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Influence of Mineral Admixtures on the Permeation Properties of Self-Compacting Concrete at Different Ages

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Abstract Due to its specific properties, the study of selfcompacting concrete (SCC) represents an area of research that has strong potential for development. However, in spite of the interest of researchers in this new material, SCC has not yet gained universal acceptance as a construction material, and its application remains limited. The development of an economical SCC with interesting properties in the fresh and hardened state is important for the acceptance of such a concrete. Algerian natural source Pozzolan is rarely used in SCC due to the absence of any thorough study of its properties. This study investigates the permeation properties of SCC mixtures made with this Algerian natural Pozzolan, compared with conventional vibrated concrete and other SCC mixtures containing fly ash or limestone filler. Additionally, the correlations between chloride diffusion and sorptivity, and between apparent gas permeability and chloride diffusion, were investigated. Results indicate that, despite its economic benefits, SCC incorporating natural Pozzolan presents very low permeation properties (for example: lower migration coefficients compared to the SCC mixtures with lime-

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stone filler or ordinary vibrated concrete (difference >50 % for results of the 30 MPa strength class). The relation between chloride diffusion and sorptivity, and between apparent gas permeability and chloride diffusion, is also confirmed.

Keywords Self-compacting concrete · Permeation properties · Natural Pozzolan · Economical concrete · Mineral addition

الخلاصية

تعتبر دراسة الخرسانة ذاتية الدمك (خ ذ د) مجالا خصبا للبحوث التي تمتلك امكانات كبير ة للتنمية و ذلك بسبب حصائصها المتميز ة. و على الر غمّ من إهتمام الباحثين بهذه المادة الجديدة، فإن (خ ذ د) لم تحظ حتى الآن بالقبول العالمي كمادة بناء، ونطبيقها لا يزال محدودا إن العمل على إيجاد (خ ذ د) اقتصادية تمتلك خصائص مقبولة في الحالتين الصلبة والسائلة جد مُهِم لقبول هذا النوع من الخرسانة.

البوزلان الطبيعي في الجزائر نادر الإستخدام في (خ ذ د)، وهذا نظرا لعدم وجوددراسة مستفيضة خصائصه. حول تبعَّث هذه الدراسة خَصَّائصُّ النفاذية لمختلف مخاليط (خ ذ د) المصنوعة من البوزلان الطبيعي الجزائري ومقارنته مع الخرسانة النقليدية ومخاليط أخرى من (خ ذ د) تَحتوي على الرماد المتطَّاير أو غبار الحجر الجيري. بالإضافة إلى ذلك، لقد ، تمت مناقشة إن العلاقة بين انتشار الكلوريد ومعذل الامتصاص وبين نفاذية الغاز وانتشار الكلوريد.

تشير النتائج إلى أنه زيادة على فوائدها الاقتصادية، ان (خ ذ د) التي تحتوي على البوزلان الطبيعي تتميز بخصائص نفاذية منخفضة جدا على سبيل المثال ، ان معاملات انتشار الكلوريد أقل بالمقارنة مع مخاليط (خ ذ د) المحضرة من الحجر الجيري أو مع الخرسانة التقليدية (فرق < 50 ٪ بالنسبة لنتائج المخاليط ذات درجة قوة 30 ميجا باسكال). وتم تأكيد العلاقة بين انتشار الكلوريد ومعدّل الامتصاص وبين نفاذية الغاز وانتشار الكلوريد.

1 Introduction

Self-compacting concrete (SCC) describes a concrete with the capacity to compact itself only by means of its own weight

without the requirement of vibration. It fills all recesses, reinforcement spaces and voids, even in highly reinforced concrete members and flows free of segregation nearly to balance level. Flowing in the formwork, SCC is able to de-aerate almost completely.

It is believed that full realization of the benefits that SCC can bring to the concrete industry will only occur when the material becomes perceived as a cost-effective viable technology. Due to its higher binder and chemical admixture content, SCC is usually associated with 20–50 % higher material cost compared to ordinary concrete of comparable compressive strength $[1]$ $[1]$. Martin $[2]$ $[2]$ found that depending on the fly ash content of SCC, its material cost is 10–17 % higher than that of conventional concrete. However, there is still need to make SCC more cost competitive. The concept of developing economic SCC is not new; several methods have been adopted to reduce the cost of this concrete $[1,3-5]$ $[1,3-5]$ $[1,3-5]$ $[1,3-5]$. The valorisation of local natural materials seems to be one of the possible solutions for the future. However, the durability of SCC in which ordinary commercial materials are replaced with natural local materials needs to be established. The main objective of this study is to systematically assess durability by investigating the capillary absorption, water porosity, mercury porosity, chloride diffusivity, and gas permeability of different SCC mixtures incorporating Pozzolan from a natural source in Algeria compared with other SCC mixtures with limestone filler and fly ash. Additionally, the influences of cure and strength class were considered in the test program; strength classes of 30, 50 and 70 MPa, and curing periods of 28, 90 and 360 days were studied. To confirm the relation between compressive strength and permeation properties, results were presented as a function of compressive strength. In this study, we investigated the relation between chloride diffusion coefficients and sorptivity coefficients, and between apparent gas permeability coefficients and chloride diffusion coefficients.

2 Materials

One type of cement, Portland Cement 52.5 CEMI, was used in all the various compositions. The sand used in this study was a 0/4 fraction, siliceous round sand. The gravel fractions were 4/6 and 6/12.5, resulting from the crushing of silicocalcareous rocks [\[6](#page-7-4)].

Three types of additions were used in our study:

- A natural Pozzolan from a volcanic deposit in the western region of Algeria. Natural material in abundance that does not require a lot of energy in its extraction and use.
- "BETOCARB ®P2" limestone filler, characterized by its great fineness [\[6](#page-7-4)] (commercialized materials).
- A silico-aluminate class F fly ash (commercialized materials).

Table [1](#page-1-0) [\[6](#page-7-4)] describes the various physical and chemical characteristics of the cement and of each addition.

3 Experimental Program

3.1 Mixtures

The mix design of the SCC was determined using the "BétonLab.Pro" software [\[7](#page-7-5)]. All of the various data related to the materials used were taken into account. Twelve mix designs were studied in this investigation, thus covering three different strength classes (30, 50 and >70 MPa), as well as four types of concrete [ordinary vibrated concrete (OVC), SCC with limestone filler addition (SCC LF), SCC with natural Pozzolan (SCC PZ), and SCC with fly ash (SCC FA)]. A fixed amount of binder (cement + mineral additions) equal to 520 kg/m^3 was selected. In each strength class, the concrete mixtures were formulated starting from the same compo-

Table 1 Chemical composition and physical properties of cement and additions [\[6](#page-7-4)]

Table 2 Mixture proportions and compressive strengths of the concretes investigated

Concretes	Composition $(kg/m3)$									W/B
	\mathcal{C}_{0}^{0}	LF	PZ	FA	S ₀ /4	G4/6	G6/12	W	SP	
OVC ₃₀	260				830	171	1000	183	1.0	0.7
SCC30LF	260	260		-	807	164	661	199	2.7	0.7
SCC30 PZ	260		260	-	781	159	640	215	3.9	0.7
SCC30 FA	260	-	-	260	757	155	620	215	4.0	0.7
OVC ₅₀	350			$\overline{}$	777	159	936	182	1.0	0.52
SCC50LF	350	170		-	814	166	666	198	7.1	0.52
SCC50 PZ	350		170	$\overline{}$	790	161	647	214	6.4	0.52
SCC50 FA	350	-	-	170	774	158	634	214	4.2	0.52
OVC70	450	-		-	746	154	900	176	3.1	0.39
SCC70LF	450	70		-	839	171	687	183	10.3	0.4
SCC70 PZ	450		70	-	831	170	681	188	11.1	0.4
SCC70 FA	450			70	825	168	676	188	12.2	0.4

nents, with the same granular skeleton and constant water to binder ratio (*W*/*B*). A comparison was then carried out with the same mechanical strength.

A superplasticizer was used to obtain a slump flow as close as possible to 66 ± 3 cm for all of the SCC mixtures. The same strength classes as those of the SCC were targeted, and the formulation of the OVC was determined using the Dreux-Gorisse method.

Table [2](#page-2-0) shows the mixture proportions and the compressive strength of the concretes developed in this study.

3.2 Testing of Permeation Properties

Depending on the process and the nature of the transported matter, the transport of fluids into concrete is usually classified into three main mechanisms, namely capillary absorption, diffusion and permeation.

3.2.1 Capillary Water Absorption Test

The test determines the sorptivity or rate of water absorption through the concrete surface. The capillary absorption test was carried out on the moulded side faces of the 10-cm cube specimens. The specimens were preconditioned in an oven at 105 \pm 5 °C to constant weight.

The uptake of water by capillary absorption was measured through the weight gain of the specimen at set time intervals of 10, 30 min, 1, 4, and 24 h of concrete surface in contact with water.

The sorptivity coefficient (*S*) was obtained using the following expression:

$$
I = St^{1/2}(\text{mm/h}^{1/2})
$$
 (1)

where *t* represents time (h), *I* represents the cumulative water absorption per unit area of inflow surface (mm3/mm2) and *S* represents the sorptivity index (mm/h^{1/2}).

3.2.2 Water Porosity Test

Water porosity was calculated from three masses (weighed hydrostatically or in air):

Apparent mass of saturated concrete samples (3 cylinders \varnothing 11 × H5 cm) after immersion (liquid saturation under vacuum);

(*M*water), mass in the air while still saturated; and,

 (M_{air}) , mass of dry samples (dried at 105 \pm 5 °C until they reached a constant mass);

 (M_{dry}) , water porosity (ε) is given by the following equation:

$$
\varepsilon = \frac{M_{\text{air}} - M_{\text{dry}}}{M_{\text{air}} - M_{\text{water}}} \times 100 \tag{2}
$$

3.2.3 Chloride Diffusivity Test

The rapid test developed by Tang and Nilsson [\[8\]](#page-7-6) was used to determine the chloride diffusivity of the concrete (Fig. [1\)](#page-3-0). An external electrical potential was applied axially across the specimen, forcing external chloride ions to migrate into the specimen. After a certain test duration, the specimen was axially split and a silver nitrate solution was sprayed onto one of the freshly split sections. The chloride penetration depth could then be measured from the visible white silver chloride precipitation, after which the chloride migration coefficient could be calculated from this penetration depth. The

Fig. 1 Experimental chloride ions diffusivity test [\[8\]](#page-7-6)

Sample (\varnothing 110×60 mm)

Fig. 2 Experimental device used for measuring gas permeability

relationship used was as follows:

$$
D_{\text{nssm}} = \frac{0.0239(273 + T)L}{(U - 2)t}
$$

$$
\times \left(X_{\text{d}} - 0.0238\sqrt{\frac{(273 + T)LX_{\text{d}}}{U - 2}}\right)
$$
 (3)

where D_{nssm} is the chloride migration coefficient, $(\times 10^{-12})$ m^2 /s), *U* the absolute value of the applied voltage (V), *T* the average value of the initial and final temperatures in the analyte solution $(^{\circ}C)$, *L* the thickness of the specimen (mm), X_d the average value of the penetration depths (mm), and t the test duration (h).

3.2.4 Gas Permeability Test

The gas used in this study was helium. The test for determination of gas permeability of concrete, recommended by Cembureau [\[9\]](#page-7-7) and RILEM [\[10\]](#page-8-0), was adopted (Fig. [2\)](#page-3-1). For each concrete mixture, three disc specimens (\varnothing 110 \times 60 mm) were conditioned in an oven at 80 ◦C for 28 days and 105 ◦C for 5 days, then wrapped in cling film to cool down before testing. The test was carried out by applying a constant pressure head to the test specimen, and the flow rate of gas through the specimen at steady state under pressure was mea-

sured. This allows the permeability coefficient of the tested concrete to be determined. According to Carcasses et al. [\[11](#page-8-1)], the comparison between several mixtures can be based on the gas permeability of completely dry samples at one value of the inlet pressure. A test pressure of 2.0 bars was used in this study. The apparent coefficient of permeability (K_A) was calculated for laminar flow of a compressible viscous fluid through a porous material using the Hagen–Poiseuille relationship [\[10](#page-8-0)[,12](#page-8-2)].

$$
K_{\rm A} = \frac{2\mu Q P_1 L}{A (P_0^2 - P_1^2)}\tag{4}
$$

Where *Q* is the measured gas flow (m^3/s), μ the dynamic viscosity of helium (N s/m^2), *L* the thickness of the sample (m), *P*⁰ atmospheric pressure (Pa), *A* the cross-sectional area of the sample (m^2) , and P_1 the absolute pressure applied (Pa).

4 Results and Discussions

4.1 Concrete Properties in the Fresh State and Compressive Strength in the Hardened State

The preliminary results shown in Table [3](#page-4-0) relate to fresh concrete properties and compressive strength at 28, 90 and 360 days.

Experimental results show that, generally, the SCC mixtures present a good filling ability (slump-flow was equal to 67 ± 2 cm, V-funnel flow times ranged 5.8–7 s) and an acceptable passing ability (L-box results ranged 0.8–0.9, fill-box values were between 79 and 96.2 %). Therefore, fresh SCC properties complied with SCC recommendations, which is an indication of good deformability.

From the compressive strength results at 28, 90 and 360 days, we can see that the 28-day values are closer to the desired strength classes. This result confirms the reliability of the mix design method used.

A significant difference was recorded for other maturities. For the results obtained after 360 days of curing in particular, the difference is of 20.1, 16.8 and 3.5 MPa between SCC30 FA and OVC30, SCC30 LF and SCC30PZ, respectively. This

Fig. 3 Variation of sorptivity coefficient as a function of compressive strength

is probably due to the pozzolanic reaction of fly ash and Pozzolan from a natural source in Algeria.

4.2 Sorptivity

Figure [3](#page-4-1) shows the sorptivity coefficient results as a function of compressive strength

The comparison between different mix designs shows two almost parallel spindles. The sorptivity coefficients of SCC PZ and SCC FA are systematically lower compared to the SCC LF and the OVC. The significantly lower sorptivity of the SCC mixtures with Pozzolan from a natural source in Algeria or fly ash may be attributed to their less porous interfacial zone, and also the refined pore structure of the paste matrix. According to Ghrici et al. [\[13\]](#page-8-3), the capillary pores are reduced by the formation of secondary C–S–H gel due to the pozzolanic reaction.

We note also that the sorptivity coefficient normally decreases with the increase of compressive strength. For all concretes, we can maintain a polynomial relation between

Fig. 4 Variation of water porosity as a function of compressive strength

sorptivity and compressive strength. Turk et al. [\[14\]](#page-8-4) have already reported a correlation between the development of compressive strength and the decrease in capillary absorption. According to these authors, a longer curing time promotes the formation of a large amount of hydrates that fill and further fracture capillary porosity.

4.3 Water Porosity

According to Assié [\[15\]](#page-8-5), the first characteristic that should be represented as a function of mechanical strength is water porosity. This parameter is directly related to the compressive strength of concrete [\[16](#page-8-6)].

Figure [4](#page-4-2) shows the variation of water porosity as a function of compressive strength.

From the results obtained from SCC mix designs, we can see that the water porosity of SCC PZ (Pozzolan from a natural source in Algeria) and SCC FA is lower than that of SCC LF. The difference tends to increase with the increase in compressive strength, especially for greater strengths $(\geq 60$ MPa); the difference between SCC PZ–SCC FA and SCC LF varies between 1.3 and 3.1 %.

Fig. 5 Comparison of pore size distribution for 70 MPa strength class concretes

Fig. 6 Comparison of pore size distribution for 50 MPa strength class concretes

Fig. 7 Comparison of pore size distribution for 30 MPa strength class concretes

However, compressive strength is not the only parameter that governs the water porosity of concrete. Despite the low resistance of OVC to the corresponding SCC, the porosity of OVC mix designs was lower compared to SCC with various admixtures. The pore-size distribution is different in these two types of concrete [\[15\]](#page-8-5). The high volume of superplasticizer in the composition of SCC (presented in Table [2\)](#page-2-0) may explain the high water porosity of this concrete.

4.4 Mercury Intrusion Porosimetry

Porosity distribution curves as a function of pore size are presented in Figs. [5,](#page-5-0) [6](#page-5-1) and [7.](#page-5-2)

Fig. 8 Apparent chloride diffusion coefficient as a function of compressive strength

From the curves of Figs. [5,](#page-5-0) [6](#page-5-1) and [7,](#page-5-2) we observe the existence of the same porous mode in all concrete strength classes. The main peak corresponds to the diameter located between 0.01 and 0.1 μ (the peak is located between 0.04 and 0.05μ for the 70 MPa strength class, from 0.03 to 0.04 μ for the 50 MPa class and between 0.03 and 0.08 μ for the 30 MPa strength class). This family corresponds to an intrinsic characteristic of pore distribution in cement pastes. Despite the class 30 MPa, in which we can see a small shift for the SCC FA, the SCC PZ (Pozzolan from a natural source in Algeria) shows a porous distribution quite similar to that of SCC FA and SCC LF, especially for the 50 and 70 MPa strength classes.

4.5 Chloride Diffusivity

Figure [8](#page-5-3) shows the apparent chloride diffusion coefficient results as a function of compressive strength obtained at the age of 28, 90 and 360 days of curing.

The apparent chloride diffusion coefficient results clearly indicate that chloride diffusivity was strongly affected by the change in compressive strength. Normally, the diffusion coefficient decreases with the increase in compressive strength. This conclusion is in agreement with results reported in previous studies [\[17](#page-8-7)[–19\]](#page-8-8). Roziere [\[19\]](#page-8-8) adds that for the binder, increased compactness, expressed in terms of water/binder, always leads to a decrease in apparent chloride diffusion.

When comparing concrete as a function of the mineral admixtures, we can see two different results. For the concretes with a compressive strength of <65 MPa, the SCC mixtures containing fly ash or Pozzolan from a natural source in Algeria showed much lower migration coefficients compared to those of SCC mixtures with limestone filler or OVC (difference $>50\%$ for results of the 30 MPa strength class). The difference tends to stabilize with the increase in compressive strength. At strengths >65 MPa, the chloride diffusion coefficient curves for the different concretes form almost a single spindle. However, we can say that SCC PZ and SCC FA perform better than SCC LF and OVC. The relatively

Fig. 9 Apparent gas permeability as a function of compressive strength

low permeability coefficient of the SCC mixtures with fly ash is in agreement with previous findings [\[17](#page-8-7),[20\]](#page-8-9) to the effect that the permeability of SCC at the same strength level could be halved with the use of fly ash as addition in SCC paste. The significant reduction in chloride diffusivity due to the incorporation of fly ash may be partly explained by the improvement of density, both in the matrix and in the interfacial zone. Also, the pozzolanic reaction of Pozzolan from a natural source in Algeria and fly ash leads to the formation of pozzolanic C–S–H, which fills a portion of the capillary porosity and leads to a decrease in the diffusion coefficient [\[21](#page-8-10)].

The difference between the results of SCC mixtures with natural Pozzolan addition and SCC mixtures with fly ash addition is negligible (0.09, 0.19 and 0.31 m^2 /s respectively for 70, 50 and 30 MPa strength classes).

4.6 Gas Permeability

The average coefficient of gas (helium) permeability, obtained from three identical specimens, and measured at 2 bars pressure head, are shown as a function of compressive strength, in Fig. [9.](#page-6-0)

From Fig. [9,](#page-6-0) we note, firstly, that the gas permeability normally decreases with the increase in compressive strength. A lower compressive strength will lead to a more accessible pore structure and greater gas transport properties [\[22](#page-8-11)]. According to Torrent and Jornet [\[23\]](#page-8-12), regardless of concrete composition (*W*/*B* ratio, mineral admixtures), compressive strength seems to be a good indicator of the level of air permeability.

Also, the results clearly indicate that for the different concretes, all SCC mixtures had significantly lower permeability coefficients than OVC concrete mixtures. This was particularly noticeable for concretes with a compressive strength lower than 65 MPa. The K_{app} of OVC is more than two times higher compared to SCC mixtures with different additions. This important difference between SCC and OVC may be due to the difference in pore structure, and in relation to this, to the assumption of laminar flow through the pore system [\[24](#page-8-13)]. This important difference between SCC and OVC is in agreement with those of Assié [\[16](#page-8-6)] and Boel et al. [\[24\]](#page-8-13). Boel et al. [\[24\]](#page-8-13) add that the most likely answer can be found in the pore structure (size, density, etc.) and the appearance of several types of flows, such as sliding, laminar and turbulent flow of gas within the intermediate pore system. We can also explain this difference with the amount of paste. Tahlaiti et al. [\[25](#page-8-14)] found that, even for several formulations of SCC, the Kapp decreases as a function of the increase in paste.

When comparing SCC mixtures of the same strength class, we notice that the results curves are more or less close. However, the mix designs of SCC PZ and SCC FA have a slightly lower permeability compared to those of SCC LF. The difference seems to be stable (parallel curves) for all different compressive strengths. The incorporation of Pozzolan from a natural source in Algeria or fly ash in SCC paste, generally allows for a decrease in the gas permeability of SCC. This is probably related to the denser microstructure, explained by the refinement and segmentation of the capillary porosity caused by the hydration of natural Pozzolan and fly ash particles (seed crystals).

The gas permeability results of SCC PZ and SCC FA are very close. The curves are nearly superimposed.

5 Relation Between Chloride Diffusion Coefficients and Sorptivity Coefficients

Figure [10](#page-6-1) illustrates the relationship between chloride diffusion coefficients and sorptivity coefficients.

The results reveal an exponential relation between the chloride diffusion coefficients and those of sorptivity. The coefficient of correlation equals 0.87. It can be concluded, therefore, that the chloride diffusion coefficients increase with the increase in sorptivity coefficients (lower sorptivity indicates lower chloride ion diffusion). This result is in agreement with that of Ganesan et al. $[26]$, who found a good correlation between diffusivity and sorptivity. This can be explained by the method of chloride penetration into concrete. According to

Fig. 10 Variation of chloride diffusion coefficients as a function of sorptivity coefficients

Fig. 11 Variation of gas permeability coefficients as a function of chloride diffusion coefficients

GCI [\[27](#page-8-16)], chloride ions as a solution are first "drawn" into the pores along with the water absorbed. Beyond the absorption zone, chloride penetration occurs by diffusion.

6 Relation Between Apparent Gas Permeability Coefficients and Chloride Diffusion Coefficients

Previous studies [\[28,](#page-8-17)[29\]](#page-8-18) have already mentioned the existence of a high correlation between gas permeability and chloride-ion diffusions.

Figure [11](#page-7-8) illustrates the relationship between gas permeability coefficients and chloride diffusion coefficients of concretes studied.

First, we can see that gas permeability increases with the increase in chloride ion diffusion. This confirms the trend seen in other studies and the existence of a close relationship between permeability and diffusion. Independently of the type of mineral admixtures in SCC and the type of concrete, the correlation coefficient is equal to 0.92. However, one point deviates significantly from the trend line. This is the OVC 30, which has a relatively high permeability with a separation of approximately two orders of magnitude compared to the trend. According to Sugiyama et al. [\[29\]](#page-8-18), the correlation between permeability coefficients and those of diffusion is due to the fact that these two physical properties are largely related to the same factor (*W*/*B*).

7 Conclusion

The objectives of this paper were to valorise the Algerian natural Pozzolan in an economical SCC and to compare the permeation properties of this concrete to other SCC mixtures with limestone filler and fly ash.

From the multiple results obtained, several correlations have been made.

The following conclusions are drawn from the experimental results reported in this paper:

- 1. The type of mineral admixture considerably influences the permeation properties of SCC. The SCC PZ had coefficients very similar to those of SCC FA and significantly lower than those of SCC LF.
- 2. There is a close relationship between permeation properties and compressive strength of SCC. The sorptivity, water porosity, chloride diffusion, and gas permeability coefficients decrease with the increase in compressive strength.
- 3. There is an exponential relationship between the chloride diffusion and sorptivity coefficient results, and a polynomial relationship between gas permeability and chloride diffusion.
- 4. Comparing the results of SCC and OVC, we can conclude that sorptivity, chloride diffusion and permeability are less important for SCC than for the corresponding OVC. The difference increases with the decrease in compressive strength.
- 5. Lastly, the incorporation of Pozzolan from a natural source in Algeria is very beneficial in reducing the permeability properties of SCC. Even with very large amounts of natural Pozzolan and high *W*/*B*, SCC PZ helps reduce sorptivity, water porosity, chloride diffusion, and gas permeability coefficients. The pozzolanic reaction has certainly led to the formation of a finer and less porous microstructure.

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