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Particle size distribution and property of bacteria attached to carbon fines in drinking water treatment

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Abstract: The quantitative change and size distribution of particles in the effluents from a sand filter and a granular activated carbon (GAC) filter in a drinking water treatment plant were investigated. The average total concentration of particles in the sand filter effluent during a filter cycle was 148 particles/mL, 27 of which were larger than 2 μm in size. The concentration in the GAC effluent (561 particles/mL) was significantly greater than that in the sand filter effluent. The concentration of particles larger than 2 μm in the GAC filter effluent reached 201 particles/mL, with the amount of particles with sizes between 2 μm and 15 μm increasing. The most probable number (MPN) of carbon fines reached 43 unit/L after six hours and fines between 0.45 μm and 8.0 μm accounted for more than 50%. The total concentration of outflowing bacteria in the GAC filter effluent, 350 CFU (colony-forming units) /mL, was greater than that in the sand filter effluent, 210 CFU/mL. The desorbed bacteria concentration reached an average of 310 CFU/mg fines. The disinfection efficiency of desorbed bacteria was lower than 40% with 1.5 mg/L of chlorine. The disinfection effect showed that the inactivation rate with 2.0 mg/L of chloramine (90%) was higher than that with chlorine (70%). Experimental results indicated that the high particle concentration in raw water and sedimentation effluent led to high levels of outflowing particles in the sand filter effluent. The activated carbon fines in the effluent accounted for a small proportion of the total particle amount, but the existing bacteria attached to carbon fines may influence the drinking water safety. The disinfection efficiency of desorbed bacteria was lower than that of free bacteria with chlorine, and the disinfection effect on bacteria attached to carbon fines with chloramine was better than that with only chlorine.

Key words: *drinking water treatment*; *particles*; *size distribution*; *bacteria attached to carbon fines*; *inactivation*

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1 Introduction

The pollution of source water has received more and more attention in China in recent years. It has become common to treat drinking water, and the advanced treatment process has long been regarded as a means of ensuring drinking water quality. The granular activated carbon (GAC) technique has been applied in developed countries and in some parts of China (Dussert et al. 1994). Various researchers have shown that biological behavior is common in GAC beds and particles with bacteria easily penetrate treatment filters and enter the effluent due to hydraulic forces. The bacteria attached to carbon particles are highly resistant to chlorination and may influence drinking water safety. Over the long term, drinking water safety would be ensured by using turbidity as the control index for the effluent water quality

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(Hatukai et al. 1997). In the past, outbreaks of waterborne disease caused by protozoan parasites *Cryptosporidia* and *Giardia* have occurred in the United States and other countries. Some research has shown that there may be a high probability of *Cryptosporidia* and *Giardia* appearing in drinking water when the concentration of particles larger than 2 μm is greater than 100 particles/mL (Hargesheimer et al. 1998), so particles in water treatment effluent are being investigated as a substitute index of the Protozoa (Cook 1995). Most American water treatment plants require that the concentration of particles with a size larger than 2 μm remain under 50 particles/mL, and in Pennsylvania the concentration of particles with a size ranging from 3 to 18 μm must be less than 10 particles/mL. Pathogenic protozoa, which has already brought potential danger to drinking water safety, has been found in micro-polluted drinking water sources in some areas, such as Shandong and Anhui Provinces in China. GAC filter effluent with more particles and microorganisms attached to particles would potentially endanger drinking water safety. In this study, the quantitative change and size distribution of particles in the effluent from a sand filter and a GAC filter in a drinking water treatment plant are investigated. In addition, the inactivation effects of chlorine and chloramine of varying concentrations on bacteria attached to carbon fines are examined.

2 Materials and methods

2.1 Experimental parameters

The experiment was conducted in a southern drinking water treatment plant whose raw water quality is shown in Table 1. The drinking water treatment process is shown in Figure 1. Water samples were collected from the Jiangsu Reach of the Yangtze River. The process includes preozonation, coagulation, flocculation, sedimentation, sand filtration, ozonation and GAC filtration. It consists of the conventional process (before the V-shaped sand filter) and the advanced treatment process (after the O_3 contact tank). The annual average turbidity of the effluent from the V-shaped sand filter remains steadily under 0.5 NTU. The process parameters of the V-shaped sand filter and the GAC filter are shown in Tables 2 and 3.

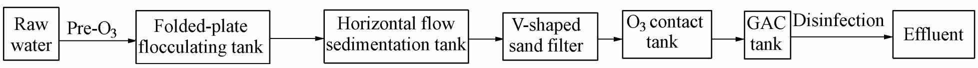

Figure 1 Drinking water treatment process in the plant

Table 1 Raw water quality during the experiment

Turbidity (NTU)	Water temperature (℃)	COD_{Mn} ($mg \cdot L^{-1}$)	pH	NH_3-N ($mg \cdot L^{-1}$)	Total bacterial count ($CFU \cdot mL^{-1}$)
10–15	20–25	6–7	7.9–8.0	0.3–0.5	150–300

Table 2 Parameters of the V-shaped sand filter

Filtration rate ($m \cdot h^{-1}$)	Sand depth (m)	Sand size (mm)	Water wash		Air wash		Surface washing ($L \cdot m^{-2}s^{-1}$)
			Wash rate ($L \cdot m^{-2}s^{-1}$)	Time (min)	Wash rate ($L \cdot m^{-2}s^{-1}$)	Time (min)	
11	1.15	0.95–1.35	3–6	5	15	4	2

Table 3 Parameters of the GAC filter

Filtration rate ($m \cdot h^{-1}$)	GAC depth (m)	Gravel depth (m)	Water wash		Air wash	
			Wash rate ($L \cdot m^{-2}s^{-1}$)	Time (min)	Wash rate ($L \cdot m^{-2}s^{-1}$)	Time (min)
12	1.80	0.50	15–16	5–7.5	15–16	5–6

2.2 Methods

(1) The particle-counting instrument was produced by the IBR Company in New York, USA. Its detection capability covers particles with diameters from 2 to 100 μm and the maximum detection range is 18 000 particles/mL. Its operating voltage is 220 V.

(2) The most probable number (MPN) of activated carbon fines was detected by counting the number of particles of different sizes with a microscope. A GAC filter effluent sample of 1.0 L was filtered through a membrane with a 0.45 μm pore size and the trapped substance was collected in a rinse solution (10 mL) of distilled water. The enriched sample was examined with a microscope and the carbon fines were identified and counted based on their special characteristics.

(3) Bacteria were cultivated in an R2A culture medium in order to find the total bacteria number.

(4) The bacteria attached to fines were desorbed by high-speed centrifugation with some eluant. The centrifuge speed, temperature and time were, respectively, 18 000 r/min (revolutions per minute), 4℃ and five minutes.

(5) Other water quality monitoring methods were adopted according to national standards.

3 Results and discussions

3.1 Particles in effluents from different treatment processes

The changes and size distribution of particles in the effluents from different processes were investigated. The experimental results are shown in Table 4. The percentages of particles of different sizes in pre-O_3 effluent were similar to those in raw water, but the total number of particles decreased slightly. The particle removal efficiency of sand filtration was better, with a 180-fold drop compared to raw water. The concentration of particles larger than 2 μm in the GAC effluent, 120 particles/mL, which exceeds the American standard of 100 particles/mL, was higher than that in sand filter or O_3 effluent. At the same time, the percentage of particles larger than 5 μm in size increased to some extent, and particles of 20 μm were even detected in the effluent.

3.2 Particles in sand filter effluent

3.2.1 Change of particle concentration in sand filter cycle

The changes in particle concentration and turbidity in the effluent from the V-shaped sand filter during the 70-hour filter cycle are shown in Figures 2 and 3.

Table 4 Size distribution of particles in effluents from different processes

Size distribution	Raw water (particles/mL)	Preozonation (particles/mL)	Sedimentation (particles/mL)	Sand filtration (particles/mL)	Ozonation (particles/mL)	GAC filtration (particles/mL)	Final effluent (particles/mL)
≤2 μm	21 795	20 448	4 798	454	483	466	282
2–5 μm	7 027	6 512	725	48	61	65	35
5–7 μm	4 149	3 824	415	21	30	37	17
7–10 μm	1 292	1 128	126	3	8	14	5
10–15 μm	446	349	839	1	2	3	2
15–20 μm	155	104	13	0	0	1	0
20–25 μm	54	32	5	0	0	0	0
25–50 μm	1	3	0	0	0	0	0
>2 μm	13 124	11 952	2 123	73	101	120	59

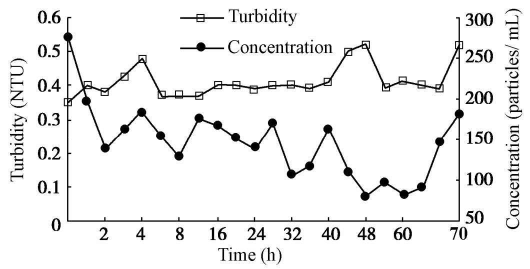

Figure 2 Changes of particle concentration and turbidity in sand filter cycle

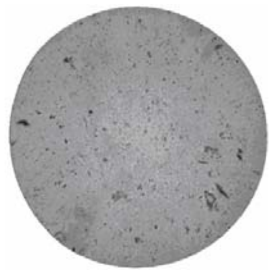

Figure 3 Particles after sand filtration

The average total concentration of particles in the sand filter effluent during a filter cycle was 148 particles/mL. The concentration was greater in the initial filtration process, 277 particles/mL at the beginning and 197 particles/mL after one hour. The concentration in the effluent decreased with filtration and fluctuated within a range of 80 to 160 particles/mL after one hour, but began to increase to a level of 180 particles/mL after 60 hours. The turbidity of the effluent mostly remained below 0.5 NTU during the filter cycle and there was no correlation ($R^2 < 0.1$) between the turbidity value and particle concentration, so the particles in drinking water treatment would not be effectively controlled by the use of turbidity as a control parameter for treated water.

Particle size distribution is an important control target in appraising the biotic danger of treated drinking water. The average concentration of particles larger than 2 μm in the filter effluent was 27 particles/mL, less than the safety standard in most American water treatment plants, which was below 50 particles/mL (data not shown). But the size of particles (larger than 2 μm) in the effluent mainly fell within a range of 5 to 15 μm (accounting for 48.1% of the total), exceeding the safety standard established in Pennsylvania, which stipulates that the concentration of particles of 3 to 18 μm be less than 10 particles/mL.

In addition, the change in turbidity of the effluent came later than that of particle concentration. The particle concentration began to increase after a filtration time of six hours, when the turbidity was still around 0.4 NTU. It increased to 180 particles/mL at the end of filtration, while the turbidity had only reached 0.52 NTU. Such results are in accordance with many other studies and actual operation data from many American waterworks, in which the particle-counting instrument is usually five to ten hours ahead of the turbidity indicator in revealing quality change. Particle concentration change is thus a more effective control parameter than turbidity for assessing the water quality of sand filter effluent.

The variation of the particle concentration in the filtrated water may be explained as follows: the greater amount of particles in the initial filtrated water resulted from the poor purification capacity at the beginning of filtration. The particle removal efficiency, especially for particles larger than 2 μm, was enhanced with filtration. There was a distinct drop in the number of particles larger than 2μm during the filter cycle, but the number of particles less than or equal to 2 μm fluctuated continually. Particles began to increase in number in the final period of filtration and there was no difference in the rises of particles with different sizes. Control of particles larger than 2 μm is thus vital to ensuring filtrated water quality.

3.2.2 Changes of particle concentration in initial sand-filtered water

The changes of sizes of particles in the initial filtrated water are shown in Figures 4 and 5. Figure 4 shows that the concentration of particles (larger than 2 μm) was greater in the earliest stage of filtration, over 50 particles/mL in the initial ten minutes. After a filtration time of one hour, it decreased to a level of 50 particles/mL, close to the average value for the filter cycle. This demonstrates that it is essential to control the filtrated water in the initial ten minutes. As can be seen in Figure 5, the number of particles with sizes between about 2 μm and 7 μm accounted for 91% of the total (larger than 2 μm) and the composition of particles in the initial filtrated water accorded with that of the whole filter cycle, which indicates that particles' removal may be dominated by attractive forces such as van der Waals forces and chemical bonding.

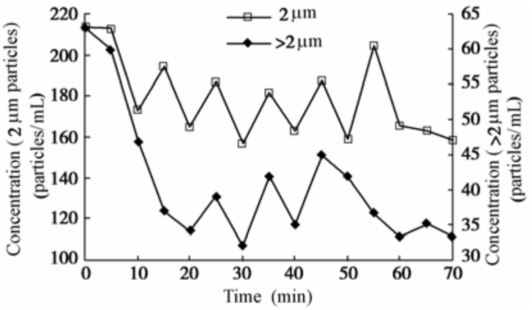

Figure 4 Changes of particle concentration in initial sand-filtered water

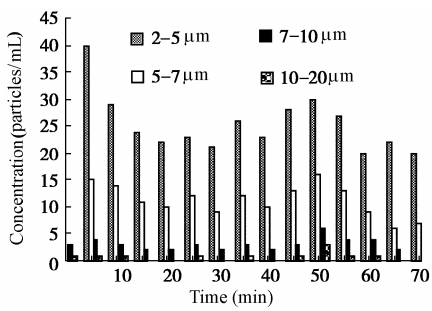

Figure 5 Distribution of particle size in initial sand-filtered water

3.3 Particles in GAC filter effluent

3.3.1 Changes of particle concentration in GAC filter cycle

The turbidity range was 0.23 to 0.45 NTU in the GAC effluent. Particle concentrations during a filter cycle of 24 hours under backwash conditions are shown in Figure 6. The concentration of particles was 460 particles/mL in the initial filtrated water after backwash and the concentration of particles larger than 2 μm was well beyond 50 particles/mL. The concentration of particles in the effluent decreased along with the operation of the GAC filter and stayed within the range from 220 to 800 particles/mL until the beginning of the next backwash. The concentration of particles larger than 2 μm in size basically stayed within the range from 50 to 100 particles/mL during operation until the largest count of 287 particles/mL in the last period of backwash. In the GAC filter effluent, the concentration of particles larger than 2 μm reached 201 particles/mL, with the amount of particles with sizes between 2 μm and 15 μm increasing. Photos of particles filtrated with a membrane with 0.45 μm pore size are shown in Figure 7.

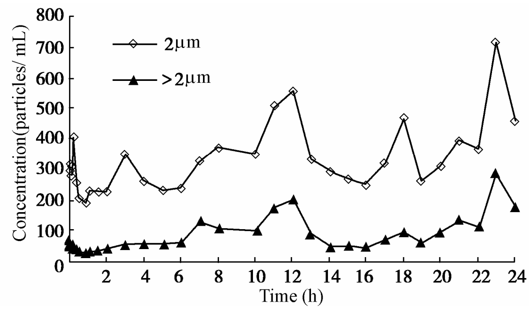

Figure 6 Changes of particle concentration in GAC filter cycle

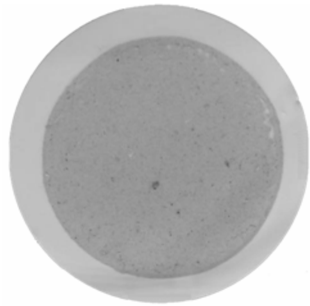

Figure 7 Particles in GAC telophase effluent in GAC filter cycle

3.3.2 Changes of particle size and concentration in the initial GAC-filtered water

Changes in the size of particles in the initial filtrated water are shown in Figures 8 and 9. The concentration of particles with a size larger than 2 μm was greatest in the earliest stage of filtration, more than 50 particles/mL in the initial nine minutes. After a filtration time of ten minutes, it decreased to about 50 particles/mL, close to the average value for the filter cycle. It is essential to control the water filtrated in the initial six minutes.

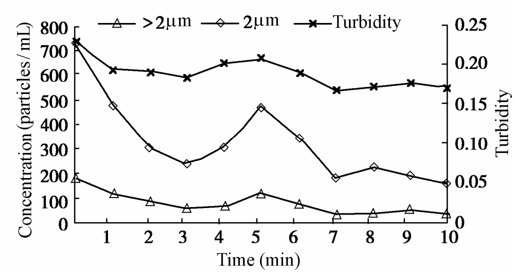

Figure 8 Changes of particle concentration and turbidity in initial GAC-filtered water

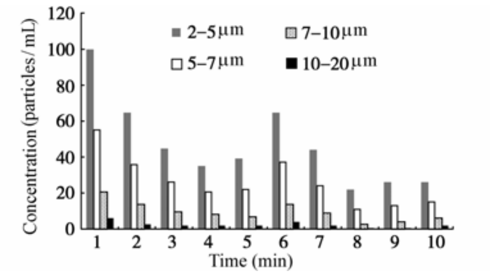

Figure 9 Size distribution of particle concentration in initial GAC-filtered water

As shown in Figure 9, particles with sizes between around 2 μm and 7 μm accounted for 90% of the total particles larger than 2 μm and the composition of particles in the initial filtrated water accorded with that of the whole filter cycle. Turbidity, on the contrary, changed less. It is therefore proposed that the outflow of water in the initial six minutes be discarded in order to reduce the amount of particles.

In order to more thoroughly investigate the particles, they were preliminarily divided into organism particles and mineral particles. A photo of particles filtrated with a filter screen (200 oculus) after incubation (20℃) of GAC effluent water for six days is shown in Figure 10. As can be seen, the majority of particles were plankton, organism congeries and so on. Organism particles basically consisted of zooplankton, algae, biology slough, organism congeries and mineral particles consisting of micro-activated carbon fines, mineral debris and other materials.

3.4 Changes of carbon fines in GAC filter

Some studies have proved that the number of bacteria increases with the use of a GAC filter as compared with sand effluent (Camper et al. 1986a). Some fines are removed from the GAC bed by the flow shearing force and these fines are of a smaller size, from several to dozens of microns. The bacteria appearing in the effluent usually have high resistance to chlorination because of their attachment to carbon fines (LeChevallier et al. 1984). In experimental results, the size of carbon fines mainly ranged from 2.0 to 5.0 μm, which was consistent with that of particles in the effluent from the GAC filter. The change in the number of activated carbon fines in the effluent during a filter cycle of 48 hours is shown in Figure 11.

Figure 10 Filtrated particles in GAC filtrt effluent

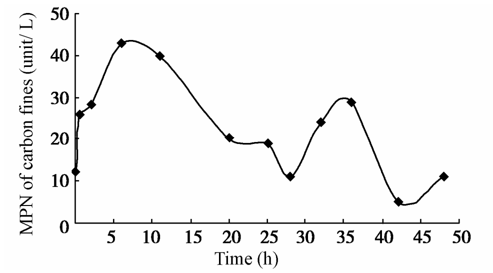

Figure 11 Changes of carbon fines in GAC filter cycle

It can be seen in Figure 11 that the MPN of carbon fines varied from 10 to 40 unit/L, comparable to other research results of 10 to 100 unit/L. Moreover, similar results were attained in other studies in China (He and Xu 2004). The experimental results indicated that there were differences in the concentration of carbon fines in the GAC filter effluent due to the activated carbon sorting and processing operation. The results also showed that the MPN of carbon fines was low at the beginning of backwash but reached the highest number, 43 unit/L, after six hours. Along with running time, the MPN gradually decreased for 30 hours, then increased again. Fines of 0.45 to 8.0 μm accounted for the majority, with the percentage of fines smaller than 1.0 μm being more than 50%. However, it should be pointed out that carbon fines accounted for less than 0.1% of all the particles in the GAC effluent and the correlation coefficient (R^2) was only 0.30. This analysis indicates that carbon fines in the effluent may not be effectively controlled only by reducing the particles.

3.5 Change of bacteria concentration in filter effluents

The microorganisms in GAC effluent consisted predominantly of heterotrophic bacillus. After incubation, the bacteria colony was tested with Shineso (China) AS-02. The size distribution, characteristics and Gram's staining of the bacteria colony are shown in Tables 5 and 6.

The experimental results in Table 5 show that the total number of bacteria in the GAC effluent was nearly two times that in the effluent from the sand filter. This shows that biological behavior in the GAC bed was prevalent and that there was an abundance of different kinds of bacteria.

Table 5 Auto-analysis of bacteria colony

Size distribution	Bacteria colony (CFU/mL)	
	Sand filter effluent	GAC filter effluent
0–0.1 mm	0	0
0.1–0.5 mm	113	204
0.5–1.0 mm	26	62
1.0–2.0 mm	42	46
2.0–5.0 mm	25	38
>5.0mm	4	0
Total	210	350

Table 6 Characteristics and Gram's staining of bacteria colony

Sand filter effluent			GAC filter effluent		
Colony color	Percentage (%)	G+/G-	Colony color	Percentage (%)	G+/G-
bright yellow	50	G+	bright yellow	0	-
ivory yellow	30	G+	ivory yellow	30	G+
ivory white	20	G-	ivory white	70	G-

3.6 Disinfection efficiency of bacteria in GAC filter effluent

Some research has shown that bacteria can attach themselves to active carbon fines and show high resistance to chlorination due to the protection of the fines (Stringfellow et al. 1993). The desorption means of the homogenization technique were used in the experiment: high speed centrifugation with eluant containing sodium pyrophosphate (0.01%), peptone (0.01%) and Tris buffer (10^{-1} mol/L, $\mathrm{pH} = 7.0$). The amount of desorbed bacteria from carbon fines reached an average of 310 CFU/mg. However, it should be pointed out that the attached bacteria may be a small part of the total, because the centrifugal force in the experiment did not provide perfect desorption effects. Observations made with a scanning electron microscope indicate that the GAC was colonized by bacteria that grew in cracks and crevices and was coated by an extra cellular slime layer (Lechevallier 1990).

Disinfection studies have indicated that organisms attached to GAC particles, including *Streptococcus pneumoniae*, *Aeromonas hydrophila*, *Serratia* and *Acinetobacter*, may continue to survive by inactivation with 2.0 mg/L of chlorine after 30 minutes of contacting time

(pH = 7.4) (Camper et al. 1986b). The disinfection efficiency of bacteria attached to carbon fines was less than 40% with 1.5 mg/L of chlorine and a contacting time of 30 minutes. This demonstrates the protection of attachment to carbon fines during the disinfection. In addition, some research has proven that the inactivated bacteria carried by activated carbon fines can penetrate the distribution system, attach themselves to the pipe surface, grow and develop into a biofilm, resulting in secondary biotic pollution of drinking water due to certain resistance to residual disinfectant in the pipe network (Morin and Camper 1997).

Several studies have shown that the disinfection efficiency with chloramine is better than that with only chlorine. The disinfection efficiency of bacteria in the effluent from the GAC with different chlorine and chloramine dosages was investigated, with a contact time of 30 minutes, a pH of 7.3 to 7.5, and an environmental temperature of 28℃. The ratio of chlorine to ammonia was 2:1 during chloramine disinfection, with a gradation of chlorine-before-ammonia and an alternation time of 30 seconds. The bacteria were cultured in an R2A culture medium. The experimental results are shown in Figure 12. The disinfection effect of chloramine was better than that of chlorine. The inactivation percentage was more than 90% with 2.0 mg/L of chloramine and 30 minutes of contact, but only 70% with chlorine under the same conditions.

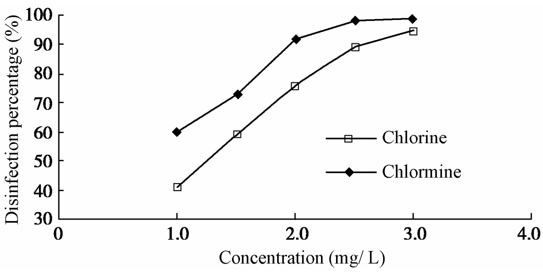

Figure 12 Effect of disinfection by chlorine and chloramine in GAC filter effluent

The auto-analyzing size distribution of the bacteria colony after disinfection with chloramine showed that the colony color was mainly ivory white and orange yellow.

4 Conclusions

Experimental results show that the high particle concentration in raw water and sedimentation effluent led to high levels of outflowing particles in the sand filter effluent. The particle concentration in the GAC filter effluent exceeded the present particle control criterion of American drinking water product standards, and could endanger the safety of the drinking water supply. The activated carbon fines in the effluent accounted for a small proportion of the total particle concentration. However, the existing bacteria attached to carbon fines may influence the drinking water safety, as proven by desorption with high speed centrifugation. The disinfection efficiency of desorbed bacteria was lower than that of free bacteria with chlorine, and the disinfection effect on bacteria attached to carbon fines with chloramine was better than that with only chlorine.

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