An Argon Recycling System for Inductively-Coupled Plasma Mass Spectrometry (ICP-MS)

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Abstract: A novel method has been proposed in this work to reduce the consumption of argon in ICP-MS devices. The method involves collecting the exhaust argon from the interface region of the ICP-MS device, removing the impurities from the collected gas stream using some filters, and feeding back the purified argon back to the ICP torch. The proposed system is, therefore, capable of significantly reducing the argon consumption.

Keywords: Inductively Coupled Plasma, Mass Spectrometry, Argon Recycling, Argon Purification

1. Introduction

Inductively coupled plasma is an unparalleled excitation/ionization source which is being used in a variety of spectrometric applications. Among these, inductively coupled plasma mass spectrometry (ICP-MS) is now the fastest growing one with exceptionally low detection limits (10 ng/L and lower), wide analytical working range, isotropic capability, short analysis time, etc. These features has turned ICP-MS into a versatile trace element analysis technique with applications in various fields such as environmental, geological and geochemical, clinical and biomedical, forensic, semiconductor technology, etc. [1].

At the heart of every ICP-MS device lies an ICP torch wherein the plasma ionizes the sample particles which are then aspirated into the sampler orifice, separated from unwanted particles using a mass separation device (e.g., a quadruple, magnetic sector, or time-of-flight separator), and finally reach the ion detectors for further analysis. Despite all the major benefits of an ICP-MS device which are the cause of fast growing interest toward these devices throughout the world, they consume a lot of argon which is mostly used for sustaining the plasma and keeping the torch from thermal damages. As argon is an expensive gas with limited/no resources in the developing/underdeveloped countries, ICP-MS market– like all the other ICP-based spectrometric technologies– have not experienced the same successful growth in these areas of the world as that of the industrial countries. Therefore, it has been of much interest to reduce the argon consumption of ICP torches.

Reed [2] was the first to build a practical ICP torch in 1961 which comprised of only one quartz tube. Later Greenfield et al. [3, 4] adapted the original design to develop a three-tube torch with the central tube serving to introduce the sample particles into the plasma. This was the first instance of an annular plasma (as opposed to a solid plasma) which was later proved to be superior in terms of analytical capabilities. At this time, ICP began attracting attention as a potential alternative emission source in atomic absorption spectroscopy (AAS), emission spectroscopy, and flame-photometry. This was mainly due to the fact that ICP was presenting higher temperature, longer particle residence time, better control over the chemical environment (in contrast to flames), minimized depressant effects of chemical interferences, and less background emission, with the absence of any electrodes and their undesired consequence—most common in dc plasmas [5-7]. In 1974, Fassel and Kniseley [8] and later Scott et al. [9] developed a modified version of the Greenfield torch with a smaller diameter, improved geometrical features, and higher figures of merit (FOM). This torch, now mostly known as the Fassel Torch, is generally considered as the most efficient version by most spectrochemists.

Table 1 shows the typical range of parameters in an ICP-MS device. It can be seen that as high as 19 L/min of argon should be fed to the torch to sustain the plasma and retain an acceptable level of analytical performance. More importantly, if the outer gas flow rate is decreased below critical levels, the torch will not be cooled effectively which most certainly leads to either devitrification or melting.

Table 1. Typical range of mass spectrometry ICP source parameters [1]

Frequency (MHz)	Power (W)	Outer gas (L/min)	Intermediate gas (L/min)	Carrier gas (L/min)
27 or 40	~ 1600	12 - 17	~ 1.2	~ 1

These drawbacks were a motivation for many researcher to find practical ways to decrease the consumption of argon. One of the most practiced methods for this purpose is torch

miniaturization and geometrical modification. Savage and Hieftje [10] proposed that a mini-torch could stably support a plasma with less than 8 L/min of argon flow. In one study, Rezaaiyaan et al. [11] developed some "plasma stability curves" to find out the most efficient operational parameters for running a stable plasma. Alternatively, Van Der Plas and de Galan [12] investigated the effect of radiation cooling on torches built from different materials. This is while some other people turn their attention to cooling the torch with water in some instances [13, 14] but mostly air [15-19] due to obvious reasons. However, none of these methods seem to have aroused lasting interest in the field and argon consumption in ICP-based analytical devices still remains at relatively high values.

Due to the mentioned reasons, alternative strategies for reducing argon consumption should be considered. In this paper, a completely new approach is followed wherein the argon is collected at the torch exit, forced through some filters to remove impurities, and finally fed back into the torch. Details of this method are elaborated in the following section.

2. Argon Recycling

A quick literature survey reveals the basic requirements of an efficient ICP source as:

1) Maintaining a stable plasma with sufficient excitation/ionization energy;
2) Low consumption of argon gas and energy per work cycle;
3) Minimal background emission and analyte signal noise;
4) Low detection limits;
5) Ease of operation.

In a typical ICP-MS system, it is observed that only about 12% of the consumed argon goes into the sampler orifice for analytical purposes. The remainder is discharged into environment without any further use. It would be thus reasonable if the exhausted argon could be collected and recycled back into the system. The purity of argon needed for ICP-MS must be as high as 99.996%. This imposes serious challenges in designing an effective purification system, considering the high concentration of water, acids, salts, and different hydrocarbons in some samples. As a result, the discharged gas will have significant amounts of H_2O, N_2, H_2, O_2, CO, and CO_2 that should be removed during the purification process.

In the first step of recycling process, the exhausted argon from the torch outlet should be collected before becoming mixed with the surrounding air. Considering the high temperature of plasma at the torch outlet, the collection system should be immune to thermal damages. The material used for building the collectors should neither introduce any additional contaminants to the collected gas. Moreover, components of the collection system should not interfere with the sampling process, induction coils, and R.F. coupling. Since, the position of the torch—with respect to sampler orifice—should be adjustable, the collection system should not hinder such a freedom. Also, the torch and plasma condition should be observable through the lens stack during the ICP-MS operation. To consider all these parameters in the design procedure, initially a numerical model was developed using the ANSYS-FLUENT software package. The model is capable of accounting for the magneto-hydro-dynamic effects along with fluid flow and heat transfer within the computational domain. Several simulations were carried out using this model to reach to a suitable design as per the aforementioned requirements. The dimensions and other working parameters were adapted from the NexION® 350 ICP-MS instrument from Perkin Elmer Corporation. Based on the predicted temperature, gas velocity, pressure, and argon concentration fields, several designs were proposed and revised before moving ahead to the manufacturing phase.

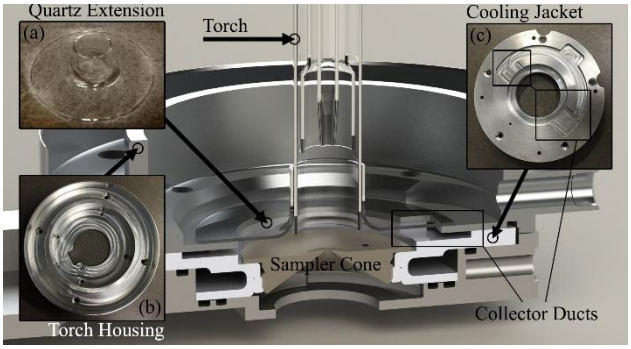

Fig. 1. Cross section of the 3D model for the designed argon collection system. The fabricated parts for quartz extension, torch housing, and cooling jacket are shown in (a), (b), and (c), respectively.

Fig. 1 shows the final design and its components. It was realized that some original parts of the NexION® system should be slightly modified in order to comply with the new collector design. Primarily, a quartz extension was built to prevent the argon gas exiting the torch to mix with the surrounding air. This component easily slides over the torch without restricting its movement or blocking the view of lens stack for monitoring the plasma. The interface components of the mass analyzer was also redesigned to be able to integrate collection ducts with the cooling jacket. These ducts are used to collect the exhaust argon in a symmetric manner around the torch exit and later cool the collected gas for further stages of the purification process. A pump was eventually used to suck the collected argon from the ducts and force the gas through all the purification filters. Fig. 2 shows the simulated temperature distribution in various locations of the collection system. It can be seen that the temperature does not exceed the permitted levels at any point of the collector walls. In fact, the temperature of the collected gas at the exit of the collection system was

obtained to be not more than 650 K, showing that the cooling jacket is working effectively to remove the heat from the collected gas through the duct walls.

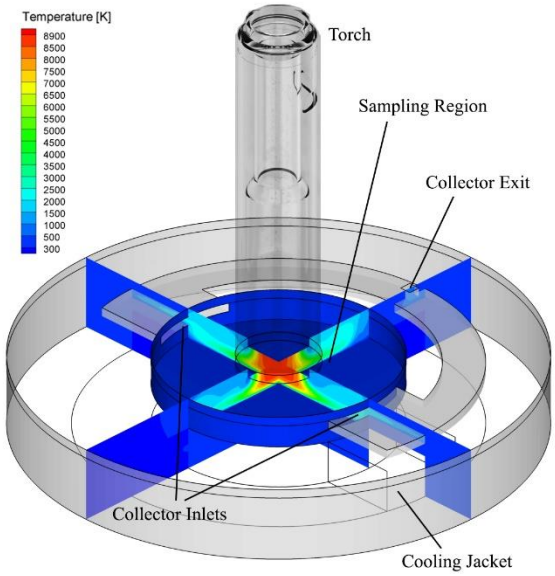

Fig. 2. 3D model of the collector integrated with the simulated temperature distribution on select cross sections.

Next, the designed system was mounted on a NexION® system to assess the performance in action. Fig. 3 shows the condition of the plasma as viewed through the lens stack. This is while the argon is being collected from the interface region. It was observed that the plasma runs stably without any issues or signs of thermal damage to the components.

Fig. 3. Operational condition of plasma with the new design, as viewed through the lens stack.

To make sure the collected gas was not contaminated with any air throughout the collection system, the collected gas was sampled and analysed with a Gas Chromatograph. The results are summarized in Table 2.

Table 2. Comparison between various species peaks measured with Gas Chromatograph for the pure bottled argon and collected exhaust argon.

	Pure argon from bottle	Collected gas
H_2 Peak (arb. unit)	0	~ 0
O_2 Peak (arb. unit)	35	32
N_2 Peak (arb. unit)	68	83

It is clear that the collected argon is as pure as the initial bottled argon fed to the torch meaning there is no air leakage within the collection system. These measurements were done while no sample was being injected into the plasma and only pure argon was fed to the torch. In the real-time operation of an ICP-MS device, however, a variety of sample types are injected for analysis. As a result, after collection, the exhaust gas should be further processed by some filters.

Fig. 4 shows the schematics of a filtration cycle. In the first stage after collecting the argon, an open-cell metal foam filter is proposed to remove the water content from the gas stream. A thermoelectric cooler is used to decrease the temperature of the filter below -10°C to freeze and capture the water content. Subsequently, the dry gas stream if forced through an open-cell porous titanium filter which is kept at elevated temperatures of up to about 800°C. This increases the rate of titanium interaction with the impurities present in the gas stream. As a result, the exiting gas will be free of any contaminations. Later the hot gas can be cooled using a heat exchanger and fed back into the torch. Various components of the proposed filtration system has been built and are currently under investigation. Initial results looks very promising and show that the filtration system is capable of removing all the contaminants from the gas stream. Detailed results will be disclosed in the future publications.

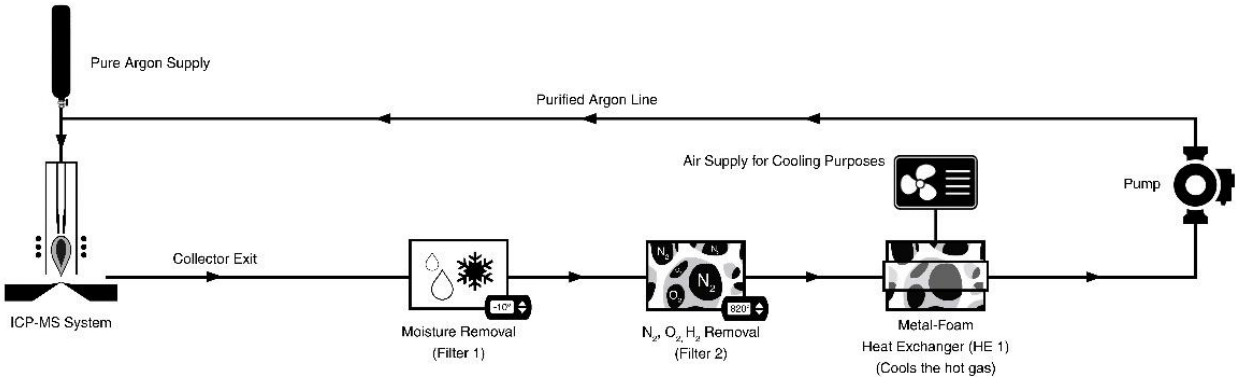

Fig. 4. Schematics of the filtration system for the collected argon

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A US patent application (US 62/479,982) has also been filed on Argon recycling for an inductively coupled plasma mass spectrometer.

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