the vessel. Before and after loading the autoclaves with liquid mercury and organic solvent, nitrogen was passed through them for  $\sim$  40 min to remove atmospheric gases. The mercury was placed in quartz ampoules, which were open at one end, weighed and inserted into the autoclaves. The latter were then partly filled with organic solvent (octane, toulene or dodecane). The experiments were conducted at temperatures between 100°C and 200°C by heating the autoclaves in an electric furnace equipped with thick aluminium walls to reduce temperature gradients. At the upper end of this range, the temperature varied by less than 1°C over the duration of an experiment. The solvents were obtained from Fischer Scientific Inc. and had a purity of 99%. All weights were determined using a Mettler M3 analytical balance.

After the experiments, the autoclaves were water-cooled to room temperature, opened and the ampoules containing the mercury removed from the autoclaves. Most of the solvent in the ampoules was aspirated using a micropipette. To eliminate any solvent remaining in the ampoules after aspiration ( $< 0.1$  mL), the latter were washed in  $~1$ <sup>-70</sup>-100 mL of acetone at ambient temperature. This effectively replaced the small quantities of solvent remaining in the ampoules with acetone, which was later evaporated at 50°C. As the solubility of mercury in acetone at 25°C is several orders of magnitude lower than that measured in our experiments (Rosenberg and Kay, 1974), we ignored any possible losses of mercury from the sample due to this washing procedure. The ampoules were then reweighed, and the measured mass losses were interpreted to represent the solubility of mercury in the solvent. Potential losses of mercury due to its volatilization during evaporation of the acetone (which never took more than 5–10 min) were also ignored, as the blank runs (heating of samples of mercury to 50°C in the absence of acetone) did not show any detectable mass losses

We did not consider the contribution of Hg dissolved in the vapor phase in calculating the solubility of Hg in the hydrocarbon liquids investigated because this contribution is predicted to be negligible. At 100°C, based on calculations using the Antoine Equation (Lindstrom and Mallard, 2003), the vapor pressure of the solvent in our experiments would have ranged from 0.02 bars with dodecane to 0.53 bars with toluene and at 200°C the corresponding values would have been 0.68 and 6.2 bars. At these two temperatures, the mercury vapor pressure would have been 0.0004 and 0.024 bars, respectively. Thus the mole fractions of Hg in the vapor phase would have been  $4.83 \times 10^{-1}$ and  $1.21 \times 10^{-8}$  in dodecane and  $1.96 \times 10^{-10}$  and  $2.61 \times 10^{-9}$  in toluene at 100°C and 200°C, respectively. These vapor pressures would have corresponded to a maximum Hg solubility in the vapor phase ranging from 5.48 ppt to 14 ppb, i.e., about five orders of magnitude lower that the solubilities measured in the experiments (see below). We therefore conclude that the solubility of Hg in the vapor phase made no significant contribution to the overall solubility, i.e., all the Hg was effectively dissolved in the hydrocarbon liquid.

To establish the time required to attain equilibrium, a set of kinetic experiments was performed at several temperatures by removing one autoclave from the oven per day over a period of 15 days. From Figure 2, it can be seen that the concentration of metallic mercury did not change significantly after 5 d. We therefore interpret this to represent the time needed for the solvent to reach equilibrium with respect to mercury concentration. All subsequent experiments had run-times of at least two weeks.

#### **3. RESULTS**

The results of our experiments are summarized in Table 2 and Figures 3 to 5. These data show that the solubility of liquid mercury in octane and dodecane increases with increasing temperature, and is appreciable at the highest temperature investigated (e.g., 821 ppm Hg in dodecane at 200°C). The solubility of metallic mercury in toluene also increases with increasing temperature. However, the measured concentrations were significantly lower than in the alkanes, and reached a maximum of only 280 ppm at 200°C. In both types of solvent,  $log (X_{Hg})$  is a linear function of reciprocal temperature (Figs.



Fig. 2. Plot of log( $X_{Hg}$ ) in octane at 100°C vs. duration of experiment showing that octane saturates with liquid mercury after  $\sim$  5 d.

3–5) and consequently Hg° solubility can be predicted using the following equations, which were fitted to the experimental data:

> Dodecane:  $Log(X_{Hg}) = -1943.1/T[K] + 0.8884$ Octane:  $Log(X_{Hg}) = -2346.3/T[K] + 1.6017$ Toluene:  $Log(X_{Hg}) = -2768.3/T[K] + 1.9726$

These equations are considered accurate to  $\pm 2\%$  in the temperature range of 100 to 200°C.

Although our experimental method differs quite significantly from those of the studies mentioned earlier, our results, when extrapolated down to the appropriate temperatures, agree well with those of these studies, notably Okouchi and Sasaka (1981) and Clever and Iwamoto (1987).

The main potential source of error in the experimental method employed in our study is the loss of mercury due to adsorption of Hg on the walls of the autoclaves. This error is considered to be negligible, because the measured concentration of mercury in repeated experiments was reproducible to 98%. The weighing error is estimated to be less than 0.00005 g, which results in a maximum deviation of 2 ppm in the determined concentration of dissolved Hg, and an overall experimental error of less than 2%.

# **4. DISCUSSION**

The linear correlation of the logarithm of mercury solubility with  $1/T$  [K] indicates that the mercury speciation at elevated temperatures does not differ significantly from that at 25°C, and that Hg<sup>o</sup> was the dominant mercury species in our experiments. As concentrations of other mercury complexes were negligible, it should, in principle, be possible to model our data using Scaled Particle Theory (SPT). This theory was developed to describe the solubility of inert gases in polar and nonpolar solvents (Pierotti, 1976), and assumes that the gas dissolves by occupying cavities created in the latter. The dissolution of metallic mercury into the hydrocarbon solvent can be expressed by the following reactions:

$$
Hg^{\circ} \leftrightarrow Hg_{(gas)} \tag{1}
$$



Table 2. Solubility of Hg in organic solvents as determined experimentally in this study.

<sup>4</sup> Also tabulated is the calculated solubility based on an unmodified Scaled Particle Theory model modified to take into account the temperature dependencies of the mean interaction depth of ₩ mean interaction depth  $\mathbf{e}$ đ the temperature dependencies and as ppm. solute and solvent and the mean hardsphere diameters (see text for further detail). Solubilities are reported as the logarithm of the mole fraction of Hg in the solvent and as ppm. account in the solvent into take  $Hg$ modified to  $\sigma$ fraction model logarithm of the mole Theory Particle<sup>'</sup> further detail). Solubilities are reported as the led scal and a model Theory Particle Scaled unmodified text for ty based on an u<br>diameters (see solubility hardsphere calculated mean h tabulated is the c:<br>1 solvent and the and solvent Also solute:



Fig. 3. Plot of  $log(X_{Hg})$  in toluene vs. reciprocal temperature comparing experimentally determined values with those predicted by modified SPT (solid line). Dashed line represents best fit to experimental data (log( $X_{\text{Hg}}$ ) = -2768.3/T[K] + 1.9726).

$$
Hg_{(gas)} \leftrightarrow Hg_{(diss)} \tag{2}
$$

which can be simplified to an overall reaction:

$$
Hg^{\circ} \leftrightarrow Hg_{(diss)} \tag{3}
$$

This process of solvation is the same as that required for the application of SPT theory. The latter involves first creating a cavity that is large enough to introduce a solute atom into the liquid solvent. The size of the cavity and the energy required to form it are determined by the geometry of both solvent and solute molecules. Intermolecular interactions are then taken into account in a second step using, for example, the Lennard Jones model. In this thermodynamic approach, the partial molar Gibbs free energy for the creation of a cavity,  $G_C$  is expressed as follows:



Fig. 4. Plot of  $log(X_{Hg})$  in octane vs. reciprocal temperature comparing experimentally determined values with those predicted by modified SPT (solid line). Dashed line represents best fit to experimental data (log( $X_{\text{He}}$ ) = -2346.3/T[K] + 1.6017).



Fig. 5. Plot of  $log(X_{Hg})$  in dodecane vs. reciprocal temperature comparing experimentally determined values with those predicted by modified SPT (solid line). Dashed line represents fit to experimental data ((log( $X_{Hg}$ ) = -1943.1/T[K] + 0.8884).

$$
\bar{G}_C = RT \Bigg\{ 6 \frac{y}{1-y} \Bigg[ 2 \Bigg( \frac{a_{12}}{a_1} \Bigg)^2 - \frac{a_{12}}{a_1} \Bigg] + 18 \Bigg( \frac{y}{1-y} \Bigg)^2 \Bigg[ \Bigg( \frac{a_{12}}{a_1} \Bigg)^2 - \frac{a_{12}}{a_1} + \frac{1}{4} \Bigg] - \ln(1-y) \Bigg\} \quad (4)
$$

where  $a_1$  and  $a_2$  are the hard-sphere diameters of solvent and solute,  $a_{12} = (a_1 + a_2)/2$  is the mean hard-sphere diameter and  $y$  ( $y = \pi a_1^3 \rho/6$ ) describes the geometry of the solvent and *R* is the gas constant. In the latter relationship,  $\rho$  represents the number density of the solvent.

Following the approach of Wilhelm and Battino (1971), the Gibbs free energy of the intermolecular interactions,  $G<sub>p</sub>$ , can be described as follows:

$$
\bar{G}_I = -3.555 \cdot \pi \cdot a_{12} \cdot \rho \cdot \frac{\varepsilon_{12}}{k} \tag{5}
$$

where *k* is the Boltzman constant and  $\varepsilon_{12}/k$  is the mean interaction depth of solute and solvent, which is calculated as:  $\varepsilon_{12}/k$  $= (\varepsilon_1 \times \varepsilon_2)^{0.5} \times 1/k$ . The Gibbs free energy of dissolution,  $G_s$ , is related to  $G_C$  and  $G_I$  as well as to  $K_H$  (Henry's Law constant) as follows:

$$
G_S = G_C + G_1 + RT \ln \left( \frac{RT}{V_1^o} \right) = RT \ln(K_H) \tag{6}
$$

in the equation above *R* represents the gas constant and  $V_1^{\circ}$  the molar volume of the solvent. This model assumes that the hard-sphere diameters and the interaction depth do not change with temperature. The hard-sphere diameters were taken from De Ligney and Van der Veen (1972) and the interaction depths from Pierotti (1976), while the number density of the solvent was calculated in the manner proposed by the latter author. Predictions of mercury solubility made using this model match the experimental data very well at room temperature but at temperatures above 100°C the experimentally determined concentration of metallic mercury in ppm is underestimated by more than 95% (cf. Table 2).

Solvent	$a1^{\circ}$ (cm)	$da1/dT$ (cm/K)	$\varepsilon$ 1/k	$(d\varepsilon 1/k)/dT$		$V1^{\circ}$ [cm <sup>3</sup> ]
Toluene	$5.68 \times 10^{-8}$	$-0.9 \times 10^{-11}$	535	$-0.1$	0.49	106.84
	De Ligney and Van der Veen $(1972)$	This study	Pierotti (1976)	This study	Calculated according to Pierotti $(1976)$	De Ligney and Van der Veen $(1972)$
Octane	$6.58 \times 10^{-8}$	$-2.5 \times 10^{-11}$	606	$-0.22$	0.542	163.54
	De Ligney and Van der Veen $(1972)$	This study	De Ligney and Van der Veen (1972)	This study	Calculated according to Pierotti $(1976)$	De Ligney and Van der Veen $(1972)$
Dodecane	$7.56 \times 10^{-8}$	$-7 \times 10^{-11}$	685	$-0.12$	0.4	228.60
	De Ligney and Van der Veen $(1972)$	This study	De Ligney and Van der Veen (1972)	This study	Calculated according to Pierotti $(1976)$	De Ligney and Van der Veen (1972)
Mercury	$2.9 \times 10^{-8}$	$-7 \times 10^{-11}$	851	$-0.1$	Not required	Not required
	De Ligney and Van der Veen $(1972)$	This study	De Ligney and Van der Veen (1972)	This study		

Table 3. Modeling parameters used in this study. Parameters for other solvents used in the hydrocarbon mixture have been interpolated based on this table.

To better predict our data, we introduced temperature dependencies for the mean interaction depth of solute and solvent, and mean hard-sphere diameters. Wilhelm (1973) showed that the hard-sphere diameters  $a_1$  and  $a_2$  decrease linearly ( $a_{1,2}$ )  $a_{1,2}^{\circ}$  +  $(da_{1,2}/dT) \times (T - 298.15)$  with increasing temperature. Optimization to best fit showed that the hard-sphere diameter of  $Hg^{\circ}$  decreases by a factor of  $7 \times 10^{-11}$  cm/K. The interaction depth  $\varepsilon_{12}/k$  of the hard spheres was also allowed to decrease linearly with temperature, as it has been shown by Nasehzadeh et al. (2002). A behavior, that has been attributed to the higher kinetic energies of the atoms at higher temperature by this author. Following Nasehzadeh et al. (2002), we therefore also added a negative linear temperature dependency to the interaction depth parameter  $(\epsilon_{1,2}/k = \epsilon_{1,2}^{\circ}/k + [(\mathrm{d}\epsilon_{1,2}/k)/\mathrm{d}T] \times (T -$ 298.15) and optimized both temperature terms manually. All parameters and modifications are presented in Table 3.

As can be seen from Figures 3–5 and Table 2, the mercury solubility calculated using SPT with adjustable hard-sphere and interaction depth parameters agrees better with the measured mercury solubility than that calculated using fixed values of these parameters, although the differences are still significant. Nevertheless, in the absence of experimental data, SPT appears to provide a reasonably satisfactory approach to predicting the solubility of metallic mercury in light hydrocarbons at elevated temperatures.

## **5. GEOLOGICAL APPLICATION**

To evaluate the potential of natural hydrocarbons to transport mercury in concentrations sufficient for them to constitute ore fluids, we used the data presented earlier to model the solubility of Hg at temperatures between 100°C and 200°C in an analog of natural petroleum, based on the composition of crude oil from the Bradford Oilfield, Pennsylvania. The composition of the Bradford crude oil was taken from Levorsen (1954). All calculations were performed using the SPT model described above. Given that Hg° solubility in heavy hydrocarbons cannot be satisfactorily predicted with SPT (De Ligney and Van der Veen, 1972), only species with  $C_N < 10$  were considered. Aromatic compounds were represented by toluene and benzene. This hydrocarbon mixture was assumed to behave ideally, and Hg° solubility was calculated for temperature steps of 10°C in the interval, 110 to 200°C. The modeling parameters were taken from De Ligney and Van der Veen (1972), Pierotti

(1976), and Wilhelm (1973), and are presented in Table 3. Mercury solubility reached a maximum of 1190 ppm at 200°C (Table 4), which is considerably higher than the predicted metal concentration of many ore fluids (Barnes, 1997), but decreased sharply with decreasing temperature, and at 120°C was only 25 ppm Hg. As our calculations were based on a relatively simple analog of a crude oil, it must be emphasized that the values reported here only crudely predict the capacity of complex hydrocarbon mixtures like petroleum to dissolve metallic mercury.

The organic solvents investigated in this study are able to dissolve substantially more metallic mercury than water (Krupp, 1988; Fein and Williams-Jones, 1997), which is normally the solvent assumed to transport metals in nature. Indeed, predicted concentrations of mercury are so low in most aqueous liquids, that the latter probably do not represent viable agents for the formation of economic mercury deposits (Fein and Williams-Jones, 1997). However, mercury is an extremely volatile element, and can attain very high concentrations as Hg° in water vapor. In fact, values for mercury solubility in the vapor phase are comparable to those reported here for organic solvents (Varekamp and Buseck, 1983). Thus, in environments where hydrocarbons are absent and the vapor phase important, e.g., epithermal settings, vapor may well be the principal means of transporting Hg to the site of ore deposition.

Table 4. Composition of crude oil from the Bradford Oilfield, Pennsylvania, as reported by Levorsen  $(1954)^{a}$ 

Characteristic	Weight $%$	Mol/kg
Hydrocarbon		
Toluene $C_6H_5CH_3$	0.572	0.06
Benzene $C_6H_6$	0.0389	0.005
Hexane $C_6H_{12}$	3.4	0.39
Octane $C_8H_{18}$	3.04	0.29
Nonane $C_0H_{20}$	2.69	0.21
Temperature $(^{\circ}C)$		ppm Hg <sup>o</sup> /kg mixture
120		25
200		1190

<sup>a</sup> Only hydrocarbons lighter than  $C_{10}$  have been considered due to the limitations of the SPT model. All calculations were performed using a modified SPT model (see Table 2).

Although it is probably premature to discuss the factors that may control the formation of mercury deposits from hydrocarbon liquids, the relatively simple speciation (Hg°) and the strong dependence of mercury solubility on temperature documented by this study, suggest that cooling is likely to be an important control. Indeed this might be the only control on the deposition of liquid mercury, and coupled with a high activity of  $H_2S$  may prove to be the means by which deposits of cinnabar, the principal ore of mercury, form. Clearly, much remains to be done before a hydrocarbon-based model for the formation of economic mercury deposits is more than mere speculation, but there is now strong justification for proceeding with such research.

### **6. CONCLUSIONS**

The three investigated solvents (octane, dodecane and toluene) dissolve significant amounts of mercury at temperatures experienced by liquid petroleum in nature, and the solubility is prograde. Moreover, as suggested by the lower temperature experiments of Okouchi and Sasaka (1981), mercury solubility in alkanes increases with increasing carbon number. Aromatic hydrocarbons, represented by toluene, dissolve lower concentrations of mercury than alkanes. Mercury solubility  $(log(X_{Hg}))$ in both the alkanes and toluene increased linearly with reciprocal temperature, indicating that mercury speciation did not change with temperature, and that Hg° is the dominant mercury species in hydrocarbons at temperatures from 25 to 200°C. From these data and model calculations using an analog based on the composition of natural occurring petroleum, we conclude that crude oil can transport mercury in concentrations sufficient to form an economic mercury deposit.

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