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USE OF CLINOPTILOLITE NATURAL ZEOLITE IN AQUACULTURE - A REVIEW

Steluța Camelia SURMELI (SAVA)[1], Monica Paula MARIN[1], Gratziela Victoria BAHACIU[1], Nela DRAGOMIR[1], Bogdan Alexandru SAVA[2], Carmen Georgeta NICOLAE[1]

[1]University of Agronomic Sciences and Veterinary Medicine of Bucharest, 59 Marasti Blvd, District 1, Bucharest, Romania
[2]National Institute for Lasers, Plasma and Radiation Physics, 409 Atomistilor Street, Magurele, Romania

Corresponding author email: savacamelia64@yahoo.com

Abstract

In 2016, the global aquaculture was 46.82%of total fish production, which means 80 million tonnes of 170.9 million tonnes. Latest researches highlighted that using the natural zeolites in aquaculture in order to maximize the use of resources (water, food, species) and to ensure the lowest negative impact on the environmentwas the most viable solution. The studies on zeolites use, clinoptilolitein particular, were focused on their use as feed additives (up to 2.5% concentration) and also as water quality improvers; this is mainly due to their ability to remove ammonia, its compounds and heavy metals, to reduce water hardness and to consequently prevent diseases and decrease the losses on fish population. A practical and efficient use of natural zeolites in aquaculture will determine increased economic efficiency.

Key words*: ammonia, feed, fish, water quality.*

INTRODUCTION

Aquaculture has the fastest growing rate among the food-producing sectors, especially those producing animal protein (Simeanu et al., 2015). At present, over 50% of the total fish production used as human food comes from this sector and it is expected to increase to 62% by 2030 due to the drastic decrease of wild fishery catches (FAO 2014). In order to ensure a higher productivity, aquaculture technologies need to be more effective and also to reduce the negative impact on the environment with regard of toxic contaminants decreasing both in effluents from recirculating systems and pond wastewater.

Since 1756, over 60 types of natural zeolites have been discovered and more than 150 different types have been synthesized (Ghasemi et al., 2016). In 1857, the reversible dehydration zeolites property was discovered, and in 1858, it was determined that zeolites also have the propriety of cationic exchange in aqueous solution. Due to their porous structure, the specific gravity of zeolites is low. Natural zeolites have considerable variations of chemical composition: water content, cations and the Si / Al ratio (Mishra and Jain, 2011). Due to their accesibility, lower cost and ecological compatibility, zeolites are very important in environment preserving (Bedelean and Avram, 1991). One of the best zeolites used for ammonia removal is clinoptilolite.

Since 2007, zeolite-based additives have been declared safe for end-users of meat, milk or eggs from animals that have received zeolite in feed or mannure. Clinoptilolite zeolite is registered by the European Community as food additive DIN 53 770.

The global use of zeolites (clioptilolitein particular) in bioeconomy has shown a major development. In Romania, since 2000, researchers started to study the use of zeolites in animal husbandry andhave shown favorable effects of their use as nutrients and of feed conversion coefficients (Pogurschi et al., 2017). The use of the Romanian clinoptiloliterich volcanic rock as a feedadditive showed the capacity to improve the milk quality and production, animal health and welfare, ensuring optimal technological conditions for the environment (Marin et al., 2018).

Researchers studied clinoptilolite as an ion-exchange and adsorbent in waste water purification in recirculation systems due to its affinity for certain cations - especially ammonia, but also for its ability to capture toxic heavy metals. Applications of zeolites, especially clinoptilolite, in the transport of live fish as well as their use as potential feed additives have to be also considered.

MATERIALS AND METHODS

This paper presents a review of the most relevant literature regarding the influence of natural zeolites, clinoptiloliteespecially, on the assurance of the best medium conditions for fish activity. It was also investigated the effect of clinoptiloliteadding into the diet on the rate of fish growth. The experiments were conducted mainly on fresh water fish species, but marine species have also been studied.

RESULTS AND DISCUSSIONS

One of the main parameters determining the water quality of water in a fish farm is the concentration of ammonia.

The amount of ammonia in fish ponds is influenced by the feed intensity, the amount of feed protein and wastedfeed that decomposes into the system (Cristea et al., 2002).

Compared to ponds, the carrying capacity of a recirculating system is higher, but the filtering capacity is limited.

Fishes can assimilate the ammonia and the remnant concentration may exceed 0.02 ppm, resulting in a lethargic state and ultimately the death of the fish population.

Even when the concentration of ammonia in the pools does not reach lethal values, a series of sub-lethal effects occur: lower feed conversion, lower growth rate and low immunity to various diseases (Ghasemi et al., 2016).

Total ammoniacal nitrogen (TAN) has two forms: non-ionized ammonia, NH_3, and ionized ammonia, NH_4^+.

The non-ionized form is extremely toxic for most fish species and its share of TAN depends on pH and water temperature. TAN is initially transformed by nitrifying bacteria into nitrites NO_2^-, then nitrates NO_3^-, a nitrogen compound with less toxicity for fish.

By combining nitrite with hemoglobin in fish blood, methaemoglobin is formed, thus preventing the transport of oxygen from gills to tissues (Marin et al., 2015).

In order to decrease the level of ammonia from contaminated waters, several processes can be used in farms: replacement of a volume of waste water with fresh water, nitrification or use of ion exchange processes.

The zeolites used in the aquaculture water purification process show high selectivity to ammonia (Beler-Baykal et al., 1996; Booker et al., 1996; Pansini, 1996).

Researches have shown that the ion exchange reaction between zeolite and water components is influenced by the zeolite type, particle and pores size, Si / Al ratio and chemical composition of contaminated water (Koon and Kaufman, 1975; Jorgensen et al., 1976; Klieve and Semmens, 1980; Curkovic et al., 1997).

The kinetic of the ion exchange reaction is influenced by the particle size of the zeolite; the cations from the solution will not reach the inner positions of larger zeolite particles if the contact time between the solution and the adsorbent is not long enough. 0.7-1.0 mm clinoptilolite and mordenite particles showed a greater absorption capacity of ammonia in a recirculating system compared to larger particles, 1.0-1.4 mm, at a certain contact time (Mwale, 2000).

Improving water quality by using clinoptilolite

Clinoptilolite is among the zeolites with higher affinity to ammonia, along with mordenite and chabazite.

It is a hydrated sodium and potassium / calcium aluminosilicate. It has a unique crystalline structure, "clino" type, and special properties no metter the size of the particle (Emadi et al., 2001).

This structure resists at extreme pressure, it requires high temperatures to break, similar to those that melt the glass and is not chemically attacked, except for extreme acid or alkaline conditions.

Degradation over time is impossible if one of the above conditions is not met.

The ideal simplified formula of clinoptilolite is (Na, K) $6Si_{30}Al_6O_{72}$_nH_2O (Figure 1).

Removal of ammonia from fish ponds

Clinoptilolite is a highly effective adsorbent in removing ammonia from water due to the relatively high cation exchange capacity (Figure 2) and alsoto a high selectivity for ammonium ions in the presence of other competing cations (Peters and Bose, 1975).

The differences among natural zeolites of the same type are due to the different environmental conditions, in which they formed, as well as the quantities and types of impurities they include (Hawkins, 1983).

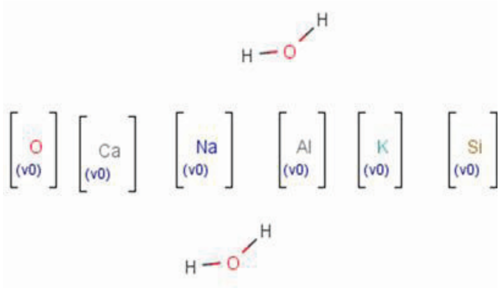

Figure 1. Chemical formula of clinoptilolite (Mutlu et al., 2016)

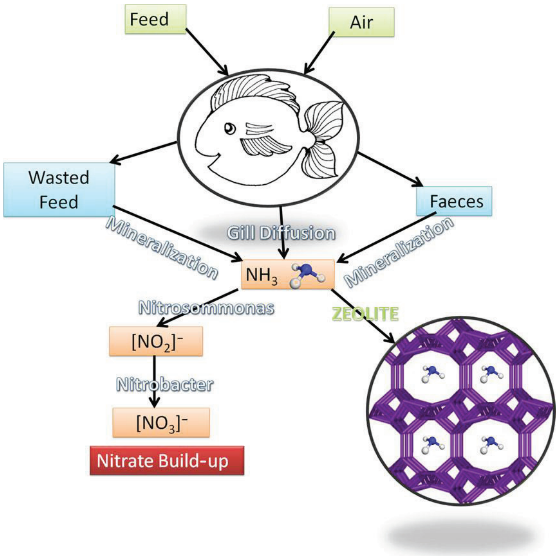

Figure 2. Nitrogen cycle in fish ponds (Durborow et al., 1997)

By using 12 g L^{-1} and 15 g L^{-1},respectively, the survival rate to lethal ammonia concentrations in the case of beluga *(Huso huso)* (Asgharimoghadam et al., 2012; Farhangi et al., 2013) and Persian sturgeron (*Acipenser persicus*) has increased (Farhangi and Rostami-Charati, 2012). The aquarium growth of scalars (*Pterophyllum scalare*) with the addition of 10 g L^{-1} of clinoptilolite was considered optimal for ammonia and water hardness decreasing (Ghiasi and Jasour, 2012). Research on rainbow trout (*Oncorhynchus mykiss*) showed that 15 g L^{-1} of natural clinoptilolite decreased the mortality rate when reaching a lethal concentration (Farhangi and Hajimoradloo, 2011). At thesame time, the effectiveness of a 2.5% natural zeolite diet was demonstrated by lowering the concentration of ammonia by 24% compared to the control group (Ergun et al., 2008).

Clinoptilolite was also used in aquatic growth systems of tilapia (*Oreochromis sp.*) and salad (*Lactuca sativa* var. *Longifolia*) as a bed for salad seedlings, added 10 g each in small cotton bags.

The results have shown that zeolite can improve water quality by lowering the total ammonia concentration (Rafiee and Saad, 2006).

Researchers tried to replace the nitrifying biological filters with natural clinoptilolite and the results are promising.

In a carp recirculating system, the level of nitrate compounds was determined after the filterring of the effluent through a filter of clinoptilolite and nitrifying bacteria. The ammonia values were significantly lower and the amount of nitrates significantly higher in effluent treated with zeolite and bacteria (Motesharezadeh et al., 2015). Studies on recirculation systems (Bergero et al., 1994) have shown a greater effectivenessin ammonia removal of clinoptilolite and phillipsite compared to chabazite-rich volcanic tuff.

In the experiments conducted at USAMV Bucharest, in an aquarium with crucian carp (*Carassius carassius*), the biological filter was replaced with clinoptilolite, an amount of 6.7 g L^{-1} and particle size of 1-3 mm (Sava et al., 2017).

The results were encouraging and it was found that the admissible level of ammonia and nitrite were exceeded only after 32 hours of experimentation (Table 1).

Table 1. Chemical analysis of the samples (Sava et al., 2017)

Time, hours	pH	NH_4 [mg/l]	NO_2 [mg/l]	NO_3 [mg/l]	P_{total} [mg/l]	Ca [mg/l]
0	7.67	0.035	0.026	6.643	0.096	33.5
4	7.72	0	0.030	6.643	0.799	37.5
8	7.71	0	0.076	5.757	1.332	42.3
12	7.72	0	0.056	6.200	1.788	43.1
16	7.76	0	0.033	7.971	2.239	44.7
20	7.77	0	0.033	8.414	2.230	45.5
32	7.66	0.146	0.079	11.514	4.967	48.7

By using acolumn of regenerated clinoptilolite, a decrease from 0.28 mg L^{-1} to 0.08 mg $L^{-1}$of the ammonium content was found in the first 12 hours (Nicolae et al., 2017).

In order to increase the anion retention capacity of the clinoptilolite in water from a recirculating system, researches have been conducted on modifying thezeolite with a surfactant. It has been observed that by increasing the temperature from 10°C to 15°C the retention capacity of the modified zeolite increased from 10 to 20 mg g^{-1} in the case of nitrates and from about 0.0 to 1.2 mg g^{-1} in the case of nitrites (Shokouh et al., 2010).

Researches were also been carried out on the possibilities of using synthetic zeolites as such or with natural ones in fish farms (Ariful Islam et al., 2014). Regarding theammonia removal from freshwater culture systems, the results have shown that the overall efficiency of synthetic zeolites is lower and their cost is higher compared to natural clinoptilolite.

Use of clinoptilolite as a feed additive

Clinoptilolite is approved by FDA and EU for birds and animals feeding additive, classified as safe from the health and safety point of view. Up to now, no toxic effect of accidental ingestion of zeolite is known. Including of 2-5% clinoptilolite in fish feed showned an increasing of biomass, an encouraging result to be more investigated.

By adding a 2% zeolite in rainbow trout (*Oncorhynchus mykiss*) feed, the biomass productivity increasedof up to 10%. The study was conducted on 100 trouts that were given normal feed with 48% of proteins for 48 days (Ghasemi et al., 2016). By adding a 5% clinoptilolite into the diet of common carp (*Cyprinus carpio*), the apparent digestibility coefficients and biomass growth were positive (Khodanazary et al., 2013). Feed enhanced with 1-2% clinoptilolite was used for feeding redbelly tilapia (*Coptodon zillii*), and this determined an improved feed conversion and growth performance (Yıldırım et al., 2009). There were also researches that shown no influence on fish development after the use of 5% and 10% clinoptilolite in the diet of silver salmon (*Oncorhynchus kisutch*) (Edsall and Smith, 1989), but these can be explained by the use of zeolites from different geological sources, particle size, or zeolite conditioning.

Improvement of growth indices has been associated with the zeolite detoxification effect (Ortatatli&Oguz, 2001; Rizzi et al., 2003), but also by the slower passage of fodder through the intestine, which has led to an improvement in the use of the nutritive substances in feed (Dias et al., 1998, Eya et al., 2008). Therewith, clinoptilolite captured ammonia, toxic to celular level, which led to a better use of nutrients (Papaioannou et al., 2005).

Other uses of natural zeolites

Researches were also directed towards the investigation of the ion exchange capacity of zeolites in order to remove the various cations of toxic heavy metals from the wasted waters in fish farms. By adding 10 g L^{-1} of clinoptilolite to water, the accumulation of cadmium in the common carp (*Cyprinus carpio*) body was reduced (Ghiasi et al., 2011). Prussian carp (*Carassius gibelio*) tissues analysis revealed the reduction of cadmium bioaccumulation by using clinoptilolite which was added in doses of 0.5, 2.0 and 4.0 g L^{-1}, the rate of reduction of heavy metal being correlated with the dose used (Nicula et al., 2010).

Clinoptilolite has been successfully used to transportation of live fish. In order to choose the most effective solution for controling the ammonia level in a fish transporting tank, it has to consider a series of parameters, including fish species, fish density, tank volume and transportation time. Studies have shown that the addition of 7 g L^{-1}clinoptilolitezeolite is sufficient to reduce ammonia in the transport of rainbow trout (*Oncorhynchus mykiss*) (Oz et al., 2010). By adding the same amount of clinoptilolite, high water quality and reduced ammonia concentration in frycarps transport (*Gibelion catla*, *Labeo rohita* and *Cirrhinus mrigala*) were maintained, which resulted in a higher survival rate (Singh et al., 2004). In the case of Victoria lake ciclide (*Haplochromis obliquidens*), a mixture of clove oil and clinoptilolite introduced into the transportation bags reduced the ammonia level by 82% compared to the bags in which the mixture was not used (Kaiser et al. 2006). Similar results were also reported with the use of clinoptilolite at doses of 10, 20 and 40 g L^{-1} for carriage in polyethylene bags of goldfish (*Carassius auratus*) when the ammonia concentration was reduced by 73%, 87% and 93% (Bower and Turner, 1982). The study was carried out during the transportation of ornamental fish, *Ancistrustriradiatus*, at high temperatures, and showed that the use of 22.7 g $L^{-1}$of clinoptilolite reduced ammonia levels from 51.6mg L^{-1} to 15.5 mg L^{-1} (Ramirez-Duarte et al., 2011). Clinoptilolite has also been successfully used for carp larve transportation in the Kemerevoregion (Polyakov et al., 2009).

Clinoptilolite has a low ammonia absorption capacity in salty waters;this disappears after about 8 hours (Emadi et al., 2001). However, researcheshave shown that clinoptilolitezeolite can be used to transport marine fish over a 24-hour period. The reduction of TAN levels by 18.33% at 8 hours, 34.08% at 16 hours, and 20.96% at 24 hours, was observed after using 40 g L^{-1} (Kanyilmaz et al., 2014). In order to be effective in saline water, clinoptilolite should be added in bigger quantities and the maximum absorption capacity is reached 16 hours after introduction into water.

It can be concluded that for transportation of live fish and the elimination of TAN from fish farms, clinoptilolitezeolite is more effective in freshwater than salty water.

CONCLUSIONS

The purpose of the present work was to present different applications of clinoptilolitezeolite in the aquaculture industry. Despite its low efficiency when used in salty water, researches and studies in freshwater and fish feed have proven the utility of using this zeolite in fish farms due to its unique structure and physico-chemical properties, in particular the cation exchange. Starting from its high affinity for ammonia, but also for other cations, it is possible to determine the main directions of using of this zeolite in aquaculture. Due to the selective ion exchange process, this is an alternative to the biological filters in the recirculation systems. Clinoptilolite has shown an important role played in improving the water quality in the growth basins and implicitly in the health of fish. In the transportation of live fish, its use contributes to the maintenance of total ammoniacal nitrogen (TAN) in normal limits and along with other zeolites, clinoptilolite offers the possibility of removing heavy metals from the water used in aquaculture but also from the fish body. The use of zeolite in fish diet has beneficial effects and determines the increasing of biomass and maintaining the health of fish populations. Other benefits that recommend using clinoptilolite and other zeolites in fish are low cost, good accesibility (deposits found in many parts of the world) and that theyt are environmental friendly materials. Researchers will continue to study these "stones of the future" to use them in environmental protection technologies and to obtain high aquaculture production at low cost.

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