

Effect of stable weak magnetic field on Cr(VI) bio-removal in anaerobic SBR system

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Abstract To study the impact of stable weak magnetic field on the Cr(VI) removal efficiency of predominated strains in ASBR system, the choice of the optimum magnetic density and its effect should be considered chiefly. At different magnetic densities, the growth and propagation rates of predominated strains in solid or liquid mediums and their capabilities of removing Cr(VI) were compared. The results showed that the optimum magnetic density was 6.0 mT. To meet the state first-class standard of effluent discharge, it took 2–5 h more in the plant wastewater treatment than in the synthetic wastewater treatment, but the presence of magnetic field made the reaction time up to par to decrease 1 and 2–3 h, respectively, compared with that of the control. The magnetized magnetic powder could improve the sludge sedimentation capability, turbidity of outflow water and efficiency of bio-system.

Keywords Cr(VI) bio-removal · Magnetic field · Microorganism · Wastewater containing Cr(VI)

Introduction

Presently, accumulation of heavy metals by microbe is a widely concerned phenomenon, and toxic metals removal or valuable metals enrichment and regeneration from industrial wastewater by living or abiotic biomass are also one of the most interesting research fields (Ferraz et al. 2004.). The mechanisms of those processes are associated with complexation, ion exchange, coordination, adsorption, chelation, and microprecipitation which may be synergistically or independently involved. It is generally believed that microbial heavy metal accumulation involves a combination of active and passive transport phases. An initial rapid phase involves physical adsorption or ion exchange at cell surface and a subsequent slower phase involves active metabolism-dependent transport of metal into bacterial cells, such as particulate metal granules formed in specific organelles (Tadashi and Yoshiko 2003). While biosorbable uptake by using non-living biomass is a whole passive process (Zouboulis et al. 2004).

As a trace metal element, chromium is an essential nutrient for numerous organisms, but it is toxic, mutagenic, and carcinogenic with the uptake levels elevating. Cr(VI) is more toxic than Cr(III) and requires more concern. Strong exposure of Cr(VI) causes cancer in digestive tract or lung and may cause epigastric pain, nausea, vomiting, severe diarrhea, and hemorrhage (Shakoori et al. 2004). Cr(VI) causes two types of dermatological toxicities: allergic

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contact dermatitis (ACD) and skin ulcers. Repeated exposure to Cr(VI) in concentrations of 0.004–0.025 mg l⁻¹ can both induce sensitization and elicit chromium ACD. Exposure to 0.020 mg l⁻¹ Cr(VI) can cause skin ulcers in nonsensitized people. Some investigators recommend reducing the Cr(VI) concentrations in consumer products, such as detergents, to less than 0.005 mg l⁻¹ (Shelnutt et al. 2007). If untreated wastewater containing Cr(VI) was directly discharged to environment, the environment and ecosystem would be polluted and people's health would be endangered seriously (Trumble and Jensen 2004). Therefore, it is essential to remove Cr(VI) from wastewater before disposal. Due to severe toxicity of Cr(VI), the EU Directive and US EPA have set the maximum contaminant concentration level for Cr(VI) in domestic water supplies to be 0.05 mg l⁻¹.

The main sources of Cr(VI) in environment include the effluents discharged from electroplating, tan, pottery, and paint, ink, dye manufacturing plants. In general, Cr(VI) is present mainly as either dichromate (Cr₂O₇²⁻) in acidic solution or as chromate (CrO₄²⁻) in the alkaline (Srinath et al. 2002; Rajender et al. 2007). The wastewater with low concentrations of Cr(VI) is usually treated with ion-exchange resins, but the high cost of the resin limits its application (Rengaraj et al. 2001). And a lot of sludge due to chemical sedimentation may cause secondary pollution (Jang et al. 2001). As compared, bio-removal of Cr is one of the most respective methods with the merits of high efficiency, low cost and safety, and which is becoming a hot research field.

To improve cell accumulation capability, researchers have taken several methods, such as screening high-efficiency strains, altering morphological and physiological features by genetical manipulation (Zouboulis et al. 2004). According to the toxicity of Cr(VI), it is not enough to remove the Cr(VI) from liquids only, to reduce it to nontoxic or less toxic valence, such as Cr(III), and eliminate them from the liquids can be considered as the most thorough method (Melo and D'Sousa 2004). The accumulated Cr(VI) may act as terminal electron acceptor and the reduced Cr(III) then binds to cell wall (Guan et al. 1993a, b). Cr(VI) was quantitatively transferred to Cr(III), which mainly existed in the external medium of *E. coli* ATCC 33456 cultures. The dissimilatory

Cr(VI) reduction was found to be a largely soluble reductase activity as a result of cometabolism in *E. coli* ATCC 33456. The respiratory-chain-linked electron transport to Cr(VI) was also involved in Cr(VI) reduction, although it requires the presence of soluble reductase to mediate electron transport to Cr(VI) (Shen and Wang 1993).

There were still some problems with previous work on Cr(VI) bio-removal, for example, the instability of treatment efficiency, the slowness of the strains growing and reproducing in the anaerobic environments. When the Cr(VI) concentration reached 75.53 mg l⁻¹, there was a little decrease in treatment efficiency, and with increase of Cr(VI) concentration from 64.66 to 95.47 mg l⁻¹, the treatment efficiency decreased from 92.64 to 71.18%, and the treatment time up to par also increased from 3.5 to 7.5 h (Deng et al. 2004; Xu et al. 2005a, c; Xu and Sun 2005b). Of course, the toxicity of Cr(VI) is still the most important reason. It can restrain the breeding of microorganism and make the system running unsteadily. As a result, the effluents hardly meet to the stricter and stricter environmental protection demand.

A great deal of researches showed that magnetized water has higher pH and electric conductivity than general tap water. In addition, magnetization can cause a higher osmotic pressure of water and stronger permeability through cell membrane (Boleslaw 1985; Lednev 1991). Static magnetic fields may cause DNA damage and strong magnetic fields can induce orientation phenomena in cell culture (Junji 2005). The soil magnetized at 0.15–0.35 T showed higher respiration rate, invertase activity, and fertility capability (Zhang and Yu 1999). Magnetic powder of 4000 mg l⁻¹ can increase the growth rate and Cu removal rate of the predominated strain (Tu et al. 2004). Moreover, direct function of some magnetic field on intracellular water and substance could activate cytoenzyme and accelerate the bio-chemical reaction in creature bodies (Liboff et al. 2003). The biodegradation capability of the activated sludge reaches the max. at 18 mT (Yavuz and çelebi 2000). The impacts of low-level electromagnetic environment and strong magnetic field on human health have also been concerned widely (Martin and Nino 2006). Duration of exposure varied up to 24 min, a 50 Hz 10 mT magnetic field decreases the number of yeasts *Saccharomyces cerevisiae* at laboratory temperature

(24–26°C), and the result is similar to the experiments with bacteria *E. coli*, *S. aureus*, and *L. adecarboxylata* (Novák et al. 2007). So for some microorganism, only a suitable magnetic field could promote microorganism abilities of nutrients absorption and utilization, and consequently accelerate their growth rate.

As an effective method, magnetic separation technique has been applied to the treatment of many wastewaters, such as oil-bearing effluent, mine effluent, and there are also some new complex processes, such as magnetic chemistry process (Zhao et al. 2003). Few researches have been focused on the application of magnetic field to bio-system (Yavuz and Çelebi 2003; Tomska and Janosz 2004) and some preponderant strains (Shu et al. 2005; Hu et al. 2006) showed that it was possible to improve the stability and efficiency of Cr(VI)-containing wastewater treatment by setting a special magnetic field in Cr(VI) bio-removal system. It was obvious that the magnetic field might be one of the most hopeful bio-strengthening methods applied to enhance the functions of living cells.

A survey of literatures indicated that not much work had been done so far on magnetic field for Cr(VI)-containing wastewater bio-treatment. Therefore, our study aimed to select optimum magnetic density, at which the high-efficiency bacteria reached the maximum growing and reproducing rate, and apply it in the bio-treatment of wastewater containing Cr(VI).

Experiment materials

Strains and sludge for tests

Strains A (*Brevibacillus* sp.) and B (*Bacillus* sp.) are two high-efficiency Cr(VI) removal bacteria, which were screened in our previous researches (Deng et al. 2004; Xu et al. 2005a, c; Xu and Sun 2005b) and some of their main morphologic characters are listed in Table 1. The anaerobic sludge used in the experiments below was obtained from a municipal sewage

Table 1 The colony configurations of these two predominated strains

	Color	Shape	Edge	Size	Location
A	Buff	Round	Slick	Needle	Surface
B	Buff	Erose	Lacelike	About 0.2 cm	Surface

plant in Guangzhou, to which the enough predominated strains was inoculated.

Sources and qualities of Cr(VI)-containing wastewater

Synthetic wastewater made of potassium dichromate contains some nutrients and supplements, such as glucose, KH_2PO_4 , MgSO_4 , and trace metal. The water qualities are about Cr(VI) concentration of 60 mg l^{-1} , COD_{Cr} of 180 mg l^{-1} and pH value of 6.6. The concentration of nutrient N and P was controlled according to the ratio of 100:6:1 (COD:N:P). This kind of wastewater was used in the tests of magnetic density screening.

Plant wastewater was fetched from a hardware factory in Zhongshan city, of which the Cr(VI) concentration, COD_{Cr} and pH are 21.62, 200 mg l^{-1} , and 4.5, respectively, and was used in the treatment effect tests of magnetic field-anaerobic SBR system.

Culture medium (g l^{-1})

KH_2PO_4 0.5, $(\text{NH}_4)_2\text{SO}_4$ 2.0, NH_4Cl 0.1, Na_2SO_4 0.5, NaHCO_3 0.5, CaCl_2 0.1, MgSO_4 0.1, $\text{C}_3\text{H}_5\text{NaO}_3$ 3.0; yeast extract 2.0, trace metal supplement solution 6 ml, distilled water 1 l. Used in enrichment culture of strains A and B.

Magnetic powder

The Fe_3O_4 powder was purchased from GuangZhou Magnetic Material Factory.

Test devices

Our improvised devices were two anaerobic serial batch reactors (ASBR) with the same specs—20 cm (length) \times 20 cm (breadth) \times 55 cm (high). The MLSS value of both reactors was 3000 mg l^{-1} , and the anaerobic sludge was stirred slowly in the reactors by pumps at the flux of 80 ml min^{-1} . The magnetic powder was magnetized to form an average magnetic density of 6.0 mT and added into reactor II at a ratio of magnetic powder:MLSS = 1:1, meanwhile, a

magnetization device was set around the backflow pipe to stabilize the magnetism of magnetic powder mixed in the returned-sludge, while as a control, there was no magnetic powder and magnetization device in reactor I.

The time of each steps in ASBR, such as wastewater inflow, anaerobic reaction, sludge sedimentation, water discharge, and system unused was 0.5, 13, 2, 0.5, 6 h, respectively.

Methods

Determination of the optimum magnetic density

Influence of different magnetic densities on strains in solid medium

Plates inoculated equal quantity of strain A or B were set at different magnetic densities (0, 2.4, 6, 10, 17.4 mT), respectively. After a 72 h anaerobic culture at 37°C, the differences in colony and individual morphology were compared.

Influence of different magnetic densities on strains in liquid medium

The liquid medium inoculated equal quantity of strain A or B were held in plates and set at different magnetic densities (0, 2.4, 6, 10, 17.4 mT), respectively. After a 24-h static anaerobic culture at 37°C, the quantity varieties of live strains A or B in medium and the absorbency at the wavelength of 640 nm were analyzed and compared. To get more exact results, for all tests mentioned in this paper, two groups of samples were tested synchronously.

Influence of different magnetic densities on Cr(VI) removal efficiency of strains A and B

About 100 ml synthetic wastewater containing equal quantity of strain A and B (about 10^6 bacteria per liter wastewater) were held in five plates averagely, and then set at different magnetic densities (0, 2.4, 6, 10, 17.4 mT), respectively. After a 24-h static anaerobic culture at 37°C, the residual concentrations of Cr(VI) in the supernatant liquids were analyzed and compared.

Influence of magnetic field on treatment effect in ASBR

Influence of magnetic field on synthetic wastewater bio-treatment

The equal volume of the synthetic wastewater was added into ASBR I and II, the concentrations of Cr(VI) and COD_{Cr} in the supernatant liquid were determined corresponding to the reaction time of 0, 3, 5, 6, 7 h.

Influence of magnetic field on plant wastewater bio-treatment

The equal volume of the plant wastewater were added into ASBR I and II, the concentrations of Cr(VI), COD_{Cr} and total Cr in the supernatant liquid were determined corresponding to the reaction time of 0, 1, 2, 4, 7, 9, 12 h.

Influence of magnetic powder on the sludge settlement capability

Every other day, SV_{30} values were measured at the same time before sludge sedimentation until the systems run 30 d stably.

Results and discussion

Determination of the optimum magnetic density

Influence of different magnetic densities on strains in solid medium

After 24 h culture, colonies could be observed only on the mediums at 6.0 and 17.4 mT. Results of 72 h culture showed that the influences of weak magnetic field on the growth of strain A and B were different, strain A had the strongest adaptability at 6.0 mT, the macroscopic colonies appeared earliest and its quantity was the largest one, and with the culture time further extending, the size of the colonies did not increase obviously. While colony of strain A formed at 17.4 mT was bigger than that of 6.0 mT ultimately, though the adaptability at 17.4 mT was worse at

initial culture stages, and the colonies at 2.4 and 10.0 mT had no obvious differences with the control in size and forming rate.

As compared with strain A, the influence of weak magnetic field on strain B was more obvious, the colony diameter at 17.4, 6.0, and 2.4 mT was about nine times larger than the control. And the colony at 6.0 mT was the largest one, which illuminated that 6.0 mT had the strongest growth improvement for strain B.

Influence of different magnetic densities on strains in liquid medium

The absorbencies of culture liquid varied with the culture time are shown in Fig. 1. At the initial stages of magnetic field introduction, the absorbency of culture liquid has no obvious difference, and after 4 h culture, the tendency begins to change, and this difference becomes more and more obvious. All the absorbencies of the testing exceed that of the control, and the amplitude at 6.0 mT is the biggest one. Thus it is concluded that the influence of magnetic field on bacteria' growth is of accumulation and hysteresis.

Figure 2 shows that all these testing magnetic densities improve the quantity of mixed bacteria in the liquid medium obviously, except the magnetic density of 10 mT, compared with the control. After 24 h culture, the density of mixed bacteria increases by 32.5–65%, and it reaches the maximum when the surface magnetic density of the medium is 6.0 mT,

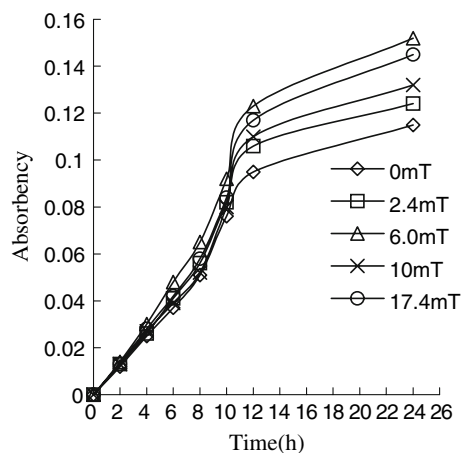


Fig. 1 Accumulation and hysteresis of the magnetic bio-effect

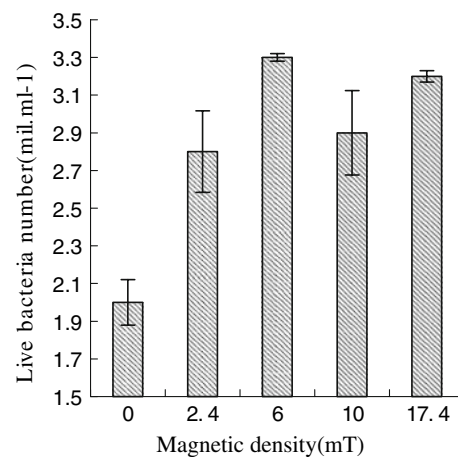


Fig. 2 Influence of magnetic density on the quantity of living bacteria

that is to say, the propagation rate of mixed bacteria is the biggest one at 6.0 mT.

Comparison of Cr(VI) removal efficiency of strains at different magnetic density

After 24-h anaerobic static treatment, all the residual concentrations of Cr(VI) in the supernatant liquid at different weak magnetic field are lower than that of the control, as shown in Fig. 3. And the

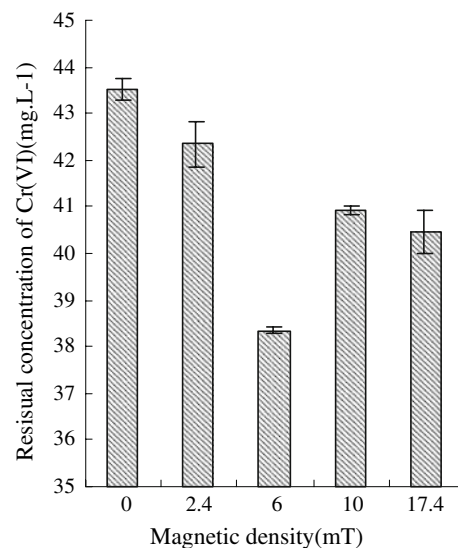


Fig. 3 Comparison of Cr(VI) removal effects at different magnetic densities

Cr(VI) removal rate is 36.17% at 6.0 mT, which is the maximum and 8.84% higher than that of the control.

Therefore, 6.0 mT is determined as the optimum magnetic density and applied in experiments below.

Influence of magnetic field on bio-treatment effect in ASBR

Influence of magnetic field on bio-treatment effect of synthetic wastewater

The contribution of Cr(VI) to the concentration of total Cr is above 99.9% in the synthetic wastewater prepared with potassium dichromate, so only the influences of magnetic field on the removal rates of Cr(VI) and COD_{Cr} were considered in this experiment. As Figs. 4 and 5 shown, when the Cr(VI) concentration of the inflow wastewater ranges between 60 and 70 mg l⁻¹, the removal rates of Cr(VI) and COD_{Cr} in reactor II (reactor in presence of magnetic field) are about 8 and 15% higher than those of reactor I (the control, reactor in absence of magnetic field). Moreover, to meet the first-class effluent discharge standard of Cr(VI) (0.5 mg l⁻¹) and COD_{Cr} (90 mg l⁻¹), compared to reactor I in absence of magnetic field, the presence of magnetic field could reduce 1 h of the reaction time.

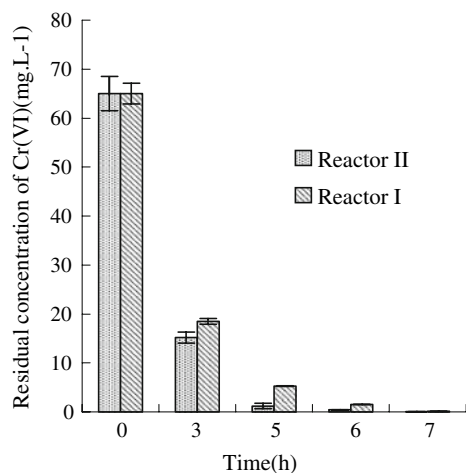


Fig. 4 The influence of magnetic field on Cr(VI) bio-removal

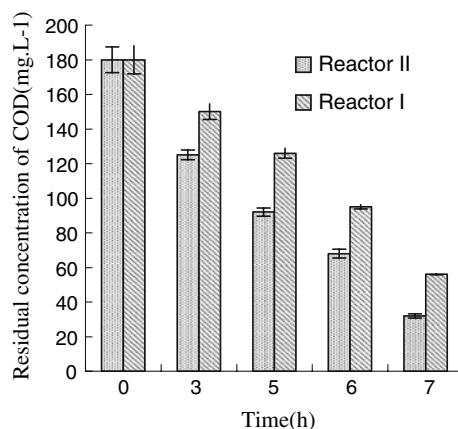


Fig. 5 The influence of magnetic field on COD_{Cr} bio-removal

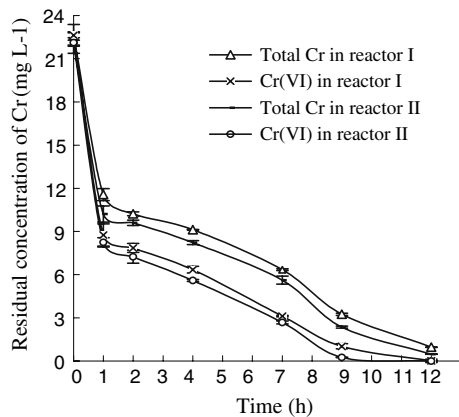
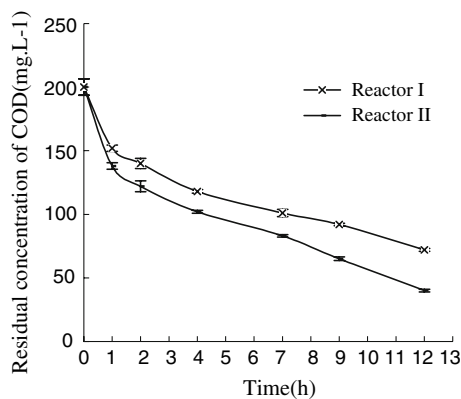
Influence of magnetic field on bio-treatment effect of plant wastewater

By comparison between the plant wastewater and the synthesis wastewater, it is concluded that Cr(VI) concentration of the plant wastewater is much lower, COD_{Cr} value is a little higher and pH value is lower remarkably. The treatment efficiency of the plant wastewater in both reactor I and reactor II is lower than that of the synthetic wastewater, and the reaction time up to par of Cr(VI) and COD_{Cr} increased 2–5 h, as listed in Table 2, which relate to the complexity of the plant wastewater composition, the bio-available degree of organic compounds and the acidic water quality.

Figures 6 and 7 show that the magnetic field of 6.0 mT can improve the bio-purification efficiency of the Cr(VI)-containing electroplating wastewater, the reaction times up to par of Cr(VI), total Cr and COD_{Cr} decrease 2–3 h. It is also found that pH of the outflow water promotes to about 6.8, which may relate to the ion exchange between OH⁻ on surface of biomass and anionic Cr(VI) species. Meanwhile, no matter introduction of magnetic field or not, during the plant wastewater treatment, the decrease of residual total Cr concentration always lags behind that of Cr(VI), and when the water quality is up to par, the ratio of Cr(VI) and total Cr in reactor I and II were reduced from 97.79 to 1.56 and 1.25%, respectively. Therefore, it was considered that both the removal processes of Cr(VI) and total Cr behave transformations from resolvable states to non-resolvable states, and the deoxidization of Cr(VI) is especially important.

Table 2 Comparison of the reaction times meeting the state one class discharge standardI[#] means reactor III[#] means reactor II

		Cr(VI)		Total Cr		COD _{Cr}		Summation	
		I [#]	II [#]	I [#]	II [#]	I [#]	II [#]	I [#]	II [#]
Reaction time up to par (h)	Synthetic wastewater	7	6	–	–	6	5	7	6
	Plant wastewater	12	9	13	10.5	9	7	13	10.5

**Fig. 6** Comparison of Cr(VI) and total Cr residual concentration in the two reactors**Fig. 7** Comparison of COD_{Cr} residual concentration in the two reactors

Influence of magnetic powder on the sludge settlement capability

By comparison of the sludge concentration variations in two reactors, it is found that the presence of magnetic powder could improve the sludge settlement capability obviously. After addition of magnetic powder, the value of SV₃₀ decreased from 12.0 to 8.2%. But the value of SV₃₀ in both reactors changed indistinctly after a stable running period of 30 d. So,

the addition of magnetic powder can play an important role in reducing the sludge loss, decreasing the turbidity of outflow water, which is very helpful to stabilize and enhance the treatment efficiency of bio-system.

Conclusions

A suitable weak magnetic field is benefit to the growth and propagation of predominated strains A and B and the magnetic bio-effect is of accumulation and hysteresis. All experimental results of the strains culture in solid or liquid medium and static bio-treatment show that the optimum magnetic density for strains A and B is 6.0 mT. To reach the first-class effluent standard of Cr(VI) (0.5 mg l⁻¹) and COD_{Cr} (90 mg l⁻¹), compared to the situation in absence of magnetic field, magnetic field can reduce the reaction time of the synthetic wastewater and the plant wastewater by 1 and 2–3 h, respectively. The functions of magnetized magnetic powder include the stimulation to strains growth and propagation, the improvement on sludge toleration, removal, deoxidization of Cr(VI) and the promotion of sludge sedimentation capability. Addition of a suitable weak magnetic field is an effective method in toxic compounds bio-treatment field, and the detail mechanism still needs further research.

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