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Migration and Transformation of Phosphorus Forms in Field Ditch **Sediments**

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Abstract: A broad attention has been paid to the field ditches as the sewage interception channel for the agricultural nonpoint source pollution, however, rare studies have involved in the phosphorus migration and transformation between the ditch sediments and overlaying water, especially, from the ditch sediments to the water. The present paper aims to explore with XPS binding energy data how pH, dissolved oxygen, sunlight affect the process of phosphorus migration and transformation in ditch sediments to the water by methods of changing pH, aerating nitrogen and oxygen in water and changing the sunlight; and hopes to provide a guidance on how to give full play to the ditch's sewage interception capacity and avoid ditch becoming pollution source reversely. The experimental results with XPS binding energy data show that no electron transfer occurs insignificantly at different pH values in sediments, i.e. no electron transfer reaction occurs on surface. This reveals that the phosphorus absorption form HPO₄²⁻ in sediment substitutes OH state on the sediment surface. The amount of phosphorus migration and transformation is the minimum under the neutral condition, and the maximum under the alkaline condition. Under aerobic atmosphere, the data of XPS binding energy shows the surface charge of iron after oxygen aeration reduces, the surface charge of oxygen increases, i.e. Fe²⁺ is oxidized into Fe³⁺, the phosphorus is absorbed increasingly; the experimental results with XPS binding energy data disclose there exists electron transfer reaction significantly, that is the illumination affects the process of phosphorus migration and transformation through dissolved oxygen (DO) and the pH impacted on the biological processes under the presence of sunlight in sediments, indirectly. The influence of illumination on the sediment phosphorus migration and transformation is regular insignificantly. Conclusively, the amount of migration and transformation in sunlight-free group is greater than the sunlight group.

Keywords: Dissolved oxygen, ditch sediments, light intensity, migration and transformation, pH, phosphorus fraction, XPS.

1. INTRODUCTION

Over-fertilization during the agricultural production causes a large number of nutrients enter the rivers and lake via drainage or runoff, directly resulting in water eutrophication. Poyang Lake region is the main producing areas of double cropping of rice in Jiangxi [1]. Although the absorption rate did not improve (the absorption rate of phosphate fertilizer does not exceed 35%) [2], the unit fertilizer applied has increased year by year. According to statistics, Jiangxi Province had the average farmland fertilizer 355.8 kg/hm² in 1990, which reached 623.4 kg/hm² in 2006 with an increase of 75.2%. The resultant agricultural nonpoint source pollutants were flushed into the rivers along with drainage and precipitation through field ditches, and finally rushed to Poyang Lake and affected the lake water quality. As a primary sewage interception channel in agricultural nonpoint source pollution control pathway, the ecological ditches have been investigated by many researchers [3-8] on their ability to intercept the nutrients. The research findings showed that the ditch sediments have a stronger nutrient-removing ability. However, due to seasonal field drainage, the ditches especially the traditional ones can increase the phosphorus content in sediments in a period. Owing to shallow water and sunlight penetration to bottom, the diurnal dissolved oxygen concentration and pH varies significantly [9-11]. With the change in oxygen concentration, illumination and other external environmental factors. the transformation and decomposition accumulated organic matters and nutrients in the sediments will be speeded up [12] so that the phosphorus absorbed, precipitated and carried in the ditch sediments is resolved and entered the downstream water, thus increasing the risk of eutrophication. A majority of present studies have explored the nutrient removal ability of ecological ditch sediments and phosphorus release process in lake sediments. Rare studies have discussed the phosphorus migration and transformation rule of the ecological ditches. The present selects the ecological ditch sediments from a small basin in Qiaoxiali

Villa, Bailu Town, Xingzi county in Poyang Lake basin as a demonstrative area to examine the effect of pH, dissolved oxygen, and light intensity on migration and transformation of phosphorus forms decided by graded extraction method and combine with XPS binding energy data, hopes to find a nutrient transformation rules of the ecological ditches, fully utilizes the sewage interception ability of ecological ditches in Poyang Lake area, reduces the nutrients that enter the lake and finally reduces the risk of eutrophication in Poyang Lake.

2. DEMONSTRATIVE EXPERIMENTAL AREA

The demonstrative area in the study is less than 2.3 km away from Poyang Lake in the straight-line distance based on the traditional planting mainly, covers an agricultural area about two hectares, mainly includes rice fields, vegetable (peanuts, beans, sweet potato) field and a few of wastelands. The average fertilizer applied per mu is 401 kg. The soil is Quaternary red soil. The field ditches are crisscrossed in the region and become the primary channel for the field drainage and rainfall runoff. After the convergence, the water goes into Poyang Lake *via* the downstream rivers. Within ditches, the depth is 40-50 cm, the overall water flow rate is smooth, 20-30 cm-thick mud is accumulated in the ditch bottom and a small amount of emergent aquatic plants are dotted.

3. MATERIALS AND METHODOLOGY

3.1. Sample Collection and Analysis for the Test

Sediments in 6 sampling points were selected from the demonstrative farmland ditches (see Fig. 1). The sediments (mainly from the soil carried by runoff in farmland and from the suspension caused by the ditch water flowing) were gathered using cylindrical sampler, sealed, crushed, dried, ground, and screened by 16-mesh for use purpose in laboratory.

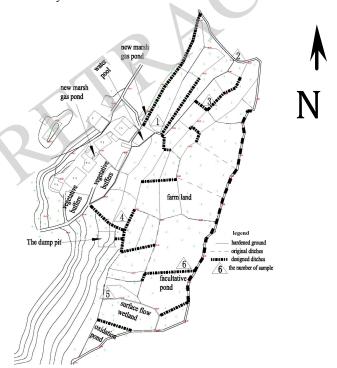


Fig. (1). Sampling location.

The ditch water pH was measured $6\sim9$ on site, the ditch sediment pH was measured $5.8\sim7.2$, and the dissolved oxygen (DO) was measured $4.2\sim8.9$ mg/L on site. With ASTM classification method [13], and the Mastersizer 2000 laser particle size analyzer, the particle distribution was measured: claye particle ($<4~\mu m$) 9.6%, silt particle ($4\sim63~\mu m$) 4.9%, sand particle ($63\sim2000~\mu m$) 15.5%.

3.2. Analysis Items and Analysis Method

3.2.1. Overlying Water

The analysis items included the dissolved total phosphorus (DTP), pH, dissolved oxygen (DO), etc. in the overlying water and all items were measured in line with the Monitoring and Analysis Method for Water and Wastewater (the 4th edition).

3.2.2. Sediments

The phosphorus forms in the sediments were decided by Specification for Lake Eutrophication Survey (the 2nd edition) and SMT [14] (graded extraction method). After determination, the sediment background values are NaOH-P 57.76 mg/kg, HCl-P 6.01 mg/kg, IP 63.77 mg/kg, Or-P 12.73 mg/kg and TP 76.49 mg/kg, accounted for the 75.51%, 7.86%, 83.37%, 16.63% and 100% of TP respectively. It is clear that the sediments are mainly composed by inorganic phosphorus, and the inorganic phosphorus mainly contains NaOH-P component, suggesting that the agricultural water body pollution is serious [15].

3.3. Experimental Methods

3.3.1. PH and Rule of Phosphorus Migration Transformation

15 g previous processed samples and 200 ml deionized water were added in each of 18300 mL absorption bottles. The bottles were divided into both Group A and B. Group A was sterilized 30 min. Group A and B were divided into 3 subgroups each, in which NaOH and HCl solution was added to regulate pH to 5, 7 and 9.18 samples were sealed and placed in a shaker $(25 \pm 0.5)^{\circ}$ C and oscillated at 150-160 r/min. The experimental time was 30 days. The samples were collected from overlying water in predetermined period to analyze. A stable pH was maintained in each bottle during the experiment. Final distribution of phosphorus forms in the sample was determined after completion of the experiment.

3.3.2. Dissolved Oxygen and Rule of Phosphorus Migration and Transformation

300 g previous processed sample was gently laid in a 1000 mL tall beaker, and 750 mL deionized water was added in mud-water height ratio about 1:5. The sample was cultured in a constant temperature incubator culture (25°C). During the culture, dark time and sunshine time were 12 hours a day separately. The sample was divided into 2 groups (anaerobic and aerobic), each group included 3 parallel sub-samples and all were put under a sealed state. The air was aerated daily under the aerobic conditions and the nitrogen (at purity 99%) was aerated daily under the anaerobic conditions about 2 h (in such an amount that the

bottom mud does not overflow upwards). The sample was sealed with plastic film to prevent against the contact and circulation between air and overlying water. Samples were taken from overlying water to analyze DO and DTP in a period 30 days. Final distribution of phosphorus forms in the sample was determined after completion of the experiment.

3.3.3. Sunlight and Rule of Phosphorus Migration and **Transformation**

15 g previous processed sample was gently laid in a 500 mL tall beaker and 200 mL deionized water was added in a mud-water height ratio about 1:2. The samples were tested separately under sunlight-free illumination (0 lx), low illumination (3000 lx) and high illumination (6000 lx). 3 repeated experiments were done in each illumination. The sunlight time and sunlight-free time were 12 hours each. During the experiment, the beaker is mount-open so the overlying water is contacted and circulated with the air. The experimental temperature was controlled at 25°C for continuously 30 d. Samples were taken from overlying water regularly. The beaker was wrapped with black card-paper tightly for the sunlight-free group. Final distribution of phosphorus forms in the sample was determined after completion of the experiment.

4. RESULTS AND DISCUSSION

4.1. PH and Phosphorus Migration and Transformation

As illustrated in Figs. (2, 3), DTP in overlying water is at minimum in a neutral state, pH rise or fall will cause the increase of phosphorus concentration in overlying water, however, pH rise is prone to increase the phosphorus concentration in overlying water, namely pH increase will facilitate the phosphorus migration and transformation from soil to water. The phosphorus in sediments will migrate to water regardless of acidic, neutral or alkaline state of sediments. The NaOH-P (Fe/Al-P) migrates in a best significant way at alkaline state whilst HCl-P (Ca-P) is prone to migrate in acidic conditions. The results agreed with findings of Gao Li, et al. [16-18].

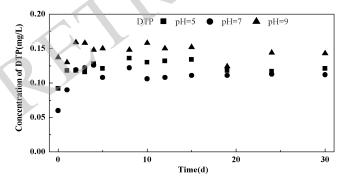


Fig. (2). Influence of pH on phosphorus concentration in overlying

XPS characterization of sediments before and after experiment further proofs the above phenomenon (Figs. 4, 5). The narrow scan spectrum on surface Fe (2p) before phosphorus release in sediments shows the binding energy of surface Fe (2p) is 711.41 ev. Based on XPS chemical-state

database, it is suggested that the main object phase of surfaceiron is Fe₂O₃; the binding energy of surface

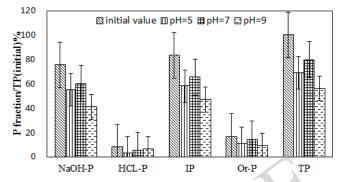


Fig. (3). Migration and transformation of phosphorus forms in ditch sediments in original state and at different pH values (%).

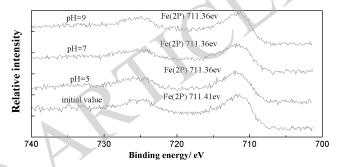


Fig. (4). Fe (2p) binding energy at different pH value.

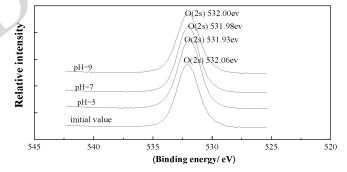


Fig. (5). O (1s) binding energy at different pH value.

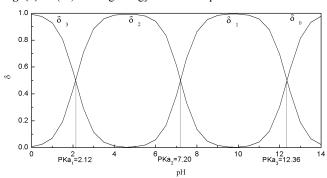


Fig. (6). The relationship between the four forms of phosphate distribution coefficients and the pH.

Fe (2p) is all 711. 36 ev after the sediment releases the phosphorus under all pH conditions, and does not change comparing to the findings prior to experiment. It is clear from the relationship deduced by Ding [19] between the Fe

energy charge level and the binding energy that the surface charge of iron atom has almost not changed. From the relationship constructed by Jong [20] between O(1s) binding energy and surface charge, the surface charge calculated on the oxygen atoms has almost not changed as well (the binding energy of sediments before and after experiment are 532.06 ev, 531.93 ev, 531.98 ev and 532.00 ev respectively), namely, they change in an identical trend, suggesting that no electron transfer occurs insignificantly during the process of phosphorus release in sediments, i.e. no electron transfer reaction occurs on surface. This further proofs that the phosphorus absorption form HPO₄²⁻ in sediment substitutes -OH state on the sediment surface. According to Fig. (6), the free OH in the solution is high in the concentration under the alkaline conditions, which is prone to compete for absorption with phosphate anions bound by Fe³⁺, Al³⁺, etc. Eventually Fe³⁺ and Al³⁺ in sediments are transformed to more stable hydroxyl compound. Phosphates binding to Fe³⁺ and Al³⁻ migrate to water due to the ion exchange so that the phosphorus concentration rises in water solution [21-23]. After computing, Fig. (3) also shows that the reduced NaOH-P amount is up to 49%, 31% upwards of HCl-P 18%. Contrarily, the reduced HCl-P amount is up to 39% under the acid state, 11% upwards of NaOH-P 28%, suggesting the increasing pH produces a larger influence on phosphorus migration and transformation, the alkaline state is beneficial for NaOH-P migration to water and the acidic state is beneficial for HCL-P migration to the water. The reasons may be (1) The ratio of NaOH-P to HCl-P is up to 9.61 in original sedimentary soil; (2) The phosphorus migration and transformation in sediments and water are subject to the phosphate solubility in the case of smaller pH. The calcium phosphate increases in solubility, thus leasing to the phosphorus migration to water better [16, 23].

4.2. Dissolved Oxygen and Phosphorus Migration and Transformation

The rules derived under the aerobic and anoxic experiments are the same as the previous study results, namely, the hypoxic environment facilitates migration from the overlying water to sediments and aerobic environment does in an opposite manner [24].

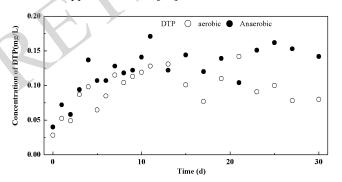


Fig. (7). Dissolved oxygen (DO) influence on phosphorus migration and transformation in the overlying water.

Fig. (7) shows that the DTP concentrations in overlying water under the aerobic state are lower than under anaerobic state.

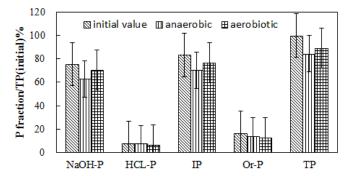


Fig. (8). Mgration and transformation of phosphorus forms in ditch sediments under differential state (%).

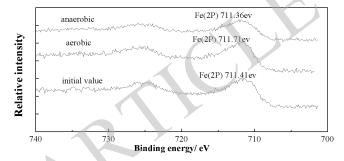


Fig. (9). Fe (2p) binding energy at different state.

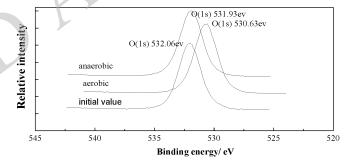


Fig. (10). O (1s) binding energy at different state.

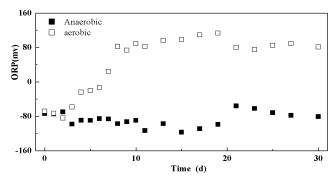


Fig. (11). Change rule of ORP in overlying water.

Fig. (8) shows NaOH-P in sediments decreases under anaerobic and aerobic conditions, but it releases significantly under anaerobic conditions, whilst HCl-P and Or-P decrease more under aerobic conditions. Fig. (8) also shows NaOH-P is the main source of phosphorus release, which is agreed with what was derived by Wang Shengrui *et al.* [25]. XPS characterization data in sediments before and after the experiment is agreed with the above conclusion. The narrow

scan spectrum on its surface Fe (2p) shows that the binding energy on surfaces Fe (2p) is 711.41 ev and 711.71 ev respectively whilst the binding energy on O (1 s) is 532.06 ev and 530.63 ev before and after aeration respectively. The binding energy on surface Fe (2p) and O (1 s) after nitrogen aeration is 711.36 ev and 531.93 ev respectively. Similarly, computing by the model [19, 20], it is clear that the surface charge of iron after oxygen aeration reduces, the surface charge of oxygen increases, suggesting that the electron transfer occurs on the sediment during aeration, namely, the oxidation-reduction reaction occurs on the surface. No electron transfer occurs before and after nitrogen aeration, namely, no redox reaction occurs.

4.3. Sunlight **Phosphorus** and Migration and **Transformation**

4.3.1. PH and ORP (Oxidation-Reduction Potential) Change Rule in Overlying Water

Fig. (12) shows that pH changes significantly in overlying water in sunlight group, but the illumination is correlated to the pH value insignificantly. The pH keeps at 6.5~7.0 in initial 5 d and then starts to rise. Because algae growth in the sunlight group, CO₂ is absorbed, the oxygen is released and the change of pH is significantly higher than sunlight-free group. The presence of algae cell can be observed microscopically. The pH whenever under sunlight or sunlight-free groups, the pH reaches a turning point at the 21st day and then gradually stabilizes. The pH in the sunlight group increases to 8.3 around (alkaline), while the maximal pH is only 7.1 in the sunlight-free group.

The change of DO is similar to pH under different sunlight conditions. Fig. (13) shows that the DO changes significantly in overlying water in sunlight group, but the illumination is correlated to DO insignificantly. It keeps at 0.1-0.8 mg/L in initial 5 d and then starts to rise. The increased DO in sunlight group is significantly higher than sunlight-free group. The DO in the sunlight group reaches a turning point at the 21st day and then gradually stabilizes whilst DO in sunlight-free does not change obviously. The DO in sunlight group increases to 8.3 mg/L around and is 7 mg/L in sunlight-free group at the end of experiment.

The above results show that illumination affects the dissolved oxygen concentration by affecting biological processes, and ultimately affect the change of pH and DO in the overlying water [26]. Under the presence of sunlight, the pH and DO in the overlying water will increase. The equation is specified as follows:

$$CO_2 + H_2O \xrightarrow{\text{llum in ance \& } \bullet \text{ chlorphyll}} [CH_2O] + O_2$$
 (1)

Due to the immaturity of the interfaced benthic algal communities during the early experimental period, pH is at 6.5~7.0, whilst DO maintains at 0.1-1.0 mg/L. Under sunlight conditions, the algae thrive in the photosynthesis gradually and cause the significant rise of DO. At about 21 d, the biological growth under sunlight conditions reaches a peak, the interfaced communities are at a dynamic equilibrium, and the pH and DO are thus stabilized. However, Figs. (12, 13) also show the effect of illumination does not a significant rule on the pH and DO, suggesting that there are a plurality of algae in the ecosystems under the study whilst the saturated light intensity is different as required by the maximum specific growth rate in different algae [27, 28].

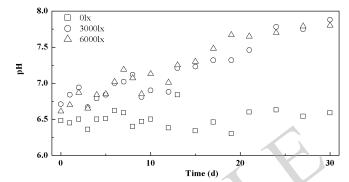


Fig. (12). Change of pH in overlying water.

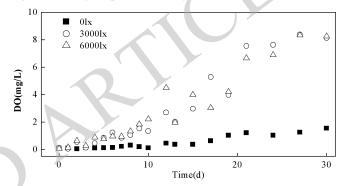


Fig. (13). Change of DO in overlying water.

Sunlight Migration and **Phosphorus Transformation**

Figs. (14, 15) show that the amount of phosphorus migration and transformation to the overlying water in the sunlight group is generally lower than the sunlight-free group.

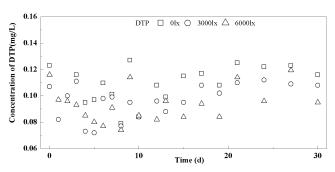


Fig. (14). Influence of sunlight on the phosphorus concentration in the overlying water.

XPS data from Figs. (16, 17) show that the binding energy on sediments surface Fe (2p) is 711.41 ev, 711.71 ev, 711.72 ev and 711.70 ev respectively whilst the binding energy on O (1 s) is 532.06 ev, 531.98 ev, 530.507 ev and 530.591 ev respectively before and after the experiment. Computing by the model [19, 20] gives that the surface charge of iron reduces under the sunlight action, the surface charge of oxygen increases, suggesting that the electron transfer occurs on the sediment during sunlight action significantly, namely, the oxidation-reduction reaction occurs on the surface. But it is correlated to the illumination insignificantly. No electron transfer occurs under 01x illumination, namely, no redox reaction occurs. This finding is agreed with above conclusion. That is, the illumination affects NaOH-P transformation indirectly by changing pH and DO [29] mainly affected by biological process. Due to this reason, the phosphorus assimilation by algae is not appropriate to the corresponding relation between algae proliferation and sunlight intensity in Figs. (14, 17). Based on the conclusion in pH and DO change rule in overlying water, this will be conducive to phosphorus migration and transformation from sediments to the water. However, Figs. (14, 15) show that the phosphorus concentration in the overlying water under sunlight condition is not higher than under the sunlight-free group. It is obvious that the biological effect may equal or surpass the pH influence. The specific mechanism remains to be further research.

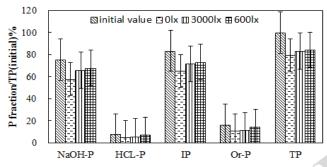


Fig. (15). Rule of migration and transformation of phosphorus forms in original state and under different sunlight intensity (%).

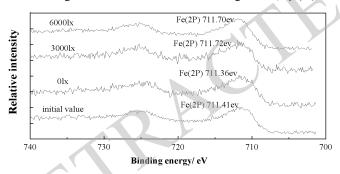


Fig. (16). Fe (2p) binding energy at different sunlight intensity.

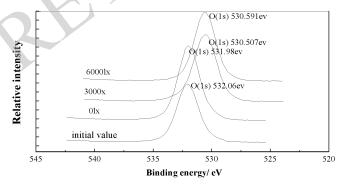


Fig. (17). O (1s) binding energy at different sunlight intensity.

HCl-P and Or-P is far more vulnerable to the biological effects than NaOH-P because Zhou Yiyong [30] et al. found

that the mineralized phosphorus can be effectively dissolved in organic acid synthesis in microbes. The large number of strains that can produce organic acids and gluconic acid are the typical medium. The organic phosphorus mineralization is also an acting process of phosphatase. All organisms in the water can produce the phosphatase as the greatest contribution is made by bacteria, phytoplankton and zooplankton [31]. Therefore, the organophosphorus and HCl-P decreased more significantly in sunlight group. But the illumination is correlated with the released phosphorus insignificantly, thus further proofing the presence of different algae systems. The phosphorus assimilation by algae is not appropriate to the corresponding relation between algae proliferation and sunlight intensity. This finding is agreed with the previous studies on phosphorus release.

CONCLUSION AND RECOMMENDATIONS

The sediment is composed mainly of inorganic phosphorus that includes NaOH-P component majorly, suggesting that the agricultural water pollution is more serious.

Many researchers has studied the process of phosphorus release of sediment influenced by pH, DO, light, but the existence of electron transfer phenomenon is to be revealed by XPS binding energy data combined with phosphorus migration process rarely. In order to reveal the migration of phosphorus into the essence, XPS binding energy data is unified with phosphorus migration process. The results are as follows.

The influence of pH migration and transformation in sediments varies with the phosphorus forms. The ditch sediments in the study can cause the rise of phosphorus concentration whenever at acidic or alkaline state, but the amount is more under the alkaline state. There is no electron transfer occurs insignificantly at different pH values in sediments, the phosphorus absorption form HPO₄²⁻ in sediment substitutes OH state on the sediment surface.

When oxygenated, the oxidation-reduction reaction occurs on the sediment surface, Fe²⁺ is oxidized into Fe³⁺ under aerobic atmosphere, the absorbed phosphorus increases. Under anaerobic atmosphere, the phosphorus in sediments is prone to migrate to the water.

The illumination affects NaOH-P transformation indirectly by changing pH and DO mainly affected by biological process. The electron transfer occurs on the sediment during sunlight action significantly, namely, the oxidation-reduction reaction occurs on the surface. But the illumination is correlated to the phosphorus migration and transformation in the sediments insignificantly. Conclusively, the migrated and transformed phosphorus in sunlight-free group is greater than the sunlight group.

Based on these conclusions, it is recommended the aquatic plants, especially submerged plants, must be grown in field ditches. This will not only be beneficial to oxygen recovery, but also lower the pH in the overlying water without any hindrance to the sunlight. However, the attention should be paid to the planting density and regular harvesting.

In this paper, there are still some limitations. Mainly, the factors of temperature and flow rate are not taken into account, and the effects of various factors are not independent of each other, but intercrossed. Therefore, the further studies must explore the other factors, compute the contribution rate of each factor and finally determine the predominant factor.

CONFLICT OF INTEREST

The authors declared that they have no conflict of interest to this work.

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