Additives Influence on the Properties of Asphalt Binders: A Case Study

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ABSTRACT

This study investigated the impact of using modified bitumen binders and additives to enhance the physical characteristics of asphalt mixtures at high, medium, and low temperatures. Both basic and rheological characteristics of the bituminous binders were tested by adding Styrene-Butadiene-Styrene, a type of thermoplastic elastomer, as a chemical additive to asphalt mixtures at various fractions (3.5%, 4%, and 4.5%), and two kinds of modified Bitumen: Kraton styrene block copolymers and Europrene (SOL T 6302). The following methods were used to test the properties of the binders: needle penetration, ductility, softening point, resistance to hardening when exposed to heat and air, Pressure Aging Vessel test dynamic (PAV), Shear Rheometer (DSR), Bending Beam Rheometer (BBR), direct tension test, and storage stability. The results showed that the best properties of the asphalt concentration were observed when using 4% of additives, while the penetration of Bitumen was reduced to 27.2% and 30.3% when adding Kraton and Europrene SOL T6302, respectively. The influence of adding 4.0% additives decreased the bulk specific gravity to 2.395. Laboratory tests showed how the mentioned chemical additives directly affect the properties of the mixture and the binder by increasing their viscosity.

Keywords-performance grade; Kraton styrene; Europrene styrene; bitumen; asphalt

I. INTRODUCTION

Additives are frequently used to increase the physical and mechanical characteristics of materials such as oil and asphalt at various temperatures [1-2]. As asphalt is made to sustain outside conditions such as low and high temperatures, water, or loads, some additives are mixed usually with it to increase its strength, such as two distinct types of the Styrene-Butadiene-Styrene (SBS) chemical additives: Kraton styrene block copolymers and Europrene (SOL T6302). SBS is considered a good choice to mix with concrete as it exhibits elastic behavior at ambient temperatures and can be treated similarly to plastic at higher temperatures (thermoplastic elastomer). SBS and other thermoplastics can be stored as rubbers and easily molded into useful objects [3]. In [4], the deformation of rubberized asphalt blended with repeated load conditions was simulated, concluding that adding rubber crumbs to the asphalt mixture can reduce its rut depth by 55%. An experimental analysis was carried out in [5] to improve the mechanical properties of asphalt binders by mixing chemical WMA additives with nano clays, showing that the addition of these modifiers can postpone the aging of asphalt binders. In [6], the free surface energy method was used to examine the impact of additives on asphalt mixtures. The proposed method was validated and can be used accurately to quantify the influence of additives on the asphalt mixtures. Adhesion promoters were used in [7] to reduce moisture-related pavement degradation such as cracking and raveling, and to strengthen the bond between asphalt and aggregates. The performance, structural stability, damage growth, and durability of asphalt pavement are strongly affected by the adhesive bonding between the modified asphalt binder and the aggregates. Furthermore, the interaction between the bitumen and the aggregates can be improved, but the performance of binders in bituminous mixtures is not impaired, as shown in [8].

The cracking resistance and the long-term deformations of modified asphalt mixtures can be assessed using several tests and methodologies. This study aimed to investigate the physical and mechanical properties of asphalt binders after blending them with a certain number of chemical additives and examining how this process can improve compatibility. The geological properties of conventional and modified binders and the properties of the asphalt concrete mixture were evaluated using different tests to determine how to improve the quality of the binder and the asphalt concrete mixture. Furthermore, the different ratio percentages of the additives were compared to choose the best ratio and kind of additives. The findings of this study will benefit the road and pavement industry.

II. EXPERIMENTAL MATERIALS AND FRACTIONS

A. Bitumen

This study used conventional bitumen (AC 40/60) with 0.1mm penetration and two types of modified bitumen: Kraton styrene block copolymers (KRA 40/60) and Europrene SOL T 6302 (EUR 40/60). Both types were mixed with the conventional bitumen and were chosen as a sample of the asphalt binder. The conventional binder type 40-60 was initially assessed using the following tests: Penetration, Ductility (PD), Rolling Thin-Film Oven (RTFO) test, and softening point test. The aim of the chosen bitumen 40-60 was to confirm these chemical additives under evaluation. The bitumen was analyzed to understand its mechanical behavior and adaptability. The modified bitumens by KRA 40/60 and EUR 40/60 were tested by: RTFOT, Pressure Aging Vessel (PAV) test, Dynamic Shear Rheometer (DSR) test, Bending Beam Rheometer (BBR) test, and direct tension test (Recovery). Table I shows the results of these tests.

B. Additives

Kraton SBS (KRA 40/60) and Europrene SOL T 6302 (EUR 40/60) were used as additives, as shown in Figure 1. These additives work at the point where aggregates and bitumen meet, reduce internal friction forces, and ensure that the mixtures are more workable and compatible at lower temperatures [9]. The characteristics of both additives were examined at various percentages (3.5%, 4.0%, and 4.5%) of the binder weight. These additives are classified as thermoplastic elastomers, a type of substance that is similar to plastic but functions more like rubber. These additives were added to the binder. Kraton SBS copolymers act as a surface-active agent that can increase manufacturing temperatures by approximately 180°C, which is required during melting with bitumen AC 40/60 in solid form with a density ranging from 0.864 to 0.878gr/cm^3 at a temperature of 56°C and a flashpoint of 198°C. This additive was considered a helper to avoid strong adhesion between aggregates in [9-10], acting as a lubricant during the mixing, laying, and compacting processes.

TABLE I. EXPERIMENTAL TEST RESULTS FOR UNMODIFIED AND MODIFIED BITUMEN

Properties	AC 40/60	Modified Bitumen					
		Kraton KRA 40/60			Europrene EUR40/60		
		3.5%	4%	4.5%	3.5%	4%	4.5%
Pen at 25^{0}C- ASTM D5-13 (1/10mm) (%)	41.8	31.2	27.2	25	34	32.6	30.3
Softening point- ASTM D36-09 (^{0}C)	52.5	62.3	65.9	67.2	63	69.2	70.9
Bitumen ductility - ASTM D113 (cm)	160	58	35	30	56	34	32
Penetration after heating- ASTM D1754-04 (%)	63.2	-	-	-	-	-	-
Ductility after heating- ASTM D1754-04 (cm)	91	-	-	-	-	-	-
RTFO test (gm)	0.24	-	-	-	-	-	-
Resistance of hardening under the influence; Change in weight (Δ Mass %) of heat and air (RTFO test) -ASTM D1754-04		-0.28	-0.3	0.33	0.33	-0.3	-0.285

Kraton Europrene SOL T6302

Fig. 1. Chemical additive materials.

The chosen chemical additives do not affect the properties of bitumen [11] and do not contract its viscosity [12]. Several tests were conducted to study the performance of hot mixes with these additives, including dynamic modulus, tensile strength, fatigue, flow, and rutting. The addition of KRA 40/60 reduced the aging of the mixture, increased rutting depth without eliminating it at a flash point of 198°C and density of approximately 0.875g/cm^3 [13]. According to [14], this ingredient acts as a lubricant during mixing and compacting operations to prevent strong adhesion between the aggregates. The performance of the hot bituminous mixture including KRA 40/60 was evaluated and compared with the standard additive stated in [15]. This additive was tested for rutting resistance, fatigue life, and moisture sensitivity, showing that hot bituminous mixes performed similarly, and occasionally even better, in terms of fatigue resistance [16]. The EUR 40/60 is a surfactant agent with active adhesion-promoting qualities, has an antioxidant impact, and is capable of lowering manufacturing and compaction temperatures.

C. Mineral Aggregates

Mineral aggregates were examined according to the ASTM C136-06 using: Los Angeles test, sieve analysis of fine and coarse aggregates, and specific gravity and absorption of coarse or fine aggregate. Three different sizes of coarse aggregates, 25, 19, and 12.5mm, were used in this study. The coarse aggregate fractions according to the mentioned nominal sizes were 10, 17, and 27%, respectively, and the crushed aggregate percentage reached 88.3, 94.0, and 96.7%. In addition, the coarse aggregates were mixed with 42% crushed sand and 4% filler. The specific gravity of the three different sizes of coarse aggregates was 2.656, 2.660, and 2.674, and the specific gravity of the crushed sand and the filler was 2.618 and 2.710, respectively, following ASTM C127-12 and C128-12.

III. RESEARCH METHODOLOGY

A. Characterization of Binders

The following tests were used to evaluate the properties of the binders: needle penetration at 25°C by ASTM D5-13, ductility test by ASTM D113-07, softening point (ring and ball method) by ASTM D36-09, resistance to hardening by ASTM D1754-04, PAV dynamic test by ASTM D6373/ASTM D8239, DSR by ASTM D7175, BBR by ASTM D6648, direct tension test elastic recovery of modified bitumen by ASTM D6084, and storage stability by ASTM D6927-06.

The collected data were translated into kinematic viscosity measurements using a rotational viscometer approach [17] on conventional and modified bitumen additives with 1.035 and 1.043Kg/m^3 density, respectively. The test samples were prepared using ASTM D7175 and heated at 175°C for both KRA 40/60 and EUR 40/60 additives. Homogenization was carried out using a glass rod and manually stirring for 2 or 3 minutes after the melting process was finished, which could take up to 4 hours. Finally, the samples were transferred to containers in homogenous liquid form. The experimental protocol was used to determine whether the additives affect the properties of the binders, and the tests were completed quickly to avoid heating-cooling cycles and bitumen aging. Table II shows the test results.

TABLE II. EXPERIMENTAL TEST RESULTS FOR MODIFIED BITUMEN

	Properties	Modified bitumen					
		Kraton KRA 40/60			Europrene EUR40/60		
		3.5%	4%	4.5%	3.5%	4%	4.5%
1	**Viscosity (135C°) - ASTM D4402; (CP; Max 3000 CP).**	1938	2365	2401	2005	2138	2625
2	**Direct tension (Recovery) ASTM D6084 (%).**	77	83.5	85	74	78.5	79.5
3	**DSR ($G^*/\sin\delta$) ASTM D7175 (KPa)** **at 82^{0}C** **at 88^{0}C**	1.22 0.66	1.93 0.80	2.1 0.95	1.22 0.82	1.88 0.87	1.97 0.88
4	**RTFOT (change in weight) (%) ASTM -D1754-04**	-0.28	-0.3	0.33	0.33	-0.31	-0.28
5	**DSR ($G^*/\sin\delta$) ASTM D7175 (KPa)** **at 82^{0}C** **at 88^{0}C**	3.45 1.69	3.81 1.69	4.5 2.3	2.7 1.7	3.36 1.75	3.97 1.85
6	**DSR ($G\times\sin\delta$) ASTM D7175 (KPa) at 40^{0}C**	993	980.9	1000	1380	1500	1372
7	**BBR test - ASTM D6648 Stiffness(MPa)** **at 0^{0}C** **M-Value**	63.7 0.33	76.9 0.31	82 0.29	62 0.35	64.7 0.344	60.8 1.85

B. Mixture Characterization

KRA 40/60 and EUR 40/60 additives were selected to create a bituminous mixture at lower temperatures to confirm their impact. Three additive percentages, i.e. 3.5, 4.0, and 4.5%, were mixed with AC 40/60 bitumen, as shown in Figure 2, and were evaluated using: theoretical maximum specific gravity by ASTM D2041-11, bulk specific gravity by ASTM D2726-11, and Marshall test by ASTM D6927-06.

Fig. 2. Marshall specimens for three different additive percentages.

This study examined the modulus of asphalt concrete which is utilized in the base and intermediate course of road pavements. The bituminous mixture was created using limestone aggregates and an AC 40/60 conventional bitumen at 0.1mm, as shown in Table III.

TABLE III. VALUES OF UNMODIFIED AND MODIFIED ASPHALT CONCRETE MIXTURE

#	Properties	Unmodified	Modified	
		Ref.	Kraton	Europrene
		AC 40/60	KRA40/60	EUR40/60
1	**Theoretical maximum specific gravity - ASTM D 2041 -11 (Gmm)**	2.512	2.512	2.494
2	**Bulk specific gravity (Gmb)- ASTM D2726-11 (Gmb)**	2.409	2.395	2.394
3	**Marshall test ASTM D6927-06** **Optimum asphalt content Pb (%)** **Air Void (%)**	4.45 4	4.6 4	4.6 4
4	**Voids in mineral aggregate VMA (%)** **Void filled with asphalt VFA (%)**	13.05 68	13.75 70	13.65 72
5	**Stability (N)** **Flow (0.25mm)**	17600 11	23900 11.4	24000 11.2
6	**Tensile Strength Ratio (TSR) (%)**	85.76	90.75	90.3

IV. RESULTS

This section presents the characterization results of unmodified and modified bitumen with various percentages of KRA 40/60 and EUR 40/60.

A. Using Standard Bitumen

As seen in Table I, the vast majority of the test results were within the limits, satisfying the requirements for bitumen certification. The penetration was less than 50%, the ductility was more than 100cm, and the softening point was less than 55°C. After the penetration was raised to more than 60%, ductility decreased to less than 100cm (ASTM D1754-04).

B. Using KRA 40/60 with Asphalt

Bitumen samples were tested with additive percentages of 3.5, 4.0, and 4.5%. Only AC 40/60 bitumen samples were used and mixed with asphalt for 180 minutes. The mixture sample was put in the oven for 8 hours at 180°C. Table I shows the results from using the needle penetration and the softening point of all the examined samples. The results show that when the percentage of additives increased, the ductility and penetration of the samples decreased slightly and the softening point slightly increased. The elastic recovery percentage of the samples increased as the additive percentage increased. Table II shows that the viscosity at 135°C increased at the same additive percentages, and Table III shows that the storage stability increased as the percentage of additives increased. This test is important to predict how bitumen will behave after storage and transportation. Finally, the results showed that the additive was still homogenous.

C. Using EUR 40/60 with Asphalt

These samples were also tested at various additive percentages of 3.5, 4.0, and 4.5% after 180 minutes of mixing and 8 hours in the oven at 180°C. Table I shows that penetration decreased slightly and softening point increased when the additive percentage increased. The elastic recovery percentage increased as the additive percentage increased. The risk of polymer compaction during the dynamic viscosity test is high because the normal stability significantly affects the viscosity of the additive. All percentages of additives increased in the RTFO test in terms of penetration and softening point. As the samples were as original bitumen, they cracked before reaching 100mm of extension elastic recovery. The viscosity of the reference AC 40/60 is more affected by the examined samples of EUR 40/60. Figure 3 shows the direct tension test used.

Fig. 3. Marshall specimens for three different additive percentages.

V. RESULT ANALYSIS

The penetration of the specimens with EUR 40/60 showed a significant influence over those with KRA 40/60, and both decreased as the proportion of additives increased. The softening point for both additives increased as their percentage increased, but samples with EUR 40/60 increased more than those with KRA 40/60. The complex shear modulus G^* can be considered the sample's total resistance to deformation when repeatedly sheared. The phase angle δ is the gap between the applied shear stress and the resulting shear strain. An asphalt binder appears stiffer to sustain rutting; therefore, $G^*/\sin\delta$ showed a higher complex shear modulus elastic part. The stiffer the asphalt binder is, the greater the G^* is. To evaluate the rutting of the two additives compared to the standard bitumen, the values of $G^*/\sin\delta$ of fresh asphalt were recorded as greater than or equal to 1.0KPa. For the RTFO test residue, the stated values of $G^*/\sin\delta$ were greater than or equal to 2.2KPa. The lower value of δ lied to the greater elastic portion of G^*. Figure 4 shows the relations of the elastic part of the shear complex modulus. For the KRA 40/60 at 82°C, the shear complex modulus increased more than for EUR 40/60 with increasing additive percentage and pressure to more than 1KPa. The curve graph of KRA 40/60 raised less than EUR 40/60 when pressure was less than 1.0KPa at 88°C.

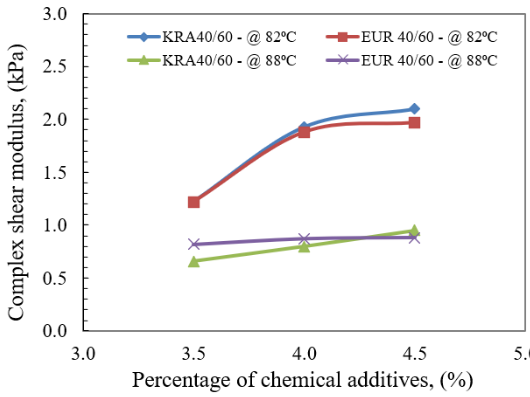

Fig. 4. Variations of complex shear modulus elastic portion with chemical additive percentage.

The shear complex modulus of KAR 40/60 gradually increased with increasing the additive percentage, as shown in Figure 5. At 82°C, the KRA 40/60 results were higher than those of EUR 40/60 and more than 2.2KPa. In other words, the graph line of the shear complex modulus of KRA 40/60 gradually raised with increasing its percentage. At 88°C, the shear complex modulus of KRA 40/60 for 3.5 and 4% was less than 2.2Kpa and EUR 40/60. An asphalt binder became elastic while it is still reasonably flexible to withstand fatigue cracking.

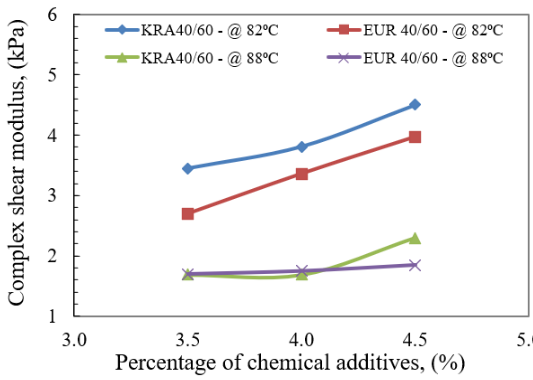

Fig. 5. Variations of complex shear modulus elastic portion with chemical additive percentage after changing weight percent.

The values of $G^*\times\sin\delta$ of the complex shear modulus viscous part were at lower values. The maximum value of the viscous component of the complex shear modulus is used when fatigue cracking is most concerning. Figure 6 shows that the $G^*\times\sin\delta$ value was less than or equal to 5000KPa when evaluating the fatigue distress of typical pavement temperature in the field and the lab at a temperature of 40°C. The fatigue behavior of KRA 40/60 was lower than that of EUR 40/60, and both gradually increased with increasing additive percentages. The complex shear modulus of 4% EUR 40/60 decreased, as shown in Table II.

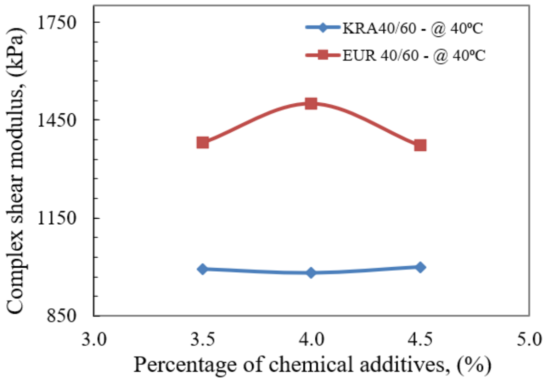

Fig. 6. Variations of complex shear modulus viscous portion with chemical additives percentage.

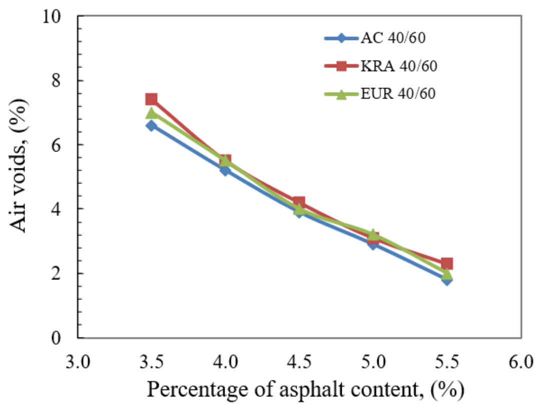

Fig. 7. Variations of air voids with asphalt content percentage.

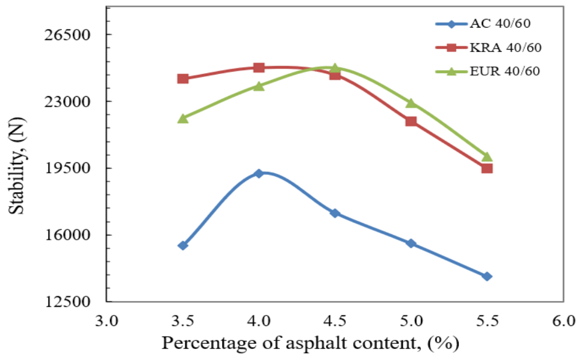

Fig. 8. Variations of stability with asphalt content percentage.

Creep stiffness was computed based on the recorded deflection and the standard beam characteristics. The creep stiffness values observed were less than or equal to 300MPa, and the changing rate of stiffness (*M*-value) was more than or equal to 0.3. The direct tension test was performed to determine compliance with the Performance Grade (PG) standards for asphalt cement if the maximum stiffness is between 300 and 600MPa and the changing stiffness rate is greater than 0.30. The EUR 40/60 samples increased to 4% of additive and then started to decrease. The creep stiffness values of KRA 40/60 samples were greater than those of EUR 40/60. The creep stiffness of both additives was less than 300MPa. The changing rate of the stiffness additives was greater than 0.30, and the *M*-value of the KRA 40/60 samples was lower than that of the EUR 40/60, as shown in Table II. Comparing KRA 40/60 with AC 40/60, its values of variation air voids with the percentage of asphalt content were larger.

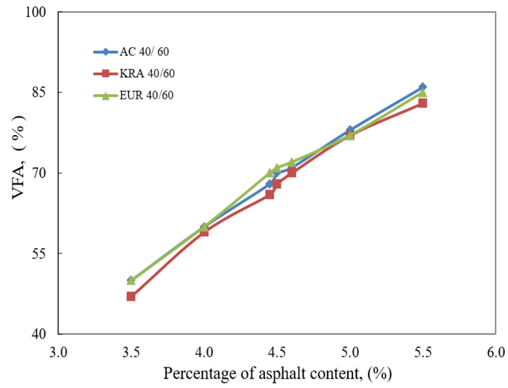

Fig. 9. Variations of percent voids filled with asphalt content.

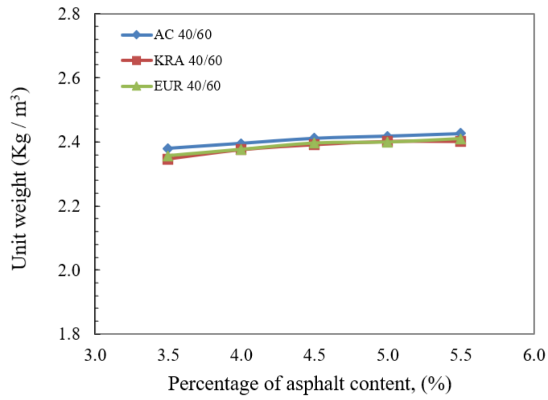

Fig. 10. Variations of unit weight with asphalt content.

The crack propagation of asphalt is rutting at high temperatures, fatigue cracking at intermeddle temperatures, and thermal cracking at low temperatures. According to Figure 7, all curves of air voids decreased as the proportion of asphalt content increased. Figure 8 compares the percentage values of asphalt of the reference bitumen AC 40/60 to the variance stability of the two other additives. As shown in Figure 8, the flow value (0.25mm) of the additives is higher than the reference bitumen AC 40/60. Figure 9 shows that the variance Voids Filled with Asphalt (VFA) rise at increasing values of asphalt percentages, and when this result is compared to the AC 40/60, as shown in Figure 10, that significantly raises the weight of both additive types.

VI. CONCLUSIONS

Based on the outcomes of this study, the tensile strength ratio of modified asphalt with two types of chemical additives

KRA 40/60 and EUR 40/60 showed better properties compared to unmodified asphalt with AC 40/60 bitumen. Adding 4.0% of chemical additives to the AC 40/60, increased its stiffness in parallel with the penetration index and reduced the brittleness of the binder and its temperature sensitivity. This improves rutting resistance, thermal cracking resistance, and resistance to moisture damage. The best additive concentration was observed at 4% additive, which led to less penetration and ductility, and an increased softening point. The penetration of bitumen was 41.8% without additives, reduced to 27.2% with adding KRA 40/60 and 30.3% EUR 40/60, as shown in Table I. The degree of softening increased in both additives, at 65.9ºC for Kraton and 70.9 °C for Europrene SOL T6302, compared to 52.5°C for the AC 40/60. Adding 4.0% of additives increased the asphalt content to 4.6% and decreased bulk specific gravity to 2.395. The maximum specific gravity of Eurprene SOL T6302 decreased to 2.494Kg/m^3, while it was not affected for Kraton. Air voids remained unchanged by adding additives while stability increased.

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