

## Measurement of $^{129}\text{I}$ in ferromanganese crust with AMS

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### Abstract

In the present study, the analytical method for  $^{129}\text{I}$  in ferromanganese crusts is developed and  $^{129}\text{I}/^{127}\text{I}$  ratio in ferromanganese crusts is measured by the accelerator mass spectrometry (AMS). The developed method is applied to analyze  $^{129}\text{I}/^{127}\text{I}$  ratio in two ferromanganese crusts MP5D44 and CXD08-1 collected from the Mid-Pacific Ocean. The results show that  $^{129}\text{I}/^{127}\text{I}$  ratio in MP5D44 and CXD08-1 crusts varies from  $7\times 10^{-14}$  to  $1.27\times 10^{-12}$ , with the lowest value falling on the detection limit level of AMS reported by previous literatures. For the depth distribution of  $^{129}\text{I}/^{127}\text{I}$ , it is found that both MP5D44 and CXD08-1 crusts have two growth generations, and the  $^{129}\text{I}/^{127}\text{I}$  profiles in two generations all displayed an approximate exponential decay. According to the  $^{129}\text{I}/^{127}\text{I}$  ratio, the generate age of bottom layer of MP5D44 and CXD08-1 was estimated to be 54.77 and 69.69 Ma, respectively.

**Key words:** ferromanganese crust, AgI,  $^{129}\text{I}/^{127}\text{I}$  ratio, accelerator mass spectrometry,  $^{129}\text{I}$ -dating

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### 1 Introduction

Iodine is a hydrophilic element with only stable isotope,  $^{127}\text{I}$ , and one long-lived radioisotope,  $^{129}\text{I}$ , which has a long half-life ( $T_{1/2}=15.7$  Ma). Its half life falls between that of  $^{53}\text{Mn}$  ( $3.7\times 10^6$  a) and  $^{40}\text{K}$  ( $1.28\times 10^9$  a), which fills the blank of time scales of a tens millions. In nature,  $^{129}\text{I}$  has two nature sources: formation in the atmosphere by the interaction of cosmic-ray with xenon isotopes, and production in the geosphere as a product of the spontaneous or neutron-induced fission of uranium isotopes (Fehn et al., 1992). In sea water, dissolved inorganic iodine is known to be mainly present form, which is strongly absorbed on iron oxide, clay, various organic compounds (Behrens, 1982) and phosphates to sediments or ferromanganese crusts. Based on the geochemistry, it can be found that  $^{129}\text{I}$  can be used to dating ferromanganese crusts.

Ocean ferromanganese crusts recorded important information of paleoceanography and paleoclimate change (Klemm et al., 2005), such as bottom water activity, development of oxygen-minimum zone, productivity. The reliable age dating of the crusts is considered as one prerequisite for paleoceanography reconstruction.  $^{10}\text{Be}$  ( $^{10}\text{Be}$ ) with a half-life of 1.5 Ma has been used for the age dating, which could only determine the timescale within 10 Ma from surface to about 2 cm of the crusts. At ages greater than 10 Ma, it has to be estimated from the extrapolation of growth rates based on  $^{10}\text{Be}/^9\text{Be}$  ratios, leading to possible significant deviation. However,  $^{129}\text{I}$ , with a half-life of 15.7 Ma, allows age determinations up to 80 Ma (Fabryka-Martin et al., 1985; Fehn et al., 1992, 2000; Liu et al., 1997), mainly covering the growth period of crusts, hence  $^{129}\text{I}$  could be a better tool for the dating of ferromanganese crusts (Liu and Ji, 2010). Although  $^{129}\text{I}$  has been widely applied in the dating of gas hydrate, fore arc

fluids, hydrothermal fluid, volcanic, geothermal fluids and organic matters with high concentration of iodine (Fehn et al., 1987, 1990, 2007; Moran et al., 1995; Muramatsu et al., 2001; Snyder and Fehn, 2002; Fehn and Snyder, 2003; Lu et al., 2007, 2008; Tomaru et al., 2009a, b), the research on the analytical method for the determination of  $^{129}\text{I}$  in ferromanganese crusts is still lack, and up to now no report has been found about dating ferromanganese crust by  $^{129}\text{I}$ .

In the present study, the analytical procedures for the determination of  $^{129}\text{I}$  in ferromanganese crust by AMS was developed and applied to analyze two ferromanganese crusts in the Mid-Pacific Ocean. We report results of the study to test application of  $^{129}\text{I}$  in the ferromanganese crusts.

### 2 Samples

Crust CXD08-1 was collected by towing net from the Mid-Pacific Mountains ( $19^{\circ}57.9'\text{N}$ ,  $172^{\circ}55.1'\text{E}$ ; water depth, 1 908 m) during cruise DY105-11 in 2001 on R/V *Dayang 1*. The crust MP5D44 was sampled by dragnet from Line Islands ( $10^{\circ}24.2'\text{N}$ ,  $167^{\circ}36.5'\text{W}$ ; water depth, 3 100 m) during cruise DY105-15 on R/V *Haiyang 4* in 2003. The CXD08-1 and MP5D44 are plate-like and with knobby surface texture, whose thickness is about 8.0 cm and 7.0 cm, respectively. Three main layers in the two crusts could be clearly identified from the bottom to the surface.

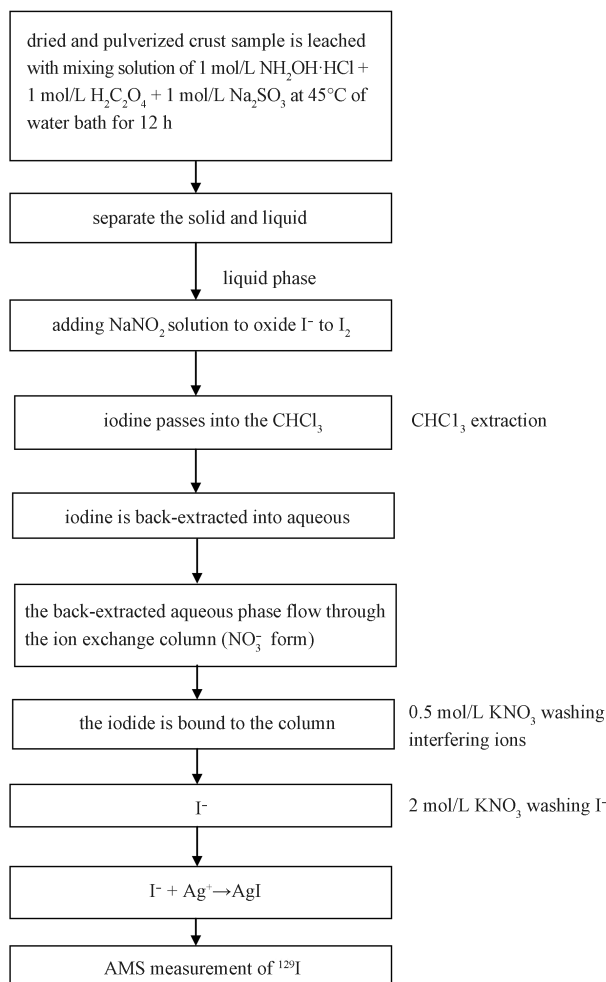
### 3 Methods

#### 3.1 Separation iodine from the ferromanganese crust

To the best of our knowledge, the AMS is the only detector instruments for the determination of  $^{129}\text{I}/^{127}\text{I}$  ratio in the crust as the natural  $^{129}\text{I}/^{127}\text{I}$  ratio is as low as  $10^{-12}$  (Hou et al., 2009; Luo et

al., 2011; Chen et al., 2014). For the measurement of  $^{129}\text{I}$ , iodine separated from the crust needs to be converted to silver iodide (AgI) for AMS analysis.

The procedure in the study was schematized in Fig. 1 as following shown. The analytical method for  $^{129}\text{I}$  in ferromanganese crusts included the following steps: (1) separation of iodine from the ferromanganese crust, (2) purification of iodine through extraction, back-extraction and anion exchange chromatography, (3) preparation of the target component (AgI) for accelerator mass spectrometry (AMS), and (4) measurement of  $^{129}\text{I}$  by AMS. The mixture  $\text{Na}_2\text{SO}_3\text{-H}_2\text{C}_2\text{O}_4\text{-NH}_2\text{OH}\cdot\text{HCl}$  was applied to leach iodine from the Fe-Mn crust powder in water bath for 12 h. The excessive  $\text{NaNO}_2$  solution was added to oxidize the iodide to molecular iodine ( $\text{I}_2$ ), followed by the extraction of chloroform, back-extraction of  $\text{NaHSO}_3$  and separated through anion exchange column<sup>①</sup>. After purification, in order to prepare the AgI for AMS measurement,  $\text{AgNO}_3$  was added to the iodine eluate, and the mixed solution was stand overnight for AgI precipitate. After filtration, the AgI precipitate was washed with deionized



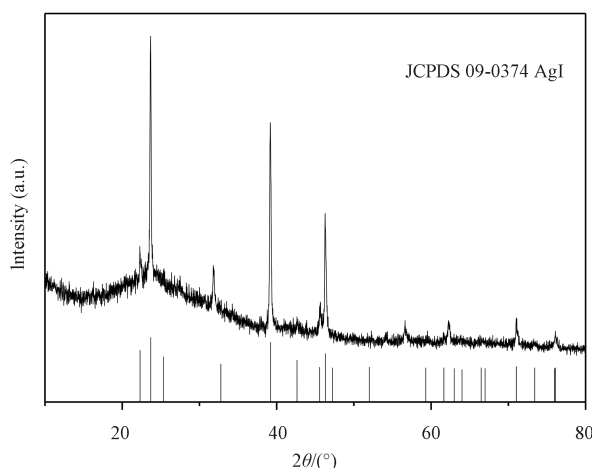
**Fig. 1.** The schematic diagram of separation iodine from the crust samples for AMS measurement.

water and alcohol, respectively, and then dried in an oven at 60°C. The AgI sample we prepared for AMS measurement is yellow color when the size is enough, but the color of AgI is yellow-

green or gray-yellow when only a little. AgI is light sensitive and decomposes into Ag and  $\text{I}_2$ . Therefore, AgI samples are stored in the dark and in small glass bottles to reduce cross contamination.

### 3.2 Identification of AgI

Figure 2 depicted the X-ray diffraction (XRD) spectrum of the sample for the measurement by AMS. The upper spectrum is AgI sample made from crust sample and the lower line segments are the XRD spectrum of standard AgI (Fig. 2). The XRD spectrum indicated that the sample to be measured by AMS was pure AgI component.



**Fig. 2.** The X-ray diffraction spectrum of the AgI sample.

### 3.3 Measurement of accelerator mass spectrometry

The measurement of  $^{129}\text{I}$  was performed by China Institute of Atomic Energy Accelerator Mass Spectrometer (CIAE-AMS). The system was developed based on tandem accelerator of CIAE in the late of 1980s (Jiang et al., 1989). The AMS system consists of ion source, injection system, accelerator, high energy analyzer and detector. The ion source with 40 target positions is multi-cathodes source of negative ion by cesium sputting (MC-SINCS), from NEC company, America. The injection system is comprised of a 90° spherical electrostatic analyzer and a 112° magnetic analyzer of double focus. Tandem accelerator is model HI-13 from HVEC company, America, with the highest terminal voltage of 14 MV. The high energy analysing system was made of magnetic analyzer and static analyzer.

In the study, we chose the terminal voltage of 7.83 MV and the charge state of 9<sup>+</sup>.  $^{129}\text{I}$  was detected by a silicon semiconductor detector, and stable  $^{127}\text{I}$  was measured at the Farady cup.  $^{129}\text{I}/^{127}\text{I}$  atomic ratio was evaluated by reference laboratory standard as following equation shown,

$$I_{\text{sa}} = \frac{I_{\text{st}}}{R_{\text{st}}} R_{\text{sa}}, \quad (1)$$

where  $I$  is the  $^{129}\text{I}/^{127}\text{I}$  atomic ratio;  $R$  is the ratio of  $^{129}\text{I}$  count rate; subscript sa denotes samples and st represents laboratory standard sample. The laboratory standard sample of CIAE-AMS was used, which had a  $^{129}\text{I}/^{127}\text{I}$  ratio of  $4.99 \times 10^{-12}$ . All samples were measured for three cycles. For each crust sample, the final result was the average of three measured  $^{129}\text{I}/^{127}\text{I}$  ratio values, and the error was derived from mean the standard deviation.

<sup>①</sup> Ji Lihong, Liu Guangshan, Huang Yipu, et al. The distribution of iodine and effects of phosphatization on it in the ferromanganese crusts from the Mid-Pacific Ocean. (accepted)

## 4 Results and discussion

### 4.1 Crust sample quantity needed for $^{129}\text{I}$ measurement with AMS

The iodine contents and  $^{129}\text{I}/^{127}\text{I}$  ratios in the two crusts are shown in Table 1, with the quantity of crust samples and AgI for the measurement of AMS. Previous literature demonstrated that the detection limit of  $^{129}\text{I}$  atom number (atoms per sample) for AMS fell on the order of magnitude of  $10^6$ , and the lowest iodine quantity was estimated to be 2 mg (Hotchkis et al., 2000). More iodine was needed for higher counting values with lower counting error. The lower  $^{129}\text{I}/^{127}\text{I}$  ratio was, the more iodine was needed. According to the atom number of 1 mg  $^{127}\text{I}$  ( $4.74 \times 10^{18}$ ) and the initial value of  $^{129}\text{I}/^{127}\text{I}$  ratio ( $1.5 \times 10^{-12}$ ), there was evaluated to be a  $7.11 \times 10^6$   $^{129}\text{I}$  atoms in 1 mg iodine that is similar with detection limit level of AMS. The age of the newer crust layer was near morden time, thus the  $^{129}\text{I}/^{127}\text{I}$  ratio in the newer crust layer might be approximately falling on the level of the initial value. For the older crust layer, the  $^{129}\text{I}/^{127}\text{I}$  ratio might be lower, leading to the  $^{129}\text{I}$  atom number in 1 mg iodine lower than the detection limit of AMS. Therefore, more iodine from the crust samples was needed for AMS measurement. The weights of AgI sample in the present study varied from 3.31 to 36.88 mg, containing iodine more than 2 mg (Table 1). Because some amount of  $^{129}\text{I}$  always exists in the stable iodine carrier, the carrier method cannot be

used for the analysis of crust sample with ultralow level  $^{129}\text{I}$ . For older crust layer, the  $^{129}\text{I}/^{127}\text{I}$  ratio was low but rich in iodine (Table 1, Fig. 3), indicating that AgI sample prepared from the older crust layer could satisfy the requirement of AMS measurement in the present study.

### 4.2 $^{129}\text{I}/^{127}\text{I}$ ratio in two marine ferromanganese crusts

The  $^{129}\text{I}/^{127}\text{I}$  ratio in the two crusts are shown in Table 1 and depicted in Fig. 3. The result showed that the  $^{129}\text{I}/^{127}\text{I}$  ratio in two crust samples varies from  $7 \times 10^{-14}$  to  $1.27 \times 10^{-12}$  (Table 1), lower than  $^{129}\text{I}/^{127}\text{I}$  initial value ( $1.5 \times 10^{-12}$ ) (Moran et al., 1998; Fehn et al., 2007; Lu et al., 2008; Hou et al., 2010; Fehn, 2012), which demonstrated that our results are reliable with ignored contamination from anthropogenic iodine. Within two samples, the  $^{129}\text{I}/^{127}\text{I}$  ratio in the MP5D44 crust fall on a range of  $1.3 \times 10^{-13}$ – $4.8 \times 10^{-13}$ , with an average of  $3.0 \times 10^{-13}$ , while for the CXD08-1 crust,  $^{129}\text{I}/^{127}\text{I}$  ratio ranges from  $7 \times 10^{-14}$  to  $1.27 \times 10^{-12}$ , with an average of  $4.5 \times 10^{-13}$ . The lowest  $^{129}\text{I}/^{127}\text{I}$  ratio found in the study was  $7 \times 10^{-14}$ , similar with the detection limit level of AMS reported by previous literatures (Fabryka-Martin et al., 1987; Hotchkis et al., 2000; Zhou et al., 2013). The value counted for 1 h was considered as the  $^{129}\text{I}/^{127}\text{I}$  ratio detection limit of the present study.

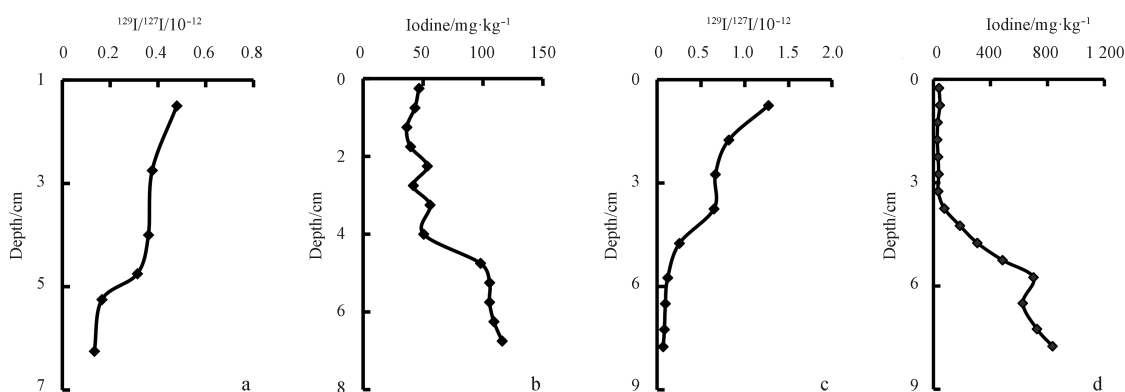


Fig. 3. The distributions of  $^{129}\text{I}/^{127}\text{I}$  ratio in two ferromanganese crust. a and b. Sample MP5D44 and c and d. Sample CXD08-1.

The two crusts have similar distribution of  $^{129}\text{I}/^{127}\text{I}$  ratio, approximately showing an exponential decay with increasing depth (Fig. 3), and thus two generations (a young generation and an old generation) could be identified for two crusts. Based on the distribution of iodine and  $^{129}\text{I}/^{127}\text{I}$ , the boundaries between young and old growth generation in CXD08-1 and MP5D44 locate at the depth of 3.75 and 4.00 cm, respectively, which is the same depth of iodine mutation (Ji, 2011). The iodine contents in the MP5D44 and CXD08-1 crust were both enriched in the old crust layer compared to the young crust layer, while the opposite was found for  $^{129}\text{I}/^{127}\text{I}$  ratios (Fig. 3).

### 4.3 The effective diffusivity for iodine within the ferromanganese crust

The prerequisite for isotope dating methods is that the element in the ferromanganese crusts is stable, with lower diffusion rate. The method of Henderson and Burton (1999) is used to calculate the effective diffusivity of iodine in the ferromanganese crusts. According to the method, the effective diffusivities of iodine in MP5D44 and CXD08-1 crusts are  $1.4 \times 10^{-6} \text{ cm}^2/\text{a}$  and  $4.7 \times 10^{-7} \text{ cm}^2/\text{a}$ , respectively. Henderson and Burton (1999) gave

the effective diffusivity of U, Th, Nd, Pb, Be and Hf in the ferromanganese crusts. Comparing with their results, the paper finds that the effective diffusivity of iodine in CXD08-1 crust is one order of magnitude more slowly than that of U, and it is close to Hf. The value of MP5D44 crust is close to U. These results suggest that  $^{129}\text{I}$  dating in the crust is feasibility and reliability.

### 4.4 $^{129}\text{I}$ dating of the ferromanganese crust

The  $^{129}\text{I}/^{127}\text{I}$  ratios could be converted to ages according to the following equations:

$$R_{\text{mea}} = R_{\text{init}} e^{-\lambda_{129} t}, \quad (2)$$

$$t = \frac{1}{\lambda_{129}} \ln \left( \frac{R_{\text{init}}}{R_{\text{mea}}} \right), \quad (3)$$

where  $R_{\text{init}}$  is the  $^{129}\text{I}/^{127}\text{I}$  initial ratio ( $1.5 \times 10^{-12}$ );  $R_{\text{mea}}$  is measured  $^{129}\text{I}/^{127}\text{I}$  ratio of the crust;  $\lambda_{129}$  is the decay constant of  $^{129}\text{I}$  ( $\lambda_{129} = 4.4 \times 10^{-8} \text{ a}^{-1}$ ). Without considering the potential contributions from spontaneous fission of  $^{238}\text{U}$  ( $^{238}\text{U}$ ), the ages derived from this calculation are considered to be minimum ages.

The estimated age of two ferromanganese crusts in the present study is listed in Table 1, indicating the generate age of bottom layer of MP5D44 and CXD08-1 was 54.77 Ma and 69.69 Ma, respectively. The ages of boundaries between young generations and old generations in MP5D44 and CXD08-1 crusts are 32.34 Ma and 19.00 Ma, respectively.

The  $^{129}\text{I}$  dating in ferromanganese crusts provides a time-

scale for marine environment change during the Tertiary recorded in the ferromanganese crusts. The two ferromanganese crusts growth was probably initiated by increasing flow of the Antarctic bottom water as a result of major global cooling in the Eocene and again in the Miocene (Ji, 2011). A more detailed paleoceanography and paleoclimate change will be reported elsewhere together with these data.

**Table 1.** The sample quantity, AgI quantity, iodine concentration and  $^{129}\text{I}/^{127}\text{I}$  ratio in the crust sample

Ferromanganese crusts	Depth zone/cm	Iodine/mg·kg <sup>-1</sup>	AgI sample/mg	$^{129}\text{I}/^{127}\text{I}$ ratio/ $10^{-12}$	Age/Ma
CXD08-1	0.5–1.0	45.4±4.0	4.31	1.27±0.30	3.77
	1.5–2.0	27.1±3.2	5.21	0.82±0.25	13.75
	2.5–3.0	36.9±3.7	4.71	0.67±0.16	18.45
	3.5–4.0	77.0±14	4.68	0.65±0.17	19.00
	4.5–5.0	308±48	18.06	0.26±0.04	40.18
	5.5–6.0	620±49	36.88	0.12±0.01	57.08
	6.0–7.0	627±56	31.17	0.09±0.03	62.71
	7.0–7.5	727±58	31.39	0.08±0.03	65.47
	7.5–8.0	836±70	32.39	0.07±0.02	69.69
Range		27.1–836	4.31–36.88	0.07–1.27	
Mean		367.2	18.76	0.45	
MP5D44	1.0–2.0	38±11	3.31	0.48±0.15	25.89
	2.5–3.0	42±11	7.43	0.38±0.08	31.37
	3.5–4.5	51±10	5.54	0.36±0.05	32.34
	4.5–5.0	98±10	6.39	0.31±0.07	35.47
	5.0–5.5	105±9	7.98	0.17±0.04	49.98
	6.0–6.5	109±7	7.58	0.13±0.06	54.77
Range		38–109	3.31–7.98	0.13–0.48	
Mean		74	6.37	0.30	

## 5 Conclusions

The measurement method of  $^{129}\text{I}$  in ferromanganese crust from marine environment is developed and applied to analyze two crust samples from Mid-Pacific Ocean in the present study. The  $^{129}\text{I}/^{127}\text{I}$  ratio in the MP5D44 crust varied from  $1.3\times 10^{-13}$  to  $4.8\times 10^{-13}$ , with an average of  $3.0\times 10^{-13}$ , while  $^{129}\text{I}/^{127}\text{I}$  ratio in the CXD08-1 crust ranged from  $7\times 10^{-14}$  to  $1.27\times 10^{-12}$ , with an average of  $4.5\times 10^{-13}$ . Additionally, the  $^{129}\text{I}/^{127}\text{I}$  ratios in two crusts are both lower than the initial value, suggesting that the contamination could be ignored during the separation and purification procedure and the presented results are reliable in the work.

This is the first report of a successful  $^{129}\text{I}$  dating of ferromanganese crusts. Based on the  $^{129}\text{I}/^{127}\text{I}$  ratio and derived decay equation, the generate age of bottom layer of MP5D44 and CXD08-1 was 54.77 Ma and 69.69 Ma, respectively. The  $^{129}\text{I}$  dating in ferromanganese crusts provides a time-scale for marine environment change during the Tertiary recorded in the ferromanganese crusts.

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