Study on CO₂ removal method in recirculating aquaculture waters

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Abstract

The dissolved CO₂ accumulation has become an important factor restricting production in the high-density recirculating aquaculture system in which pure oxygen injection is used. In this paper, a novel CO₂ removal device is designed for the recirculating aquaculture water environment based on the principle of gas exchange. In terms of experiments, the DOE (design of experiment) method is applied to design three factor two level orthogonal experiment. Further, significance effect of gas to liquid ratios (G/L), inlet CO₂ concentration, the water flow rate (Q_w) on CO₂ removal efficiency is analyzed. Results show that G/L has the most significant influence on the CO₂ removal efficiency. Influences of the latter two on CO₂ removal efficiency are not apparent. Tests results of G/L effect on CO₂ removal efficiency show that, when G/L=1~5, CO₂ removal efficiency increases rapidly with the increase of G/L; when G/L=5, CO₂ removal efficiency=80%~88%; when G/L=8, CO₂ removal efficiency=86%~92% , when G/L>8, CO₂ removal efficiency increases gently with the increase of G/L. Considering both system energy saving and effective removal of carbon dioxide, G/L=5~8 is considered to be the best for the aquaculture water CO₂ removal device running, CO₂ removal efficiency=80%~92%.

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This work is supported by National Flounder Species industrial system project(nycytx-50-G04) and key technology research and major equipment development of Fishery energy-saving(201003024)
1. Introduction

The high concentration of CO$_2$ is harmful to the fish in aquaculture waters environment. When the concentration of CO$_2$ exceeds the safe level, the amount of oxygen that the blood hemoglobin of fish can carry is reduced significantly and respiration distress can occur, even with high concentrations of dissolved oxygen in the water. Meanwhile, the whole system’s pH also decreases dramatically and the performance of biological purification is affected [1]. In the traditional culture model, because of the low stocking density, CO$_2$ doesn’t accumulate excessively, which doesn’t make fish dangerous. In the recirculating aquaculture system, the stocking density raises and the water exchange rate drops (about 10%) [2]. Consequently, large amounts of dissolved carbon dioxide will greatly restrict production. When stocking densities were less than 30 to 60kg/m$^3$, conventional aeration systems would generally provide sufficient removal of CO$_2$ through transferring oxygen into the water with airstones, surface agitation and water falls. However, with the increase of the fish density to 100kg/m$^3$ or higher, in order to make the aquaculture system more productive, pure oxygen systems become a widely used aerobic way to meet the demand of the normal growth of fish for dissolved oxygen,. For every 10mg/L of oxygen consumed, approximately 13–14 mg/L of CO$_2$ excreted through fish gills. As a result, The CO$_2$ accumulates to a high concentration through respiration of the fish and biological nitrification [3-5], which is great toxic to fish. The safe operating levels of CO$_2$ depend on the species, development stage, and overall water quality [6]. In general recommendation, the CO$_2$ concentration of aquaculture water should be less than 10mg/L [7].

CO$_2$ removal technology of aquaculture waters in china is still in the pilot study stage, generally, a large-scale recirculating aquaculture system does not set CO$_2$ removal link. However, the United States and Europe have used CO$_2$ removal device in the intensive recirculating aquaculture systems successively [8-10], the effective CO$_2$ removal has been achieved and fish production per unit of water has been rised. Currently, the mainstream CO$_2$ removal devices are stripping columns [11]. Due to a lot of factors, accurately predicting the removal rate is very difficult. In this paper, through three factor two level orthogonal testing, the effect of gas to liquid ratios(G/L), inlet CO$_2$ concentration, water flow rate(Q$_w$) and their interactions on the CO$_2$ removal rate are studied and the best level combinations are discovered to achieve the effective CO$_2$ removal.

2. Materials and methods

2.1. The testing device and principle

CO$_2$ removal testing device is a vertical cylinder in which the pickings are irregular or trims are piled up in the supporting plates near to the bottom of the column. Fans blow gas to the bottom. The liquid is poured into the packing shed layer surface by the distributor at the top of the tower, disperses to film in the packing surface, and flows down through the gap between the packing. The packing surface is to be the mass transfer surface of gas and liquid two-phase contact. CO$_2$ solubility in water is in line with Henry's law, that is, in a certain temperature, the gas solubility in water is proportional to gas partial pressure on the liquid surface, so as long as CO$_2$ partial pressure in the gas is very small, CO$_2$ will escape from the water, this process is known as desorption. There is few CO$_2$ in the air. Its partial pressure is about 0.03% of atmospheric pressure [12-13]. Therefore, air is commonly used as the medium of CO$_2$ removal device, which is sent to the bottom of CO$_2$ removal device by the blower. In the packing surface,
air fully contacts with water, and then it discharges from the top of tower together with the escaping CO₂. Water with CO₂ enters into the upper part of the tower and flows down through the liquid distributor. After water is in full contact with air in the packing surface and degassed CO₂, it is drained from the water outlet of the bottom. Finally the process of the CO₂ removal is finished. The structure of the CO₂ removal device is shown in Figure 1.

Fig.1. Structure of CO₂ removal device

It is composed of the cylinder made of acrylic material, water outlet, air inlet, liquid distributor, packing support plate, packing and so on. The opening porosity of liquid distributor is 15.6%, the height of packing is 1m, the type of packing is Φ25X25 Pall ring, made of polypropylene plastic.

Fig.2 The flow of CO₂ removal testing
2.2. Testing System

The flow of CO₂ removal testing is shown in Figure 2. The testing system includes a CO₂ cylinder, two adjusting pools, a 25m³/h pump, two copper floodgate valves, a water glass rotameter whose measuring range is from 0.4m³/h to 5m³/h, two gas flow meters whose measuring ranges were from 1 m³/h to 10 m³/h and from 5 m³/h to 50 m³/h respectively, and the CO₂ removal testing device. The clean tap water, room temperature air and CO₂ gas of 99.99% purity are also utilized in the testing.

2.3. Testing procedures

1) Clean water is injected in the adjusting pool 1 with the given depth. The valve of CO₂ cylinder is opened for 5 minutes aeration. Water in the adjusting pool 1 is stirred to mix evenly and mixture should be stabilized for 30 minutes.

2) Sample bottles with 200ml volume are adopted for water sampling. In order to prevent the escape of dissolved CO₂ from the water, sampling bottles are fully filled and 10 groups are sampled.

3) With the preparation of 1mol/L and 0.5mol/L NaOH solution, CO₂ concentration is determined by using Metrohm automatic titrator Titrando809. The average value of 10 groups of water samples serves as the final result of the titration.

4) According to the titration results, a certain level of water in the adjusting pool 1 is injected into the adjusting pool 2 and diluted to the desired test concentration. Subsequently 10 groups of water samples are titrated again for the amendments of the inlet CO₂ concentration.

5) When the blower is opened, air enters into the tower by opening copper floodgate valve 2. The rate of the gas flow can be adjusted by the gas flow meters. Likewise, the water flow rate is adjusted by the glass rotameter after the pump is opened.

6) After the system is running for 2 minutes, the potentiometric titration should be carried out after 10 water samples bottles are taken out of the water.

2.4. The determination of DOE test factors and their levels

The Western classical DOE test factorial design method [14-15] is adapted to analyze the impact factors of CO₂ removal efficiency.

The main impact factors of CO₂ removal efficiency are gas to liquid ratios (G/L), inlet CO₂ concentration, water flow (Q₆), the type of packing, the height of packing and so on. In this test, since the type and height of packing are determined by empirical data, gas to liquid ratios (G/L), inlet CO₂ concentration and water flow (Q₆) were selected as DOE test factors. According to the research results of Timmons and others [1], in the design of CO₂ degassing tower, the reference range of G/L is from 5 to 20, and that CO₂ removal efficiency could reach 100% when G/L=20, the hydraulic loading are from 17kg/m²s to 24kg/m²s. A three factor two level orthogonal test is designed based on DOE. The level values of DOE test factors are shown in Table 1, DOE factorial design sheet is shown in Table 2.

| Test factors | G/L | Water flow Q₆/ (m³/h) | Inlet CO₂ concentration/ (mg/L) |
|--------------|-----|----------------------|-------------------------------|
| Level data   | 1,20| 1.2                  | 25,77                         |
Table 2. DOE factorial design sheet

| Standard order | Running order | Central point | Blocks | G/L | Qw | CO2_{in} |
|----------------|---------------|---------------|--------|-----|----|----------|
| 7              | 1             | 1             | 1      | -1  | 1  | 1        |
| 6              | 2             | 1             | 1      | 1   | -1 | 1        |
| 2              | 3             | 1             | 1      | 1   | -1 | -1       |
| 1              | 4             | 1             | 1      | -1  | -1 | -1       |
| 5              | 5             | 1             | 1      | -1  | -1 | 1        |
| 8              | 6             | 1             | 1      | 1   | 1  | 1        |
| 4              | 7             | 1             | 1      | 1   | 1  | -1       |
| 3              | 8             | 1             | 1      | -1  | 1  | -1       |

2.5. Evaluation indexes

CO2 equilibrium system in the natural water is very complex and important, mainly including the following equilibrium processes [16].

- **Balance of dissolved and escaping CO2:**
  \[ \text{CO}_2 \text{ (gas)} \rightleftharpoons \text{CO}_2 \text{ (dissolved)} \]  

- **Hydration balance of dissolved CO2:**
  \[ \text{CO}_2 \text{ (dissolved)} + \text{H}_2\text{O} \rightleftharpoons \text{H}_2\text{CO}_3 \]  

- **Ionization equilibrium of carbonic acid:**
  \[ \text{H}_2\text{CO}_3 \rightleftharpoons \text{H}^+ + \text{HCO}_3^- \]  
  \[ \text{HCO}_3^- \rightleftharpoons \text{H}^+ + \text{CO}_3^{2-} \]

The important difference between CO2 and other gases is that when CO2 dissolved in water, it undergoes chemical reactions, forming carbonates and H+. Hence, CO2 removal process involves both mass transfer and chemical equilibria reactions. The carbonate system is unable to instantaneously achieve equilibrium during the degassing process because the dehydroxylation of HCO3− to CO2(aq) is a slow process in terms of chemical equilibria, taking over 1min to reach full equilibrium[17]. The slow dehydroxylation rate means that as a body of water passes through a degassing unit (it often only takes a few seconds), there is no effective replacement of the HCO3− in the pool with a fraction of the degassed CO2. The CO2 fraction is not replenished until the water re-establishes equilibrium some time after leaving the degassing unit, and the amount of CO2 that is re-formed depends on the ionization fraction [18].

According to the complex processes, the foreign researchers proposed two evaluation standards. One is CO2 removal efficiency [19], the other were the standard CO2 transfer rate (SCTR, g CO2 h⁻¹) and CO2 transfer rate (CTR, gCO2h⁻¹) [9].

**CO2 removal efficiency (%)**
\[ \text{CO}_2 \text{ removal efficiency} = \frac{\text{CO}_2 \text{ (in) } - \text{CO}_2 \text{ (eff)}}{\text{CO}_2 \text{ (in)}} \times 100\% \]  

Where CO2_{in} = influent CO2 concentration(mg/L), CO2_{eff} = CO2 concentration of effluent water after equilibration reactions(mg/L).

**SCTR**
\[ \text{SCTR} = \left( \frac{K_{L}a_{CO2}}{C_m - (C_{CO2})_{eff}} \right) V 60 \times 10^{-3} \]
\[ CTR = \left( K_L a \right)_{20} C_m - (C_s)_{20} V \times 60 \times 10^{-2} \theta^{\text{Temp}-20} \]  \hspace{1cm} (6)

Where \((K_L a)_{20}\) is the mass transfer coefficient at 20℃(h⁻¹), \((Cs)_{20}\) is saturation concentration at 20℃(mg/L), \(C_m\) is standard measured concentration (arbitrarily defined as 1mg/ L), \(V\) is volume of water (L), 60 is conversion from min to h, and \(10^{-3}\) converted from mg to g.

\[ (K_L a)_{20} = \frac{\ln(CO_2^{eq}(2\text{air}) - CO_2^{eq}(2\text{in})) / CO_2^{eq}(2\text{air}) - CO_2^{\text{eff}}(2\text{eff}))}{t} \]  \hspace{1cm} (7)

Where \(CO_2^{eq}(2\text{air})\) = CO₂ concentration of water in equilibrium with the air entering the air-lift(mg/L), \(CO_2^{eq}(2\text{in})\) = influent CO₂ concentration(mg/L), \(CO_2^{\text{eff}}(2\text{eff})\) = CO₂ concentration of effluent water prior to kinetic equilibration reactions(mg/L), \(t\) = test time(h).

Equation (4) is different from formula (5) and formula (6), because CO₂ removal efficiency is calculated with the use of CO₂ concentration of effluent water after equilibration reactions, the process involves both mass transfer and chemical equilibrium reactions. Equation (4) considered comprehensively, so CO₂ removal efficiency is adapted in this paper to be the system evaluation index.

3. Results and Analysis

3.1. Significant Analysis of test factors

According to the running order in table 2, 8 groups of test were carried out. The testing results are shown in Table 3.

Table 3. CO₂ removal efficiency testing results

| Test order | 1    | 2    | 3    | 4    | 5    | 6    | 7    | 8    |
|------------|------|------|------|------|------|------|------|------|
| influent concentration | 77.20 | 77.20 | 25.07 | 25.07 | 77.20 | 77.20 | 25.07 | 25.07 |
| Effluent concentration | 33.38 | 5.66  | 0.95  | 7.58  | 16.63 | 9.76  | 4.34  | 11.99 |
| CO₂ removal efficiency | 56.75% | 92.60% | 96.20% | 69.75% | 78.45% | 87.36% | 82.66% | 52.13% |

Using DOE Factorial design analysis, standard Pareto diagram is obtained, as shown in Figure 3.

Figure 3 shows the significant sizes of all impact factors and absolute values of the response intuitively. Any effect extending to the reference line (red line) at the default level (α=0.05) is significant, α value used in the hypothesis testing is the maximum acceptable level when the original hypothesis is rejected, with the probability between 0 and 1 [20-21]. It could be seen from Figure 3 that the most significant factor is G/L, while the effects of the water flow \(Q_w\), inlet CO₂ concentration, the interactions of two factors and the interactions of three factors on CO₂ removal efficiency were not significant. Therefore, in the actual operation process of CO₂ removal device, adjusting water flow \(Q_w\), inlet CO₂ concentration and the interactions between factors is not effective in improving CO₂ removal efficiency, G/L should be adjusted for higher CO₂ removal efficiency.

To quantify the relationship between G/L and CO₂ removal efficiency, the testing of effect of G/L changes on CO₂ removal efficiency is implemented.
3.2. The effect of G/L changes on CO₂ removal efficiency

Gas to liquid ratios (G/L) is the volume ratio of air inflow and Water treatment capacity per unit time in the working state of CO₂ removal device. The water flow Q_w is altered 1 m³/h by glass rotameter. The value of G/L has six levels which are 1, 5, 8, 10, 15, and 20 respectively. Thus, air flow rates are 1 m³/h, 5 m³/h, 8 m³/h, 10 m³/h, 15 m³/h and 20 m³/h correspondently. According to recirculating aquaculture water quality criterion [1], for the tolerant species (tilapia), the CO₂ concentration in culture water should be less than 60 mg/L, for the sensitive species (salmon), the CO₂ concentration in culture water should be less than 20 mg/L. Therefore, in this testing, the influent CO₂ concentration has three levels, which are 20 mg/L, 60 mg/L, 80 mg/L. Under these conditions, 18 groups of testing should be conducted and ten parallel tests should be implemented each group. The results are presented in Figure 4, when G/L=1-5, CO₂ removal efficiency increases rapidly with the increase of G/L; when G/L=5, CO₂ removal efficiency=80%-88%; when G/L=8, CO₂ removal efficiency=86%-92% when G/L>8, CO₂ removal efficiency increases gently with the increase of G/L. At the same time, the effects of G/L on CO₂ removal efficiency at different CO₂ concentrations changes a little bit, it proves once again that inlet CO₂ concentration is not an important factor.
4. Discussion

4.1. Selection of removal methods

In addition to the classic way of stripping column, CO₂ in the recirculating aquaculture waters can also be removed by chemical methods. According to CO₂ concentration, the proper amounts of alkali can be put into water.

But this method has many disadvantages. For example, CO₂ is part of the chemical equilibrium in the water, accurately calculating the amount of alkali to be cast is quite hard. Practically, amounts of alkali can easily exceed the normal level without cautions when it is cast, which results in harm to fish. Additionally, the high CO₂ concentration exists in the intensive aquaculture system in which a large quantity of alkali is required every day. The full response needs a period of time. In consequence, the cycle of CO₂ removal takes much more time. Nevertheless, the classical way of stripping column employs physical methods to remove CO₂. It doesn’t produce additional side effects on the system. Furthermore, its principle is simple and its performance is satisfactory. CO₂ in the culture water is removed in a short time. Other researches have shown that gas stripping and flotation tank can be in charge of removing CO₂ to some extent [9].

4.2. The economic measures of CO₂ removal device running

Considering current level of economy of China, the scale of investment in the existing mode of recirculating aquaculture is large with high cost especially in terms of energy [22]. Reduction of the energy consumption and system optimization has become popular research directions. Although water pumps and thermoregulation are the main energy consumption components in the system, the energy consumption of technical procedures such as removal of particulate matter, oxygenation, and CO₂ removal cannot be ignored as well.

The energy consumption of the classic stripping column consists of head loss and the blower. In the system design and actual production, the following measures can be taken to reduce energy consumption of the process:

1) The proper allocation of volume of water circulation through CO₂ removal device. Summerfelt [10] and others have studied on carbon dioxide production within biofilters contains in recirculating salmonid culture systems. Their research results have showed that the biofilter actually accounted for 37% of the total CO₂ produced within this recirculating salmonid system. Therefore, the CO₂ stripping unit should be placed immediately after the biofilter simply as common sense to optimize water quality. In the recirculating aquaculture system, the volume of water circulation should be designed in terms of meeting demand for dissolved oxygen and ensuring the rapid removal of the soluble ammonia nitrogen in water in general[23]. Larger the amount of water circulation in the system becomes, higher the stocking density is. Relative to oxygen consumption and ammonia nitrogen accumulation, the concentration of CO₂ in the aquaculture water rises slowly. It costs a few hours to reach the tolerance level of the fish [1]. Therefore, the volume of water circulation through CO₂ removal device do not have to be too large, CO₂ removal device may be as a spur track to parallel in the main loop after the biological filter, the ball valve can control its amount of water circulation. In the design of system, the number of CO₂ removal device should be determined by the size of culture load. In the beginning period, system is generally in a non-full-load working condition, with low-density, small fish and low CO₂ concentration in culture water, you can choose not to open the CO₂ removal device. In the middle and later periods, with the density increases, the concentration of CO₂ in culture water will also climb, you can choose the number of CO₂ removal device running as required.
2) Reduce air supply energy consumption. G/L required in the CO₂ stripping column is larger, the blower is the main energy-consuming equipment. The test results show that when G/L > 8, the CO₂ removal efficiency increases gently with the G/L increased. When water flow Q_w is a fixed value, the higher G/L means that the more volume of air is entered into the CO₂ removal device, the blower energy consumption is more serious. If the CO₂ concentration in aquaculture water is in the safe level, it will not cause harm to the fish, therefore, CO₂ removal device does not require the removal rate of 100%, it just need to control the CO₂ concentration at safe levels. In actual operation, both system energy saving and effective removal of carbon dioxide should be considered, to select the appropriate G/L. Not only the effective CO₂ removal is realized, but also system can be running with lower energy consumption of air supply. According to the testing results, it is proposed that G/L of CO₂ removal device should be in the range of 5 to 8 in the actual conditions. In addition to blowers, we can take full advantage of natural conditions to meet the CO2 removal device for air demand. The air outlet of CO₂ removal device will be designed to be the tube of “Stubbs wind” extended out of the roof, the hood will be installed on the top of the tube. In the winter or outside windy conditions, with combined effects of “Stubbs wind” and the hood, the indoor air will be pumped into the tower from the air inlet, and discharge from the tube of “Stubbs wind” through packing, to realize unpowered ventilation with the blower replaced. Theoretical results show that the generating air volume with combined effects of “Stubbs wind” and the hood can meet the demand, the actual performance remains to be further verified.

4.3. The application prospects

In recent years, recirculating aquaculture system has been large-scale employed in many areas [3] and is continually being developed at an amazing speed. The performance of CO₂ removal is satisfying. If it is applied to the intensive recirculating aquaculture system, not only the problem of high CO₂ concentration accumulation will be solved, but also PH in water will be stable. Hence, the research on the CO2 removal methods in culture waters is playing an important role in promoting the development of the intensive recirculating aquaculture system and technological progress of fish-farming in china.

4.4. Further works

Besides the three impact factors are studied in this paper, packing or without, water drop height, the type of packing, the height of packing, the structure of CO₂ removal device, air speed and so on are also included. The CO₂ removal performance has been carefully investigated under the conditions that the type of packing is Φ25X25 Pall ring, the height of packing is 1m and the opening porosity of liquid distributor is 15.6%, so only three of the more important factors have been considered. For a more comprehensive and precise study on the effects of impact factors on CO₂ removal efficiency, we will take more influencing factors into account in the design of experiment in the future.

5. Conclusions

Based on principle of gas exchange, CO₂ removal device has been designed for recirculating aquaculture water in this paper, DOE (design of experiment) method has been used to study on CO₂ removal efficiency. The testing results of three factor two level orthogonal testing show that, G/L has the most significant effect on CO₂ removal efficiency, the effects of the latter two on CO₂ removal efficiency are not significant. Therefore, in actual operation of CO₂ removal device, G/L should be regulated to improve CO₂ removal efficiency. The testing results of the effect of G/L changes on CO₂ removal efficiency show that, when G/L=1~5, CO₂ removal efficiency increases rapidly with the increase of G/L;
when G/L>8, CO₂ removal efficiency increases gently with the increase of G/L. Considering both system energy saving and effective removal of carbon dioxide, G/L=5~8 is considered to be the best for the aquaculture water CO₂ removal device running, CO₂ removal efficiency=80%~92%.

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