Online monitoring technology for Pu-239 aerosol based on ICP-MS

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Introduction

Continuous air monitor (CAM) based on planar implanted passivated silicon (PIPS), with the detection limit at about 1E-1Bq/m^3 and the measurement time over 30mins for single measurement, can't meet the requirements in China very well.

Therefore, it is urgent to develop a Pu-239 aerosol real-time monitoring system that is more effective and sensitive, and has a lower detection limit. Based on the analysis of ICP-MS measurement principle and the development of aerosol direct introduction device, a new online monitoring technology for Pu-239 aerosol was established.

Methods

As shown in the fig 1, a Pu-239 aerosol real-time monitoring system based on aerosol direct introduction device, membrane desolvation nebulizer and ICP-MS was established.

The aerosol direct introduction device(fig 2) was used to exchange the air with argon which was used to carried the aerosol into the ICP-MS, due to the ICP-MS can't work when the air flow was over 50mL/min. When the aersol introduced rate is <900 mL/min , the gas exchange rate is 100%.

Pu-242 standard solution (1.24×10^{-3}Bq/m^3) was introduced by a membrane desolvation nebulizer for the quantitative measurement[1].

Two kinds of methods for the estimation of detection limit (LOD) were used, one was calculation from the signal-to-noise ratio according to the HJ 186-2010, the other was calculation from the calibration line at low concentrations according to the ISO 11843-2-2000.

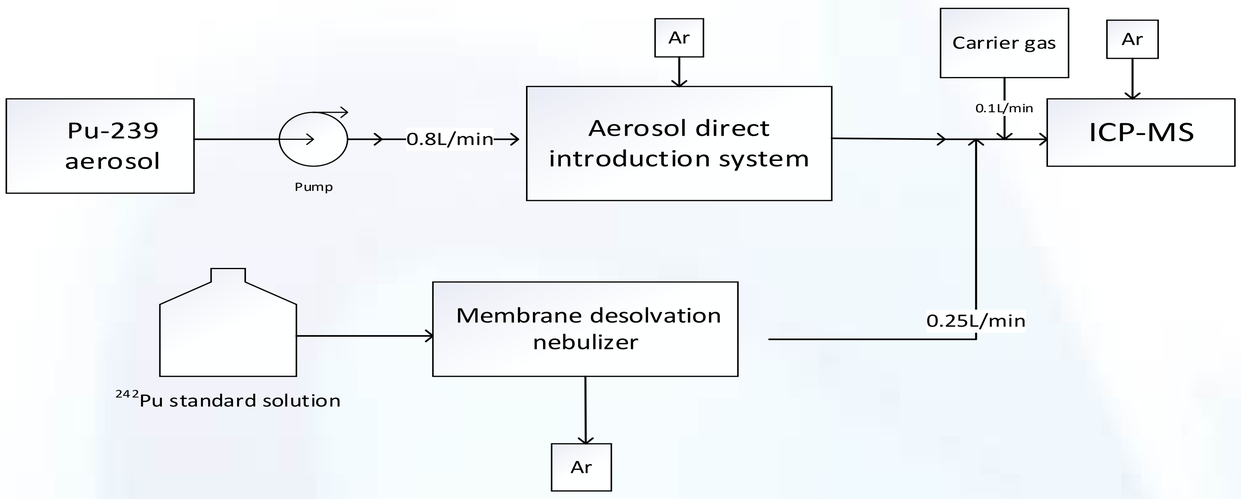

Fig 1 Aerosol real-time monitor system based on ICP-MS

Fig 2 Aerosol direct introduction device

Table 1 The background counts of the system

number	Counts of Pu-239 （30s）	Counts of Pu-242 （30s）
1	1	7539
2	0	7245
3	0	7439
4	0	7614
5	0	7364
6	0	7752
7	0	7946
8	0	7949
9	1	7815
10	0	7542
average	0.2	7621
RSD （%）	200	2.9

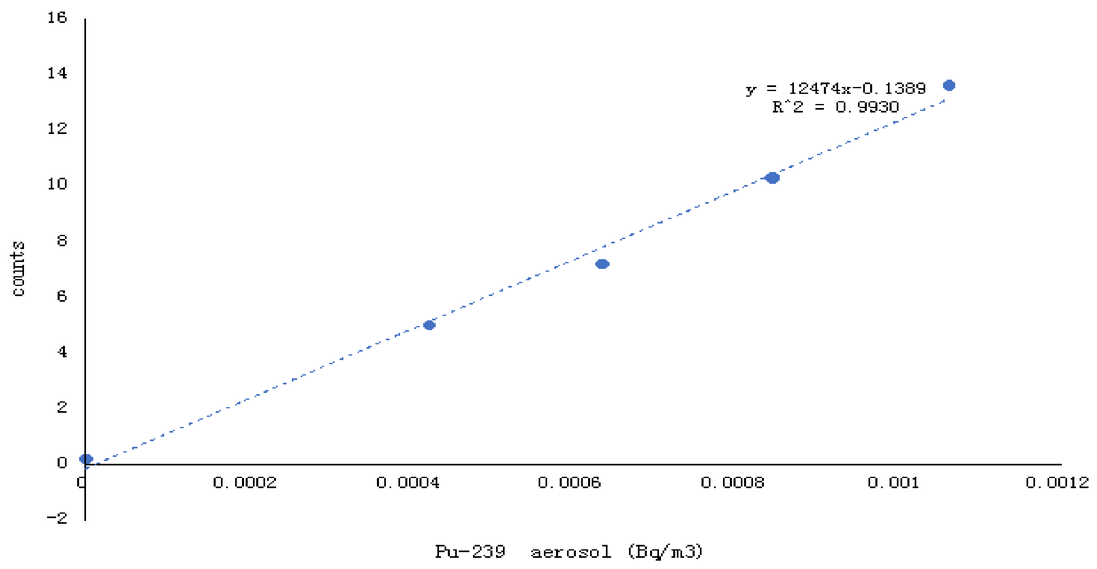

Fig. 3 calibration line at low concentrations

Results

The aerosol direct introduction device established could ensure the gas exchange rate for oxygen and nitrogne with argon above 99.9%, when the aerosol introduction volume <0.9L/min and the argon volume is 20L/min, which could make the air samples directly introduced into the ICP-MS instruments possible. According to the background from table 1, the LOD is 8.48E-04Bq/m^3, and according to the calibration line from fig 3, the LOD is 1.13E-03Bq/m3. And 1.13E-03Bq/m^3 was chosen as the LOD of this technology, the measurement time was 1min which was much quicker than the CAM (table 2). The LOD chosen for workplace monitoring is about 1/186 DAC for M type Pu-239 aerosol and 1/708 DAC for S type Pu-239 aerosol, and 1/11150 DAC-h for M type Pu-239 aerosol and 1/42478 DAC-h for S type Pu-239 aerosol, which shows great superiority to the CAM base on PIPS.

Table 2 LOD of sevral kinds of CAM

System	Company	LOD	Meaurement time	notes
iCAM	Canberra	4.8Bq/m^3	5mins	^{222}Rn is 10Bq/m^3
SmartCAM	Lab Impex Systems	0.1Bq/m^3	30mins	-
alpha 7a	Thermo	0.2 Bq/m^3	1h	^{222}Rn is 30Bq/m^3
Our work	CIAE	1.13E-03Bq/m^3	1min	-

Conclusion

An aerosol direct introduction method for aerosol directly measured by ICP-MS was established， and a new online monitoring technology for Pu-239 aerosol based on aerosol direct introduction device, membrane desolvation nebulizer and ICP-MS was established， with the measure time is 1min and the detection limit is 1.13E-3Bq/m3, which was an effective technical support for the radiation protection and process safety early warning.

Reference

1. Nomizu T, Kaneco S, Tanaka T, et al. Determination of Femtogram Amounts of Zinc and Lead in Individual Airborne Particles by Inductively Coupled Plasma Mass Spectrometry with Direct Air-Sample Introduction[J]. Analytical Sciences, 1993, 9(6):843–846.

2.Vincent T, Nishiguchi K, Utani K, et al. Online Multi-elemental Monitoring of Environmental Atmospheric Gases with a Gas Exchange Device Coupled to the High Sensitivity Thermo Scientific iCAP Qs ICP-MS[J]. 2012