## ALUMINIUM CONTAMINATION VIA ASSISTED LEACHING FROM METALLIC ALUMINIUM UTENSILS AT NEUTRAL PH

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Abstract. Fluoride ion in trace concentrations is found to leach metallic aluminium from utensils even at neutral pH, releasing aluminium hydroxide which is readily soluble in dilute acids. The levels of leaching at different  $F^-$  concentrations are given and compared with leaching levels with equivalent concentrations of Cl<sup>-</sup>. It is suggested that consumption of water boiled in aluminium utensils may contribute to cumulative aluminium toxicity.

## 1. Introduction

Aluminium pollution is receiving increasing amount of attention because of the new evidence suggesting its cumulative toxicity. (Shore and Wyatt, 1982; Wisniewski et al., 1982; Brusewitt, 1984; Candy et al., 1986; Prescott, 1989). Aluminium ingestion could result from: (1) contamination of food via leaching from cooking utensils; (2) storage of food in contact with aluminium; (3) aluminium salts added to water during purification; (4) aluminium compounds added to food, e.g. aluminium in baking powder; (5) aluminium in vegetables (plants assimilate aluminium to varying degrees depending on species, the availability of aluminium in the soil, soil pH etc.); (6) use of aluminiumcontaining drugs. In our earlier work we discussed the leaching of aluminium from cooking utensils in the presence of the fluoride ion (Tennakone et al. 1987, 1988). However, this investigation was limited to conditions of acidic and alkaline pH. Neutral or nearly neutral pHs are more prevalent in cooking than wide deviations from the neutral. Furthermore aluminium utensils are frequently used in boiling water for making tea, coffee or drinking. In this work we present our measurements on leaching under conditions of neutral pH, in the presence of the fluoride ion for comparison the leaching at neutral pH in the presence of chloride is also studied. Fluoride assisted dissolution of aluminium becomes important because water consumed in several localities contains 10  $\mu$ g g<sup>-1</sup> or more fluoride. Furthermore, some food materials are known to contain excessive amounts of fluoride (Tennakone et al., 1988).

## 2. Experimental

The following procedure was used to simulate leaching under cooking conditions and to

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obtain reproducible results. Aluminium plates ( $8 \times 6$  cm thickness = 2 mm) were etched in 0.05M HCl for 45 min. and washed with distilled water to remove all chloride. Plates were then agitated ultrasonically in distilled water to remove the loose deposit of alloying metals and aluminium hydroxide formed during etching (commercial aluminium used in this investigation contains approx. 0.3% Cu and 0.1% Fe). Ultrasonically cleaned plates were washed again and kept immersed under distilled water. Water (400 ml) in glass beakers containing varying amounts of fluoride (sodium fluoride was used) was kept boiling and the plates were dropped into each beaker (plates were fully submerged). A constant volume was maintained by addition of water and the plates were removed after a measured interval of time. To dissolve aluminium hydroxide 1 ml of conc. HCl was added to each beaker and the aluminium concentration was estimated using an inductively coupled plasma spectrophotometer (Shimadzu ICPS – 1000 II) for each set of conditions (i.e. F<sup>-</sup> concentration and boiling time) the experiment was repeated 10 times and in each case freshly cleaned plates were used. The experiment was also conducted in the presence of the chloride instead of the fluoride ion. The source of chloride used was NaCl.

## 3. Results and Discussion

Table I gives the concentration of leached aluminium in the solution at different concentrations of F<sup>-</sup> when the duration of boiling is 0.5 or 1 h. In the absence of fluoride, the leaching is insignificantly small and with the increase of the F<sup>-</sup> concentration  $(1 - 10 \,\mu g \, g^{-1})$  a progressive increase in the amount leached is noticed. At neutral pH the product of dissolution is the transparent gel of aluminium hydroxide readily soluble in dilute mineral and organic acids. At a given concentration of F<sup>-</sup> leaching increases with time approaching a saturation in about an hour. The saturation leaching level is independent

Concentrations of leached aluminium in the presence of F <sup>-</sup> .					
[F⁻]µg g⁻¹	Boiling Time/h	Leached [Al]/ $\mu$ g g <sup>-1</sup>			
0	0.5	$0.05\pm0.01$			
1	0.5	$0.47\pm0.02$			
2	0.5	$0.78 \pm 0.03$			
4	0.5	$1.42 \pm 0.11$			
6	0.5	$2.45\pm0.07$			
8	0.5	$3.11\pm0.17$			
10	0.5	$4.03 \pm 0.65$			
0	1	$0.07 \pm 0.01$			
1	1	$0.67 \pm 0.04$			
2	1	$1.03\pm0.04$			
4	1	$1.87\pm0.02$			
6	1	$2.53\pm0.03$			
8	1	$3.7 \pm 0.02$			
10	1	$4.4 \pm 0.27$			

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