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## Effects of common inorganic anions on the rates of photocatalytic degradation of sodium dodecylbenzenesulfonate over illuminated titanium dioxide

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**Abstract:** Experiments were carried out to study the effects of several anions on the photocatalytic degradation rates of sodium dodecylbenzene sulphonate (DBS) with  $\text{TiO}_2$  as catalyst. The anions were added as  $\text{Na}_2\text{SO}_4$ ,  $\text{NaNO}_3$ ,  $\text{NaCl}$ ,  $\text{NaHCO}_3$ ,  $\text{NaH}_2\text{PO}_4$  and  $\text{Na}_3\text{PO}_4$ , and two levels of anion content, i.e. 12 mmol/L and 36 mmol/L in terms of  $\text{Na}^+$ , were studied. The results revealed that:  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$  and  $\text{HCO}_3^-$  retarded the rates of DBS degradation to different degrees;  $\text{PO}_4^{3-}$  increased the DBS degradation rate at low concentration and decreased the rate at high concentration;  $\text{H}_2\text{PO}_4^-$  accelerated the rate of DBS degradation. The mechanism of the effects of anions on DBS degradation was concluded as the following three aspects: anions compete for the radicals; anions are absorbed on the surface of catalyst and block the active site of catalyst; anions added to the solution change the pH value and influence the formation of  $\cdot\text{OH}$  radicals and the adsorption of DBS on catalyst.

**Keywords:** surfactant; anion; photocatalytic degradation; water environment

## Introduction

Since early 1990s, the remediation of water organic pollution with advanced oxidation process has aroused attentions from worldwide (D' Oliveira, 1990; Ollis, 1991; Hidaka, 1995; Sunada, 1998; Tanaka, 2000). Remarkable progress has been obtained over the application of photocatalyzed oxidation process to the treatment of organic polluted water (Blake, 1991; Bedford, 1994; Hofstadler, 1994; Vinodgopal, 1995; Tsukasa, 1996; Xu, 1997; Bahnemann, 1997; Xia, 1999;). Photocatalysis is a process in which the illumination of catalyst, sometimes with the combination of oxidant (such as  $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ ), produces radicals with strong activity, e.g.  $\cdot\text{OH}$ , and these radicals go on to attack organic contaminants and cause mineralization through chemical reactions of addition, substitution and electron transfer. Recently, research on the rate and mechanism of single organic contaminant photodegradation has been conducted in detail (Reeves, 1992; Spadaro, 1994; Martin, 1996), while little attention has been paid to the effect of water environmental conditions on the photodegradation of contaminants in waste water (Prairie, 1992; Parent, 1996). Some associated organic contaminants and various salts often exist in waste water in addition to a certain or several organic contaminants with high concentration. For example, the sodium sulfate and sodium triphosphate which are additives of detergent often exist in waste water with high concentration of anionic surfactants. Therefore, it is important to take into account the effects of various chemical compositions on the photodegradation of the target organic contaminant so that the experimental data accord well with the waste water conditions. Some research has been conducted on the effects of inorganic anions on photocatalytic oxidation rates of several organics including salicylic acid, ethanol, 2-propanol, phenol and aniline under a fixed pH value (Abdullah, 1990). However, the addition of some anions will change the pH value in the system. So, the experiments without adjusting the pH value may comprehensively reflect the effects of anions on organic photodegradation.

In this paper, the study on the effects of anions including  $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{PO}_4^{3-}$  and  $\text{H}_2\text{PO}_4^-$  on photocatalyzed degradation rates of sodium dodecylbenzenesulfonate (DBS) over illuminated

titanium dioxide were conducted without fixing pH value, and the mechanism of the effects of anions on DBS degradation was also discussed.

## 1 Material and methods

### 1.1 Material

The catalyst used in the experiments were  $\text{TiO}_2$  bought from ACROS of Germany. X-ray diffraction analysis indicated an anatase to rutile ratio of 99 : 1, and the surface area is  $7 \text{ m}^2/\text{g}$ . Sodium dodecylbenzenesulfonate (DBS) was of standard reagent grade obtained from National Research Center of Standard Chemicals in China. All other chemicals were of analytical reagent grade supplied by Beijing Chemical Plant.

### 1.2 Degradation experiments

A dispersion consisting of 400 ml solution with 20 mg/L DBS, inorganic anions of certain content and  $\text{TiO}_2$  was contained in a 500 ml glass vessel with a surface area of  $55.4 \text{ cm}^2$  and was illuminated with a high-pressure mercury lamp (250W) under continuous magnetic stirring. Cool water passed through the glass vessel continuously. The UV irradiation intensity was  $53 \text{ W/m}^2$ , which was close to the maximum solar UV intensity at Beijing in summer. Oxygen was replenished by opening the reaction vessel when the dispersion was sampled after a certain irradiation time. The 5 ml aliquot was sampled for analysis at each interval. Degradation rates for DBS were determined by measurement of the loss of DBS or TOC from the reaction solution. In the experiments for studying the effects of inorganic anions on DBS photodegradation, two levels of concentration, i. e. 12 mmol/L (level A) and 36 mmol/L (level B) in terms of  $\text{Na}^+$  for NaCl,  $\text{Na}_2\text{SO}_4$ ,  $\text{NaHCO}_3$ ,  $\text{NaNO}_3$ ,  $\text{Na}_3\text{PO}_4$  and  $\text{NaH}_2\text{PO}_4$ , were used. The photocatalytic degradation of DBS was fitted with first-order kinetics.

### 1.3 Analysis

The pH values of the dispersions in the photodegradation processes were determined with pHB-4 meter produced by Shanghai Leici Instrument Plant, China. Samples withdrawn from the reaction vessel were centrifuged to remove  $\text{TiO}_2$  particles, and the solution was analyzed for DBS concentration and TOC. For the determination of DBS contents, methylene blue was added to the solution, leading to the formation of a new blue ionpair compound with a strong absorbency at 652 nm. Then, the blue compound was extracted with chloroform and the content of DBS was determined by measuring the absorbency of chloroform solution at 652 nm with spectrophotometer of Beckman DU-8B. The total organic carbon (TOC) was determined by a Shimadzu TOC 500 (China State Environmental Protection Administration, 1989).

## 2 Results and discussion

### 2.1 The effect of amounts of catalyst on DBS degradation

DBS disappearance curves are shown in Fig. 1. Those curves followed first order kinetics. The rates of DBS photodegradation increased with the increase of the amount of  $\text{TiO}_2$ . When  $\text{TiO}_2$  were 0.5 g/L, 1.0 g/L and 1.5 g/L, the photodegradation rate constants of DBS were  $0.0107 \text{ min}^{-1}$ ,  $0.0158 \text{ min}^{-1}$  and  $0.0178 \text{ min}^{-1}$  respectively. The rate constant increased slightly when  $\text{TiO}_2$  increased from 1.0 g/L to 1.5 g/L. From this relationship we chose to use 1.0 g/L  $\text{TiO}_2$  in the following experiments.

### 2.2 Total organic carbon elimination and pH

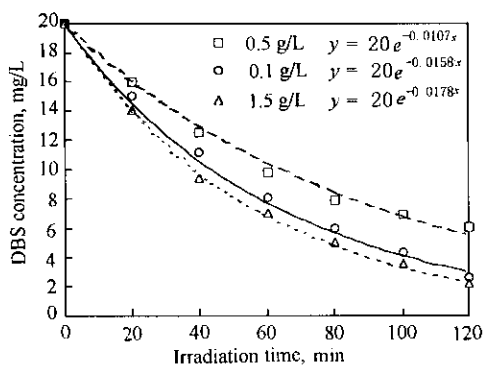


Fig.1 The effect of the amount of catalyst on DBS photodegradation rate (y: DBS concentration; X: irradiation time)

change

Total organic carbon decreased gradually in the photodegradation process (Fig. 2). Its elimination rate was significantly less than DBS disappearance rate. In other words, the complete mineralization of organic compounds requires a much longer illumination time than the disappearance of DBS. In the first three hours, TOC elimination extent was only 0.75 mg/L. But TOC decreased more quickly in the second three-hour than in the first, and the disappearance rate was 2.3 mg/(L·h). The pH value decreased from 6.76 to 4.01 after 3h irradiation, then it increased gradually to 4.10 at 6h irradiation. The decrease of pH value was due to the production of organic acids. In the initial irradiation stage, DBS might be decomposed to organic acids, leading to the significant decrease of pH value and insignificant decrease of TOC. After a certain period of irradiation, the organic acids were degraded to little molecular compound and CO<sub>2</sub>, causing the significant decrease of TOC and increase of pH value.

## 2.3 Effect of inorganic anions on the rate of DBS photodegradation

### 2.3.1 Effect of chloride

As shown in Fig. 3, sodium chloride decreased the rates of DBS degradation. When the content of sodium chloride increased from level A to level B, the degradation rate constant of DBS decreased from 0.0138 min<sup>-1</sup> to 0.0109 min<sup>-1</sup>. The experiment results in our research were consistent with what reported by Pichat and Abdullah (Abdullah, 1990; Pichat, 1993). The effects of chloride on the photodegradation rate of DBS over illuminated TiO<sub>2</sub> may be caused by two factors: firstly, Cl<sup>-</sup> will compete with DBS for the oxidizing radicals and influence the electron transfer (Sunada, 1998; Conceicao, 2000); secondly, they will be absorbed on the surface of the catalyst and block the active sites of the TiO<sub>2</sub> catalyst (Abdullah, 1990).

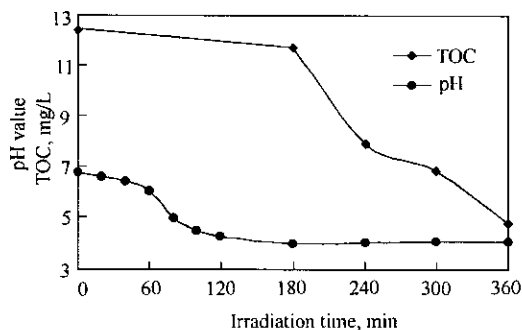


Fig. 2 Total organic carbon elimination and pH value curves of 20 mg/L DBS photocatalyzed degradation

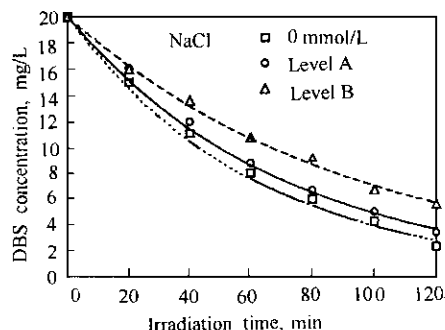


Fig. 3 Effect of NaCl on the oxidation rates of DBS

### 2.3.2 Effect of nitrate

The addition of nitrate to the solution decreased the photocatalyzed degradation rate of DBS (Fig. 4). The rate constant decreased from 0.0158 min<sup>-1</sup> without nitrate to 0.0107 min<sup>-1</sup> with level A nitrate. However, when nitrate content increased from level A to level B, the degradation rate of DBS did not decrease significantly. The result in our experiments were consistent with that of Abdullah (Abdullah, 1990). According to his research, the effect of increasing concentration of nitrate added as NaNO<sub>3</sub> from 10 mmol/L to 40 mmol/L on rates of oxidation of salicylic acid to CO<sub>2</sub> was trivial.

### 2.3.3 Effect of sulphate

The addition of sulphate with low content (level A) did not exert significant impact on the photodegradation rate of DBS (Fig. 5), i. e., the rate constant just decreased from 0.0158 min<sup>-1</sup> to 0.0146 min<sup>-1</sup>. However, the degradation rate constant decreased remarkably from 0.0146 min<sup>-1</sup> to 0.0015 min<sup>-1</sup> when the content of sulphate increased from level A to level B. This may be due to the

increase of absorbing quantity of sulphate on  $\text{TiO}_2$  surface from level A to level B, giving rising to the block of active site  $\text{TiO}_2$  surface.

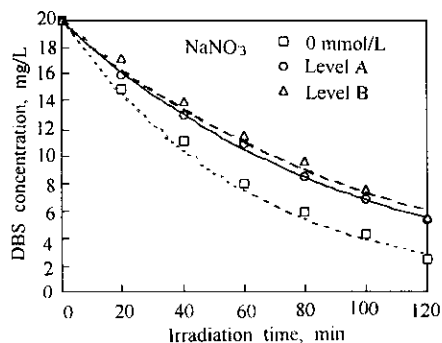


Fig.4 Effect of  $\text{NaNO}_3$  on the oxidation rates of DBS

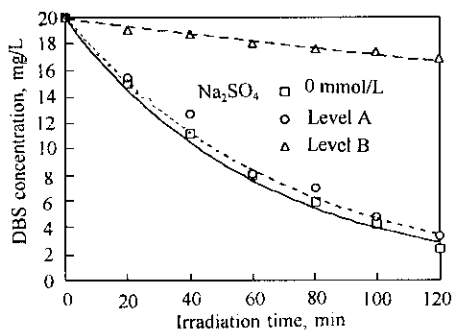


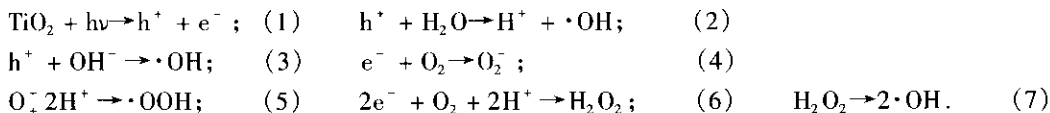
Fig.5 Effect of  $\text{Na}_2\text{SO}_4$  on the oxidation rates of DBS

### 2.3.4 Effect of bicarbonate

The existence of bicarbonate retarded the degradation rates of DBS (Fig.6). The rate constant was reduced from  $0.0158 \text{ min}^{-1}$  to  $0.0107 \text{ min}^{-1}$  when adding level A bicarbonate to the solution. But the degradation rate did not decrease remarkably when the content of bicarbonate increased from level A to level B. The addition of sodium bicarbonate to the solution will lead to the increase of pH. According to the zero zeta potential experiments, the isoelectric point of ACROS  $\text{TiO}_2$  is 4.5. The increase of pH value would make the surface of  $\text{TiO}_2$  more negatively charged, which was infavourable for the DBS's absorption on  $\text{TiO}_2$ , leading to the inhibition of the degradation of DBS. The pH value increased from 6.76 to 8.34 when adding level A  $\text{NaHCO}_3$  to the solution. Therefore, the existence of  $\text{HCO}_3^-$  lessened the degradation rate of DBS. But the pH value only increased from 8.34 to 8.38 when the content of  $\text{HCO}_3^-$  increased from level A to level B, so the degradation rate constant of DBS was reduced only  $0.0008 \text{ min}^{-1}$ .

### 2.3.5 Effect of phosphate

The addition of level A  $\text{Na}_3\text{PO}_4$  to the solution increased the DBS degradation rate from  $0.0158 \text{ min}^{-1}$  to  $0.0233 \text{ min}^{-1}$  (Fig.7). While the concentration of  $\text{PO}_4^{3-}$  increased to level B, the DBS degradation rate constant was reduced to  $0.011 \text{ min}^{-1}$ . The following reactions take place in  $\text{TiO}_2$  dispersion under the UV illumination.



The increase of  $\text{OH}^-$  content is not helpful for the adsorption of DBS on  $\text{TiO}_2$ , but according to Reaction (3), it is favorable for the formation of  $\cdot\text{OH}$ . In this experiment, the pH value of the degradation system was very high, it was enhanced from 6.76 to 11.28 with the existence of level A  $\text{Na}_3\text{PO}_4$ , and the formation of  $\cdot\text{OH}$  controlled the DBS decomposition process. Therefore, the addition of level A  $\text{Na}_3\text{PO}_4$  to the system promoted the degradation of DBS. However, the pH value of the system only increased from 11.28 to 11.71 when the content of  $\text{Na}_3\text{PO}_4$  increased from level A to level B (Fig.7). The absorption of

$\text{PO}_4^{3-}$  on  $\text{TiO}_2$  surface dominated the process, so the photodegradation rate was reduced.

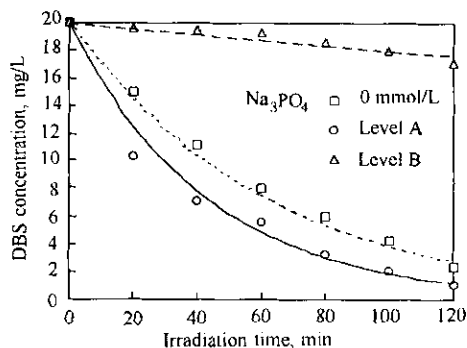


Fig.7 Effect of  $\text{Na}_3\text{PO}_4$  on the oxidation rates of DBS

respectively, which would make  $\text{TiO}_2$  particles more positively charged and promoted the adsorption of DBS on the surface of  $\text{TiO}_2$ . In addition, according to the above mentioned Reaction (5), the formation of  $\text{HO}_2\cdot$  radicals from  $\text{O}_2^-$  and  $\text{H}^+$ , giving rise to  $\text{H}_2\text{O}_2$ , has been proposed as one way of producing  $\cdot\text{OH}$  radicals. This reaction will be enhanced by decreasing the pH value (D' Oliveira, 1990). Therefore, the decrease of pH caused by the addition of  $\text{NaH}_2\text{PO}_4$  led to the increase of DBS photodegradation rate. On the other hand,  $\text{NaH}_2\text{PO}_4$  added to the solution would be absorbed on the surface of  $\text{TiO}_2$  and blocked the active site of  $\text{TiO}_2$  and retarded the DBS oxidation rate. So, the net effect of  $\text{NaH}_2\text{PO}_4$  on DBS decomposition rate depended on the increase of rate caused by pH value change and the decrease of rate attributed to adsorption of  $\text{NaH}_2\text{PO}_4$  on  $\text{TiO}_2$ . In our experiments, pH value change dominated the process, so the DBS photodegradation was accelerated.

### 2.3.7 Comparison of effects of anions on DBS photodegradation rate

As shown in Table 1, when adding level B  $\text{NaCl}$ ,  $\text{NaNO}_3$ ,  $\text{Na}_2\text{SO}_4$ ,  $\text{NaHCO}_3$ ,  $\text{Na}_3\text{PO}_4$  and  $\text{NaH}_2\text{PO}_4$  to the DBS degradation system, the sequence of photocatalyzed degradation constant of DBS was  $\text{NaH}_2\text{PO}_4 > \text{NaCl}$ ,  $\text{NaHCO}_3$ ,  $\text{NaNO}_3 > \text{Na}_2\text{SO}_4 > \text{Na}_3\text{PO}_4$ . The sequence of adsorption tendency was  $\text{PO}_4^{3-} > \text{SO}_4^{2-} > \text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$ ,  $\text{H}_2\text{PO}_4^-$  (Chen, 1987), which was accordance with the sequence of photocatalyzed degradation constant of DBS as a whole. Among  $\text{HCO}_3^-$ ,  $\text{NO}_3^-$ ,  $\text{Cl}^-$  and  $\text{H}_2\text{PO}_4^-$ , the photocatalyzed degradation of DBS with  $\text{H}_2\text{PO}_4^-$  was the highest. As shown in Fig.9, the pH values in the degradation system with  $\text{NaH}_2\text{PO}_4$  were below 4.6 which was the lowest among the anions. As above mentioned, decreasing pH value is helpful to the degradation of DBS, therefore, the photocatalyzed oxidation rate of DBS was the highest with the presence of  $\text{NaH}_2\text{PO}_4$ .

### 2.3.6 Effect of biphosphate

As shown in Fig. 8, the existence of  $\text{NaH}_2\text{PO}_4$  led to the increase of DBS degradation rate. But the rate did not increase with the increase of  $\text{NaH}_2\text{PO}_4$  content. The DBS degradation rate constant increased to  $0.0235 \text{ min}^{-1}$  with the addition of level A  $\text{NaH}_2\text{PO}_4$  to the system, while it was only  $0.0193 \text{ min}^{-1}$  with the addition of level B  $\text{NaH}_2\text{PO}_4$ . On one hand, the addition of  $\text{NaH}_2\text{PO}_4$  changed pH value of the system. It was decreased from 6.76 to 4.87 and 4.68 when adding level A and level B  $\text{NaH}_2\text{PO}_4$  to the solution

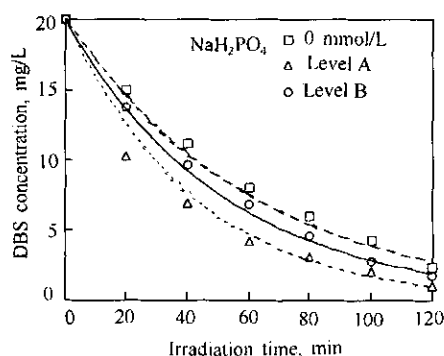


Fig.8 Effect of  $\text{NaH}_2\text{PO}_4$  on the oxidation rates of DBS

**Table 1** The effects of NaCl, NaNO<sub>3</sub>, Na<sub>2</sub>SO<sub>4</sub>, NaHCO<sub>3</sub>, Na<sub>3</sub>PO<sub>4</sub> on the photodegradation rate constant (min<sup>-1</sup>) of DBS fitted with first order kinetics

Contents in terms of Na <sup>+</sup> , mmol/L	NaCl	NaNO <sub>3</sub>	Na <sub>2</sub> SO <sub>4</sub>	NaHCO <sub>3</sub>	Na <sub>3</sub> PO <sub>4</sub>	NaH <sub>2</sub> PO <sub>4</sub>
0	0.0158	0.0158	0.0158	0.0158	0.0158	0.0158
12 (Level A)	0.0138	0.0107	0.0146	0.0107	0.0233	0.0235
36 (Level B)	0.0109	0.0098	0.0015	0.0099	0.0011	0.0193

Among NaCl, NaHCO<sub>3</sub> and NaNO<sub>3</sub>, the photocatalyzed decomposition rate of DBS was the highest with the addition of NaCl, which was caused by the lowest pH values in the degradation system with the presence of NaCl.

3 Conclusion

The existence of Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, HCO<sub>3</sub><sup>-</sup>, NO<sub>3</sub><sup>-</sup>, PO<sub>4</sub><sup>3-</sup> and H<sub>2</sub>PO<sub>4</sub><sup>-</sup> added as Na<sub>2</sub>SO<sub>4</sub>, NaNO<sub>3</sub>, NaCl, NaHCO<sub>3</sub>, Na<sub>3</sub>PO<sub>4</sub> and NaH<sub>2</sub>PO<sub>4</sub> had effects on the photocatalyzed degradation rate of DBS. Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup> and HCO<sub>3</sub><sup>-</sup> all retarded the rates of DBS degradation at different degrees with ACROS TiO<sub>2</sub> as catalyst. PO<sub>4</sub><sup>3-</sup> had positive effect at low concentration and negative effect at high concentration; H<sub>2</sub>PO<sub>4</sub><sup>-</sup> accelerated the rate of DBS degradation under the experimental conditions. The influences of anions on photocatalyzed oxidation rate of DBS were attributed to the following three factors: (1) the anions compete for the radicals or assist recombination of photogenerated electrons and holes; (2) anions will be absorbed to the surface of catalyst and block the active site of catalyst; (3) anions added to the solution change the pH value and influence the formation of ·OH radicals and the adsorption of DBS on catalyst. The effect of a certain anion on DBS photodegradation rate depended on the three factors synthetically.

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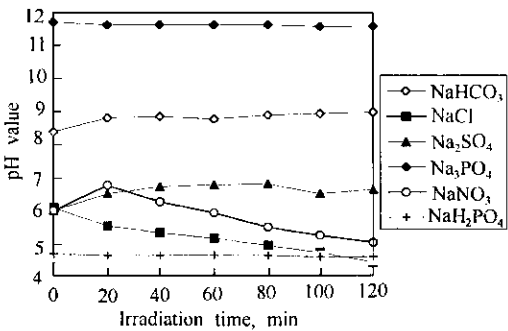


Fig. 9 The pH value curves of DBS photocatalyzed degradation with the presence of anions

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