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Effect of different ores on water quality adjustment of seawater desalinated by reverse osmosis

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Abstract: In many places, reverse osmosis seawater desalination plays an important role in solving freshwater crisis. In spite of its superior quality, instability of desalinated water may be corrosive to water distribution systems, thus may pose a threat on human health. To solve this problem, four ores were used to adjust the quality of the desalinated water. The physical and chemical properties of maifanshi, dolomite, limestone and an imported ore, including composition, XRD (X-ray Diffraction), porosity and pore size distribution, surface morphology were analyzed. Based on these properties, the effects of the four ores on water quality adjustment of the desalinated water by reverse osmosis were studied. The results indicate that the porosity and dissolution of the imported ore are improved obviously after the artificial process. The hardness, alkalinity, Mg^{2+} contents of the import ore are 50% higher than any other ores, while the pH value of the effluent is too high, the Ca^{2+} content in the effluent is lower than that of limestone. Still, there is a certain gap compared with the tap water. In the future, research should focus on the development of modifying agents with large surface areas and faster dissolution rate, which can adjust the desalinated water without acidifying the influent.

Key Words: reverse osmosis desalinated water; water quality adjustment; dissolution of ores

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不同矿石对反渗透淡化水的调质效果研究 . 浙江大学学报(理学版),2016,43(2):231-236,252

摘 要:反渗透海水淡化已成为解决淡水危机的一种有效方法,但反渗透淡化水的化学稳定性差,使用过程中会腐蚀管网,作为饮用水存在一定的健康风险. 采用溶解矿石法对反渗透淡化水的水质进行调节. 对麦饭石、白云石、石灰石、进口矿石的理化性能(包括成分、物相组成、孔隙率及孔径分布、表面形态)进行了分析,并在此基础上对比分析了 4 种矿石对反渗透淡化水的调质效果. 结果表明,进口矿石经人为加工后孔隙增多,溶出速率明显变快,调质后出水的硬度、碱度、Mg^{2+} 质量分数均高出其他 3 种原矿石 50%以上,但其出水 pH 值过高,Ca^{2+} 质量分数低于石灰石,与自来水相比仍有一定差距,未来应致力于研制一种比表面积大、溶解速率快、无需对淡化水酸化而直接调质的调质剂.

关 键 词:反渗透淡化水;调质;溶解矿石
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0 Introduction

Reverse osmosis has become one of the most important methods of seawater desalination in the world[1]. However, the removal rates vary with different ions in the seawater. That is, the removal rates of divalent ions such as Ca^{2+}, Mg^{2+} and

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SO_4^{2-} are higher than that of monovalent ions such as Na^+ and Cl^-. Besides, since CO_2 cannot be eliminated by RO membrane, pH value of desalinated water would be lowered. As a result, the chemical stability of desalinated water by reverse osmosis is poor[1]. Consequently, reverse osmosis desalinated water would be corrosive to the water distribution systems. Meanwhile, low mineral content in the reverse osmosis desalinated water may pose a threat on human health[2].

Therefore, it is essential to adjust the water quality of desalinated sea water by reverse osmosis. Currently, three types of adjustment methods for desalinated water exist: (1) methods based on direct dosage of chemicals[3]; (2) methods based on mixing desalinated water with multiple water sources[4]; (3) methods dissolving ores to offer Ca^{2+} and Mg^{2+} for alkalinity[5]. Compared with the former two methods, the last one is more cost-effective for wide application. XIAO[6] reported that the water alkalinity, hardness, and pH value of desalinated water were improved after the dissolution of limestone. Hence, the chemical stability of water was accordingly improved. LIU[7] discovered that the post-treatment of carbon dioxide by dissolving calcite could improve the Ca^{2+} content, alkalinity and chemical stability of desalinated water. BIRNHACK et al[8] used a combination of dolomite and calcite to adjust the quality of desalinated water. It was found possible to produce water with the following quality criteria: alkalinity=75 mg·L^{-1} as $CaCO_3$, $[Mg^{2+}]$ =12.4 mg·L^{-1}, $[Ca^{2+}]$ =48 mg·L^{-1}, pH=8.17. Therefore, dissolving ores can be used to adjust the water quality of desalinated sea water.

In this paper, effects of maifanshi, dolomite, limestone and an imported ore with different physical and chemical properties on the adjustment of reverse osmosis desalinated water quality are compared. Mechanisms of the effects on the adjustment of reverse osmosis desalinated water quality are explained. Also, references for the practical applications are provided.

1 Materials and methods

Maifanshi, dolomite and limestone were purchased from Zibo (Shandong Province, China), Laizhou (Shandong Province, China) and Shijiazhuang (Hebei Province, China), respectively. In comparison, the imported ore purchased from Germany was applied in practice, which was used to compare the effects of water quality adjustment on desalinated water with the other three natural ores. Water quality parameters of reverse osmosis desalinated water were listed in table 1.

In this experiment, the reverse osmosis desalinated water flowed through a tank filled with ores of 50 mm in diameter and 250 mm in height. By regulating the flow of influents, the retention time was controlled at levels of 1, 3, 5, 10, 15, 20, 25, 30, and 60 min. Hardness, alkalinity, Ca^{2+} and Mg^{2+} contents and pH value in the effluent of different ores were recorded and analyzed. Then, the dissolution effects of different ores were compared.

Table 1 Water quality parameters of RO desalinated water

Test items	Test results
pH value	6.70
Na^+ / (mg·L^{-1})	76
Ca^{2+} /(mg·L^{-1})	0.19
Mg^{2+} / (mg·L^{-1})	0.44
Cl^- / (mg·L^{-1})	101
SO_4^{2-} / (mg·L^{-1})	0.83
Turbidity/NTU	<0.5
TDS/ (mg·L^{-1})	218
Total Hardness/ mg·L^{-1} $CaCO_3$	3.12
Total Alkalinity/ mg·L^{-1} $CaCO_3$	3.81

2 Results

2.1 Physical and chemical properties

2.1.1 Composition analyses

As demonstrated in table 2, SiO_2 is the main component of maifanshi sharing a proportion with Al_2O_3 for over 80%, while the total proportion of calcium and magnesium are lower than 2%. Calcium content in the imported ore is close to that of dolomite, while magnesium content is the highest among the four ores. Calcium content in limestone is higher.

Table 2　Composition analyses of four different ores

Main component	Content/%			
	Maifanshi	Dolomite	Limestone	Imported ore
Fe_2O_3	0.17	6.12	0.19	0.01
SiO_2	60.88	1.07	17.06	0.96
Al_2O_3	25.14	0.26	0.35	0.04
CaO	0.85	30.58	35.25	31.02
MgO	0.81	21.42	17.20	24.58
K_2O	2.450	0.052	0.093	0.022
Na_2O	0.240	0.080	0.160	0.025
P_2O_5	0.060	0.013	0.036	<0.001
SO_3	0.037	0.012	0.014	0.025
Cd	<0.000 2	<0.000 2	<0.000 2	<0.000 2
Pb	0.003 9	<0.001 0	<0.001 0	<0.001 0
Mn	0.030 0	0.006 6	0.016 0	0.001 6
Ag	<0.000 5	<0.001 0	<0.001 0	<0.000 5
Cr	0.024	<0.001	<0.001	0.017
Sb	<0.001 0	<0.001 0	<0.001 0	<0.001 0
Be	<0.001 0	<0.001 0	<0.001 0	<0.001 0
Ba	0.048 0	<0.001 0	0.003 0	<0.001 0

2.1.2　X-ray diffraction(XRD) analyses

XRD analyses of maifanshi, dolomite, limestone and imported ore are shown in fig. 1.

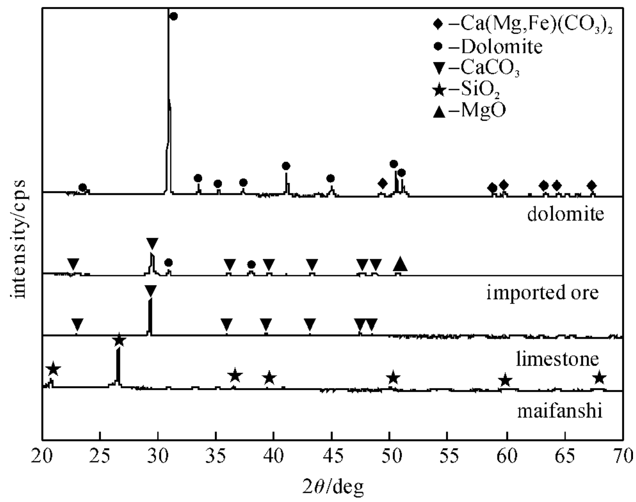

Fig. 1　XRD analyses of ores

As shown in fig. 1, the main phase in maifanshi is SiO_2 while in limestone is $CaCO_3$. Besides, $CaMg(CO_3)_2$ is the main phase in dolomite as iron is also included and $Ca(Mg,Fe)(CO_3)_2$ may be the possible existing form. Furthermore, there is not only $CaMg(CO_3)_2$, but also $CaCO_3$ and MgO in the imported ore. Accordingly, it can be preliminarily judged that the imported ore is an artificial ore made by semi-burnt dolomite. The thermal decomposition of dolomite follows the following two steps[9]:

$$CaMg(CO_3)_2 \longrightarrow MgO + CaCO_3 + CO_2\uparrow, \quad (1)$$

$$CaCO_3 \longrightarrow CaO + CO_2\uparrow. \quad (2)$$

The imported ore is made by controlling the decomposition of dolomite in reaction(1), which results in uncomplete decomposition of $CaMg(CO_3)_2$ in the dolomite.

2.1.3　Porosity and pore size analyses

Analyses of porosity and pore size of the four ores are demonstrated in table 3.

As demonstrated by table 3, maifanshi with average pore size of 13.72 nm has a porosity approaching 20%, which is higher than that of dolomite and limestone. Maifanshi has been reported to have a porous and spongy structure[10], which has a large specific surface area, excellent adsorption performance, and can capture heavy metal ions, bacteria, nitrite etc. in the water. In contrast, dolomite and limestone develop high degrees of crystal growth, hard textures, low porosities, and large internal pore sizes during natural formation. The imported ore has a lower porosity than that of maifanshi, but with smaller pore size and larger surface area. The micro-porous structure of the imported ore is mainly caused by the release of CO_2 during the process of burning.

Table 3　Porosity and pore size analyses of ores

	Maifanshi	Dolomite	Limestone	Imported ore
Surface area/ $(m^2 \cdot g^{-1})$	1.07	0.13	0.18	4.60
Pore volume/ $(mL \cdot g^{-1})$	0.004 4	0.000 5	0.000 7	0.018 4
Pore size/nm	13.72	16.84	15.65	7.56
Porosity/%	19.36	0.36	0.37	7.55

2.1.4　Surface morphology analyses by SEM

The surface morphology analyses of four ores by SEM are demonstrated in fig. 2.

As demonstrated in the fig. 2, the surface of maifanshi is clastic and the surfaces of limestone and dolomite are blocky, while the imported ore is granular. There are few pores in limestone and dolomite and textures of both are hard. The imported ore and maifnshi have relatively obvious porous structures. The mean pore size of the imported ore is smaller than that of maifanshi, which is consistent with the results of porosity analysis.

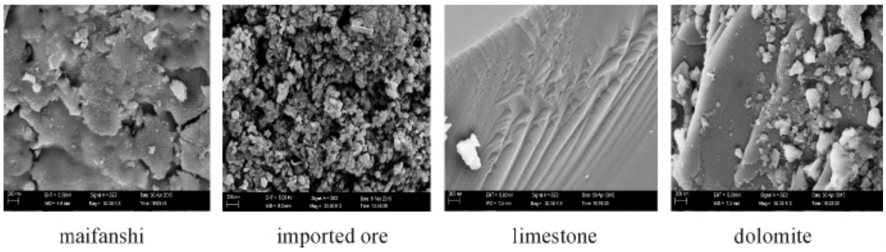

Fig. 2 SEM analyses of ores (×30K)

2.2 Adjustment effects on RO desalinated water

2.2.1 Hardness, Ca^{2+} and Mg^{2+} contents in effluents with varying retention times

Hardness, Ca^{2+} and Mg^{2+} contents in effluents varied with retention times, as shown in fig. 3, 4 and 5, respectively.

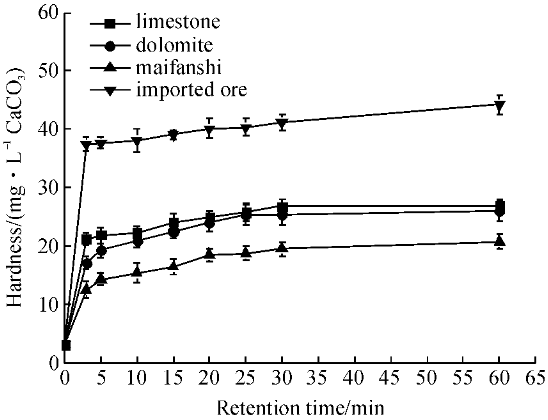

Fig. 3 Hardness contents of effluents

As shown in fig. 3, hardness contents in effluents of four ores firstly increase with retention time and kept in balance over time. This is mainly because the driving force of mass transfer decreases with the retention time and the dissolving rates of minerals keep decreasing until reaching a stable level. The dolomite and limestone in the experiments are both raw ores, and the influents are not acidified by H_2SO_4 or CO_2. As manifested in table 3, both of the ores have hard textures and low porosities, so the contact areas for ores and the desalinated water are limited. In spite of the high porosity, maifanshi is low in calcium and magnesium contents, resulting in the low hardness content of effluent. The contact areas of the imported ore and the desalinated water are expanded after artificial processing. In addition, the imported ore is rich in calcium and magnesium. Therefore, the imported ore has the highest hardness content in the effluent followed by limestone, dolomite, and maifanshi in order. When the retention time is controlled at 5 min, hardness contents in the effluents of maifanshi, dolomite, limestone and the imported ore are 14.35, 19.30, 21.88 and 37.68 mg · L^{-1} $CaCO_3$, respectively. When the retention time is controlled at 60 min, hardness contents in the effluents of maifanshi, dolomite, limestone and the imported ore are 20.80, 26.03, 27.04 and 44.23 mg · L^{-1} $CaCO_3$, respectively. In practice, the reverse osmosis desalinated water passes through a tank filled with ores. If the retention time is too long, the required tank volume will increase[11], which is especially unfavorable on board. Through tests and investigations, the hardness content of tap water ranged from 100 to 250 mg · L^{-1} $CaCO_3$ in Northern China, and 30 to 150 mg · L^{-1} $CaCO_3$ in Southern China[12-16]. The hardness content in the effluent of the imported ore is higher than any other ores, but there is still a certain gap compared with the tap water.

As shown in fig. 4 and 5, Ca^{2+} and Mg^{2+} contents in the effluents firstly increase with the retention time and then keep in balance. Among the four ores, Ca^{2+} contents in the effluent rank as: limestone > dolomite > the imported ore > maifanshi. Mg^{2+} contents rank as: the imported ore > dolomite > limestone > maifanshi. The main components in maifanshi are SiO_2 and Al_2O_3. The total percents of calcium and magnesium are less than 2%, so the Ca^{2+} and Mg^{2+} contents in the effluent of maifnashi are relatively low. However, when compared with dolomite and limestone, the dissolution rate of Ca^{2+} in maifanshi is higher due to high porosity. Moreover, the decay rate of maifanshi is high in application, leading to short service life. Limestone has a higher Ca^{2+} content while the imported ore has a higher Mg^{2+} content in the effluents, which corresponds to the high calcium and magnesium contents in the raw ores. $CaCO_3$ in the imported ore survives from burning while $MgCO_3$ decomposes into MgO, so the Ca^{2+} content in the effluent is practically the same as that of dolomite and Mg^{2+} content is higher than any other ores. Through tests and investigations, Ca^{2+} content of the tap water ranges from 10 to 50 mg・L^{-1} in China, mostly between 20 to 30 mg・L^{-1}. Mg^{2+} content of the tap water ranges from 2 to 20 mg・L^{-1} in China, mostly 5 to 10 mg・L^{-1}[14-17]. Although Ca^{2+} and Mg^{2+} contents in the effluent of the imported ore have been improved, there is still a certain gap when compared with the tap water.

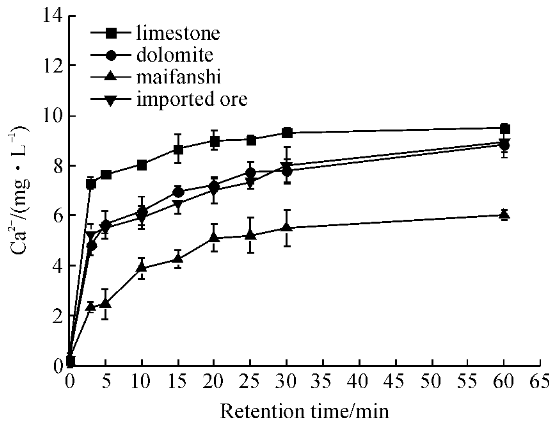

Fig. 4　Ca^{2+} contents of effluents

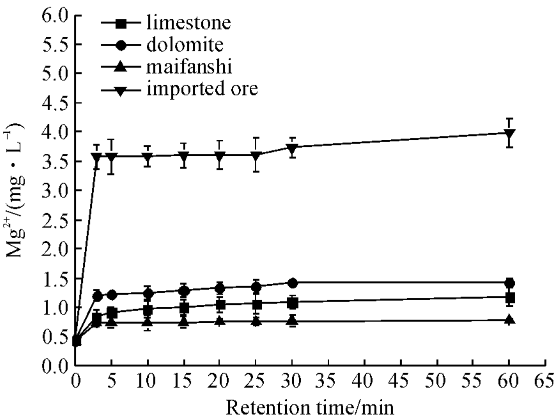

Fig. 5　Mg^{2+} contents of effluents

2.2.2　Alkalinity contents and pH values in effluents with varying retention times

Alkalinity content and pH values in effluents of the four ores vary with retention times, as demonstrated in fig. 6 and 7.

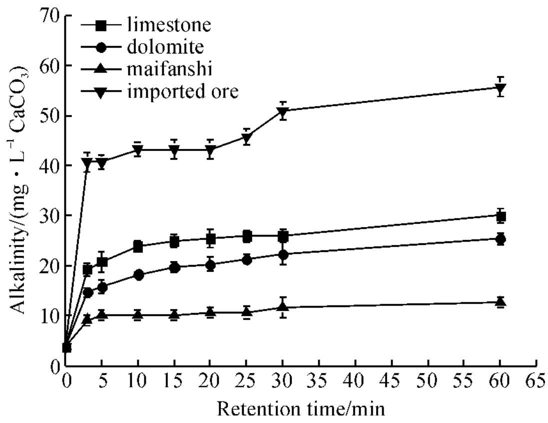

Fig. 6　Alkalinity contents of effluents

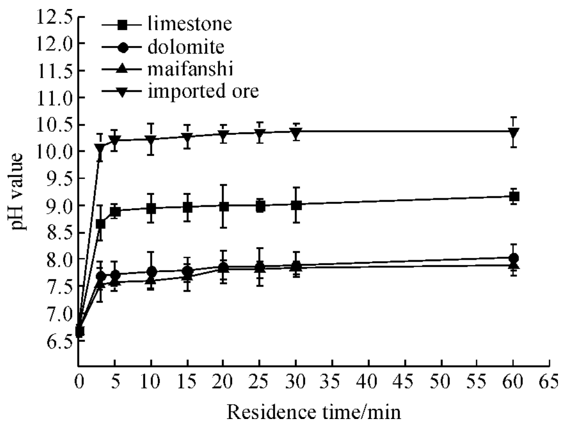

Fig. 7　pH of effluents

As fig. 6 and 7 indicate, the relationship between retention time and alkalinity content or pH in effluent are similar. Alkalinity is defined as the sum of all ions that can react with protons. Alkalinity$=2[CO_3^{2-}]+[HCO_3^-]+[OH^-]-[H^+]$. Buffer capacity of the solution is defined as the ability of resisting the disturbance from the changing acidity or basicity. Within a certain pH range, the higher the alkalinity of the solution is, the higher the contents of HCO_3^- and CO_3^{2-} are included in it. Consequently, a buffer system of the solution forms and the buffer capacity of the solution is improved. Generally, the improvement of the alkalinity in water can inhibit the corrosion process of pipes. In the early times, it was thought that HCO_3^- could react with Ca^{2+} to generate $CaCO_3$ precipitation, thus forming protective layers on the inner pipes, protecting pipes from corrosion[18]. Later, some researchers put forward that when HCO_3^- exists in water of pipes, iron compounds with lower solubility than $Fe(OH)_2$ form, such as $FeCO_3$[19]. Furthermore, considering the relations between alkalinity and human health, drinking of alkaline water in a long term can balance the pH level in human body, which is beneficial for human beings[20]. The existence of Al in maifanshi enables the bidirectional adjustment of the pH in the effluent. The main component in limestone is $CaCO_3$, which has a pH value of 9.84 in saturated solubility[21]. When dissolved in water, CO_3^{2-} hydrolyzes as follow: $CO_3^{2-} + H_2O \longrightarrow HCO_3^- + OH^-$. The production of OH^- results in a higher pH value and alkalinity of effluent. The main component in dolomite is $CaMg(CO_3)_2$ with a relatively low dissolution rate. Moreover, there is iron in dolomite, making the pH value of effluent lower than that of limestone after hydrolysis. The existence of $CaCO_3$ and MgO in the imported ore results in a higher pH value of the effluent than that of limestone.

3 Conclusions

The physical and chemical properties of maifanshi, dolomite, limestone and the imported ore were analyzed. Adjustment effects of the four ores on the quality of desalinated water by reverse osmosis by were studied. The calcium content in the imported ore and the magnesium in the limestone is the highest among the four ores. Limestone and dolomite have a similar porosity, which are 98% lower than maifanshi and 95% lower than the imported ore. The hardness, Mg^{2+} and alkalinity contents in the effluent of the imported ore are 50% higher than any other ores, while the Ca^{2+} content is lower than limestone and the pH value of the effluent is too high. These observations closely related to the decomposition of $MgCO_3$, minerals contents and porosities in ores. However, there is still a certain gap compared with the tap water.

Dissolving ores can be a useful way to adjust the quality of desalinated water. The pH value, hardness, alkalinity and mineral contents in the effluent increase significantly after adjusting. Chemical stability of the osmosis reverse desalinated water is also improved. Therefore, the tendency of the pipes corroded by desalinated water is alleviated, and thus is also more safe and healthy for drinking.

By comparison, the imported ore can adjust the quality of reverse osmosis desalinated water quickly, while the pH value of the effluent is too high. In the future, emphasis should be put on the development of modifying agents with large surface area and fast dissolution rate, which can adjust the desalinated water without acidifying the influent.

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