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^{39}Ar ENRICHMENT SYSTEM BASED ON A 2.45 GHz ECR ION SOURCE*

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Abstract

Aimed at improving the ATTA's (Atom Trap Trace Analysis) dating efficiency with ^{39}Ar radioactive isotope, an isotope enrichment system has been developed at IMP (Institute of Modern Physics) to increase the abundance of ^{39}Ar in the incident sample gas. In this enrichment system, a 2.45 GHz ECR ion source was designed to ionize sample gas and produce isotopes beams with several mA, and the isotopes beam is transported and separated in the separation beam line, which is consisted of two quadrupoles and an analysis magnet. The separated isotopes are collected by a rotated aluminium foil target. According to the recent cross-checked results with ATTA, high enrichment factor of ^{39}Ar isotope has been successfully reached. This paper will give a general introduction to the platform setup. The isotope enrichment efficiency is the critical issue for such a platform and will be specially discussed.

INTRODUCTION

^{39}Ar (half-life is 269 yr) is an ideal isotope for water or ice dating on the time scale of 50 ~ 1,000 years [1]. ^{39}Ar is produced in the atmosphere through cosmic ray induced nuclear reactions and equilibrates at an isotopic abundance of 8×10^{-16}, which makes it extremely challenging for all analytical methods [2]. Up to now, only three effective methods have been successfully applied to environmental sample analysis, i.e. low-level decay counting (LLC) [3], accelerator mass spectrometry (AMS) [4], and the recently developed ^{39}Ar-ATTA [5].

In term of research convenience and feasibility for small samples, ^{39}Ar-ATTA has demonstrated itself the most promising method for ^{39}Ar dating research with significantly higher selectivity [6]. However, typical applications of ATTA such as ocean ventilation would expect Ar sample of only 2 mL STP (standard temperature and pressure) of argon, which can be extracted from 5 L of water [7]. To improve the counting efficiency and counting rate of Ar-ATTA, a variety of improvements such as gas recirculation has been realized. But to improve the counting rate of ^{39}Ar by a factor of 10~100 to lower the analytical uncertainties for practical analyses, ^{39}Ar enrichment might be a more straightforward technical solution.

Noble gas enrichment with an electromagnetic separation system has been previously validated as a successful approach [8]. As ^{39}Ar has a very low abundance in sample Ar gas, to have efficient enrichment within hours, a high intensity beam mass separator system is needed. At IMP, we developed an electromagnetic separation system integrated with a high current 2.45 GHz ECR ion source capable of producing 1~10 mA Ar^{+}.

SYSTEM DEVELOPMENT

To enrich ^{39}Ar and increase its concentration with high efficiency, crucial factors are intense Ar^{+} beam production and effective separation of the Ar isotopes on the target plane. A high-resolution spectrometer system can separate Ar isotopic ion beam well on the target plane. A 2.45 GHz ECR ion source can produce mono-charged ion beams up to 100 mA. Besides, an ECR ion source can realize a very high ionization efficiency of the incident sample gas compared to other types of high current ion sources, which is very advantageous for limited Ar sample gas. Therefore, a 2.45 GHz ECR ion source [9, 10] is utilized as the Ar ion beam production ion source. High beam current can obviously shorten the needed enrichment time, but the beam transport efficiency in the spectrometer system will become lower as a result of beam lose, which is mainly caused by space charge effect of the intense ion beam. Increasing ion beam extraction high voltage can mitigate the influence of space charge effect and therefore improve the transport efficiency. However, higher ion source extraction voltage means higher beam energy and that will imply higher rigidity of the magnetic separator system, and consequently a bulkier system, more power consumption, higher project budget and so on. Additionally, higher beam energy will cause trouble in unwanted beam and en richment target design. The final design of this enrichment system is a compromise of all the afore crucial factors.

The layout and picture of the IMP ^{39}Ar enrichment system is given in Fig. 1. In addition to a 2.45 GHz ECR ion source and a high-resolution spectrometer system consisting of a 95-degree dipole and two quadrupoles, a target chamber and a gas circulating system are also shown in the picture. The total length of the system is about 6 meters. All elements of this system have been meticulously designed to satisfy the projects goals.

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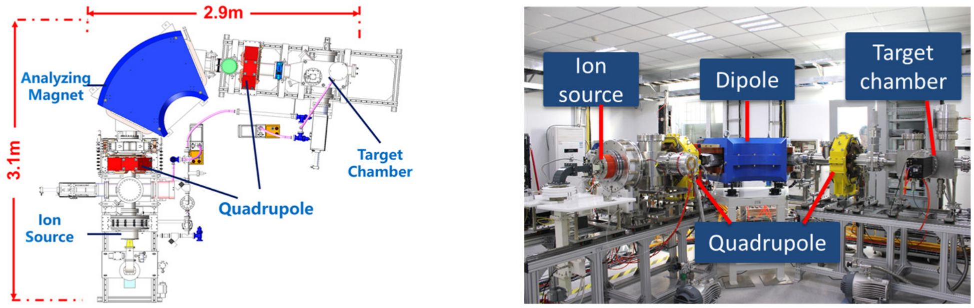

(a) (b)

Figure 1: Layout (a) and picture (b) of ^{39}Ar enrichment platform.

Ion Source

The typical parameters of the ion source are given in Table 1. A 2.45 GHz ECR ions source has been developed for this system. This ion source features a permanent magnet structure and ϕ 200 mm×100 mm dimension for compactness and easy-operation. Although maximum 5 mA Ar^+ is required for the routine operation, more than 10 mA Ar^+ beam extraction has been tested to demonstrate the ion source capacity.

Table 1: Typical Parameters of the Ion Source

Ion Species	Ar^+
Maximum Current	> 10 mA
Operation Model	DC
Ion Energy	40 keV
Beam Stability	≤ 1%

When treating real samples, only limited gas (typically < 5 ml STP) is available, and thus a high ionization efficiency is desired to improve the enrichment efficiency (as only the ionized Ar atoms can reach the target). Figure 2 shows the ionization efficiency and beam current at different gas consumption rates with microwave power of 300 W, which indicates that this ECR ion source could reach an ionization efficiency of >40% with Ar gas.

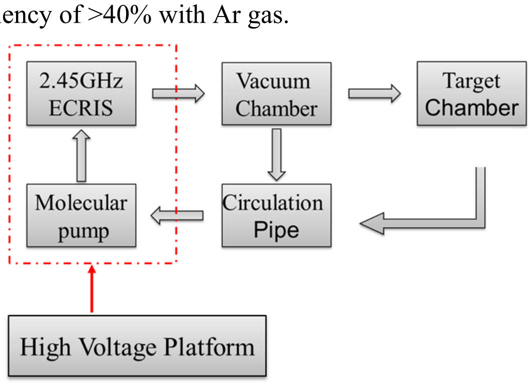

Figure 2: Sketch map of gas circulation.

As presented in Fig. 3, to have a reasonable ionization efficiency, the ion source needs to be operated with an extraction beam current of 1 mA to 3 mA.

Gas Circulation

As the typical Ar ionization efficiency is 50% ~ 60%, to maximize the enrichment efficiency up to 80%, Ar recirculation is necessary. The sketch map of gas circulation is shown in Fig. 3. The working gas that is suctioned from vacuum chamber and target chamber will be injected into ECR ion source to ionize again. In addition, the assistant molecular pump and ECR ion source are set on a high voltage platform.

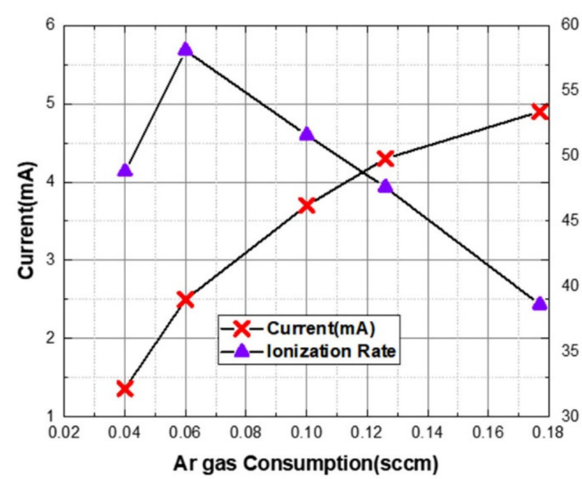

Figure 3: Ionization rate of ion source at different Ar gas consumption and beam current.

Moreover, Non-Evaporable Getter (NEG) pumps have been utilized in the ^{39}Ar enrichment platform vacuum system. Once the enrichment system starts to enrich ^{39}Ar, the vacuum evacuation system will be switch to recirculation operation mode, and the vacuum condition will be maintained via the NEG pumps. More information about this system has been indicated in former report [11].

EXPERIMENTS

According to the project schedule, some experiments have been finished to check this system's properties. Test experiment was finished to check Ar isotopic positions and some enrichment experiments to check project indicators. All these samples were enriched with Al foil, which had been heated in vacuum enrichment to remove gas on the surface of Al foil.

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Test Experiment

In order to cross-check the positions of the ^{39}Ar isotopic positions and indicate them directly, a temporary target was made to do this. A piece of heat-sensitive paper fixed on Al foil has been used to detect the Ar isotopic ion beams at the target plane. Ion beams of ^{36}Ar, ^{38}Ar and ^{39}Ar will bombard the heat-sensitive paper, while the high power ^{40}Ar beam will be indicated by the Al foil. As shown in Fig. 4, the resultant burning marks by bombarded ion beams are consistent with simulation results.

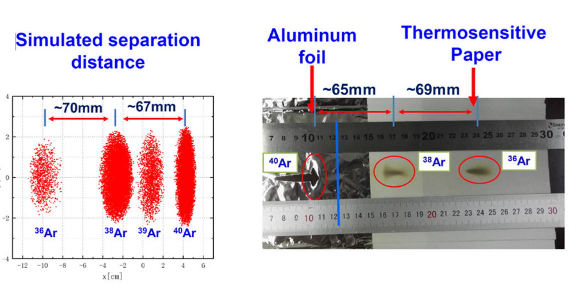

Figure 4: Analysis of the Ar isotopic positions: simulation results (left) and burning marks test (right).

Enrichment Experiments

Based on previous work, serval experiments have been finished to check system enrichment effect. And all enrichment results were validated by ATTA test. The enrichment system was operated for 6-hours and the Al foil containing the enriched samples were sent to USTC for ATTA analysis. Figure 5 shows count rate of natural abundance Ar and after enrichment Ar. Compared with natural Ar's count rate, enrichment samples' have been improved larger than 100 times.

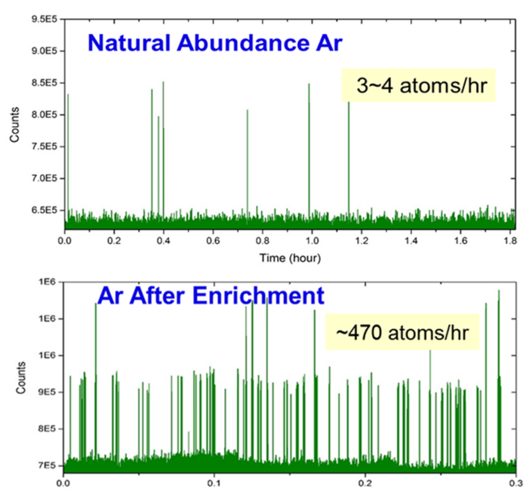

Figure 5: ^{39}Ar detection counts rate of natural abundance Ar gas and enriched samples.

Recently, two experiments with constant sample gas (natural Ar, 5 mL STP) were finished. As shown in Fig. 6, within the error of ATTA, the ^{39}Ar/^{38}Ar ratio of enriched samples are consistent with natural Ar's, which means these enrichment process can preserve sample's dating information completely.

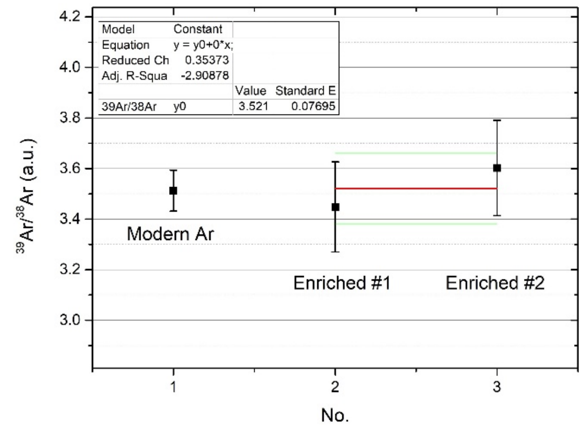

Figure 6: ^{39}Ar/^{38}Ar ratio detection with ATTA of Enriched samples and modern Ar.

CONCLUSION

This ^{39}Ar enrichment has been developed successfully. Moreover, recent experiments show that the ^{39}Ar abundance could be improve more than 100 and this system can preserve dating information well in enrichment process.

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