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An integrated device for coprecipitation and filtration of radiocesium in seawater

Jianhua He

The Third Institute of Oceanography, State Oceanic Administration, Xiamen 361005, China

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1. Introduction

 $^{134}Cs(T_{1/2}=2.1years)$ and $^{137}Cs(T_{1/2}=30.2years)$ (Shozugawa et al., 2016)are anthropogenic radionuclides that are produced as a result of nuclear fission at thermal neutron fission yields of 5.4% and 5.9% (Cai, 1998), respectively, and large amounts of them derived from Fukushima Dai-ichi Nuclear Power Plant (FDNNP) accident on March 11, 2011 were widespread released into seawater. Due to the high selectively absorption to radiocesium, ammonium phosphomolybdate (AMP) is usually used to coprecipitate ¹³⁴Cs and ¹³⁷Cs in seawater (Aoyama et al., 2008, 2016a,b; Inoue et al., 2012, 2013; Kaeriyama et al., 2013; Yu et al., 2015, Men et al., 2015), but this method is not suitable onboard due to large volume seawater requested and the consequence of unfeasible. And with the development of technology, some new methods have been introduced into the analysis of radiocesium in seawater, such as AMP-Pan method (Buesseler et al., 2012, 2014), in-situ enrichment method (He et al., 2011; Ramzaev et al., 2014), and underway detection method (Caffrey et al., 2012), although these methods have the advantage of operating onboard, but also exists the disadvantages of time consuming (more than 20 h), high cost, only suitable for surface seawater, or with higher MDA (Minimum Detecting Activity).

ABSTRACT

To improve the pretreatment efficiency of radiocesium in seawater, a prototype machine based on a new design of an integrated device for coprecipitation and filtration was developed and tested in the laboratory for its possibility and efficiency, the results show the that the efficiency of pretreatment of radiocesium can be improved more than one orders of magnitude compared to traditional methods. And the results from experiments on effect of settle time show that immediately filtration of solution after reaction will not affect the absorption of AMP to radiocesium in seawater.

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As coprecipitation with AMP is still a commonly used and lowcost pretreatment method for radiocesium (Aoyama et al., 2008, 2016a,b; Inoue et al., 2012, 2013; Kaeriyama et al., 2013; Yu et al., 2015, Men et al., 2015), an optimization for this method is necessary.

In this work, a new design of an integrated device, which improve the pretreatment efficiency of large volume seawater for radiocesium, was introduced and developed. With this device, the time of pretreating 60 L seawater for radiocesium will reduce to less than one hour, and the coprecipitation onboard would be possible.

2. Device description

This device (He et al., 2016a,b) consists of a reaction container, connected with a semi-sealed cover, an air supplier for air mixing and inflation, a side-filter and a bottom-filter covered by a electromagnetism cover, respectively, for filtration of sediment, a power supply module, and a bolster module. The Schematic of the integrated device is illustrated in Fig. 1.

The volume of the reaction container would be ranged from 20 to 100 L based on the requirement, and the semi-sealed cover with a little hole for reagent adding will prevent the overflow of the seawater when operated onboard.

The first function of air supplier is used to mix the seawater, instead of stirring needed by traditional method, and the second function is use to prevent the retention of sediment particles on the





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E-mail address: hejianhua@tio.org.cn.

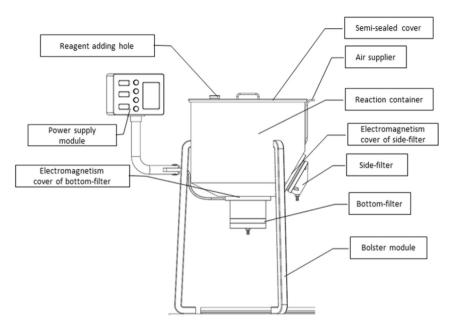


Fig. 1. Schematic of the integrated device.

membrane on side filter (Fig. 2), and improve the filtration speed.

The design of side-filter is used to realize the immediately filtration of solution after reaction, instead of siphoning after settle for 1-2 days required by traditional methods, due to the disturbance of air from air supplier preventing the retention of the sediment particle and avoiding the blocking of the membrane.

During the side-filtration process, the direction of reaction container will be changed into suitable position to keep the surface of seawater above the up-line of side-filter (Fig. 3), controlled by power supply module.

The cover of the bottom filter will be removed when the seawater left in the reaction container is less enough or the side filtration can't work well, then brush the inter-surface of the reaction container with suitable filtered seawater 2-3 times. After filtration, the membrane from side-filter and bottom filter would be collected together for the next step, then the pretreatment of the seawater for radiocesium is finished. The section view of the bottom-filter is illustrated in Fig. 4.

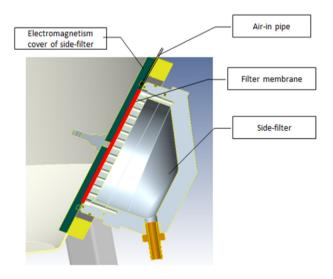


Fig. 2. Section view of the side-filter.

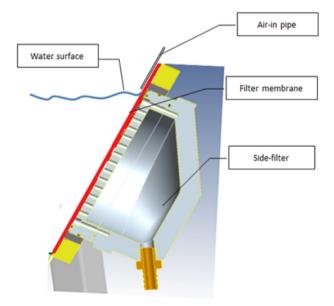


Fig. 3. Schematic of operating of side filtration.

3. Experimental

3.1. Possibility and efficiency of the prototype device

In order to test the possibility of this design, a prototype device based on the above description was developed (Fig. 5), then three parallel experiments were conducted as following steps.

- (1) Step 1: Inject about 60 L seawater in the reaction container, after covered the side-filter and bottom-filter with covers.
- (2) Step 2: Start the air supplier by air compressor to mix the seawater.
- (3) Step 3: Add 1:1 HNO₃ from the hole on semi-sealed cover to adjust the pH to ~2, then add stable Cs carrier (CsCl) and AMP to coprecipitate the radiocesium.

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