

# THE STUDY OF DROPLET ARRAY COMBUSTION ON TEXUS-46 – PRELIMINARY SCIENTIFIC RESULTS OF THE NITROGEN OXIDE PRODUCTION

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## ABSTRACT

Lean, partially premixed, swirl-stabilized spray flames are discussed for novel combustion concepts to allow a further abatement of the nitrogen oxides (NOx) emissions. The relevant knowledge for those concepts is based on combustion research of manageable and observable droplet regimes.

This paper initially presents the experiment setup for the combustion of a linear n-decane droplet array under microgravity conditions. After pointing out all relevant aspects of the associated exhaust gas sampling and analysis processes, the final focus is set on the results of the exhaust gas production. A wide range of the degree of droplet pre-vaporization is investigated. The PHOENIX experiment (“Investigation of Partial Pre-vaporization Effects in High Temperature on Evolution of Droplet Array Combustion and Nitrogen Oxides Formation”) on the TEXUS-46 sounding rocket mission was conducted against this background. Three successful combustion runs could be performed during this mission. Scientific quality and consistency of the results are high. The results on CO<sub>2</sub> and CO are in line with combustion theory, and their absolute values are of the correct order of magnitude but not yet corrected for secondary effects. The NOx emissions decrease with an increase of pre-vaporization rate  $\Psi$ . However, a straightforward portability of the derived NOx production characteristics to technical applications needs to be assessed carefully on the basis of the prevailing droplet burning regime.

## 1. INTRODUCTION

In times of depleting resources, processes involving the combustion of fossil fuels have to be highly efficient. Often, a gain in efficiency is linked to a higher combustion temperature, as the efficiency of a thermodynamic process rises with the increase of its temperature differences. Apart from that, a higher

combustion temperature corresponds to a higher fuel burnout, and hence reduced carbon monoxide emissions. On the downside of this increase in combustion temperature is a higher production rate of thermal nitrogen oxide (NOx) [16]. Its formation rate, as described by the Zeldovich mechanism, is strongly related to temperature, starting at 1800 K and rising exponentially. NOx has a serious effect on the human health and on the environment. Via the formation of nitric acid (HNO<sub>3</sub>), nitrogen oxides can cause diseases of the human respiratory system. Nitrogen monoxide (NO) can attach to the hemoglobin within the human blood, and thus inhibit the absorption of oxygen. Furthermore, nitrous oxide (N<sub>2</sub>O) can cause acid rain and is an effective greenhouse gas [2, 10, 11, 16]. Consequently, nitrogen oxide emissions have become subject to stricter regulations. For these reasons, two conflicting goals have to be considered in the design of combustion applications, that is efficiency versus decreasing NOx emissions, both being directly coupled to the combustion temperature.

First experiments to study the effect of fuel pre-vaporization on NOx emissions were conducted in the early 1980s by Cooper [3]. Yet, because of technical difficulties such as flashback and auto-ignition, which may occur in premixed flames, other concepts have been preferred for the technical realization of low NOx emissions. However, as the limit of these concepts is being reached within the current and/or the upcoming development cycles, new impulses on alternative approaches, such as fuel pre-vaporization, are needed. Consequently, experiments are carried out to gain further insight into the fundamental processes of droplet and spray flames at Technische Universität München (TUM). The TEXUS-46 sounding rocket campaign was conducted, with the rocket launch on November, 22<sup>nd</sup>, 2009, having the “Japanese Combustion Module” (JCM) onboard. This facility was used for cooperative experiments on droplet array combustion of the Japan

Aerospace Exploration Agency (JAXA) and the European Space Agency (ESA) research teams, both working on the combustion properties of partially premixed droplets and sprays [4, 5, 8 - 13]. The experimental focus was set on the flame spread phenomena and the NOx production rates, both in relation to the degree of fuel vaporization.

The microgravity environment of the TEXUS facility provided ideal experiment conditions without the disturbing effects of natural convection. This allowed the study of the interacting phenomena of multi-phase flow, thermodynamics, and chemical kinetics. And this way the understanding of the physical and chemical processes related to droplet and spray combustion could be improved.

Recent work on the NOx generation of partially pre-vaporized sprays is summarized in the publication of Baessler et al. [2]. Fig. 1 displays representative results for the particular case of a global equivalence ratio of  $\phi = 0.7$ . The early model of Cooper is opposed to the model of Baessler et al. The ensuing discrepancy is apparent and substantial, and would be highly relevant in the design process of for instance aero-engines.

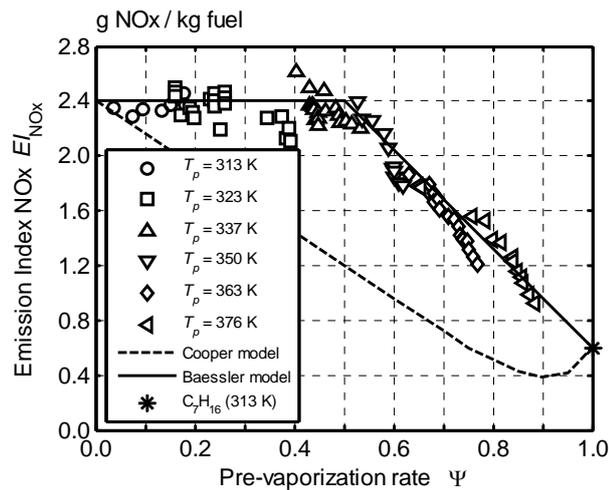


Figure 1. Influence of pre-vaporization  $\Psi$  on NOx emissions (according to [2]). The measurements are for a global equivalence ratio of  $\phi = 0.7$  and corrected to the reference temperature  $T_{ref} = 313$  K.

## 2. EXPERIMENT OVERVIEW

The investigated main experimental parameter on the reported TEXUS-46 mission was the “degree of fuel vaporization”  $\Psi$ , also denoted as the “pre-vaporization rate”. It is subject to the dimensionless droplet spacing  $S/d_0$ , with  $d_0$  being the initial droplet diameter, and has a direct impact on the local equivalence ratio  $\phi$  along the droplet array.

## 2.1. General Experiment Information

Fig. 2 shows a schematic of the investigated droplet configuration. The concept is based on a variable pre-vaporization time  $t_\psi$ . The low volatile fuel n-decane ( $C_{10}H_{22}$ ) is used due to its preference as a model fuel for kerosene and diesel. Five droplets are generated, partially vaporized, and finally burned. Droplet generation is performed at a temperature of 300 - 315 K, whereas pre-vaporization is enforced at an elevated temperature level of 500 K (+/-1 K). All combustion runs are conducted at a pressure of 1.0 bar (+/-20 mbar). Ignition is initiated by an electrically heated ignition wire [10, 11].

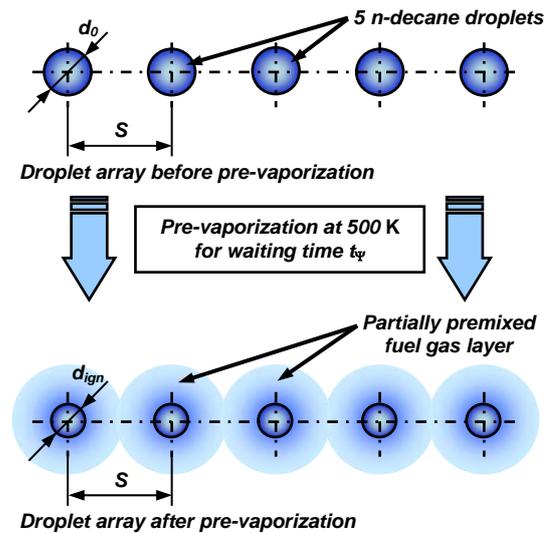


Figure 2. Schematic on fuel pre-vaporization of a linear droplet array.

Tab. 1 summarizes the parameter matrix of the originally scheduled experiments on TEXUS-46. In total four experiment runs were envisioned of which three could be performed successfully. Experiment run No. 4 got lost due to a timing/sequencing error, tripped by repeated countdown holds during the final steps of the TEXUS launch sequence.

Table 1. Nominal experiment parameters for droplet arrays on TEXUS-46. The droplet number is  $N = 5$ , and the non-dimensionless droplet spacing  $S = 18$  mm.

No.	$T_\psi$	$d_0$	$d_{ign}$	$S/d_{ign}$
1	18 s	1.5 mm	1.01 mm	17.8
2	10 s	1.5 mm	1.34 mm	13.4
3	5 s	1.5 mm	1.47 mm	12.2
4	15 s	1.5 mm	1.15 mm	15.7

The central unit of the JCM is the “Droplet Array Combustion Experiment Unit” (DCU). It is shown in Fig. 3. The DCU is built of six platforms with a total height of 810 mm and a baseplate diameter of 403 mm.

As a vacuum dome covers the DCU in order to keep it pressurized, the experiment platforms themselves measure 372 mm. Controlling devices and batteries are mounted outside of the pressurized dome, below the experiment deck. The modular design of the DCU facilitates adjustment and modification work, as for instance needed with the droplet generation setup and the “Exhaust Gas Sampling” (EGS) system.

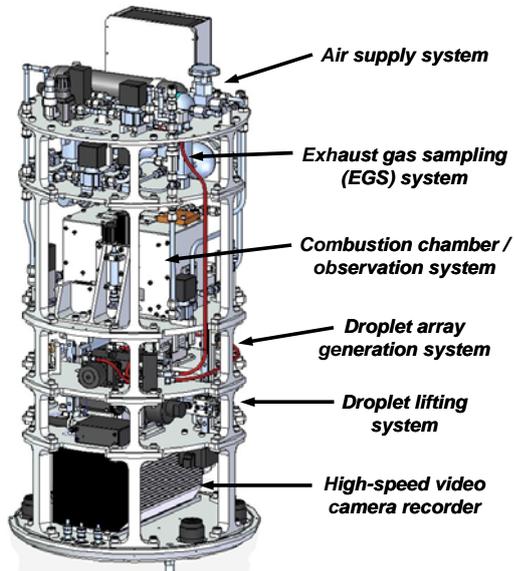


Figure 3. “Droplet Array Combustion Experiment Unit” (DCU), core section of the TEXUS experiment.

For each combustion run, the droplet array consisting of  $N = 5$  droplets was generated by the droplet array generation system on platform #3 of the DCU. The droplets were supplied through home-made glass tubes onto the droplet holder, and high-precision dosing was achieved by a servo motor controlled fuel pump. Each droplet was suspended at the intersection of a pair of X-shaped SiC fibers of  $14 \mu\text{m}$  thickness, which imparts lowest residual motion and highest sphericity [8]. The shutter on the bottom side of the combustion chamber was opened, and the droplet holder with the droplet array was elevated into the preheated combustion chamber by the droplet lifting system. Depending on the predefined degree of vaporization  $\Psi$  (cp. Tab. 1), an ignition wire ignited one end of the linear droplet array to initiate combustion. Flame spread occurred along the droplet array. Subject to the pre-vaporization rate, the combustion process was initially characterized more or less by a partially premixed flame, which was followed by a diffusion flame around the droplets until flame extinction. After flame extinction, the exhaust gas sampling was performed by the EGS system, using one evacuated gas sampling cylinder per combustion run. The samples were stored in these specially treated gas sampling cylinders until their succeeding analysis on ground.

Visual observation of the combustion process as well as temperature and pressure logging supported the scientific interpretation of the gas analysis. High efforts were put into a reliable hardware system as well as the development of the experiment procedures to ensure reproducible results of high quality. This included a thorough testing, precursor experiments, and various numerical studies.

## 2.2. Exhaust Gas Sampling and Analysis

An intrusive concept was chosen for the crucial exhaust gas sampling and analysis steps. It combines the sampling process during TEXUS flight with the successive gas analysis on ground. The related EGS system is installed on platform #5 of the DCU, in the close proximity to the combustion chamber (cp. Fig. 3) and is shown in detail in Fig. 4. The gas sampling cylinders were required to separately sample the emissions of each of the four scheduled combustion runs. All metal surfaces of the EGS system being directly exposed to the combustion products were coated with amorphous silicon to prevent negative surface effects on the gas composition and a distorting gas adsorption [1, 11, 14, 15]. Access for the evacuation of the gas sampling cylinders by a turbomolecular pump was realized through a late access port in the TEXUS module. In order to fulfill the scientific requirements, a minimum leakage rate of  $1 \times 10^{-5}$  (mbar l)/s with a remaining vacuum level  $< 1 \times 10^{-1}$  mbar inside of each sampling cylinder was postulated for the moment of the exhaust gas sampling. The achievable vacuum level before launch (at -90 min) was  $2 - 3 \times 10^{-5}$  mbar with an associated leakage rate of  $2 \times 10^{-6}$  (mbar l)/s. Thus, all requirements on the EGS system could be met from the preparatory side.

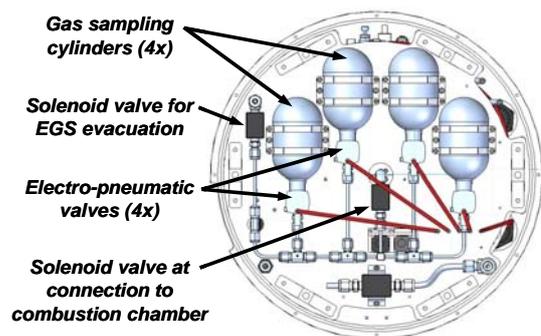


Figure 4. Exhaust gas sampling (EGS) platform with coated gas sampling cylinders and supply units.

The central issue of the sampling and analysis step was to obtain a representative gas sample from every single combustion run. Based on this general goal, a number of constructional and operational constraints could be derived. They are outlined in detail in the previous work of Moesl et al. [10, 11].

Furthermore, empirical tests in parabolic flight and drop tower were used to derive a correlation between flame extinction and the temperature readings of the combustion chamber. This correlation was used within a trigger algorithm to initiate the exhaust gas sampling. Fig. 5 illustrates front and side view of the combustion chamber during the gas sampling with a simplified model of the gas flow. Fresh air is entrained into the open combustion chamber because of the generated pressure drop and the instantaneous pressure compensation [10]. Since the linear flame propagation mode has an averaging effect on the overall combustion temperatures, the averaged production of thermal NO represents the NO quantities of a single droplet being a single link in an infinite droplet array.

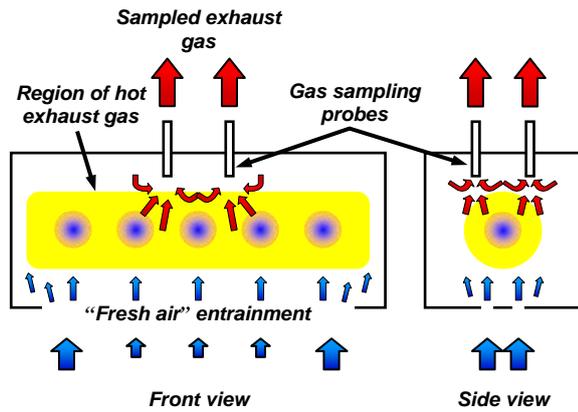


Figure 5. Sectional views of the combustion chamber during exhaust gas sampling.

The gas analysis itself was ground-based and carried out by FT-IR (Fourier Transform Infrared) spectroscopy with a NEXUS 470 unit of Thermo ELECTRON as well as by using NO/NO<sub>x</sub> chemiluminescence. For the latter a CLD 700 LEV ht unit of ECO PHYSICS was used.

Since the spectroscopic FT-IR method is ideally suited to capture the concentrations of a larger number of species, it was used to determine the major species including CO<sub>2</sub>, CO, and H<sub>2</sub>O. Oxides of nitrogen (NO, NO<sub>2</sub>, and N<sub>2</sub>O) were also determined by FT-IR as a back-up solution. However, the main NO<sub>x</sub> species (NO and NO<sub>2</sub>) were evaluated with the NO/NO<sub>x</sub> chemiluminescence analyzer, being the state of the art technology. Both analysis methods were coupled to become one unit with the chemiluminescence analyzer connected downstream of the FT-IR. After conditioning a TEXUS gas sample to the FT-IR operational parameters of  $T = 458$  K and  $p = 866$  mbar, a quasi-stationary analysis was performed keeping the sample in the 2 m optical path length cell. Subsequently, the chemiluminescence analysis was carried out with the sample being lost in consequence of the analysis process. As this fact necessitated an absolutely error free procedure, extensive calibration and test runs were conducted in preparation of the exhaust gas analysis.

### 3. EXPERIMENT RESULTS

The present experimental study was undertaken to resolve the issue of the effect of droplet pre-vaporization on the NO<sub>x</sub> generation in droplet arrays. The results may also help to derive tendencies for single droplet and spray combustion, if certain conditions are met regarding the prevailing combustion regime [6, 7]. With the TEXUS mission, a long-term microgravity possibility was provided offering sufficient  $\mu$ g time for four subsequent combustion runs (cf. to Tab. 1). Three out of these four experiments could be performed successfully.

Fig. 6 illustrates the carbon dioxide (CO<sub>2</sub>) concentrations as measured from the TEXUS flight samples. Pre-vaporization times were  $t_{\psi} = 5, 10,$  and  $18$  s. Four different analysis methods were enforced, as indicated in the legend of Fig. 6. Of course, values should coincide for each  $t_{\psi}$ , as they are determined from the very physical gas sample; but deviations within the choice of methods are small for  $t_{\psi} = \text{const}$ . This sound agreement demonstrates the quality and accurateness of the different FT-IR methods. Furthermore, Fig. 6 demonstrates an almost constant CO<sub>2</sub> production for different pre-vaporization times  $t_{\psi}$ . Since nominally identical fuel quantities are oxidized, consistent CO<sub>2</sub> concentrations have to be expected. The absolute values are of the correct order of magnitude, but the dilution effect as pointed out in Sec. 2.2 is not yet corrected for. Thus, Fig. 6 represents the raw values of the collected gas without secondary effects being accounted for. Major secondary effects are: fresh air entrainment and slightly different ambient temperatures  $T_{ign}$  at the moment of ignition.

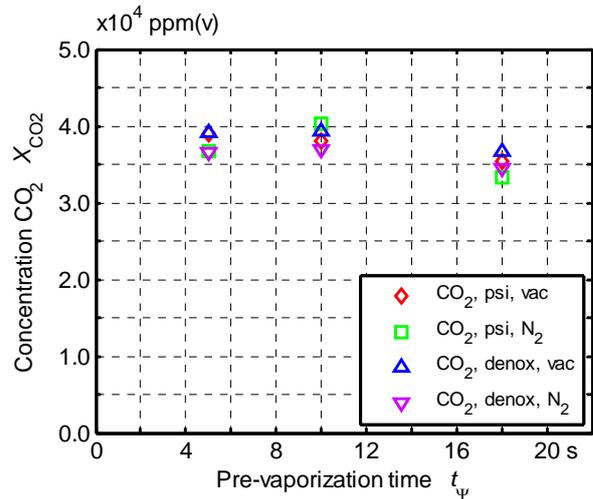


Figure 6. Carbon dioxide concentrations in ppm(v) (parts per million) of the TEXUS experiments analyzed by employing four different procedures.

Fig. 7 shows complementary data to Fig. 6 (CO<sub>2</sub>) for carbon monoxide (CO). For the low and intermediate

pre-vaporization times of  $t_\psi = 5$  and  $10$  s, the match of the four analysis methods is perfect; for the high pre-vaporization time of  $t_\psi = 18$  s, it is consistent and still very good. The exponential increase of the CO concentration with the degree of vaporization  $\Psi$  is due to the formation of a triple-flame with a rich premixed region. Its appearance could be confirmed by numerical studies, and it typically causes an excessively increasing CO production with an increase of  $\Psi$ .

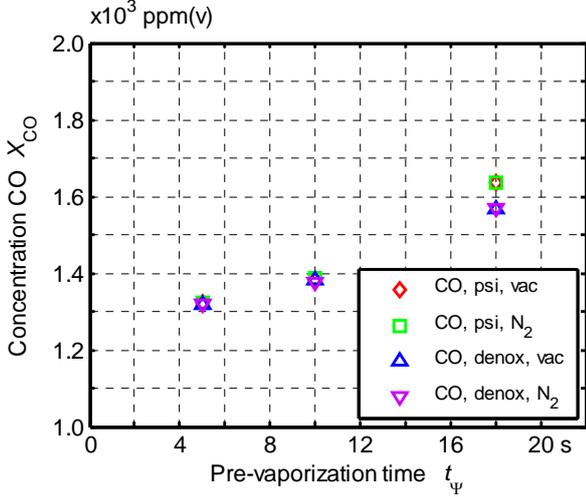


Figure 7. Carbon monoxide concentrations.

In contrast to the emissions of  $\text{CO}_2$  and CO, the nitrogen oxide emissions are expressed by the emission index  $EI_{\text{NO}_x}$ . It is defined by Eq. (1) and is the ratio of the weighted NOx masses and the fuel mass  $m_{\text{fuel}}$  with the unit  $\text{g}_{\text{NO}_x}/\text{kg}_{\text{fuel}}$ .

$$EI_{\text{NO}_x} = \frac{m_{\text{NO}} \frac{M_{\text{NO}_2}}{M_{\text{NO}}} + m_{\text{NO}_2} + m_{\text{N}_2\text{O}} \frac{M_{\text{NO}_2}}{M_{\text{N}_2\text{O}}}}{m_{\text{fuel}}} \quad (1)$$

In compliance with previous studies, and for instance Fig. 1, a decrease of  $EI_{\text{NO}_x}$  can be observed with an increase of  $t_\psi$  or  $\Psi$ . Fig. 8 clearly illustrates a decreasing, almost linear trend for the NOx emissions. Again, no secondary effects are accounted for here either. Nevertheless, the results lead to the conclusion that droplet pre-vaporization generally allows for a substantial NOx abatement. However, to be able to deploy this abatement potential in technical applications, the droplet burning regime needs to be assessed and adapted carefully.

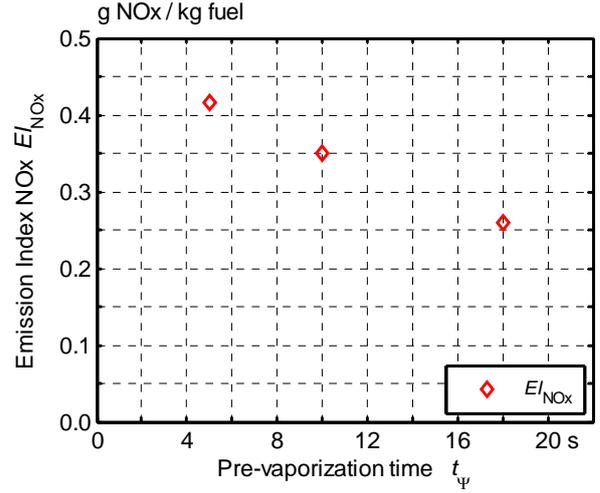


Figure 8. Emission index of nitrogen oxides  $EI_{\text{NO}_x}$  for the TEXUS experiments.

#### 4. CONCLUSIONS

Novel combustion concepts still offer a high potential for improvement towards an environmentally neutral combustion process and low exhaust gas emissions. Microgravity research can contribute to the development and market launch of those concepts. It allows the detailed observation of the most essential combustion phenomena without the disturbance of natural convection. The extended microgravity duration of a sounding rocket flight in particular allows for the investigation of the degree of droplet vaporization on the NOx emissions.

A linear array of five n-decane droplets was burned on the PHOENIX mission on TEXUS-46 (PHOENIX: “Investigation of Partial Pre-vaporization Effects in High Temperature on Evolution of Droplet Array Combustion and Nitrogen Oxides Formation”). The droplet array was suspended on SiC fibers and inserted into a preheated combustion chamber. One end of the array was ignited to initiate combustion. After flame extinction, exhaust gas was collected from the combustion chamber for three successfully conducted combustion runs. The exhaust gas analysis was ground-based and performed by FT-IR spectroscopy and NO/NO<sub>x</sub> chemiluminescence.

The scientific quality of the TEXUS experiments is very good, and it shows high consistency with precursor experiments from parabolic flight and drop tower. The results on  $\text{CO}_2$  and CO confirm quality and consistency of the different combustion runs regarding experiment performance and procedures as well as exhaust gas sampling and analysis. The measured values of  $\text{CO}_2$  are in line with combustion theory, and the absolute values are of the correct order of magnitude but not yet correct for the secondary effect of fresh air entrainment. The progressive formation of a triple-flame with an increase of the pre-vaporization rate  $\Psi$  is accompanied by an

increase of the CO production. NO<sub>x</sub> emissions decrease with an increase of  $\Psi$ . This general trend could be expected from combustion theory. However, the straightforward portability of the results at hand to technical applications needs to be assessed carefully on the basis of the prevailing droplet burning regime.

## 5. ACKNOWLEDGEMENTS

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