Numerical and Experimental Investigation of Soot Suppression by Acoustic Oscillated Combustion

Yanghui Ye, Xinjie Luo, Cong Dong, Yousheng Xu, and Zhiguo Zhang*

ABSTRACT: The soot suppression by acoustic oscillations for acetylene diffusion flames was investigated combining numerical and experimental studies. The combustion and soot formation were predicted by the finite-rate detailed chemistry model and modified Moss-Brookes model, respectively, while the turbulence was predicted by the detached eddy simulation (DES) with a low Reynolds number correction. Experimental results showed that the soot rate almost decreased linearly with the amplitude of acoustic oscillation, and the pinch-off of the flame occurred at a large acoustic oscillation. Numerical results showed that the flame structure was well predicted, while the soot rate was over-predicted at large acoustic oscillations; the consumption of O2 increased obviously with the acoustic oscillation. The soot suppression was mainly caused by the decrease of the surface growth rate when the air was pushed toward the flame.

1. INTRODUCTION

Soot usually appears in the combustion of hydrocarbon fuels due to incomplete oxidation. As a threat to the environment and humans, combustion-generated soot needs to be avoided.1−5 It can accelerate global warming6,5 and cause diseases after travelling into human organs.6 Numerical tools have been widely used to analyze the soot-emitting combustion. The difficulties mainly focus on the modeling of combustion, turbulence, and soot formation. Many experimental and numerical studies have been done toward soot formation in laminar diffusion flames.7−12 Usually, the detailed chemistry is solved for laminar combustion. Turbulent combustion is more complex due to the interactions between turbulence and chemistry. The Reynolds-averaged Navier–Stokes (RANS) models were previously used to model the turbulent combustion,13−15 but they can only produce large-scale unsteadiness, and the traditional two-equation RANS models are inapplicable for laminar flows. Large eddy simulation (LES) combined with the PDF method has been used to simulate a wide range of turbulent combustion in recent years.16−19 Although better results can be obtained by LES, its computational cost is extremely high for industrial applications since a high resolution is required for wall boundary layers. The hybrid RANS/LES models employ the unsteady RANS models in the boundary layer and the LES treatment in the separated regions. Their computational cost is between RANS and LES.

Modeling of soot formation is rather complex. It mainly includes these processes: polycyclic aromatic hydrocarbon (PAH) growth, particle inception, surface growth, oxidation, particle coagulation, and fragmentation. The formation of the first aromatic ring was elucidated in the work of Westmoreland et al.20 Wang and Frenklach examined the properties21,22 and reactions23 of aromatics and then developed the modeling of PAH formation in laminar premixed acetylene and ethylene flames.24 The PAH formation in real-fuel combustion is much more complex and needs further investigations.25,26 Many experiments27 indirectly indicated that the particle inception is mainly due to the physical28 and chemical29 coalescence of PAHs. Soot models are under continuous improvement from empirical to semi-empirical and then detailed. Detailed models30−33 use detailed chemical kinetic mechanisms to describe the formation of PAHs and soot, and thus they are expensive in the computational cost. Semi-empirical models34−36 usually include the processes of inception, surface growth, oxidation, and particle coagulation. The reaction rates are determined by semi-empirical expressions, and the model coefficients need to be adjusted according to the experimental data. Semi-empirical models usually use C2H2 as the soot precursor. However, higher C2H2 does not mean higher PAHs, and different fuels have different tendencies of PAH formation.37 Thus, different fuels need different inception coefficients. In this study, the semi-empirical model was employed since its computational cost is low and only one fuel is used.

Soot formation in unsteady counterflow diffusion flames2 has been investigated experimentally38−40 and numerically.41,42 The soot production increased at low frequency oscillations,
and it was insensitive to the acoustic oscillation at high frequency. Saito et al. investigated the soot suppression by acoustic oscillations for coflow diffusion flames. The soot production decreased by lowering the frequency and increasing the amplitude of the oscillation. This experiment was repeated in this study, and the evolutions of flame structures were recorded by a high-speed camera. The amplitudes of experimental acoustic oscillations were obtained by combining experimental and numerical analyses. With the increase of acoustic oscillation, the combustion transits from laminar to turbulent, the combustion and turbulence models applicable for both laminar and turbulent flows should be used. In this study, numerical simulations were conducted using the finite-rate detailed chemistry model and the DES turbulence model with a low Reynolds number correction. The soot formation was predicted by the Moss-Brookes model with the modification in that the effect of soot formation on the chemical species was considered by using user-defined functions (UDF). Comparisons were made between the experiments and simulations, and the soot formation was analyzed in detail.

2. RESULTS AND DISCUSSION

2.1. Effect of Soot Suppression. 2.1.1. Experimental Results. Figure 1 compares the experimental and numerical results of soot suppression by acoustic oscillations. \( \bar{m} \) denotes the time-averaged soot rate after being nondimensionalized by the initial value without acoustic oscillation, and the experimental and numerical initial values were respectively 0.3 mg/s and 0.302 mg/s.

![Figure 1](https://pubs.acs.org/doi/10.1021/acsomega.0c03107)

Figure 1. Comparison between the experimental and numerical results of soot suppression by acoustic oscillations. \( \bar{m} \) denotes the time-averaged soot rate after being nondimensionalized by the initial value without acoustic oscillation, and the experimental and numerical initial values were respectively 0.3 mg/s and 0.302 mg/s.

only decreased from 0.48 to 0.46 when the amplitude of one speaker was increased by 5%.

2.1.2. Numerical Results. The predicted soot rate without acoustic oscillation matched well with the experimental data, the corresponding surface growth rate was 0.361 mg/s, and the oxidation rates by OH and O\(_2\) were respectively 0.052 mg/s and 0.007 mg/s. Figure 2 shows the experimental observation and the numerical distributions of \( T \) (above 1300 K), \( Y_{\text{OH}} \) (mass fraction of OH), \( Y_{\text{soot}} \) (mass fraction of soot), \( \dot{M}_{\text{grow}} \) and \( \dot{M}_{\text{oxid}} \) without acoustic oscillation. It can be seen that the cross-sectional diameter of the flame was a little over-predicted. The surface growth concentrated inside the flame, while the oxidation concentrated at the boundary of the flame (e). The maximum \( Y_{\text{soot}} \) was located at 13.6 mm above the steel pipe.

The numerical results in Figure 1 show that the numerical soot rate was over-predicted at \( f = 50 \) Hz, while it was first under-estimated and then over-predicted at \( f = 100 \) and 150 Hz. The turbulent intensity increases with \( A_v \), which will accelerate the molecular diffusion. In order to analyze the influence of turbulence, the laminar model was used instead of the DES model under the largest acoustic oscillation (\( f = 150 \) Hz and \( A_v = 2.9 \) m/s). Results showed that the soot rate increased by 6.9%.

Figure 3 shows the dependences of \( \bar{m}_{\text{inc}} \), \( \bar{m}_{\text{grow}} \), and \( \bar{m}_{\text{oxid}} \) on \( A_v \). \( \bar{m}_{\text{inc}} \), \( \bar{m}_{\text{grow}} \), and \( \bar{m}_{\text{oxid}} \) decreased with increasing \( A_v \) at \( f = 50 \) and 100 Hz, while they slightly increased and then decreased at \( f = 150 \) Hz. The variation of \( \bar{m}_{\text{grow}} \) was similar to that of \( \bar{m} \). With the increase of \( A_v \), \( \bar{m}_{\text{oxid}} \) decreased more quickly than \( \bar{m}_{\text{inc}} \). \( \bar{m}_{\text{grow}} \) and \( \bar{m}_{\text{oxid}} \) first increased and then decreased at \( f = 100 \) and 150 Hz. Figure 4 shows the dependences of \( Q_{\text{m,}O_2} \) and \( \bar{m}_{\text{oxid}} \) on \( A_v \). It can be seen that the consumption of \( O_2 \) under the same \( A_v \) increased with decreasing \( f \), similar to the variation of the experimental soot suppression. Although more \( O_2 \) was consumed at a lower frequency, the predicted soot rate did not always decrease. We think the main reason for the discrepancy from the experimental results lies in the inaccuracy of the semi-empirical soot model. The predicted \( \bar{m}_{\text{oxid}} \) can be neglected in this study, but the internal burning mainly caused by \( O_2 \) will lead to the breakup of soot and the increase of the soot number concentration, which has not been considered. Thus, we think that the soot oxidation is under-estimated by the soot model in this study.

Figure 5 shows the SPL measured at the position of flames and the power consumption (\( P \)) of the two speakers. The SPL and \( P \) decreased obviously with decreasing \( f \). With the increase of \( A_v \), the SPL increased slowly since it was calculated by a logarithmic operation, while \( P \) increased rapidly.

2.2. Fluctuations of Flame and Soot Formation. 2.2.1. Experimental Results. Figures 6 and 7 show the experimental observations within one cycle under \( A_v = 0.48 \) and 0.95 m/s at \( f = 50 \) Hz, respectively. The width of the flame seen from the camera view (\( B_f \)) increased when the air was sucked into the glass tubes (Figure 6a), and \( B_f \) decreased when the air was pushed out (Figure 6c), while the height of the flame maintained to be almost constant. With the increase of \( A_v \), the minimum height of the flame decreased (Figure 7a), and the pinch-off of the flame occurred (Figure 7d). Figures 8 and 9 show the experimental observations within one cycle under \( A_v = 1.4 \) and 2.2 m/s at \( f = 150 \) Hz, respectively. It can be seen that the flame was divided into three or four sections,
which was more obvious at a large $A_v$. The variation of the flame structure was smaller comparing with that at $f = 50$ Hz.

### 2.2.2. Numerical Results

Figure 10 shows the evolutions of $\dot{m}_{\text{inc}}$ and $\dot{m}$ within one cycle under different $A_v$ values at $f = 100$ Hz. With the increase of $A_v$, the minimum values of $\dot{m}_{\text{inc}}$ and $\dot{m}$ rapidly decreased, while the variations of their maximum values were relatively small. The evolutions of $\dot{m}_{\text{inc}}$ and $\dot{m}$ at $f = 50$ and 150 Hz were similar.

Figure 11 shows the evolutions of $\dot{m}_{\text{inc}}$, $\dot{m}_{\text{gro}}$, $\dot{m}_{\text{oxid}}$, and $v_a$ within one cycle at $f = 100$ Hz and $A_v = 1.6$ m/s. $\dot{m}_{\text{gro}}$ varied in a contrary trend to $v_a$. $\dot{m}_{\text{oxid}}$ increased slowly when $v_a$ increased, and then it rapidly decreased when $v_a$ decreased. Figure 12 shows the corresponding experimental observations and the numerical distributions of $T$, $Y_{\text{OH}}$, $Y_{\text{soot}}$, $M_{\text{gro}}$, and $M_{\text{oxid}}$ at $t = 0$, 0.002, 0.004, 0.006, and 0.008 s. With the increase of $v_a$ (a1–
was extremely large, as shown in Figure 13.

Concentrated at the bottom of the flame, and the soot region also increased (b4). Meanwhile, since air was pushed out from glass tubes, and the maximum temperature increased (b4). The background was illuminated by an LED lamp.

The relative times were (a) 0 s, (b) 0.005 s, (c) 0.01 s, and (d) 0.015 s. The background was illuminated by an LED lamp.

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The distortion appears in the speakers at f < 50 Hz, the soot suppression at lower frequencies will be tested after solving this problem in the future. For numerical simulation, the detailed soot model is under consideration.

The experimental apparatus is shown in Figure 14, which is similar to that used by Saito et al.\textsuperscript{43} Two sine waves with the same frequency and similar amplitudes generated by a multifunction data acquisition device (NI USB-6356) were applied to two speakers after amplification. The amplitude of one sine wave was slightly adjusted to improve the symmetry of the flame. Several amplitudes under three frequencies (f = 50, 100, and 150 Hz) of the sine wave were tested. The speakers were connected to two quartz glass tubes, which were 30 mm in i.d., 34 mm in o.d., and 20 mm in distance. The acetylene was ejected from a stainless steel pipe with 1 mm in i.d. and 3 mm in o.d. The top of the steel pipe was tangent with the bottom of the glass tubes. The flow rate of acetylene was controlled by a mass flow controller (Alicat MC-SSLPM-D) at Q = 100 mL/min. The room temperature was around 27 °C. The measurement of the soot rate was the same as that in ref 43. The soot was collected by glass fiber filters and weighed by an electronic balance. The sampling time was 1 min under small acoustic oscillations, and a longer time was used under large acoustic oscillations. A high-speed camera (Olympus i-speed) was used to capture the flame. In order to catch the sight of soot, a white board was put behind the flame, and the board was illuminated by an LED lamp. These observations with a bright background are shown in Section 2.2. The sound pressure level (SPL) at the position of flames was measured by a sound level meter.

3. CONCLUSIONS AND FUTURE WORK

The flame structure and soot suppression under acoustic oscillated combustion were investigated combining numerical and experimental studies. For the three tested frequencies, the experimental soot rate almost decreased linearly with A, when A > 0.5 m/s, and the soot suppression under the same A decreased with f. The flame structure was well predicted by the finite-rate chemistry model except that the cross-sectional diameter was over-predicted. The soot rate predicted by the modified Moss-Brookes model decreased with A, but not linearly. The soot rate was over-predicted at f = 50 Hz, while it was first underestimated and then over-predicted at f = 100 and 150 Hz. We think that the main reason for the discrepancy from the experimental results lies in the inaccuracy of the semi-empirical soot model. With the increase of A, the consumption of O\textsubscript{2} increased obviously and the proportion of the soot oxidized by O\textsubscript{2} also increased. The decrease of the soot rate was mainly caused by the decrease of the surface growth rate when the air was pushed toward the flame after which the pinch-off of the flame might occur.

4. EXPERIMENTAL SECTION

The soot was collected by glass tubes, and the other region moved up (d1–d3) and e1–e3). Then, B\textsubscript{f} began to decrease since air was pushed out from glass tubes, and the maximum temperature increased (b4). Meanwhile, M\textsubscript{gro} moved down and concentrated at the bottom of the flame, and the other region of the flame was dominated by oxidation (e4). Finally, with the decrease of v\textsubscript{f}, the oxidation decreased (e5) and the flame began to pinch off. Thus, the pinch-off occurred when the air was sucked into tubes. At t = 0.008 s, B\textsubscript{f} was small (b5), but the flame width seen from the direction along with the glass tube was extremely large, as shown in Figure 13.

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5. NUMERICAL SECTION

5.1. Numerical Method. The numerical simulations were performed via the commercial software ANSYS Fluent using the finite-rate combustion model and modified Moss-Brookes soot model. The radiation was predicted by the discrete ordinates method. The soot–radiation interaction was considered, and the absorption coefficients of the gases were predicted by the weighted-sum-of-gray-gases model (WSGGM).\textsuperscript{49} The DES model based on the SST k–ω with
A low Reynolds number correction was used, and the DDES shielding function was applied.

5.1.1. Combustion Model. The finite-rate detailed chemistry model only computes for the Arrhenius rate without the explicit account of turbulent fluctuations on the source-term calculations. This approach is applicable for both laminar flows and turbulent flows using complex chemistry. The in situ adaptive tabulation (ISAT) was used to accelerate the computations of detailed chemistry. The conservation equations of the mass fraction of each species ($Y_i$) over the $N_R$ reactions are as follows:

$$\frac{\partial}{\partial t} (\rho Y_i) + \nabla \cdot (\rho \mathbf{v} Y_i) = -\nabla \cdot \mathbf{J}_i + \sum_{r=1}^{N_R} \dot{R}_{i,r}$$

where $\mathbf{J}_i$ is given by:

$$\mathbf{J}_i = -\left(\rho D_{i,m} + \frac{\mu_i}{S_C}\right) \nabla Y_i$$

with $\mu_i$ being the turbulent viscosity, and $S_C$ is the turbulent Schmidt number (set to 0.7). The thermal diffusion was neglected.

The chemical reaction was described by a GRI 3.0 reduced mechanism which involved 50 species and 309 reactions. The specific heat capacity of every gas species was defined by two fifth-order polynomials in temperature ($T$), while the molecular viscosity, thermal conductivity, and diffusion coefficient were defined using the kinetic theory.

5.1.2. Soot Model. The soot formation was predicted by the Moss-Brookes model with the modification in that the effect of soot formation on the chemical species was considered. The soot was supposed to incept and grow from $\text{C}_2\text{H}_2$. The reaction steps are as follows:

- Inception: $\text{C}_2\text{H}_2 \rightarrow 2\text{C}(s) + \text{H}_2$
- Coagulation: $n\text{C}(s) \rightarrow \text{C}_n(s)$
- Surface growth: $\text{C}_2\text{H}_2 + n\text{C}(s) \rightarrow (n + 2)\text{C}(s) + \text{H}_2$
- Oxidation by OH: $\text{C}(s) + \text{OH} \rightarrow \text{CO} + \text{H}$
- Oxidation by $\text{O}_2$: $\text{C}(s) + 0.5\text{O}_2 \rightarrow \text{CO}$

The transport equations of soot number concentration and soot mass fraction were solved. The production rates of the soot number density ($N$) due to inception and coagulation are given by:

$$\frac{dN}{dt} = c_1 N_A \frac{\rho Y_{\text{C}_2\text{H}_2}}{W_{\text{C}_2\text{H}_2}} e^{-21100/T} - \left( \frac{24R}{\rho_{\text{soot}} N_A} \right)^{1/2} \left( \frac{6}{\pi \rho_{\text{soot}}} \right)^{1/6} T^{1/2}$$

and

$$M_{\text{gro}} = c_2 \frac{\rho Y_{\text{C}_2\text{H}_2} e^{-12100/T(\pi N)^{1/3}}}{W_{\text{C}_2\text{H}_2}} \left( \frac{6M}{\rho_{\text{soot}}} \right)^{2/3}$$

where $c_1 = 54 \text{ s}^{-1}$, $N_A$ is Avogadro’s number, $Y$ is the mass fraction, $R$ is the universal gas constant, $\rho_{\text{soot}} = 2000 \text{ kg/m}^3$, and $M$ is the soot mass density. The mass of the incipient soot particle was set to be 1200 kg/kmol (consisting of 100 carbon atoms). The production rate of $M$ due to the surface growth was calculated by:

$$M_{\text{gro}} = c_2 \frac{\rho Y_{\text{C}_2\text{H}_2} e^{-12100/T(\pi N)^{1/3}}}{W_{\text{C}_2\text{H}_2}} \left( \frac{6M}{\rho_{\text{soot}}} \right)^{2/3}$$

where $c_2$ is the surface growth rate scaling factor, while that due to the oxidation by OH and $\text{O}_2$ was calculated by...
Figure 12. (a) Experimental observation and the numerical distributions of (b) $T$, (c) $Y_{OH}$, (d) $Y_{soot}$, and (e) $\dot{M}_{gro}$ and $\dot{M}_{oxid}$ at $f = 100$ Hz and $A_v = 1.6$ m/s. The numbers 1 to 5 denote $t = 0, 0.002, 0.004, 0.006$, and 0.008 s.
where \( \eta = 0.13 \) is the collision efficiency, \( c_4 = 8903.51 \) (kg·m)/(kmol·K\(^{0.5}\)·s), and \( Y_{O_2} \) is the mass fraction of \( O_2 \). The default value of \( c_4 = 9000.6 \) (kg·m)/(kmol·s), and it was adjusted to match the numerical soot rates without acoustic oscillations (\( \dot{m}_{soot} \)) with those of experiments. Finally, it was set to be 4000 (kg·m)/(kmol·s), which led to good agreement with the three flow rates of acetylene (see Figure 15).

The effects of soot inception, growth, and oxidation on the chemical species were added as source terms. For the surface growth, the production rates of \( C_2H_2 \) and \( H_2 \) were proportional to the surface growth rate in eq 9:

\[
\dot{M}_{C_2H_2} = c_{4\text{surf}} \eta \frac{\rho_{C_2H_2}}{\rho_{soot}} \sqrt{T} (\pi N)^{1/3} \left( \frac{6M_{C_2H_2}}{\rho_{soot}} \right)^{2/3} - c_{4\text{surf}} \frac{\rho_{C_2H_2}}{\rho_{soot}} \exp^{-\frac{19778}{T} (\pi N)^{1/3} \left( \frac{6M_{C_2H_2}}{\rho_{soot}} \right)^{2/3}}
\]

5.2. Computational Domain and Boundary Conditions. Figure 16a shows the computational domain and (b) the enlargement of the mesh at one symmetry plane. The red line denotes the vertical boundary of the hexahedral elements, and the mass flow rate of \( O_2 \) across this face was monitored.

The size of the rectangle domain was 50 mm \( \times \) 50 mm \( \times \) 250 mm, and the length of the glass tube was 200 mm. The gravity acceleration is 9.8 m/s\(^2\). Since the symmetry of the flame was well controlled during experiments, two symmetry planes were applied and only a quarter of the flame was simulated. The steel pipe inside the computational domain was 50 mm in height, and the heat transfer between the gases and the steel pipe was considered. The temperature at the bottom of the steel pipe was fixed to 320 K, which was from the experimental measurements. We tried two ways to model the speakers. One was using a wall moving at the speeds of sine functions by adopting the dynamic mesh technique; the other was using a velocity inlet of air with the speeds of sine functions. According to our simulations, these two ways led to almost the same results. In this study, the velocity inlet was adopted since it is much easier to implement. The velocity is expressed as

\[
\dot{V} = A_s \sin(2\pi t)
\]

where \( A_s \) is the amplitude of the velocity. The mass flow rate with a turbulent intensity of 0 was specified at the fuel inlet. The no-slip boundary condition was employed to all walls, and atmospheric pressure was employed to the rest of the boundaries of the computational domain. The time-step size was set to 10 \( \mu \)s after its independence verification.

Figure 16b shows the enlargement of the mesh at one symmetry plane. The total element number was 0.44 million, and special contractions of the mesh were applied around the flame. Hexahedral elements were employed inside the domain of the steel pipe and also the domain around the flame, while
tetrahedral and tri-prism elements were employed in the other regions. The consumption rate of O₂ (ṁO₂) inside the domain with hexahedral elements, together with the mass flow rate of O₂ (QṁO₂) across the vertical boundary of this domain (marked in red in Figure 16b), was monitored.

5.3. Comparisons between Experiments and Simulations. As mentioned above, the velocities in sine functions were adopted to model the speaker. In order to compare the numerical and experimental results, the experimental amplitudes of the velocity inside the glass tube were obtained as described below. First, the pressure fluctuations at the exit of the glass tube under experimental operating conditions were measured by a dynamic pressure sensor. Those pressures can be well fitted by sine functions as shown in Figure 17. After fittings, the amplitudes of the pressure fluctuation at the exit of the glass tube were obtained. Then, this measurement of pressure was simulated using a 2D axisymmetric mesh as shown in Figure 18. The left dashed box represents the glass tube, and the right dashed box represents the pressure sensor. The velocity in the sine function was employed to the left inlet, and the pressure at the exit of the glass tube was monitored and fitted by sine functions. It was found that the amplitude of the monitoring pressure almost increased linearly with the amplitude of the inlet velocity. Combining the experimental amplitudes of the pressure at the exit of the glass tube, the amplitudes of the velocity inside the glass tube under experimental operating conditions were deduced. In addition, the pressure sensor should be much smaller than the glass tube so as not to influence the vibration amplitude of the speaker.

In addition, the temperature above the flame was measured by an S-type thermocouple with 0.08 mm in the thermometer diameter to validate the simulations. When the thermocouple is put above the flame, it would be quickly covered with soot. Thus, the measurements were conducted to the flame without soot (QcH₂ = 30 mL/min). The measured temperatures at 2 and 3 cm right above the steel pipe were 929 and 727 °C, respectively. The true temperatures were higher due to the heat conduction along the thermocouple. The corresponding temperatures predicted by the numerical simulation were 998 and 766 °C, which were 7.4 and 5.4% higher than the measured temperatures, respectively, and the true errors were smaller.

ASSOCIATED CONTENT
Supporting Information
The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.0c03107.

Experimental observation at f = 50 Hz and Av = 0.48 m/s (AVI)
Experimental observation at f = 50 Hz and Av = 0.95 m/s (AVI)
Experimental observation at f = 100 Hz and Av = 0.44 m/s (AVI)
Experimental observation at f = 100 Hz and Av = 1 m/s (AVI)
Experimental observation at f = 150 Hz and Av = 1.4 m/s (AVI)
Experimental observation at f = 150 Hz and Av = 2.2 m/s (AVI)

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Notes
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