

THE EFFECT OF DRYING TEMPERATURE ON
CHEMICAL-SUBSTANCE EMISSION FROM SOLID WOOD

木材由来の化学物質放散速度に対する乾燥温度の影響について

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This study aims to correct the widespread misconception in the Japanese housing market that solid woods emit no chemical-substances, though their emissions are significantly lower than those of industrial wooden materials. Small chamber tests were carried out to compare the formaldehyde (HCHO) emission rate of Japanese Cedar and that of industrial wooden materials. Conventionally, it was clarified that Japanese Cedar emitted less HCHO than the value of the best grade, as defined by the regulations for interior materials. However, the HCHO emission rates of 2 identical specimens under the same testing conditions were significantly different, with the drying temperature of the specimens considered as a primary factor for their preparation. Japanese Cedar and German Spruce were dried under 3 different conditions and examined in small chambers. The results show that the HCHO emission rate of different species is affected by the drying temperature and that solid wood dried at high temperatures emits more volatile organic compounds (VOCs) than that dried at low temperatures.

Keywords: *solid wood, formaldehyde, volatile organic compounds, emission rate, drying temperature*

木材、ホルムアルデヒド、揮発性有機化合物、放散速度、乾燥温度

Introduction

Indoor air pollution caused by chemical substances and its adverse health effects are subjects of increasing public concern in Japan. The Building Standards Law was amended in 2003 by the Ministry of Land, Infrastructure, Transport and Tourism to address "sick-house syndrome" issues¹⁾. The amendment's main objective is the control of formaldehyde (HCHO) concentrations through the use of low-emission materials and the mandatory installation of ventilation equipment.

On the other hand, it is worth to note that in Japan solid wood, especially sugi cultivar in Kumamoto, is abundant and widely used as an architectural material. In Japan, solid woods are widely used in domestic architecture because of their soft texture, aesthetic appearance, and pleasant odor. It emits less gaseous HCHO than wood-based materials. According to the Building Standards Law, there is no restriction on the use of solid woods as interior materials.

In wood science, Meyer et al. reported that HCHO was released from solid wood²⁾. Schäfer et al. showed that thermal conditions play a role in the chemical reaction of components and that solid woods emitted variable amounts of HCHO³⁾. In addition, characteristic volatile organic compounds (VOCs) were emitted as scent and Hida et al. measured the VOC emissions from solid woods using the small chamber method⁴⁾.

Nevertheless, there are two widespread misconceptions about solid woods: firstly, that they emit no HCHO; secondly, that VOCs are not present in the emitted odor. This experimental study was conducted with the aim of collecting data to counter these misconceptions. The following experiments were conducted using small chambers to detect the emission of chemical substances from solid woods:

1. The specimens, cut from a Sugi cultivar (*Ayasugi cultivar*) felled at Oguni in Kumamoto prefecture, were examined at 3 different values of the ratio between the air exchange rate and the loading factor (N/L), and the HCHO emission rate of the solid wood was compared with that of industrial wooden materials.

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2. The above experiment showed that the HCHO emission rates of 2 identical specimens at $N/L = 1$ were scattered, and the differentiating factor between these 2 specimens was the drying temperature. Concentrating on the drying temperature, specimens cut from an Ayasugi, as same as those used in the previous experiment and from a Spruce felled at Fraunhofer Wilhelm-Klauditz-Institut Holzforschung (WKI) in Braunschweig, Germany, were investigated at $N/L = 1$.

In this paper, the typical results from these small chamber tests are reported with focus on the effect of drying temperature on the HCHO emissions, along with the VOC emissions.

Method

Testing equipment

20-liter stainless-steel airtight tanks were used as small chambers for the Ayasugi tests in Kumamoto, and six chamber sets were established in a climate room (Fig. 1). A polytetrafluoroethylene (PTFE) tube was used to line this equipment, in order to minimize emission, desorption and adsorption of chemicals. The chambers were easily adjusted to a constant temperature of 25 °C in the climate room. The air in the climate room was pumped through the small chambers by negative pressure, and the air volume was constantly maintained at 0.333 L/min ($N = 1 \text{ h}^{-1}$). A branch was connected to the outlet of the chamber to allow pump sampling to be carried out without changing the air exchange rate in the chamber. Clean air, filtered by activated carbon and silica gel, was supplied to all the chambers. The mixing ratio of the activated carbon and silica gel in the filter was determined to control the contaminants as well as the relative humidity (RH) in the chambers as 50%. The equipment used here was not as specified by JIS A 1901⁹⁾; however, simultaneously operating these 6 chamber sets made it feasible to carry out simple and inexpensive tests.

23-liter glass desiccators were used as small chambers for the Spruce tests in WKI, and one set of the equipment was placed in an air-conditioned laboratory (Fig. 2). A PTFE tube was used to line this equipment. The chambers were adjusted to a constant temperature of 25 °C by controlling the voltage of the electric heating belt around the desiccator in the isolated box and maintaining 50%RH by mixing dry air and wet air. Filtered clean air was supplied to the desiccator and the air volume was constantly sustained at 0.392 L/min ($N = 1 \text{ h}^{-1}$). A branch was connected to the outlet of this chamber as well, to allow pump sampling without changing the air exchange rate in the chamber. WKI is one of the institutes authorized by the Ministry of Land, Infrastructure, Transport and Tourism to conduct tests of chemical emissions from materials.

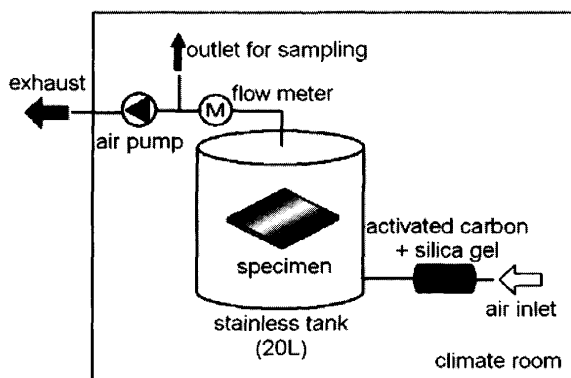


Fig.1 Model of testing equipment in Kumamoto

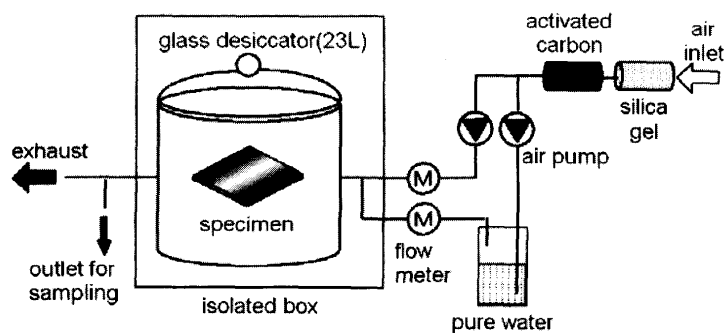


Fig.2 Model of testing equipment in WKI

Materials

Ayasugi and Spruce logs were cut into 120 × 120 × 10 mm pieces along the flat grain, and these pieces had conditioned moisture content of 9%, which is the equilibrium moisture content at 25 °C and 50%RH. In general, the solid woods used in building interiors are dried to a moisture content ranging from 9% to 15%.

Table 1. Specimens

specimen	drying condition	moisture content ϕ
A	30 °C, 50%RH	9%
B	60 °C, 60%RH	
C	105 °C (oven dried) → 30 °C, 50%RH (moistened)	

The moisture content ϕ [%] of the wood pieces is calculated by the oven-dry method, as represented by the following equation:

$$\phi = \frac{W_w - W_D}{W_D}$$

3 pieces of each species were dried at 105 °C till their weights became constant, and the constant weight W_D [g] was calculated as the average of these 3 weights. The target weight of the specimen, when $\phi = 0.09$, was determined from this equation, and the weight of the specimen after drying W_D [g] was measured as required during the drying process until the target value was achieved.

Specimen-A was dried at 30 °C / 50%RH, specimen-B was dried at 60 °C / 60%RH, and both had moisture content of 9%. Specimen-C was moistened to 9% at 30 °C/50%RH after being oven-dried at 105 °C. These drying conditions were maintained at equilibrium. It took 5-7 days to prepare each specimen.

The edges of the specimens were sealed with aluminum just before they were set in the chamber. The emission surface area of Ayasugi was 100 × 100 mm and that of Cedar was 110 × 110 mm on both sides (0.02 and 0.024 m²).

For the first chamber test, which changed the N/L value, some pieces of Specimens-A and B were prepared, and the loading factor L [m²/m³] was varied by the number of pieces in order to change the emission surface area.

For the second test which was focused on the drying temperature, all 3 specimens were prepared, and one piece was placed in the chamber.

Procedure

After cleaning the chambers, the testing equipment was operated for over 24 hours to flush out the contaminants, after which the background concentration was measured and verified to be sufficiently low. The specimens were placed in the chambers, and the air sample of each chamber was collected at least 5 times and analyzed to obtain the steady-state concentrations of the chemical substances. This procedure was applied to both the chambers in Kumamoto and WKI.

Analysis

The HCHO concentration in the chambers was measured by the authentic high-performance liquid chromatography (HPLC) method⁶⁾. The emitted VOCs in the chambers were collected in a tube filled with Tenax TA and Carbotrap. The concentration was quantified by gas chromatography/mass spectrometry (GC/MS) with a thermal desorption system^{7, 8)}. The concentrations, which are shown in the results below, were higher than the limit of determination for each sampling volume, and the emission rate (ER) [$\mu\text{g}/(\text{m}^2\text{h})$] was calculated by the equation as in JIS A 1901⁹⁾.

Table 2. Analysis conditions

	sampling	analysis		sampling	analysis
HCHO	DNPH cartridge 0.25 L/min × 2 h = 30 L	HPLC System column : 250 mm × 4 mm with a pre-column UV detector : 355 nm flow rate : 0.75 ml/min at ambient temperature with gradient injection : 25 μl without dilution	VOCs	Tenax TA + Carbotrap (original) 0.15 L/min × 40 min = 6L	GC-MS System with thermal desorption thermal desorption temp.: 300 °C desorption time : 12 min trap temp. : -30 °C

Results and Discussion

Figure 3 shows the relationship between the HCHO emission rate on the first 15 days and the N/L of the Ayasugi specimens, which were dried at 30 °C and 60 °C.

This result of the first experiment is compared with industrial wooden materials. There are chemical-emission regulations for wood-based materials in several countries. In Japan, there is a regal control on the HCHO emission rate. The best grade for emission rates, "F-four stars (F☆☆☆☆)", allows an emission rate of less than 5 $\mu\text{g}/(\text{m}^2\text{h})$ when N/L is 0.05 at 28 °C/ 50%RH¹⁾. The results in this study prove that the HCHO emission from solid wood is much less than that from wood-based materials. However, the HCHO emission rates of 2 specimens under the same value of N/L were significantly different when the drying temperature was varied.

The specimens cut from the Ayasugi and the Spruce were examined under the testing condition of $N/L = 1$, with focus on the drying temperature.

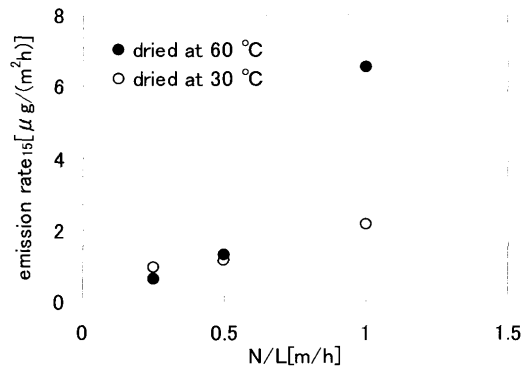

 Fig.3 HCHO emission rate and N/L for Ayasugi

Figure 4 shows the HCHO emission rate for the Ayasugi specimens. The emission rates for all specimens, except the oven-dried specimen, attained steady state levels immediately. Though the HCHO emitted from the specimen dried at high temperature was two to three times higher than that emitted from the specimen dried at 30 °C, it was still considerably low. Acetaldehyde and acetone emissions were also analyzed by HPLC. The emission rate of acetaldehyde was extremely low, less than 5 $\mu\text{g}/(\text{m}^2\text{h})$.

Figure 5 shows the acetone emission rate for Ayasugi. The specimen dried at lower temperature emitted approximately 10 $\mu\text{g}/(\text{m}^2\text{h})$ under steady-state conditions. The oven-dried sample showed a significantly higher emission rate at the beginning of the test and roughly twice the acetone concentration after 28 days. The VOC emission rate could not be obtained for Ayasugi specimens.

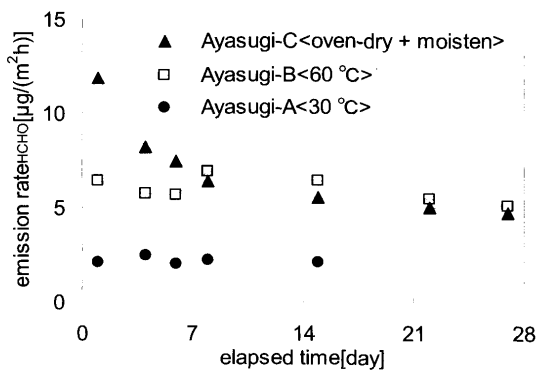


Fig.4 HCHO emission rate for Ayasugi

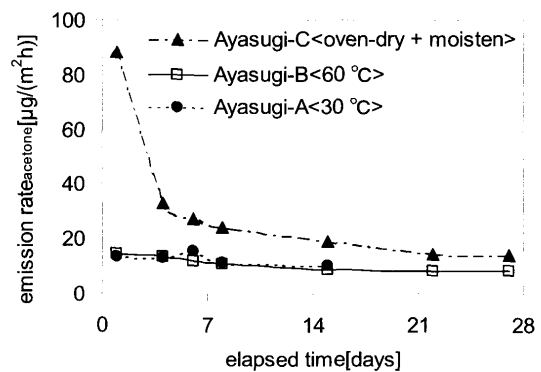


Fig.5 Acetone emission rate for Ayasugi

Figure 6 shows the HCHO emission rate for German Spruce. These tests, which were performed in Germany, were not carried out over a long term; however, the emission rate reached a steady state earlier than the case of Ayasugi. There is no significant difference between the emission rates for the specimens dried at 30 °C and 60 °C; however, the specimen moistened after having been oven-dried emitted thrice the amount of HCHO emitted by the other specimens. Acetaldehyde emission was also analyzed by HPLC, and its emission rate was approximately 10 $\mu\text{g}/(\text{m}^2\text{h})$ under steady-state conditions, as shown in Fig. 7.

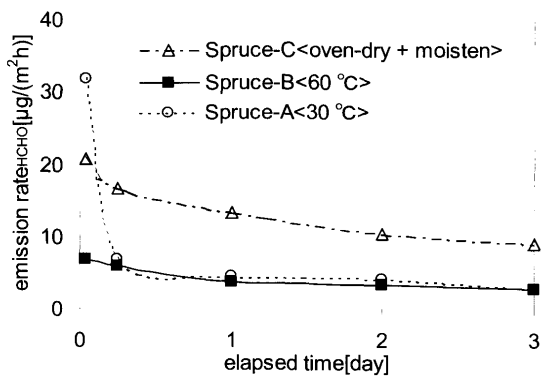


Fig.6 HCHO emission rate for German Spruce

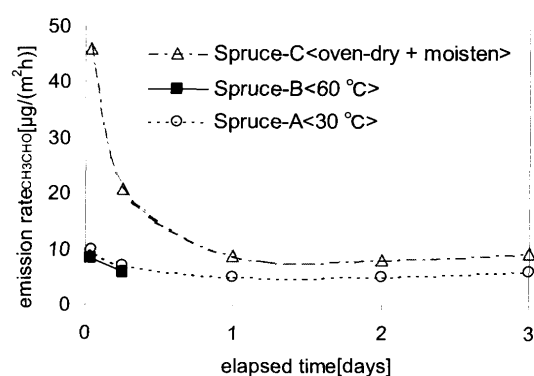


Fig.7 Acetaldehyde emission rate for German Spruce

Figure 8 shows the equilibrium HCHO emission rates of Ayasugi and Spruce based on the drying temperature from the results of the chamber tests performed in Kumamoto and WKI as described above with Figs.4 and 6. The results were the same for both the type of wood, which were moistened after having been dried at 105 °C. Conventionally, wood dried at high temperatures has been considered to emit more HCHO than that dried at low temperatures. However, the emission rates of the Ayasugi specimens dried at 60 °C were different from those of the Spruce specimens. The border of drying temperature, which significantly increases the HCHO emission rate, is dependent on the wood species and its components, e.g., lignin⁹. The lignin content of Ayasugi and Spruce could not be measured; however, softwoods have higher lignin content, and from the perspective of wood science, HCHO should be isolated from lignin by drying at high temperature. The acetaldehyde emission rate from the Spruce was about five times as that from the Ayasugi. The acetone emission rates from both were similar. The effect of drying temperature on the emission rates of acetaldehyde and acetone was similar to the effect of drying temperature on the HCHO emission rate. Different emission rates are observed for each species of wood.

Figure 9 shows the TVOC emission rate for Spruce. Here, "TVOC" means the sum of all detected VOC content, and is not the same as the ISO definition⁷). More VOCs are emitted from solid wood dried at a low temperature than from that dried at a high temperature.

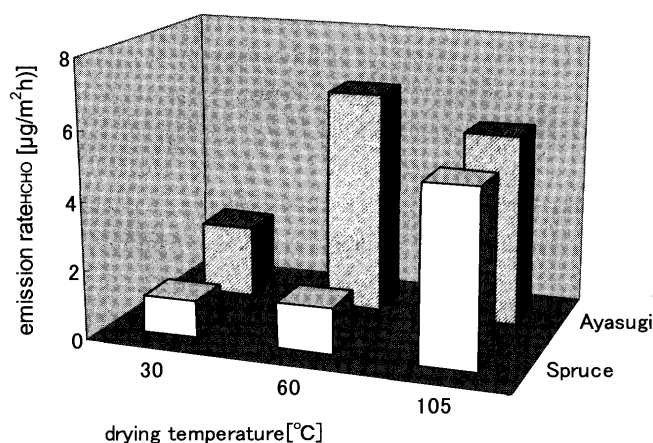


Fig. 8 HCHO emission rate and drying temperature

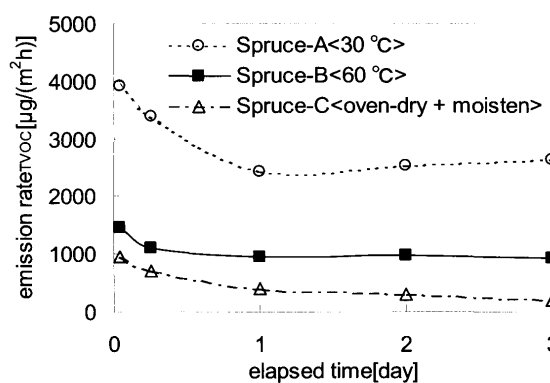


Fig. 9 TVOC emission rate for German Spruce

Table 3 VOCs emission rate for Spruce at 3 days after placing the specimens in chambers. (blank space: not detected)

Name	emission rate[$\mu\text{g}/(\text{m}^2\cdot\text{h})$]		
	A	B	C
Acetone	13		24
Acetic acid			29
Methylcyclohexane	< 1	< 1	< 1
Toluene	8	3	4
Furfural			1
α -Pinene	740	306	26
Camphene	70	13	
Benzaldehyde	5		13
β -Pinene	526	200	5
Myrcene	66	10	
3-Carene	716	258	14
Limonene	138	29	
iso-octanol	2		1
methyl(1-methylethenyl)-benzene	12		
Terpinolene	99	11	
1-methyl-4-(1-methylethenyl)-benzene	38		
p-mentha-1,5,8-triene	2		
α -Terpinene	18		
Pinocarvone	2		
Borneol	< 1		
p-Cymen-8-ol	1		
4-methyl Cyclohexanone	11		
α -Terpineol	5		
Eucarvon	1		
Longifolen	1		
others	168	96	84
TVOC	2645	931	206

The VOCs emission rate could be identified only for Spruce, and the results of the individual VOCs are shown in Table 3. Most of the VOCs emitted from German Spruce were very volatile organic compounds (VVO) such as circular monoterpenes, and they decrease with an

increase in drying temperature. More VOCs were emitted from solid wood dried at a low temperature (specimen-A) than from that dried at a high temperature (specimen-C).

In Germany, there is a labeling system called "Blue Angel", and the evaluation standard requires that the test be performed at $N/L=1$ at 23 °C/ 50%RH^{10, 11}. The "Blue Angel" label permits less than 0.05ppm of HCHO and 300 µg/m³ of VOCs after 28days have elapsed. The results in this study prove that the HCHO emission rate from solid wood is considerably less than that from wood-based materials. However, it is possible that solid wood that has not been well dried can emit more VOCs than materials which are allowed for "Blue Angel".

Conclusion

These small chamber tests were carried out to correct the prevalent misconception in the Japanese housing market that solid woods emit no chemical substances though they do emit chemicals at comparatively lower emission rates than industrial wooden materials. In summary, it was clarified that Japanese Cedar emitted significantly less HCHO than the value of the best grade specified by the regulation for interior materials established by the Ministry of Land, Infrastructure, Transport and Tourism, and the results show that the emission rate of chemical substances can be affected by the drying temperature although to different extents depending on the species of wood.

The latest trends in Japan are drying wooden materials at high temperatures for short time periods or drying at low temperature inadequately because of the imbalance in the Japanese wood market. Recent advances in the high-temperature drying process have eliminated cracking and warping. For the investigated wood species, drying at a relatively high temperature appears to be a suitable method to maintain low chemical substance concentration, i.e., to attain good indoor air quality.

In addition, measures to establish restrictions on VOC emission from building materials and to impose a restriction on indoor concentrations of TVOC are in progress. It is possible that the indoor concentrations of TVOC in residences constructed with other wood species exceed the permissible level. In order to use solid wood as a proper building material, we must seriously consider restricting TVOC concentration.

The conditioning factors, the wood grain, the species of wood, and the place of origin of specimens have been suggested as testing parameters for future research. Additionally, it would be useful to conduct further investigations into VOC emission resulting from chemical reactions between the components of the solid wood and the adhesives.

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和文要約

室内化学物質汚染とその健康影響が社会問題となり、いわゆる「シックハウス症候群」が広く認識されるようになった。建築の分野においては、2003年国土交通省が建築基準法を改正し、ホルムアルデヒド放散の少ない建材を積極的に活用することになった。特に、熊本県では県産木材の利用を推進しており、地元のスギやヒノキを内装などに用いた住宅産業が盛んである。一方、木材学の分野では、無垢の木材からホルムアルデヒドを始めとする化学物質が放散されることはよく知られている。しかしながら、一般的には、無垢の木材からホルムアルデヒドや揮発性有機化合物は放散されないかのように認識されており、「健康志向」の住宅に採用されることが多い。

そこで、本研究は、建築環境工学の立場からこういった誤解を解くためにも、小形チャンバーを用いた試験を行い、木材を対象とした化学物質放散の実態を把握することを目的としている。供試体は、内装材料として多用される樹種として、日本のスギとドイツのスプルースを採用し、それぞれ丸太の状態から同じ部位の小さな木片を切り出した。これらを、3つの温度が異なる乾燥条件で所定の重量含水率に整え、ホルムアルデヒドを主に、経過日数ごとの化学物質放散速度を求めた。

その結果、高温で乾燥させた木片のほうが、低温で乾燥させた木片よりも、ホルムアルデヒドの放散速度は大きく、揮発性有機化合物の放散速度は小さくなることがわかった。さらに、ホルムアルデヒド放散速度が極端に増える乾燥温度は、樹種によって異なることが示唆された。しかしながら、無垢の木材から放散されるホルムアルデヒドは、合板類でもっとも放散速度が少ない「F☆☆☆☆」の基準値と比較して、非常に少ないことが明らかになった。

近年、短期間に一定量の木材を供給するため高温で乾燥させることが多く、技術の進歩によりひび割れやそりも少なくなっている。今回試験に供した木材に関しては、高温乾燥により化学物質放散を抑制することができ、内装材料として用いた場合に室内空気質への影響は少ないと考えられる。しかしながら、将来的に各種揮発性有機化合物の規制が強化されれば、樹種や産地などの条件によっては木材から放散される化学物質が問題となる可能性がある。今後は、木材の乾燥および保管条件、木理、樹種、産地などの異なる場合について、同様の研究が必要であり、さらに、接着剤との組合せによる化学物質放散の実態を把握する必要がある。

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