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Predicting effect of fibers on thermal gelation of methylcellulose using Bayesian optimization

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Abstract
Understanding of the viscoelastic behavior of a polymer is a prerequisite for its thermomechanical processing beyond laboratory scale. Utilizing rheological characterization is a powerful tool to comprehend the complex nature and time-dependent properties of macromolecular materials. Nevertheless, it consumes time as rheometry involves iterating experiments under several conditions to visualize the non-linear behavior of materials under varying conditions. The work hereunder examines the rheology of cellulosic aqueous suspensions prepared using cellulose fibers as the dispersed phase (Refcell and Storacell) and methylcellulose (MC) as the polymeric matrix. Interfacial phenomena between MC and cellulose fibers arise in particle laden systems with supramolecular structures formed by non-covalent interactions. Therefore, this study elucidates the rheological evolution of these interactions as a function of temperature and fiber concentration. This study displays how researchers may reduce the number of rheological experiments and save time utilizing a novel method based on a Bayesian optimization with Gaussian processes.

Keywords: fibers, methylcellulose, rheology, Bayesian optimization

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1 Introduction

Cellulose is a key market driver in the present and future bioeconomy of polymers. As a material, cellulose has multiple benefits, such as being recyclable, chemically modifiable, and naturally available in enormous quantities (Klemm et al., 2005). Its supramolecular structure and crystallinity, together with its high surface area, make of cellulose a perfect additive to create advanced materials and structures (Kontturi et al., 2018). For instance, we have recently created a cellulose-based foam (Reichler et al., 2021) which is an alternative to replace plastics (Fig. 1). The “FoamWood” solution is a cellular and lightweight material created after drying a gel suspension of methylcellulose (MC) and cellulose fibers. Others have prepared similar composites using methylcellulose and cellulose nanofibers instead of macroscopic fibers (Church et al., 2021; Voisin et al., 2018). Then, to better understand the mechanical properties of such materials, the rheological properties of the MC suspensions must be characterized under varying stimuli (e.g., temperature and shear forces).

Fabricating the FoamWood relies on the capacity of MC to form hydrogels upon heating due to fibril network formation (Lott et al., 2013). In the sol-gel transition the fluid becomes turbid and the relaxation timescales increase.
two to three orders of magnitude (Desbrieres et al., 2000). The transition is thermoreversible and the process depends on the heating/cooling rate and the MC concentration (Kobayashi et al., 1999; Li et al., 2001). In addition, it has been shown that a small stress can have a significantly effect on the MC structure, which affects the gel temperature $T_{gel}$ and, consequently, its mechanical properties (Nelson et al., 2022). We then study the rheology of two types of particle suspensions. Those systems consist of a continuous aqueous phase of MC and a dispersed phase of cellulose fibers (never-dried or dried pulp). In rheological studies, it is well established that $T_{gel}$ is determined by looking the interception between the loss and storage modulus (Mcallister et al., 2015). However, as a consequence of the dispersed matrix in the particle laden suspension, such cross-over is not visible. Instead we introduce a new way to compute the gelation temperature by using the phase shift angle.

The macrostructure of cellulose possesses dipole-dipole associations (hydrogen bonding), which create a supramolecular system (Ioelovich, 2016). In cellulose, the dipole-dipole interactions between OH groups are additive; thus, the orientation between two dipoles affects all the surrounding medium. The idea of combining cellulose and MC involves taking advantage of the hydrogen bonding between these two. Hence, the interfaces between both materials are compatible and physically bonded to one another. In addition to the interactions arising from the OH groups, cellulose chains have hydrophobic sites. Hydrophobic interactions take more relevance for MC than cellulose since MC becomes an amphiphilic molecule (Nasatto et al., 2015). The reduction in the number of hydrogen bonds allows the solubilization of the cellulose ether in water. However, the polymer-polymer (hydrophobic) interactions among carbon chains and methyl groups make the system form a gel above room temperature (Nasatto et al., 2015). The gel transition of MC is essential for certain manufacturing processes, such as 3D printing and the FoamWood process, which allows the material to hold the 3D structure of the hydrogel during drying. The storage modulus during the solution/hydrogel transition have been shown to alter with added materials such as cellulose nanocrystals (Hynninen et al., 2018) and kappa carrageenan (Almeida et al., 2018). Here we test using rheological methods similar with macroscopic cellulose fibers. Previous research suggest that cellulose nanocrystals contribute to the percolation network of the MC fibers forming in the thermal gelation (Hynninen et al., 2018), however such result does not generalize automatically to macroscopic fibers studied here.

A considerable amount of experimental effort has been devoted to char-
acterizing the gelation of methylcellulose (Coughlin et al., 2021; Mcallister et al., 2015). The rheological method is well known to be the most direct and reliable way to determine gelation transition. However, this characterization ends up to be time consuming and requires dozens of experiments. To ease the experiment load, computer simulations (Huang et al., 2014; Queyroy et al., 2004) and phenomenological laws (Li, 2002) are the common ways to study the problem. Over the last years, significant results have been achieved with the help of the machine learning methods. Machine learning approaches have become powerful tools for revealing hidden information (Gao et al., 2022). One recent example applied neural networks that use known constitutive realtions in predicting rheological response of silica suspension in various experimental protocols (Mahmoudabadbozchelou et al., 2022). Here in particular, we use a “semi-active learning” method (Tian et al., 2021) using Bayesian optimization (Shahriari et al., 2016).

Therefore, this study aims to (i) study the rheology of methylcellulose and its gelation temperature, (ii) introduce a new way to determine the gelation temperature using the phase shift angle, and (iii) develop a Bayesian optimization approach which will reduce experiment load. We hypothesize that Bayesian optimization method utilizing Gaussian processes could capture rheological properties of cellulose materials with minimal experimental effort.

2 Materials and Methods

2.1 Materials

We used cellulose ether “Benecel MX MC-50000 cP” (50 Pa-s) from Ashland Specialties Belgium. This MC is a food-grade cellulose ether complying with the standard E461 (JECFA, 2006). According to the latter standard, the DS of methyl (CH₃) groups of the Benecel cellulose ether (BC) must be between 1.45 and 2.00. Furthermore, the maximum content of hydroxypropyl groups is 5%. BC’s average molecular weight \( M_w \) and polydispersity index \( PDI \) were inspected via size exclusion chromatography (SEC) using an 1260 Infinity II Multi-Detector GPC/SEC system. We used 0.1 M NaNO₃ as solvent and pollulan standard with \( M_w \) of 110 kg/mol. SEC analysis of BC exhibited a macromolecular structure with \( M_w \) = 534 kg/mol and \( PDI = 1.5 \). Complementing the structural characterization, BC functional groups, product of
cellulose etherification, were identified through Raman spectroscopy $\lambda = 532$ nm (Renishaw inVia™ Qontor, United Kingdom, see supplementary information Figure S1). Characteristic Raman signals of MC were detected in the wavenumber range 2750-3100 cm$^{-1}$ (i.e., $v$(CH$_3$), $v$(CH$_2$), and $v$(CH)). Here, two cellulose pulps were formulated as Refcell (RC) and Storacell (SC) to prepare the particle laden suspensions. Refcell is never-dried and bleached pine and spruce Kraft pulps. Storacell is a dried “Supreme Strong” bleached softwood Kraft pulp produced by Stora Enso Oyj (Finland). The pulps were in the form of 5% solid content aqueous suspensions.

![Graph](image1)

![Image](image2)

Figure 2: (a) Arithmetic fiber length $\ell$ and width $w$ distributions. (b-c) Optical microscope images of (b) Refcell, and (c) Storacell fibers. The images display the materials’ morphologies.

Figure 2a shows the fiber length and width distributions for Refcell and Storacell obtained with a Valmet Fiber Image Analyzer. While the number–average width of both fibers are similar, $w_{RC} = 28.23$ $\mu$m and $w_{SC} =$
Table 1: Formulations used for preparing the particle laden suspensions.

<table>
<thead>
<tr>
<th>Formulation</th>
<th>Concentration in water (wt.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sample</td>
<td>BC</td>
</tr>
<tr>
<td>BC</td>
<td>1.5</td>
</tr>
<tr>
<td>Refcell</td>
<td></td>
</tr>
<tr>
<td>BC-RC-25</td>
<td>1.5</td>
</tr>
<tr>
<td>BC-RC-50</td>
<td>1.5</td>
</tr>
<tr>
<td>Storacell</td>
<td></td>
</tr>
<tr>
<td>BC-SC-25</td>
<td>1.5</td>
</tr>
<tr>
<td>BC-SC-50</td>
<td>1.5</td>
</tr>
</tbody>
</table>

27.2 μm, the number–average fiber length for Refcell is longer ($\ell_{RC} = 1.1$ mm) than Storacell ($\ell_{SC} = 0.73$ mm). Figure 2b and c show optical microscope images of the materials at 100× magnification using a B120C Siedentopf Binocular Compound Microscope (AmScope, USA) with a digital camera Canon EOS M3. A clear morphology difference can be depicted between the employed materials. Finally, the preparation of samples used high purity Finnish tap water (pH 8.4 at 23°C) (further details about the water composition in supplementary information Table S1).

2.2 Preparation of particle laden suspension

The experiments use two particle laden suspensions with different pulp content of each type. Furthermore, the rheological study involves characterizing the BC matrix for having a comparison reference. Table 1 summarizes the formulations employed for each sample. The following process was used for the sample preparation. First, the respective amounts of BC powder and pulp suspension were mixed in a glass beaker. Then, reaching the desired mass fraction percentage in the aqueous dispersion comprised the addition of water (at 70°C). Finally, the samples’ mixing involved placing the glass containers in an ice bath and mixing them with a magnetic stirrer for one hour at 700 rpm. The suspensions were then stored overnight at 3°C before characterization.
Rheological characterization took place using a rheometer Physica MCR 302, Anton Paar (Austria). The geometry for the analysis consisted of a modified concentric cylinder: a cup (CC27) and a bob (CC17/P6), both with serrated walls to minimize wall slip issues during measurements. Figure 3 shows the setup used in the characterization. The objective of the camera was to monitor the experiment and record sources of error. Rheological measurements occurred utilizing oscillatory tests of the type Dynamic Mechanical Thermal Analysis (DMTA) and a flow curve with shear rate between 0.01 to 100 s⁻¹.

DMTA examines the effect of temperature $T$ on the rheological properties. In DMTA test, a sinusoidal oscillatory strain $\gamma = \gamma_0 \sin(\omega t)$ rises in the sinusoidal material stress $\tau = \tau_0 \sin(\omega t + \delta)$ with a phase shift angle $\delta$ and angular frequency $\omega$. The phase shift angle is a parameter measuring the dominant type of behavior in the material, being an elastic material if $\delta = 0$ and a perfect viscous liquid if $\delta = \pi/2$. A viscoelastic material has a $\delta$ between 0 and $\pi/2$. Accordingly, the change of $\delta$ in an oscillatory experiment may be used as an indicator for the transition of MC from a viscoelastic liquid to a gel. This assumes it is possible to deconvolve the shear stress signal into in-phase and out-of-phase components (Morrison, 2001)

$$\tau(\omega, t) = G' \sin(\omega t) + G'' \cos(\omega t) \quad (1)$$

Here the elastic and viscous behaviors of the material are represented by the storage modulus $G'$ and loss modulus $G''$, respectively. In MC, when $G'$ is greater than $G''$, the molecular conformation is a hydrogel (i.e., a 3D hydrated network). The in-phase and out-of-phase responses may be expressed using complex notation, and then the complex elastic modulus is $G^* = \sqrt{G'^2 + G''^2}$. Moreover, the phase shift in terms of $G'$ and $G''$ is defined as $\tan \delta = G''/G'$. To the best of our knowledge, this work proposes for the first time to use the phase shift $\delta$ as a parameter to identify the gelation temperature $T_{gel}$ of a particle laden suspension such as the one prepared for this manuscript.

We use a strain $\gamma_0$ of 1% in the Linear Viscoelastic region (LVR) that deforms the suspensions at constant angular frequency $\omega$ while heating the samples at 1°C/min (from 25 to 60°C). The characterization comprised repeating the test at three different angular frequencies (0.63, 6.30, and 63.00 rad/s). Every DMTA test employed a new sample, and $T_{gel}$ is determined from the intercept of two slopes of the phase shift $\delta$ (see supplementary information Figure S2), which is compared to the intercept temperature between
$G'$ and $G''$.

Figure 3: Illustration of the experimental assembly using (a) an Anton Paar rheometer MCR 302, (b) a serrated concentric cylinder geometry, and (c) a digital camera Canon EOS R.

In addition to DMTA, dynamic viscosity ($\eta$) experiments complemented the characterization. Isothermal flow curves of viscosity $\eta$ as a function of shear rate $\dot{\gamma}$ characterize the molecular structure of a polymer through the interpretation of its resistance to flow. In particular, for the cellulose/methylcellulose system, these curves provide information about the interaction within the mixture. Isothermal analyses carried out from 25 to 40°C (5°C stepwise) allowed to obtain flow curves. The flow curves resulted from deforming each suspension using a $\dot{\gamma}$ interval from 0.01 to 100 s$^{-1}$ setting the measuring time as the reciprocal of $\dot{\gamma}$. These tests were performed using the same assembly described in Fig. 3.

2.4 Bayesian optimization

Bayesian optimization allows us to obtain an objective functions that take a long time to evaluate, which in our case it corresponds to rheology curve. It builds a surrogate for the objective and quantifies its uncertainty using a Gaussian process regression (see supplementary information for mathematical details) (Shahriari et al., 2016). The Bayesian optimization ran using an in-house developed Python 3.8 based code. The optimization came from a regression utilizing Gaussian processes (GP) with kernels $k$ ConstantKernel, RBF, and WhiteKernel from Scikit-learn (Pedregosa et al., 2011).

This report discusses the results from rheometry after Bayesian optimization to explore the code’s feasibility in future experiments. Therefore, the
values of $\eta$ obtained from rotational tests and the outcomes of $G^*$ and $\delta$ from
DMTA tests were used to train and generate 3D surfaces. The outcomes of
the regression process assisted in discussing the rheological properties and
interactions between cellulose and BC qualitatively.

3 Results and Discussion

The Bayesian optimization used the samples’ dynamic viscosity $\eta$ to generate
the 3D surfaces in Fig. 4 (the data without fit is shown in supplementary
information Figure S3). First, the mathematical process demonstrated the
possibility of using machine learning (ML) instead of empirical rheological
models. Nevertheless, a question arises about the physical meaning of the
length-scale parameter ($l$) (Pedregosa et al., 2011) in the material’s be-
havior. Assigning a physical meaning to parameters is a common problem
when working with such high abstraction level models. Despite the previous
problem, to our knowledge, this manuscript reports a Bayesian optimiza-
tion of rheological properties for the first time. ML combined with rheology
concepts suggests being a powerful tool in materials characterization. With
further investigation, predicting the rheological properties of any system from
an experimental data set can achieve savings in time and effort. Addition-
ally, as Fig. 5 illustrates, the optimization boundaries open fitting different
rheological properties such as $G^*$ (for measurement data see Figure S4 in
supplementary information). Accordingly, the Bayesian optimization could
also fit similar viscoelastic responses, including the polymer’s electrical and
magnetic rheological responses (e.g., complex dielectric permittivity).

It is known from the provider that this BC forms a gel at temperatures
above 40°C. Figure 4a illustrates the behavior of the matrix: BC viscosity
initially follows an Arrhenius decay and then after 30°C, it starts increasing.
BC’s formation of a gel structure stems from hydrophobic polymer-polymer
interactions via methyl groups (Nasatto et al., 2015). The hydrophobic inter-
actions reduce the molecular mobility of the polymer, and as a consequence,
the viscosity increases. It is worth mentioning that all the systems showed
a shear-thinning behavior. The pseudoplastic behavior suggests the forma-
tion of no chemical cross-links; alternatively, the hydrophobic associations
cause physical entanglements with relative conformational mobility and flex-
ibility. Adding cellulose fibers to the BC matrix modifies, as expected, the
suspensions’ viscosity. The hornification’s effect in SC fibers is remarkable
when comparing samples BC-SC-25 (Figure 4d) with BC-RC-25 (Figure 4b). Over the experimental conditions, sample BC-SC-25 presented a viscosity no higher than \(\sim 100 \text{ Pa} \cdot \text{s}\); meanwhile, sample BC-RC-25 reached viscosity values one order of magnitude higher. This difference is because RC fibers’ internal volume is open, and they can create more physical interactions with the unmodified OH groups of BC (Giacomozzi and Joutsimo, 2015). Therefore, inducing shear flow in sample BC-RC-25 requires a more elevated force input than in sample BC-SC-25.

Modifying the fiber concentration to 0.50% increased \(\eta\) at temperatures no higher than 35°C for sample BC-RC-50 and at all the temperatures in the SC sample (Figure 4c and 4e, respectively). The viscosity increased because the free volume in the suspension is less after loading the system with more fibers, which start reflecting their rheological behavior in the particle laden system. Moreover, the number of cooperative interactions between cellulose fibers increased since they formed hydrogen bonding with the components of the surrounding medium (water, BC, and other fibers). RC sample’s viscosity profile (Figure 4c) is similar to the ones reported by Karppinen et al. (2012). The difference in the viscosity’s behavior observed in specimens carrying RC and SC fibers can be explained by the number of supramolecular interactions. Sample BC-RC-50 (Figure 4c) contains more cooperative interactions than the SC sample (Figure 4e). At a temperature higher than 35°C (Figure 4c), the effect of RC fibers in viscosity seemed unclear since the viscosity is lower than for sample BC-RC-25. By recalling the experiments’ videos, sample BC-RC-50 presented apparent wall slip in a higher degree than sample BC-RC-25. Therefore, it is possible to conclude that at temperatures close to \(T_{gel}\) the apparent wall slip affects the measurements’ accuracy.

Figure 5 presents experimental data (dots) together with the Bayesian optimization (surface) of \(G^*\) and \(\delta\) as a function of fiber concentration \(c\) and temperature \(T\). In Fig. 5a and 5b, at a concentration of 0.25 wt.%, RC and SC fibers affected \(G^*\) alike: they increased the shear elastic modulus of BC. Additionally, the behavior of \(\delta\) (Fig. 5c and d) at 0.25 wt.% was opposite to the one of \(G^*\). The modulus increased after adding both types of fibers since they induced an effect comparable to the one observed in fibrillated all-cellulose composites (Baghaei and Skrifvars, 2020). The fibers’ influence becomes noticeable after increasing \(c\) to 0.50 wt.%, and when BC develops a gel structure \((T > 40^\circ \text{C})\). The BC matrix may immobilize the fibers and therefore reinforce the elastic properties. Figures 5c and d support the last statement since the phase shift angle decays due to increasing \(c\) and \(T\).
Figure 4: Samples’ dynamic viscosity as a function of $\dot{\gamma}$ and $T$ fitted with a Bayesian optimization (surfaces) of the experimental data (scatter points). (a) BC, (b) BC-RC-25, (c) BC-RC-50, (d) BC-SC-25, (e) BC-SC-50. The scales of $\dot{\gamma}$ and $\eta$ are logarithmic. The data without the surface fit is shown in Figure S3 in supplementary information.
Table 2: Gelation temperature of the particle laden suspensions estimated utilizing two methodologies.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$T_{gel}$ $G'$ and $G''$ intercept</th>
<th>$T_{gel}$ $\delta$ slopes intercept</th>
</tr>
</thead>
<tbody>
<tr>
<td>BC</td>
<td>50.17°C</td>
<td>49.20°C</td>
</tr>
<tr>
<td>BC-RC-25</td>
<td>42.14°C</td>
<td>44.23°C</td>
</tr>
<tr>
<td>BC-RC-50</td>
<td>No intercept</td>
<td>43.53°C</td>
</tr>
<tr>
<td>BC-SC-25</td>
<td>44.16°C</td>
<td>45.35°C</td>
</tr>
<tr>
<td>BC-SC-50</td>
<td>No intercept</td>
<td>44.75°C</td>
</tr>
</tbody>
</table>

(i.e., the material turns gradually more elastic than viscous). Regarding the fiber type effect, it is more evident at 0.50 wt.%. RC fibers create a stiffer material since the increment of $G^*$ (Fig. 5a) and decrement of $\delta$ (Fig. 5c) were more significant than the ones observed with SC fibers. The viscosity discussion also explains these differences; the RC-BC interface possesses more cooperative associations than the SC-BC.

Table 2 collects the $T_{gel}$ of the examined suspensions. The intercept of $G'$ and $G''$ is traditionally the best route to determine the gel transition. However, in the particle laden suspensions, no intercept can be identified as a consequence of the dispersed phase effects on the matrix. Cellulose fibers increase the elastic component of $G^*$ and hide the transition as the $G'$ and $G''$ curves never intercept. Here, we used $\delta$ to determine $T_{gel}$ (see Figure S2). Considering that $\delta$ defines the viscoelastic behavior, the exponential decay of this parameter could assist in elucidating the gel transition. Identifying $T_{gel}$ from $\delta$ imitates the methodology used to determine the glass transition using the exponential decay of $G'$ (see Figure S2 in supplementary information).

In general, adding both types of fibers reduced $T_{gel}$. The fibers’ hydrophobic and OH sites may work as heterogeneous nucleation segments for BC. Adding more fibers decreased $T_{gel}$, this fact supports the idea about their nucleant effect since more fibers represent more nucleation segments. Respect the difference between RC and SC fibers, RC fibers induced a lower $T_{gel}$ than SC ones. The cooperative associations arising between RC-BC and SC-BC interfaces are again a possible explanation to the differences in $T_{gel}$.

Finally, the new method to define $T_{gel}$ and fitted surfaces provide a methodology to deduce the dependency of $T_{gel}$ of the fiber concentration. Fitting the lines as shown in Figure S2 to the predicted $\delta$ values for vari-
Figure 5: Samples’ $G^*$ and $\delta$ (at $\omega = 6.30$ rad/s) as a function of fiber concentration and $T$, fitted with a Bayesian optimization (surfaces) of the experimental data (scatter points). (a) and (c) belong to the samples containing BC and RC. (b) and (d) belong to samples containing BC and SC. The scale of $G^*$ is logarithmic. The data without the surface fit is shown in Figure S4 in supplementary information.
Figure 6: (a) The values for $T_{gel}$ may be defined at concentrations that have not been measured using the GP regressor fitted surface. The surface is the same as in Fig. 5d and the solid line shows the position of gelation. (b) The gelation temperature with respect to the fiber concentration $c$ of the Storacell fibers determined using the GP fitted surface. The circles show the values determined from the measurements.

Ous values of $c$ result in estimates of $T_{gel}$ that were not actually measured. Figure 6a shows the same surface as in the Fig. 5d with a line showing the estimated $T_{gel}$ for suspensions with $c$ from 0.0 to 0.5 %. The Figure 6b then displays the estimated gel temperature with the concentration. The circles in Fig. 6b are the values actually determined from the measurements. The estimate accords with the three points; however, these points alone do not contain enough data for such complete estimate for gel temperature. Supplementary Figure S5 shows similar plots with increasing Refcell concentration where the outcome accords with Figure 6. The examples demonstrates the power of GP regressor in successfully estimating material properties with incomplete data sets without laborious extra measurements.

4 Conclusion

Methylcellulose demonstrated being an alternative to utilizing solubilized cellulose for the manufacture of cellulosic composites. Nevertheless, the compatibility between the cellulose derivative and fibers depended on the number of supramolecular interactions. This manuscript revealed that enhancing the
interface matrix-fiber hangs on the degree of substitution of MC and fibers’ morphology. For instance, never-dried fibers possessed an uncollapsed inner structure abundant in OH groups, while dried fibers lacked it. The difference in morphology defined the properties of the particle laden suspensions, which was why RC could create more cooperative associations with the cellulose ether than SC. Accordingly, raising the concentration of RC fibers overall increased the viscosity and reinforced the mechanical properties to a higher degree when compared to SC samples.

In brief, rheology was an adequate tool to elucidate the interfacial phenomena in the studied particle laden suspensions under different stimuli and conditions (c, ˙γ, and T). Assisted with Bayesian optimization, the spectra of rheological characterization exhibited having unexplored frontiers. Our approach has effectively fill the gap between the different experimental measurements, reducing in this way the number of experimental required. Here we have used experimental data to determine the 3D surfaces, but it can be easily extrapolate to be used in simulation and reduce the computational time. It is unmistakable that using Bayesian optimization will lead to saving resources in materials characterization and research development.

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