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# Application of factorial design in optimization of ultrasonic-assisted extraction of aluminum in juices and soft drinks

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**Abstract**

A sample preparation method based on ultrasound-assisted pseudo-digestion of Al from Juices and soft drink samples under ultrasonic effect has been described. A Plackett–Burman experimental design was used as a multivariate strategy for the evaluation of the effects of varying several variables at once. The effects of five different variables preintensification time (without ultrasonic stirring), intensification time in ultrasonic bath (UB), temperature of UB, two acid mixtures ($HNO_3$–$H_2SO_4$–$H_2O_2$ and $HNO_3$–$H_2O_2$), on the recovery of Al have been investigated. From these studies, certain variable showed up as significant, and they were optimized by a using $2^3$ + star central composite design, which involved 16 experiments. The best conditions for pseudo-digestion were as follows: a preintensification time 10 min, intensification time 20 min, volume of acid mixtures 3.0 ml and temperature of ultrasonic bath 80 °C. A conventional acid digestion on electric hot plate was used to obtain total Al for comparative purpose. Final solutions obtained from both methods, were analysed by electrothermal atomic absorption spectrometry. Analytical results for the Al by ultrasound-assisted pseudo-digestion, and conventional wet digestion methods showed a good agreement, thus indicating the possibility of using ultrasonic-assisted digestion sample preparation instead of intensive treatments inherent with the acid digestion methods on electric hot plate. The procedure proposed allowed the determination of Al with detection limit (3ά/*s*) 10 μg $l^{-1}$.


*Keywords:* Ultrasonic bath; Aluminum; Factorial design optimization; Electrothermal atomic absorption spectrometer (ETAAS); Soft drink; Juices

## 1. Introduction

Aluminum (Al) is the most popular metallic element in industries and consequently in our surroundings. However, it has attracted much attention for more than two decades in discussing whether it has any relationship with serious dementia cause Alzheimer's disease. Potential health risks associated with elevated Al intake have long been discussed among physicians and Al poisoning in patients with chronic renal failure is one of the most important clinical problems involving trace metal toxicity [1].

Despite the facts that Al is present in all foodstuffs, the question of contamination with Al from the sources arises now and then. It is well known that acidic food such as apple, rhubarb or vinegar may cause corrosion of cooking pans and that, due to this, the Al content of the food may rise during preparation [2,3]. The hypothesis that Al exposure is etiological related to Alzheimer's disease has lead to much debate. Ecological studies have suggested that concentration of Al in drinking water of 0.1–0.20 mg $l^{-1}$ may increase the risk of Alzehiemer's disease with relative risk ranging from 1.35 to 2.6 mg $l^{-1}$ [4,5]. Thus special efforts should be taken to prevent contamination of food and beverages, etc., with Al either directly or during preparation with special regards to infants, old people or individual with suboptimal renal functionality.

The determination of metals in complex samples by atomic absorption spectrometry (AAS) generally requires the destruction of the sample matrix to render a solution of the analyte ready for analysis. Sample digestion techniques, such as microwave,

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and conventional wet acid digestion (CDM) for total heavy metal determination have been used widely for the dissolution of target elemental analytes [6,7]. Such digestion techniques require the use of concentrated acids and high temperatures, and often high pressures, to affect the total dissolution of elemental analytes from liquid and solid samples [8]. For elemental analysis, the sample preparation procedure employed is, apart from sample collection, ordinarily the most time-consuming step in the overall analysis [9]. The efficiency of the wet digestion procedures can be enhanced with the use of ultrasound irradiation [9,10]. Ultrasonic radiation can be considered another alternative for sample pre-treatment, since ultrasound facilitates an auxiliary energy and accelerates some steps, such as dissolution, fusion and leaching, among others [11]. In many situations, ultrasound-assisted leaching is an expeditious, inexpensive and efficient alternative to conventional extraction techniques and, in some cases, even to supercritical fluid and microwave-assisted extraction, as demonstrated by application to both organic and inorganic analytes in a wide variety of samples [12–14].

Several means of optimization can be used for the extraction of metals from different matrixes, but some methods are time-consuming: for instance, the study of each variable separately. Procedures for optimization of factors by multivariate techniques have been encouraged, as they are faster, more economical and effective, and allow more than one variable to be optimized simultaneously [15,16]. Among the different groups of designs, Plackett–Burman designs (PBDs), introduced in 1946 by Plackett and Burman [17], allow us to discover the most significant variables for a certain system with only few experiments. They are used as a screening method in order to select the variables that have influence on a system but they do not give the optimum value for each variable.

In order to obtain the optimum values for each variable involved in a certain system, central composite designs are the most widely used design framework [18]. These design structures are based on full two-level factorial design by center point replication and inclusion of an axial portion [19]. The optimization procedures based on these approaches are nowadays being applied to optimize some sample pre-treatments and operating conditions for some analytical techniques [20–23].

The aim of this work was to improve sample preparation performance, proposing the development of an ultrasound-assisted pseudo-digestion method (UDM) for fast and reproducible recovery of Al in soft drinks and juices. Factorial designs were used for optimization of the experimental variables. Parameters influencing ultrasound-assisted pseudo-digestion, such as preintensification time (without ultrasonic stirring) intensification time (in ultrasonic bath), acid mixtures, temperature of ultrasonic bath are fully investigated. Due to there being many variables that affect the acid digestion process, in order to obtain the optimal values for each variable, experimental designs, such as Plackett–Burman and central composite designs, have been used throughout the optimization.

## 2. Experimental

### *2.1. Reagents*

Through out the experimental work using ultrapure water, obtained from a Milli-Q purification device (Millipore Co., Bedford, MA, USA). For the preparation of samples, standards and blanks, Milli-Q water additionally purified by distillation. All reagents used were of analytical reagent-grade. Concentrated Nitric acid, sulphuric acid, 30% hydrogen peroxide were spectroscopic grades (Merck, Darmstadt, Germany) and were checked for possible trace Al contamination by preparing blanks for each procedure. Certified standard of Al (1000 ppm) was obtained from Fluka (Buchs SG, Switzerland) Laboratory glass wares was kept overnight in 10% (v/v) nitric acid solution, and washed with distilled water and finally with ultrapure water before use.

### *2.2. Instrumentation*

The ultrasonic-assisted digestion experiments were carried out with an ultrasonic bath Sonicor, Model No. SC-121TH, Sonicor Instrument Corporation Copiague, NY, USA with technical specifications; programmable for temperature ranging from 0 to 90 °C, timer 0–30 min, 220 V, 50/60 Hz, intensification frequency 35 kHz for the ultrasound energy and a total volume of 4 l, was used to induce the acid digestion process. Al was determined in both digests obtained by both procedures using atomic absorption spectrometer of Hitachi Ltd., Model 180-50, S. No. 5721-2, equipped with graphite furnace G-03. The instrumental parameters are shown in (Table 1).

The calibration curves (0.1–1.0 $\mu$g m l$^{-1}$) for aluminum were established with solutions prepared from a 1000 $\mu$g ml$^{-1}$ certified stock solution.

### *2.3. Samples*

Different eight brands of Soft drink and 10 juice samples were collected from the urban areas of Hyderabad city during 2004–2005. Five to ten samples of different batches of the same brand of both drinks packed on different dates were also

Table 1
Measurement conditions for ETAAS for aluminum

| Instrumental parameters | |
|---|---|
| Lamp current (mA) | 10 |
| Wave length (nm) | 309.3 |
| Slit-width (nm) | 1.3 |
| Cuvette | Tube |
| Carrier gas (ml/min) | 200 |
| Sample volume ($\mu$l) | 10 |

| Temperature programming [temperature range/time (s)] | |
|---|---|
| Dry | 80–120/15 |
| Ash | 400–700/15 |
| Atomization | 2700–2800/5 |
| Cleaning | 2800–2900/2 |