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Simultaneous optical diagnostic velocity and scalar field by molecular tagging technique

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Abstract

To understand the physical mechanisms that govern combustion based systems, detailed experimental data on parameters such as air/fuel ratio, temperature, and mean flow velocity are of interest. Simultaneous acquisition of these multiple parameters is often of interest and to date, generally requires the coupling of different diagnostics. In order to address these limitations, an optical flow visualization method using molecular phosphorescence has been implemented that allows to obtain simultaneously scalar and velocity measurements.

Introduction

Studies in the field of fluid dynamics are often complicated by the influence of numerous phenomena like chemical reactions and molecular non-equilibrium, which are present in a large range of practical cases. In case of diffusion flames, study of stabilization, leads by different mechanisms; requires the simultaneous information revealed from velocity and temperature distributions. This information could help us to improve our understanding about physics of complex aerothermochemical phenomena, which, in turn, will greatly improve the performance of devices or allows development and validation of models.

To obtain this information, the simultaneous velocity and temperature measurement in non-isothermal flows was conducted by using intrusive probes such as cold-wire sensors [1, 2]. To avoid the perturbation due to sensor introduction in flow, a large number of optical methods that are full-field, non-contact and non-invasive have been investigated. The advent of optical diagnostics such as Laser Doppler Velocimetry (LDV) and Laser Induced Fluorescence (LIF) has presented new opportunities for the non-intrusive simultaneous acquisition of velocity and temperature measurements [3, 4]. More recently, the advances in the development of whole-field flow diagnostic techniques such as Particle Image Velocimetry (PIV) and Planar Laser Induced Fluorescence (PLIF) have led to efforts to obtain simultaneous maps of velocity and temperature distributions by using a PIV-PLIF combined system [5-7]. Currently, Thermographic Particle Image Velocimetry in flames by using phosphorous particles to determined velocity and temperature was improved by Fond et al [8]. It should be noted that all the optical velocimetry techniques mentioned above use particle seeding, which measure the velocity of tracer particles, other than the velocity of working fluid directly. The velocity of the working fluid is deduced based on the assumption that the tracer particles move with the same local velocity as the working fluid. Some effects like inertia, thermophoresis forces or buoyancy forces acting on the tracer particles

should be carefully considered for each experiment in order to make a physically meaningful measurement of fluid velocity. Moreover, flow measurements determination in the near wall regions can be perturbed by laser reflection, or low particle density. In confined flows, or in two phase flow, the tracer particles may coat the windows, or the interface leading, to limited test times or even window abrasion or change flow physics completely due to the existence of the tracer particles [9].

For the temperature measurement with phosphorous particles [8] or PIV/LIF [7], the thermal responses of the particle tracer requires additional considerations.

In order to get around this limitation and to eliminate measurement artifacts, an optical technique call Molecular Tagging Velocimetry (MTV) using molecules as diagnostic tracers can be developed. This technique relies on the properties of tracer molecules, which have relatively long lifetime luminescence once excited by a laser beam with an appropriate wavelength. Typically, a laser is used to tag these molecules along a line[10-13] or a grid [14]. Molecules phosphorescence is then detected at two successive times. The analysis of the tagged line or grid displacement and deformation allows the determination of the velocity field and the decrease of signal intensity permit to obtain local scalar information. As this little-intrusive technique uses molecular tracers, instead of particles, it presents several advantages compared with PIV (homogeneous repartition of tracer, limitation of adhesion at the walls...). The efficiency of MTV has been demonstrated for liquid flows [11-13] and its application in gaseous flow with molecular seeding has been evidenced in flow configurations presented in [15-17] In gaseous flow two approaches of tagging processes have been proposed that specify the seeding molecule and visualization technique. The first type uses laser light to generate different species or fragments from seeded molecules to generate the tagged molecules, and another laser light is used to visualize the product. Several examples of this type have been demonstrate using, for instance the photodissociation of NO₂ to NO[18, 19], H₂O to

OH, or O₂ to O₃ [17]. This technique permits to make measurement un low-velocity flowfields due to long lifetime of tracerbut need two lasers. The other approach relies on phosphorescence from the seeded molecules. Seeded molecules with relative long luminescence lifetimes, such as acetone [20, 21] and biacetyl [16, 22], are excited by laser light and are traced and captured within the luminescence lifetimes. This technique needs only one laser, but a limitation is that the time interval between any two images of luminescence is short (from a few micro seconds to a few milliseconds). To overcome these limitations, several molecules were identified by literature and analyze in order to extend time intervals by long phosphorescence lifetime of molecule.

The objective of this study is to develop a simultaneously quantitative measurement of temperature and velocimetry using Molecular Tagging Velocimetry and Thermometry (MTV&T) technique. In a first time, the dependence of the phosphorescence emission of different molecular tracer and with parameters such as temperature is characterized. After, application of velocimetry measurement is demonstrated for a forced jet flow, configuration close to a jet flame.

Molecular Tagging Velocimetry and Thermometry (MTV&T) technique

MTV&T is an extension of the Molecular Tagging Velocimetry (MTV). MTV technique is devoted for flow velocity only, the MTV&T technique described in the present study can also map fluid temperature distributions simultaneously in addition to whole-field velocity measurements. In the next sections, an overview of MTV is given with more details on Molecular Tagging Thermometry (MTT) and properties of the phosphorescent tracer.

a. Molecular tagging velocimetry principle

Once tracer is added to the gaseous flow, it can then be "tagged" by a laser pulse. The region of the initially excited flow will then phosphoresce for a predefined lifetime and will deform as a function of the velocity field.

By comparing initial and deformed image marking after a time Δt , it is possible to go up to the displacement field and then to the velocity field of flow. A unidimensional marking of the flow (in the form of a line) thus makes it possible to determine the component of the velocity field of the fluid normal to this line. By increasing the complexity of the marking by creating a grid (2D), it is then possible to determine two components of the velocity field.

To deduce the velocity field, two successive visualizations of the flow are recorded, one immediately after the laser pulse, another after a delay Δt (which obviously must be shorter than the tracer phosphorescence life). These images can be

processed by an image correlation algorithm based on the classical algorithms used for PIV. The images are then divided into interrogation windows and the calculation of the correlation coefficient between the corresponding interrogation windows on the two successive images makes it possible to access the average displacement of the window and therefore the velocity fields [12]

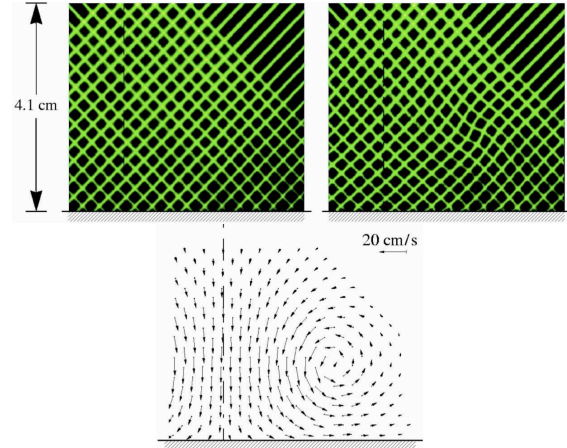


Fig. 1: MTV grid at two different times and the result velocity field [12]

b. Molecular tagging Thermometry principle

The intensity of phosphorescence process decays exponentially and can express by the relation below:

$$S_{p,t} = S_{p_0} e^{-t/\tau} \quad (1)$$

with τ the lifetime corresponding at time characteristic of the phosphorescence decay. This expression can be integrated to obtain total phosphorescence:

$$S_p = S_{p_0} \cdot \tau \quad (2)$$

The phosphorescence signal S_p , is given by the following equation and depends on different thermodynamic parameters:

$$S_p = \frac{E}{hc/\lambda} \cdot \eta_{opt} \cdot dV_c \cdot C \cdot \sigma(\lambda, T) \cdot \Phi_p(\lambda, T) \quad (3)$$

where E is the laser fluence (J/cm²), (hc/λ) the energy (J) of a photon at the excitation wavelength λ , η_{opt} the overall efficiency of the collection optics (-), dV_c the collection volume (cm³), C molecular density (molecule/cm³), σ the molecular absorption cross section of the tracer (cm²/molecule), Φ the fluorescence quantum yield (-), k the Boltzmann constant (J/K) and T the temperature (K).

Using Equation (2) and (3) we obtain:

$$S_{p_0} = \frac{E}{hc/\lambda} \cdot \eta_{opt} \cdot dV_c \cdot C \cdot \sigma(\lambda, T) \cdot \Phi_p(\lambda, T) \cdot \tau \quad (4)$$

To determine temperature by capturing signal with a gated intensified CCD camera, the first image is captured at the time delay $t_{0,a}$ after the laser pulse with the interrogation gate period of Δt . The second

phosphorescence images were captured at the time delay of $t_{0,b}$ after the laser excitation pulse with the same interrogation gate period.

During this time the phosphorescence signal S_p collected is given by:

$$S_{p,\Delta t} = \int_{t_0}^{t_0+\Delta t} S_{p,0} e^{-t/\tau} dt$$

$$= S_{p,0} \cdot \tau \cdot \left(1 - e^{-\Delta t/\tau}\right) \cdot e^{-t_0/\tau}$$

(5)

Finally, using equations (4) and (5) it obtains:

$$S_{p,\Delta t} = \frac{E}{hc/\lambda} \cdot \eta_{opt} \cdot dV_c \cdot C \cdot \sigma(\lambda, T) \cdot \phi_p(\lambda, T) \cdot \left(1 - e^{-\Delta t/\tau}\right) \cdot e^{-t_0/\tau}$$

(6)

With equal tracer concentration and laser energy in each point, the ratio of phosphorescence signals for both time delay depends only on the temperature:

$$\frac{S_{p,\Delta t,t_{0,a}}}{S_{p,\Delta t,t_{0,b}}} = \frac{\left(1 - e^{-\Delta t/\tau}\right) \cdot e^{-t_{0,a}/\tau}}{\left(1 - e^{-\Delta t/\tau}\right) \cdot e^{-t_{0,b}/\tau}} = e^{\frac{t_{0,b}-t_{0,a}}{\tau}} = f(T)$$

(7)

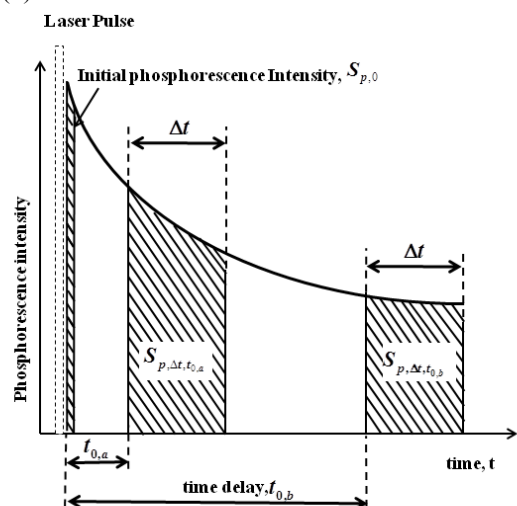


Fig. 2: Scheme of the timing offset for the MTV&T measurement

Thus, the phosphorescence signal can theoretically be used to measure the temperature.

c. Phosphorescence lifetime molecular tracers measurement

The most important parameters for selecting suitable tracers for MTV & T gas phase include:

- the absorption cross-section at convenient laser wavelengths, and scalar dependence;
- absolute phosphorescence quantum efficiency and its variation as a function of the conditions;
- Phosphorescence lifetime as a function of conditions;
- Chemical stability.

According to these parameters, two ketones molecules (biacetyl $C_4H_6O_2$, acetone C_3H_6O)[23] and an aromatic ketone (benzophenone

$C_{13}H_{10}O$)[24], have been identified. The aromatic ketone, like benzophenone, phosphoresces in the (n, π^*) triplet state as acetone or biacetyl. The phosphorescence lifetimes originating from the (n, π^*) state are usually in the millisecond range, whereas the phosphorescence lifetimes from the (π, π^*) state (case of aromatic compounds and derivatives) are 10-1000 times as long, but this state is observed in free oxygen and other triplet quenchers environment, therefore rarely observed. For these three molecules display a phosphorescence lifetime at least equal 1 ms. For them the phosphorescence lifetime are firstly measured at ambient temperature (291 K) and 0.1 MPa, in nitrogen, at an excitation wavelengths of 266, in the set-up presented below (Cf. Fig. 3).

Experimental set-up

Two different experimental measurements MTV&T experiments are conducted in an atmospheric pressure and variable temperature facility (Cf. Fig. 3.) The maximum studied temperature is 298 K at atmospheric pressure. A heating system by electrical resistances allows obtaining a uniform temperature inside the bubbling vessel. The optical system is composed of an Nd:YAG laser (266 nm – 500 mW), which excites the vaporized tracer and bath gas mixture at the center of the measurement volume. The laser beam passes through set of lens to generate sheet. The resulting laser sheet overcome a beam blocker and is split by a 50:50 beam splitter; crossing of the two resulting sheets to generate the grid pattern. The phosphorescence signal collected along the axis perpendicular to the incident light grid through quartz windows is analyzed by a photomultiplier (Hamamatsu H10721P-110) and is also collected by a 12-bit (1280 × 1024 pixels) gated intensified CCD camera (PCO) to conduct image recording. The laser and the camera are synchronized using a digital delay generator (SRS DG645), which controlled the timing of the laser sheet illumination and the CCD camera data acquisition. The phosphorescence images captured by the CCD camera are subsequently transferred to a host computer for analysis. This experimental setup is developed to follow the phosphorescence signal and to evaluate if the photophysical behavior of the tracer is adapted for an optimal application of Molecular Tagging Velocimetry diagnostic.

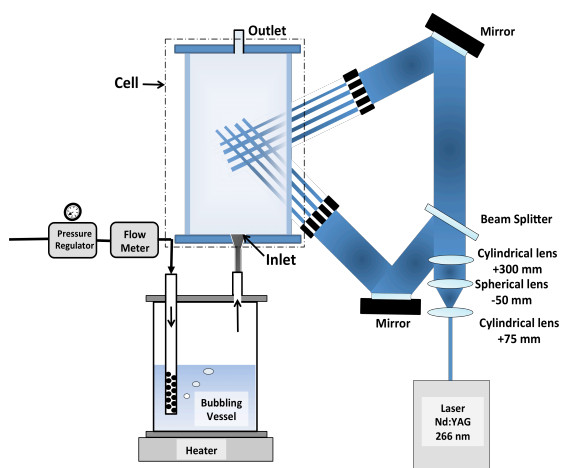


Fig. 3: Experiment facility with optical system for calibration measurement

In order to demonstrate the validity of the velocimetry measurement technique by MTV, the previous experimental set-up has been modified by the insertion of a steel tube in the inlet of the cell, as shown in the following figure:

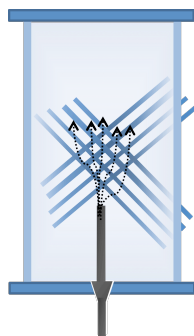


Fig. 4: Experiment setup

The flow system, illustrated in Fig. 4, operates in a blow-down mode using a high-pressure nitrogen tank. Appropriate pressure regulators and valves are used to adjust the flow velocity, whose value is constantly measured by a flow meter. The tracer is introduced by bubbling: the carrier gas into a container with liquid tracer. The resulting stream flows into the test section. In this case the flow exiting through the nozzle simulates the intake flow in a combustion chamber, in the form of an annular jet. The nozzle internal diameter is of 4 mm.

Results and discussion

a. Phosphorescence lifetime

The phosphorescence decay curves of biacetyl, acetone and benzophenone vapour in nitrogen are presented in figure below. Benzophenone is a solid component and then is dissolved in n-heptane (C_7H_{16}) (non-polar and aprotic solvent), the obtained solutions show a satisfactory dissolution, and permits to vaporize it.

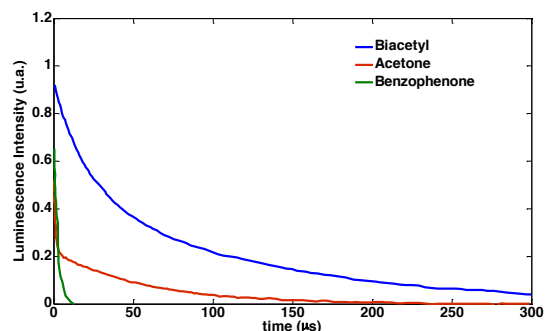


Fig. 5: Phosphorescence intensity decrease for biacetyl, acetone and benzophenone at 291 K

The tested phosphorescent tracers exhibit very fast phosphorescence decrease for the excitation wavelength of 266 nm, except biacetyl, which has a phosphorescence lifetime of 60 μs at ambient temperature.

In case of acetone and benzophenone the short phosphorescence lifetime may be attributable to the experimental conditions like preparation (bubbling), temperature, excitation wavelength, laser energy (photolyse) or impurity in solvent for benzophenone, this two molecule are more "sensitive" than biacetyl. Moreover, phosphorescence follows after the laser excitation, acetone undergoes an intersystem crossing from the first singlet state, S_1 , to the first triplet state, T_1 . There are many vibrational modes that can be accessed in the T_1 state, and each would have a different lifetime.

Finally, biacetyl is then chosen as tracer for its spectroscopic properties, satisfactory phosphorescence intensity signals, phosphorescence lifetime and for its chemical stability.

In order to obtain quantitative relation between the temperature and the phosphorescence lifetime of biacetyl a calibration is conducted. Due to limitation of using experimental set-up, the experiment is conducted at a temperature range of 291 to 298 K. The figure below depicts the measured phosphorescence lifetime for four different temperatures. It can be seen that the phosphorescence intensity decay curves can be approximated by single-exponential curves, as expected theoretically.

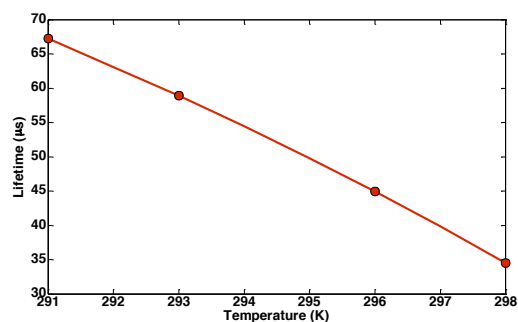


Fig. 6: Phosphorescence lifetime of biacetyl for different temperature

b. Application to a forced jet flow

In order to demonstrate the feasibility of MTV&T diagnostic, a forced jet flow in stagnant ambient fluid is studied.

Fig. 7.a displays a 20 mm x 20 mm field of view in nitrogen, biacetyl flow tagged by a laser grid. The reference image is the average of 50 images acquired 200 ns after laser firing. The maximum flow speed in the annular jet entering the cylinder is about $13,3 \text{ m}\cdot\text{s}^{-1}$. Fig. 7.b is an example of the later image of the tagged regions after a delay. In this experiment the gas flow rate is $10,0 \text{ l}\cdot\text{min}^{-1}$, corresponding at a velocity of $13,3 \text{ m}\cdot\text{s}^{-1}$ and a turbulent Reynolds number close to 4000.

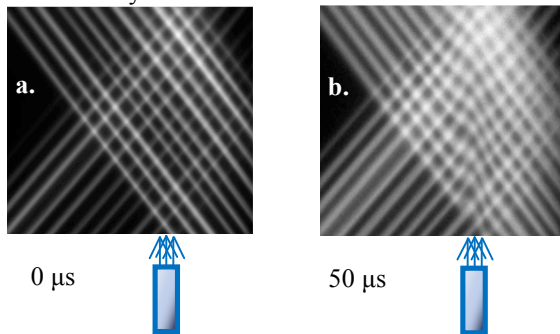


Fig. 7: a. phosphorescence image of the tagged molecules at 200 ns after the laser pulse b. phosphorescence image of the same tagged molecule acquired 50 μs later

To determine velocity, an image processing procedures are first applied including background subtraction, rotation and image normalisation, permitting to determine node position (Cf.

Fig. 8).

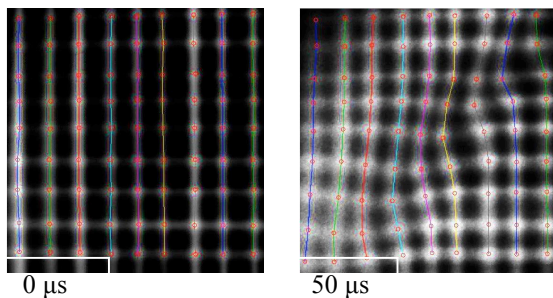


Fig. 8: Node position determination

The nodes coordinates (abscissa and ordinate) of the mass centers of the two images are determined and the dx and dy displacement vector components are calculated, using the pixel rate and the Δt from two images measurements. The velocity field shows the instantaneous velocity distribution determined from the image pair.

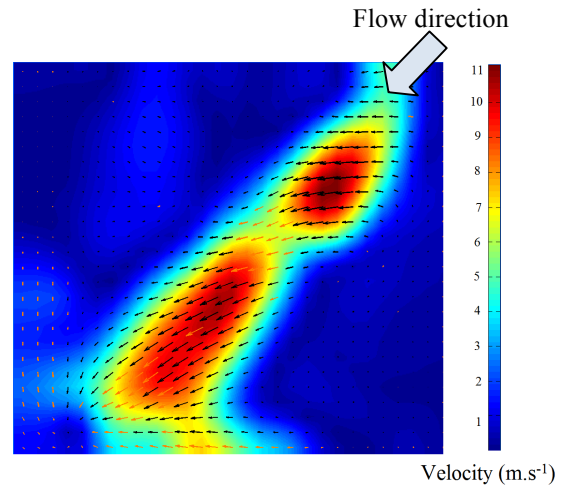


Fig. 9: velocity field derived from the image pair

The image pair in Fig. 7 also allows the determination of the temperature distribution simultaneous with the velocity field already described. Consistent with the correlation method used for the measurement of the displacement of tagged regions, the interrogation regions close to nodes were chosen in the first phosphorescence image to provide the phosphorescence intensity $S_{p,\Delta t,t_0,a}$ within those regions.

The tags molecules in each region convects to a new region in the second phosphorescence image according to their Lagrangian displacement. This displacement field is the basis of measuring the velocity field with MTV and is already available from Fig. 9. Therefore, for each interrogation window in the first phosphorescence image, the position of the corresponding window in the second phosphorescence image is determined, and the corresponding phosphorescence intensity $S_{p,\Delta t,t_0,b}$ within each region. The phosphorescence intensities, $S_{p,\Delta t,t_0,a}$ and $S_{p,\Delta t,t_0,b}$, are determined for the corresponding regions in the two phosphorescence images. The phosphorescence lifetime is calculated based on (7), resulting in the measurement of temperature according to the lifetime-versus-temperature calibration curve in Fig. 6. The simultaneous temperature field derived from the phosphorescence image pair, shown in Fig. 10, illustrates the instantaneous temperature distribution in the flow. Temperature determined at nodes is in agreement with experimental condition.

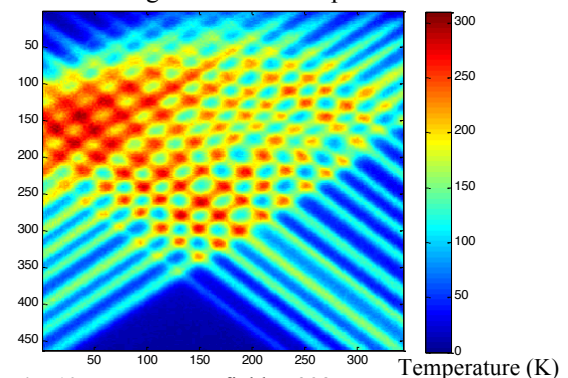


Fig. 10 :Temperature field at 298K

First promising experimental results have been obtained. The velocity profiles and local temperature are in adequacy compared to experimental conditions. The potential of the MTV&T technique for the analysis of internal gas flows is therefore demonstrated.

Conclusion

An experimental device for simultaneous velocity and temperature measurement by MTV&T has been described. This set-up has been developed with the aim of providing useful information for a better understanding of practical combustion systems, in order to optimize them.

The displacement of the tagged molecules between the two images provides the estimation of flow velocity. The phosphorescence intensity decay rate of the tagged tracer molecules between two successive images is used to determine the fluid temperature through the temperature dependence of phosphorescence lifetime. The implementation of the MTV&T technique is demonstrated in academic environment and an extension of this technique could be applied soon to a combustion system.

The MTV&T technique is therefore interesting in determining thermal boundary conditions that strongly affect the behaviour of industrial combustion systems. Moreover, in a context where simulations of combustion systems become increasingly advanced and take into account ever more complex physical phenomena, it is necessary to carry out thorough experiments in order to validate these new approaches.

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