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Spectral, Transmittance and Luminescent Properties of Sm^{3+} Doped Borophosphate Glasses for Visible Lasers and Display Device Applications

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ABSTRACT: Glass of the system: $(35-x)\text{P}_2\text{O}_5:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{V}_2\text{O}_5:10\text{ZrO}_2:10\text{K}_2\text{O}:15\text{B}_2\text{O}_3: x\text{Sm}_2\text{O}_3$ (where $x=1,1.5,2$ mol%) have been prepared by melt-quenching technique. (where $x=1,1.5,2$ mol%) have been prepared by melt-quenching technique. The amorphous nature of the prepared glass samples was confirmed by X-ray diffraction. The Optical absorption, Excitation and fluorescence spectra of three Sm^{3+} doped zinc lithium alumino boromolybdate glasses have been recorded at room temperature. The various interaction parameters like Slater-Condon parameter (F_2), Lande' parameter (ξ_{4f}), nephelauxetic ratio (β') and bonding parameters ($b^{1/2}$) have been computed. Judd-Ofelt intensity parameters and laser parameters have also been calculated.

KEYWORDS: ZLVZBPB Glasses, Transmission Properties, Optical properties, Judd-Ofelt analysis.

I. INTRODUCTION

Rare- earth ion doped glasses have attracted much attention due to some various properties such as high thermal stability, high transparency, a low melting point, a high gain density, low dispersion rates, excellent chemical, mechanical and optical properties [1-5]. Among different host matrices, phosphate glasses have wide range applications in the field of glass ceramics such as high transparency from near UV to mid-Infrared region, good physical chemical and thermal stability [6-10]. They have low melting temperature, low glass transition temperature and low softening temperature. Due to their good chemical durability, samarium-doped ceramic glasses are attractive materials for low-cost integrated optical amplifiers by using the ion-exchange technique. The addition of network modifier (NWF) Li_2O is to improve both electrical and mechanical properties of such glasses. Zinc oxide is added in the glass matrix to increase glass forming ability and to ensure low rates of crystallization in the glass system [11-15].

The aim of the present study is to prepare the Sm^{3+} doped zinc lithium vanadium zirconium potassium borophosphate glasses with different Sm_2O_3 concentrations. The Transmittance, absorption spectra, Excitation, fluorescence spectra of Sm^{3+} of the glasses were investigated. The Judd-Ofelt theory has been applied to compute the intensity parameters Ω_λ ($\lambda=2, 4, 6$). These intensity parameter have been used to evaluate optical optical properties such as spontaneous emission probability, branching ratio, radiative life time and stimulated emission cross section. Large stimulated emission cross section is one of the most important parameters required for the design of high peak power solid state lasers.

II. EXPERIMENTAL TECHNIQUES

Preparation of glasses

The following Sm^{3+} doped zinc lithium vanadium zirconium potassium borophosphate glass samples: $(35-x)\text{P}_2\text{O}_5:10\text{ZnO}:10\text{Li}_2\text{O}:10\text{V}_2\text{O}_5:10\text{ZrO}_2:10\text{K}_2\text{O}:15\text{B}_2\text{O}_3: x\text{Sm}_2\text{O}_3$ (where $x = 1, 1.5, 2$) have been prepared by melt-quenching method. Analytical reagent grade chemical used in the present study consist of $\text{P}_2\text{O}_5, \text{ZnO}, \text{Li}_2\text{O}, \text{V}_2\text{O}_5, \text{ZrO}_2, \text{K}_2\text{O}, \text{B}_2\text{O}_3$ and Sm_2O_3 . They were thoroughly mixed by using an agate pestle mortar. then melted at 1080°C by an electrical muffle furnace for 2h., After complete melting, the melts were quickly poured in to a preheated stainless steel mould and annealed at temperature of 250°C for 2h to remove thermal strains and stresses. Every time fine powder of cerium oxide was used for polishing the samples. The glass samples so prepared were of good optical quality and were transparent. The chemical compositions of the glasses with the name of samples are summarized in Table 1.

Table 1

Chemical composition of the glasses

Sample	Glass composition (mol %)
ZLVZPBP (UD)	35P ₂ O ₅ :10ZnO:10Li ₂ O:10V ₂ O ₅ :10ZrO ₂ :10K ₂ O:15B ₂ O ₃
ZLVZPBP SM (1.0)	34P ₂ O ₅ :10ZnO:10Li ₂ O:10V ₂ O ₅ :10ZrO ₂ :10K ₂ O:15B ₂ O ₃ :1 Sm ₂ O ₃
ZLVZPBP SM (1.5)	33.5P ₂ O ₅ :10ZnO:10Li ₂ O:10V ₂ O ₅ :10ZrO ₂ :10K ₂ O:15B ₂ O ₃ : 1.5 Sm ₂ O ₃
ZLVZPBP SM (2.0)	33P ₂ O ₅ :10ZnO:10Li ₂ O:10V ₂ O ₅ :10ZrO ₂ :10K ₂ O:15B ₂ O ₃ : 2 Sm ₂ O ₃

ZLVZPBP (UD) -Represents undoped zinc lithium vanadium zirconium potassium borophosphate glass specimens.

ZLVZPBP (SM) -Represents Sm³⁺ doped zinc lithium vanadium zirconium potassium borophosphate glass specimens.

III. THEORY

3.1 Oscillator Strength

The intensity of spectral lines are expressed in terms of oscillator strengths using the relation [16].

$$f_{\text{expt.}} = 4.318 \times 10^{-9} \int \epsilon(\nu) d\nu \quad (1)$$

where, $\epsilon(\nu)$ is molar absorption coefficient at a given energy ν (cm⁻¹), to be evaluated from Beer–Lambert law.

Under Gaussian Approximation, using Beer–Lambert law, the observed oscillator strengths of the absorption bands have been experimentally calculated [17], using the modified relation:

$$P_m = 4.6 \times 10^{-9} \times \frac{1}{cl} \log \frac{I_0}{I} \times \Delta\nu_{1/2} \quad (2)$$

where c is the molar concentration of the absorbing ion per unit volume, l is the optical path length, $\log I_0/I$ is optical density and $\Delta\nu_{1/2}$ is half band width.

3.2. Judd-Ofelt Intensity Parameters

According to Judd [18] and Ofelt [19] theory, independently derived expression for the oscillator strength of the induced forced electric dipole transitions between an initial J manifold $|4f^N(S, L) J\rangle$ level and the terminal J' manifold $|4f^N(S', L') J'\rangle$ is given by:

$$\frac{8\pi^2 m c \bar{\nu}}{3h(2J+1)n} \frac{1}{n} \left[\frac{(n^2+2)^2}{9} \right] \times S(J, J') \quad (3)$$

Where, the line strength $S(S', L')$ is given by the equation

$$S(J, J') = e^2 \sum_{\lambda} \Omega_{\lambda} \langle 4f^N(S, L) J \| U^{(\lambda)} \| 4f^N(S', L') J' \rangle^2$$

$\lambda = 2, 4, 6$

In the above equation m is the mass of an electron, c is the velocity of light, $\bar{\nu}$ is the wave number of the transition, h is Planck's constant, n is the refractive index, J and J' are the total angular momentum of the initial and final level respectively, Ω_{λ} ($\lambda = 2, 4, 6$) are known as Judd-Ofelt intensity parameters .

3.3 Radiative Properties

The Ω_{λ} parameters obtained using the absorption spectral results have been used to predict radiative properties such as spontaneous emission probability (A) and radiative life time (τ_R), and laser parameters like fluorescence branching ratio (β_R) and stimulated emission cross section (σ_p).

The spontaneous emission probability from initial manifold $|4f^N(S', L') J'\rangle$ to a final manifold $|4f^N(S, L) J\rangle$ is given by:

$$A[(S', L') J'; (S, L) J] = \frac{64 \pi^2 \bar{\nu}^3}{3h(2J'+1)} \left[\frac{n(n^2+2)^2}{9} \right] \times S(J', J) \quad (4)$$

Where, $S(J, J) = e^2 [\Omega_2 \|U^{(2)}\|^2 + \Omega_4 \|U^{(4)}\|^2 + \Omega_6 \|U^{(6)}\|^2]$

The fluorescence branching ratio for the transitions originating from a specific initial manifold $|4f^N(S', L') J' \rangle$ to a final manifold $|4f^N(S, L) J \rangle$ is given by

$$\beta[(S', L') J'; (S, L) J] = \sum_{S, L, J} \frac{A[(S', L)]}{A[(S', L') J' (\bar{S}, \bar{L})]} \quad (5)$$

where, the sum is over all terminal manifolds.

The radiative life time is given by

$$\tau_{rad} = \sum_{S, L, J} A[(S', L') J'; (S, L)] = A_{Total}^{-1} \quad (6)$$

where, the sum is over all possible terminal manifolds. The stimulated emission cross-section for a transition from an initial manifold $|4f^N(S', L') J' \rangle$ to a final manifold $|4f^N(S, L) J \rangle$ is expressed as

$$\sigma_p(\lambda_p) = \left[\frac{\lambda_p^4}{8\pi c n^2 \Delta\lambda_{eff}} \right] \times A[(S', L') J'; (\bar{S}, \bar{L}) \bar{J}] \quad (7)$$

where, λ_p the peak fluorescence wavelength of the emission band and $\Delta\lambda_{eff}$ is the effective fluorescence line width.

3.4 Nephelauxetic Ratio (β') and Bonding Parameter ($b^{1/2}$)

The nature of the R-O bond is known by the Nephelauxetic Ratio (β') and Bonding Parameters ($b^{1/2}$), which are computed by using following formulae [20, 21]. The Nephelauxetic Ratio is given by

$$\beta' = \frac{v_g}{v_a} \quad (8)$$

where, v_a and v_g refer to the energies of the corresponding transition in the glass and free ion, respectively. The values of bonding parameter $b^{1/2}$ are given by

$$b^{1/2} = \left[\frac{1-\beta'}{2} \right]^{1/2} \quad (9)$$

IV. RESULT AND DISCUSSION

4.1 XRD Measurement

Figure 1 presents the XRD pattern of the sample contain P_2O_5 which is show no sharp Bragg's peak, but only a broad diffuse hump around low angle region. This is the clear indication of amorphous nature within the resolution limit of XRD instrument.

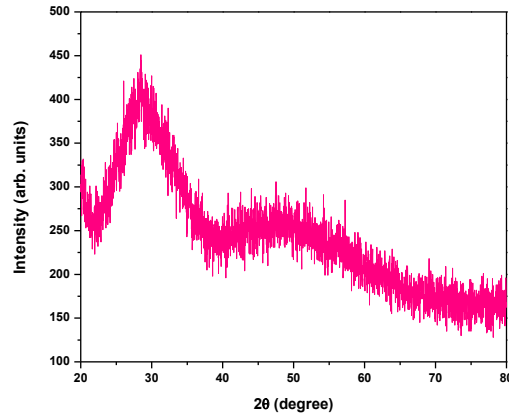


Fig. 1: X-ray diffraction pattern of ZLVZPBP SM(1.0) glass.

4.2 Transmittance Spectrum

The Transmittance spectrum of ZLVZPBP SM(1.0) doped in zinc lithium vanadium zirconium potassium borophosphate glass is shown in Figure 2.

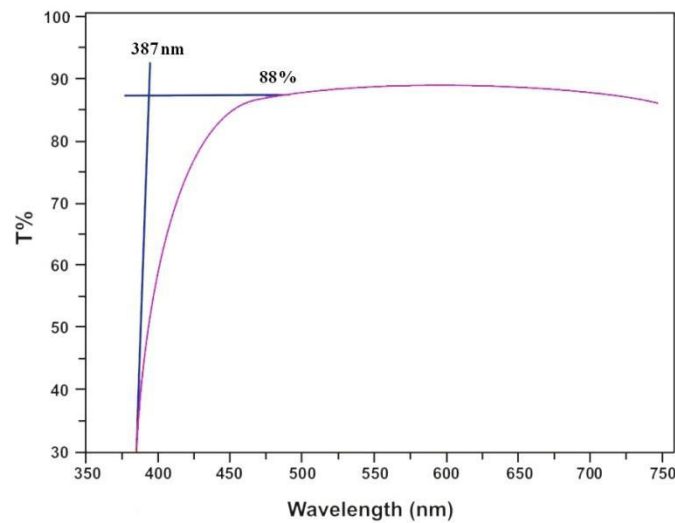


Fig. 2: Transmittance spectrum of ZLVZPBP SM(1.0) glass.

4.3 Absorption Spectrum

The absorption spectra of Sm^{3+} doped ZLVZPBP SM(1.0) glass specimen has been presented in Figure 3 in terms of optical density versus wavelength (nm). Ten absorption bands have been observed from the ground state $^6\text{H}_{5/2}$ to excited states $^6\text{F}_{1/2}$, $^6\text{F}_{7/2}$, $^6\text{F}_{9/2}$, $^4\text{G}_{7/2}$, $^4\text{I}_{9/2}$, $^4\text{M}_{7/2}$, $(^6\text{P}, ^4\text{P})_{5/2}$, $^4\text{F}_{7/2}$, $^4\text{D}_{1/2}$, and $(^4\text{D}, ^6\text{P})_{5/2}$ for ZLVZPBP SM(1.0) glass.

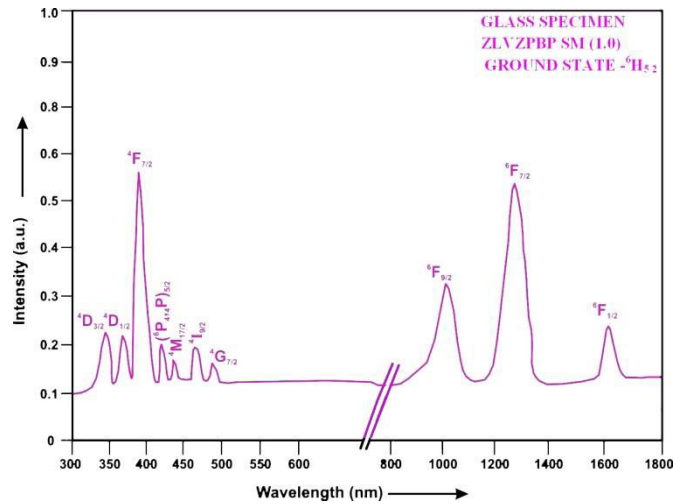


Fig.3: Absorption spectrum of ZLVZPBP SM(1.0) glass.

The experimental and calculated oscillator strengths for Sm^{3+} ions in zinc lithium vanadium zirconium potassium borophosphate glasses are given in Table 2.

Table2: Measured and calculated oscillator strength ($P_m \times 10^{+6}$) of Sm^{3+} ions in ZLVZPBP glasses.

Energy level from ${}^6\text{H}_{5/2}$	Glass ZLVZPBP SM(1.0)		Glass ZLVZPBP SM(1.5)		Glass ZLVZPBP SM(2.0)	
	P_{exp}	P_{cal}	P_{exp}	P_{cal}	P_{exp}	P_{cal}
${}^6\text{F}_{1/2}$	1.66	1.70	1.63	1.67	1.60	1.66
${}^6\text{F}_{7/2}$	5.48	5.53	5.45	5.51	5.41	5.49
${}^6\text{F}_{9/2}$	3.85	3.86	3.81	3.84	3.76	3.82
${}^4\text{G}_{7/2}$	0.18	0.12	0.16	0.12	0.14	0.12
${}^4\text{I}_{9/2}, {}^4\text{M}_{15/2}, {}^4\text{I}_{11/2}$	1.20	1.89	1.16	1.88	1.13	1.87
${}^4\text{M}_{17/2}, {}^4\text{G}_{9/2}, {}^4\text{I}_{15/2}$	0.30	0.25	0.37	0.25	0.23	0.24
$({}^6\text{P}, {}^4\text{P})_{5/2}, {}^4\text{L}_{13/2}$	1.38	1.30	1.35	1.30	1.13	1.30
${}^4\text{F}_{7/2}, {}^6\text{P}_{3/2}, {}^4\text{K}_{11/2}$	5.61	5.61	5.57	5.61	5.51	5.59
${}^4\text{D}_{1/2}, {}^6\text{P}_{7/2}, {}^4\text{L}_{17/2}$	2.54	2.45	2.51	2.43	2.48	2.42
${}^4\text{D}_{3/2}, ({}^4\text{D}, {}^6\text{P})_{5/2}$	2.67	3.47	2.62	3.46	2.57	3.45
r.m.s. deviation	0.3379		0.3526		0.3704	

Computed values of F_2 , Lande' parameter (ξ_{4f}), Nephelauxetic ratio (β') and bonding parameter ($b^{1/2}$) for Sm^{3+} doped ZLVZPBP glass specimen are given in Table 3.

Table 3. F_2, ξ_{4f}, β' and $b^{1/2}$ parameters for Samarium doped glass specimen.

Glass Specimen	F_2	ξ_{4f}	β'	$b^{1/2}$
Sm^{3+}	358.82	1258.16	0.9337	0.1821

The value of Judd-Ofelt intensity parameters are given in Table 4

Table4: Judd-Ofelt intensity parameters for Sm³⁺ doped ZLVZPBP glass specimens.

Glass Specimen	$\Omega_2(\text{pm}^2)$	$\Omega_4(\text{pm}^2)$	$\Omega_6(\text{pm}^2)$	Ω_4 / Ω_6	Ref.
ZLVZPBP SM(1.0)	5.333	4.703	4.331	1.086	P.W.
ZLVZPBP SM(1.5)	5.259	4.699	4.302	1.092	P.W.
ZLVZPBP SM(2.0)	5.208	4.682	4.278	1.094	P.W.
YZLSLCBG (TB)	5.942	2.956	2.694	1.097	[22]
ZLCMBP (DY)	2.790	1.715	1.354	1.267	[23]

4.4 Excitation Spectrum

Excitation spectra of ZLVZPBP SM (1.0) glass recorded at the emission wavelength 602 nm is depicted as figure 4. The excitation spectra consists of seven peaks corresponding to the transitions from the ground state ⁶H_{5/2} to the various excited states ⁴H_{9/2}, ⁴D_{3/2}, ⁶P_{7/2}, ⁴F_{7/2}, ⁴M_{19/2}, ⁴G_{9/2} and ⁴I_{11/2}+⁴I_{13/2} at the wavelengths of 335, 361, 375, 401, 412, 435 and 478 nm respectively. Among these, a prominent excitation band at 401 nm has been selected for the measurement of emission spectrum of ZLVZPBP SM(1.0) glass.

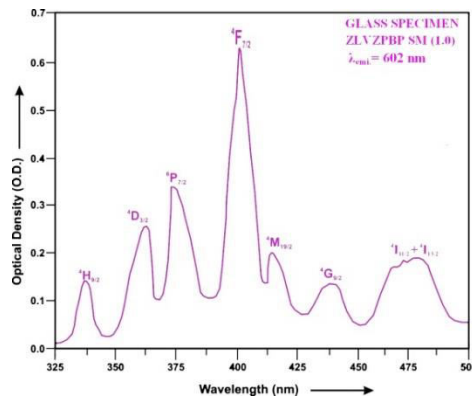


Fig.4: Excitation spectrum of ZLVZPBP SM(1.0) glass.

4.5 Fluorescence Spectrum

The fluorescence spectrum of Sm³⁺ doped in zinc lithium vanadium zirconium potassium borophosphate glass is shown in Figure 5. There are nine broad bands observed in the Fluorescence spectrum of Sm³⁺ doped zinc lithium vanadium zirconium potassium borophosphate glass. The wavelengths of these bands along with their assignments are given in Table 5. Fig. (5).Shows the fluorescence spectrum with nine peaks (⁴G_{5/2}→⁶H_{5/2}), (⁴G_{5/2}→⁶H_{7/2}), (⁴G_{5/2}→⁶H_{9/2}), (⁴G_{5/2}→⁶H_{11/2}), (⁴G_{5/2}→⁶H_{13/2}), (⁴G_{5/2}→⁶F_{3/2}), (⁴G_{5/2}→⁶F_{5/2}), (⁴G_{5/2}→⁶F_{7/2}) and (⁴G_{5/2}→⁶F_{9/2}), respectively for glass specimens.

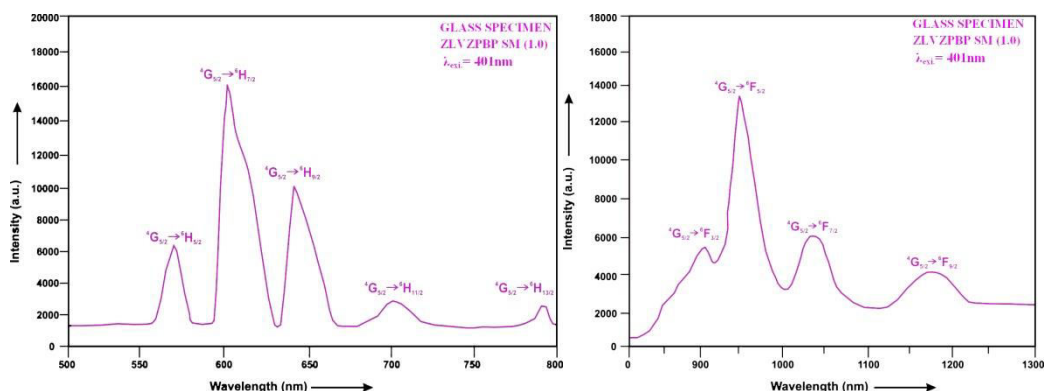


Fig.5: fluorescence spectrum of ZLVZPBP SM(1.0) glass.

Table 5. Emission peak wave lengths (λ_p),radiative transition probability (A_{rad}),branching ratio(β),stimulated emission cross-section(σ_p) and radiative life time(τ_R) for various transitions in Sm³⁺ doped ZLVZPBP glasses.

Transition	ZLVZPBP SM(1.0)					ZLVZPBP SM(1.5)					ZLVZPBP SM(2.0)				
	λ_{max} (nm)	$A_{rad}(s^{-1})$	β	$\sigma_p (10^{-20} cm^2)$	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	β	$\sigma_p (10^{-20} cm^2)$	$\tau_R(\mu s)$	$A_{rad}(s^{-1})$	β	$\sigma_p (10^{-20} cm^2)$	$\tau_R(\mu s)$		
⁴ G _{5/2} → ⁶ H _{5/2}	562	10.29	0.0367	0.0038	3567.27	10.26	0.0368	0.0041	3583.69	10.23	0.0368	0.0045	3598.31		
⁴ G _{5/2} → ⁶ H _{7/2}	602	107.51	0.3835	0.0434		107.31	0.3846	0.0463		107.02	0.3851	0.0491			
⁴ G _{5/2} → ⁶ H _{9/2}	645	105.33	0.3757	0.0330		104.57	0.3747	0.0339		104.01	0.3742	0.0350			
⁴ G _{5/2} → ⁶ H _{11/2}	705	26.36	0.0940	0.0129		26.34	0.0944	0.0134		26.28	0.0946	0.0139			
⁴ G _{5/2} → ⁶ H _{13/2}	786	2.52	0.0090	0.0017		2.51	0.0090	0.0018		2.50	0.0090	0.0018			
⁴ G _{5/2} → ⁶ F _{3/2}	915	4.41	0.0157	0.0069		4.36	0.0156	0.0072		4.33	0.0156	0.0075			
⁴ G _{5/2} → ⁶ F _{5/2}	955	19.76	0.0705	0.0314		19.56	0.0701	0.0323		19.42	0.0699	0.0327			
⁴ G _{5/2} → ⁶ F _{7/2}	1036	2.38	0.0085	0.0042		2.38	0.0085	0.0043		2.38	0.0086	0.0045			
⁴ G _{5/2} → ⁶ F _{9/2}	1180	1.77	0.0063	0.0043		1.76	0.0063	0.0043		1.74	0.0063	0.0044			

V. CONCLUSION

In the present study, the glass samples of composition : (35-x)P2O5: 10ZnO: 10Li2O: 10V2O5: 10ZrO2: 10K2O: 15B2O3: xSm2O3. (where x=1, 1.5, 2mol %) have been prepared by melt-quenching method. The Judd-Ofelt theory has been applied to calculate the oscillator strength and intensity parameters $\Omega\lambda$ ($\lambda=2, 4, 6$). The stimulated emission cross section (σ_p) has highest value for the transition (⁴G_{5/2}→⁶H_{7/2}) in all the glass specimens doped with Sm³⁺ ion. This shows that (⁴G_{5/2}→⁶H_{7/2}) transition is most probable transition, therefore these glass samples useful for Visible lasers and Display device Applications.

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