Effect of pH, Salinity and Temperature on Aluminum Cookware Leaching During Food Preparation

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The amount of aluminum intake hazardous to human beings has been under study for quite some time and has attracted particular attention from the society as it is believed to enhance diseases like the wellknown Alzheimer disease. This study measures the effect of pH, salinity and temperature of Egyptian and Indian aluminum cookware during food preparation using different water types, tap water and drinking water. In this study, the weight loss method is used to study aluminum leaching into different food solutions. Environmental scanning electron microscopy is used to study the morphology of the samples before and after exposure to the different food solutions. The Arrhenius equation is applied to find activation energies. Aluminum is found very sensitive to low and high pH as the corrosion rate increases in an alkaline environment. Corrosion rates are observed to decrease with drinking water compared to tap water. Increasing salt concentration increases the corrosion rate up to a certain value upon which a decrease is observed to reach a plateau of constant corrosion rates. This is attributed to the combination of high conductivity and oxygen solubility.

Keywords: aluminum cookware, leaching process, aluminum intake, salinity

1. INTRODUCTION

Aluminum is the third most abundant element in the earth's crust; existing mostly in the form of insoluble aluminosilicate and oxides [1]. Medical researches link aluminum to various brain, blood, and bones diseases. The cause of Alzheimer disease is still unknown, but aluminum might play a major role for the cause of it. Aluminum often binds strongly with other substances in food such as fluoride and phosphate, which may make it less absorbable. Studies report that beverages (tea, coffee and soft drinks) and cereals (cakes, puddings, biscuits, breakfast cereals, bread, flour, oatmeal and rice) are the main sources of aluminum from food. Aluminum-based food additives are widely used in bleaching,

preserving and pickling processes, and in powdered foods such as instant coffee, dried milk, and table salt. Bread, cake, biscuits and baking powders may have high aluminum levels if they contain aluminum food additives. In order to reduce the amount of unwanted aluminum intake from cookware, cooking acidic foods in uncoated aluminum pans must be avoided. Many reports suggesting that fluoride in water increases the aluminum dissolved from cooking utensils have been disproved. Even the use of aluminum foil during food preparation is assessed to have a significant risk on human health [2].

Aluminum is naturally present in some water. Also, aluminum sulfate is widely used in the tap water filtration process. Aluminum intake from water is very small; yet some studies state that the human body can easily absorb aluminum when found in water. The amount of aluminum present in drinking water has been recommended to be below 200 µg per liter by the World Health Organization [3]. Assuming that an adult consumes two liters of water per day, the aluminum intake would be only 0.4 mg; less than one-tenth of the average daily aluminum intake from food. Aluminum salts are added to water supplies in virtually most areas in the world according to the European Standard based on aesthetic considerations of water color rather than on any estimate of possible health risk. Alternative ways of water treatment are more expensive and less effective. Some water filters will remove aluminum from water though not all are effective. The largest source of aluminum actually comes from municipal water supplies. Many municipal water supplies are treated with both aluminum sulfate and aluminum fluoride. The National Institutes of Environmental Health Sciences (NIEHS) acknowledged that fluoride has been observed to have a synergistic effect on the toxicity of aluminum. Several chemical coagulants, such as iron compounds and organic polymers, can be used instead of aluminum-based coagulants [5]. The choice of the coagulant is based on a number of interrelated factors. Water chemistry (e.g., pH and temperature) is the main factor that determines which type of coagulant will perform most effectively. The most important reason why aluminum-based coagulants are chosen is that the alternatives do not always remove pathogens and particles. If a treatment plant is specifically designed to use alum with a certain type of water, it is not always possible to use an alternative without adversely affecting the water quality. Aluminum found naturally in untreated water is generally thought to be in a form that is not easily taken up by the body, and it is therefore of little concern in terms of health effects. It is only during the alum treatment process that aluminum appears to change into a form that may be more easily absorbed by the body. Like tap water, bottled waters vary in their aluminum content. Aluminum may be found in some bottled waters because it occurs naturally at the source.

Aluminum cookware, apart from other sources of dietary aluminum, is considered to be a potential source of this metal to human beings. Various research groups report aluminum leaching experimental results with food, beverages, and water under different experimental conditions modified by varying the levels of pH as well as organic and inorganic ionic content. The results reported show marked discrepancies in levels of leached aluminum. The apparent reason for such discrepancy in leached aluminum levels can be attributed to factors such as non-systematic and non-uniform experimental designs, non-standard conditions maintained during the experiments, and choice of method for aluminum analysis [6].

The complexing effect takes a very important role in the process of aluminum liberated from cooking utensils. Increased concentrations of complexing ions (organic acids, fluoride ion, OH⁻, etc.) significantly enhance the release of aluminum. The model suggests that in the pH range of most food (pH 4–8); the aluminum present is predominantly in the form of organic aluminum complexes, which is harmful to the human body [7]. Leaching of aluminum from utensils made of aluminum, indalium (alloy of aluminum), stainless steel, and hard anodized aluminum is studied under different conditions of pH and boiling time [8]. A low pH is found to enhance leaching of aluminum from the utensils. The leaching is found to be the highest during first-time preparation (new utensils) of all the foods as compared to second and third-time preparations using the same utensils. Leaching of aluminum during the preparation of various traditional Indian foods is found to be negligible in hard anodized aluminum, and indalium utensils [9].

The purpose of this study is to investigate the effect of pH, salinity, exposure time and temperature on aluminum cookware during food preparation using different water types.

2. MATERIALS AND METHODS

Egyptian and Indian aluminum cookware are chosen from the local UAE market. The cookware are cut into small rectangular shapes with dimensions of 1×1.2 cm, and each with a small hole of 1 mm diameter at one end in order to hang them in the water sample. These samples are exposed to drinking water, tap water as well as the following solutions:

Solution (1): 250 ml of 40% meat extract + 250 ml of tomato juice + 10g of citric acid Solution (2): 250 ml of 40% meat extract + 250 ml tomato juice + 10 g of citric acid+5 g of salt Solution (3): 250 ml of 40% meat extract + 250 ml tomato juice + 20 g of citric acid + 5g of salt

Tap and drinking water are used in preparation of the above solutions.

In the present work, the weight loss method (W_L) at different temperatures is used to study the aluminum leaching into different food solution samples. The aluminum specimens are cleaned by distilled water and acetone, dried, and weighed using a four-digit sensitive balance. After the test, aluminum samples are cleaned by distilled water followed by acetone and reweighed again. The pH of the solution is also measured before and after the experiment. The corrosion rate and aluminum intake per person calculations is presented in our previous work [10]. To assure consistency, all the experiments are performed in duplicates.

The samples are analyzed, before and after the experiment, using environmental scanning electron microscopy (ESEM) which is connected with an energy dispersive x-ray (EDX). This test indicates about the leaching of the metals from the initial condition. Also the picture shows the damage of the aluminum surface.

3. RESULTS AND DISCUSSION

For the Indian samples, the corrosion rate and the aluminum intake per person in drinking water, tap water, and the previous three food solutions are listed in Tables (1).

Solution		Aluminum Intake (mg/person)	Corrosion Rate (mg.cm ⁻² .hr ⁻¹ .10 ⁻²)	Corrosion Rate (mm/year)	Initial pH / Final pH
Drinking water	Pure water	100.75	10.5	3.4	7.0/9.3
	Solution (1)	140.62	14.68	4.7	3.3 / 3.0
	Solution (2)	121.11	15.58	5.06	3.0 / 2.8
	Solution (3)	181.17	18.87	6.13	2.8 / 2.6
Tap water	Pure water	301.93	31.45	10.2	7.3 / 9.1
	Solution (1)	42.65	4.43	1.44	3.5 / 3.6
	Solution (2)	61.44	6.64	2.16	3.4 / 3.0
	Solution (3)	148.8	15.5	5.03	3.2/3.0

Table 1. Effect of food solution prepared in drinking and tap water on the Indian sample after two hours of exposure at boiling temperatures

The above solutions represent the amount of aluminum leached by food in conditions close to the real cooking. People usually add tomato paste, lemon juice, table salt and other spices during cooking. The results show that leaching using drinking water alone is less than the one in tap water. The leaching is increased in food solutions prepared by drinking water while it is reduced in food solutions prepared by tap water.

The drinking water solution shows an increase in the leaching value as the salt and citric acid concentrations increase (Solutions (1), (2), & (3)). The pH of the solutions are 3.3, 3.0 and 2.8, respectively. This variation in the pH seems to be the reason for such behavior with the increase of citric acid concentration. This lies in good accord to the study done by Shuing Bi who reports that the OH⁻ ions significantly enhance the release of aluminum as the concentration of the complexing ions, such as organic acids, increase [7]. Aluminum exhibits a passive behavior in aqueous solution due to the protective compact Al_2O_3 film on its surface. However, the solubility of this protective film increases in acidic and alkaline medium. According to Bi [7], the aluminum leaching in aqueous solution may be explained by the following chemical reaction occurring on the surface of the aluminum cookware sample:

$$Al_2O_3 + 6H^+ = 2Al^{3+} + 3H_2O$$

Where, the Al_2O_3 is the protective film on the whole surface (anode and cathode). The free aluminum ions in solution react with organic acid found in food such as citric, oxalic and other complexing ligands like hydroxyl. Most previous works have largely been concerned with the physical chemistry of the aluminum dissolution and generally the results were thought to reflect chemical and electrochemical corrosion. Verissimo *et al* and Joshi *et al* report that aluminum leach more with these additives [11, 12].

Figure (1A) shows the damage on the Indian sample that is exposed to drinking water at boiling temperature for two hours. The figure clearly shows the severe localized attack which explains the lower leaching value compared to tap water.

On the other hand, Figure (1B) shows the damage of the Indian sample that is exposed to solution (1) which is prepared using tap water. The figure shows more uniform leaching.



Figure 1. ESEM of the Indian sample after two hours of exposure to boiling drinking water (A) and Solution (1) at boiling temperature in tap water (B).

The previous results indicate that the aluminum cookware is subject to destruction by some acidic and salty food. Rim Karbouj reported the same conclusion [13].

For the Egyptian samples, the corrosion rate and the aluminum intake per person in drinking water, tap water, and Solutions (1), (2), & (3) are listed in Table (2). The results show the same trend as in the Indian samples but with different leaching values.

Table 2	. Effect of food solution	prepared in	drinking w	ater on the	Egyptian	sample	after two	hours of
(exposure at boiling temp	eratures						

Solution		Aluminum Intake (mg/person)	Corrosion Rate (mg.cm ⁻² .hr ⁻¹ .10 ⁻²)	Corrosion Rate (mm/year)	Initial pH / Final pH
Drinking water	Water	21.39	2.22	0.72	7.0 / 9.1
	Solution (1)	65.5	6.3	2.04	3.5 / 3.1
	Solution (2)	100.63	10.5	3.4	3.2 / 3.0
	Solution (3)	120.77	12.58	4.1	2.8 / 3.0
Tap water	Water	403.8	42.1	13.6	7.3 / 9.4
	Solution (1)	42.65	4.43	1.44	3.4 / 3.2
	Solution (2)	42.65	4.43	1.44	3.2 / 3.0
	Solution (3)	127.62	13.3	4.3	3.2 / 2.9

Figure (2A) shows the damage on the Egyptian samples that are exposed to tap water at boiling temperature for two hours. The leaching value is the highest among all the solutions from both samples. The figure clearly shows that the damage is almost uniform on the whole sample.



Figure 2. ESEM of the Egyptian sample after two hours of exposure to boiling tap water (A) and to Solution (1) at boiling temperature in drinking water (B).

The aluminum intake resulting from using drinking water alone is 21.39 mg / person, while it is three times more in Solution (1), five times more in Solution (2), and six times more when in Solution (3).



Figure 3. ESEM of the Egyptian sample after two hours of exposure to Solution (3) at boiling temperature in drinking water (A) and in tap water (B).

Figures (2B) and (3) show the damages that occurred to the Egyptian samples after immersion in Solutions (1) and (3), respectively. It is clear that the damage occurred due to Solution (3), which

contains 20 mg of citric acid and 5 mg of salt, is more severe than the damage occurred due to Solution (1), which only contains 10 mg of citric acid. In addition to that, the figures show that both samples are subject to uniform and localized attack.

Figures (3A) and (3B) show the ESEM micrographs of the Egyptian sample after immersion in Solution (3) prepared using drinking water and using tap water, respectively. The aluminum intake values using both food solutions are almost similar; 127.62 mg per person resulting from Solution (3) prepared using tap water and 120.77 mg per person resulting from Solution (3) prepared using drinking water. Figure (3A) shows that the damage occurred due to the formation of localized corrosion with little uniform corrosion. On the other hand, Figure (3B) shows that the damage occurred due to the formation of uniform corrosion with little localized corrosion.

Using tap water, the prepared food solutions show again reduction in the leaching process compared to tap water alone. Figures (2A) and (3B) show ESEM micrographs of the Egyptian sample after immersion in tap water alone and in Solution (3), respectively. Figure (3B) shows that the sample undertook some localized leaching as well as uniform leaching, which might be the reason for the low leaching value.

The corrosion rate of the Egyptian and the Indian aluminum samples exposed to Solution (1) prepared in tap water is 1.44 mm/yr. compared to 1.5 mm/yr. reported in a previous work [14].

3.1. Effect of immersion time

The effect of immersion time of the Indian cookware using drinking water at boiling temperature is studied. The corrosion rate is increasing as the exposure time increases Figure (4).

The aluminum intake (mg/person) is recorded for test durations of 1-7h to be 40.3, 100.75, 120.90, 141.05, 161.20, 181.35 and 201.50, respectively.



Figure 4. Effect of the time of immersion on the corrosion rate of the Indian sample in drinking water at boiling temperature (solid line for mg/cm².hr, and dotted line for mm/yr).

The calculation of the average life of a pot having a thickness of 2 mm is based on the average use of two hours per day. The corrosion rate corresponding to two hours is 3.4 mm/yr (Table 2). As a result, the pots lose 7.8×10^{-4} mm from its thickness every day due to the use of pure drinking water without the addition of any food, acid, or salt during food preparation. Food solutions with additives increase the corrosion rate as seen in Table (1). If the corrosion mode is assumed to be uniform, then the pot is damaged completely in about seven years. The life span of the pot is reduced to half in real cooking conditions with the addition of meat, salt, acids, and spices. The life span is even more reduced if the corrosion mode is localized, as shown in Figure (1).

Figure (5) shows that the aluminum intake per person increases continuously with the exposure time. All the values between 40 to 200 mg per person show high levels of leaching; these values are considered unacceptable related to the limitations and indicate a high health risk. Cooking for additional hours, more than two, or using the pot to store food, after cooking, would increase the leaching rate even more.



Figure 5. Effect of time of immersion on the aluminum intake of the Indian sample in drinking water at boiling temperature.



Figure 6. Effect of time on the corrosion rate and weight loss of the Indian sample in tap water (solid line for corrosion rate, and dotted line for weight loss).

The corrosion rate and weight loss of the Indian sample in tap water at room temperature show the same behavior of the Indian sample in drinking water at boiling temperature (Figure (6)). The corrosion rate is lower at room temperature than at boiling temperature. Comparing the leaching rates of both temperatures for two hours shows that it is two and a half times more at boiling temperature than at room temperature. It is reported that aluminum leaching is significantly higher at 100°C than at the ambient temperature [13].

Figure (7) is an ESEM micrograph of the Indian sample after 408 hours of exposure. The damage seems to be localized, and pitting is very clear on the surface. This could be due to the long exposure time which might lead to scale formation. The formation of scale on the surface plays an ambivalent role; it can be positive by providing a protection of substrate or negative by forming a poorly adherent deposit accentuating pitting at pores or other voids in the scale.



Figures 7. ESEM of the Indian sample after 408 hours of exposure to tap water at room temperature.

3.2. Effect of temperature



Figure 8. Effect of temperature on the corrosion rate of the Indian sample in drinking water (solid line for mg/cm².hr, and dotted line for mm/yr).

The effect of temperature on the Indian aluminum sample is studied using drinking water after two hours of exposure (Figure (8)). The corrosion rates and the aluminum intake increase with temperature. The aluminum leaching is five times higher at 100° C than at ambient temperature; this is in agreement with the results discussed previously. At 185° C (oven work) the leaching is even more, and the corrosion rate is more than eight times than that at 100° C.

The aluminum intake (mg/person) is recorded for temperatures (°C) of 23, 70, 90, 100 and 185 (pan; pan area = 552.7 cm^2) to be 20.15, 40.3, 60.45, 100.75 and 333, respectively.

The rate of aluminum leaching to water and food solutions depends on the temperature. As the temperature increases, the molecules move faster and therefore aluminum contaminate food more frequently. It is reported that the aluminum leaching is dramatically higher at 100° C than at ambient temperature [13]. It is also reported that cooking temperature is more important in aluminum leaching than cooking time [15]. The only way to explain the relationship between temperature and the rate of corrosion is to assume that the rate of corrosion depends on the temperature at which the leaching is running.

The temperature dependence of the corrosion rate (C.R.) could be correlated by Svante Arrhenius equation:

$$C.R. = A e^{-Ea/RT}$$

A is the pre-exponential factor or frequency factor

E_a is the activation energy in J/mole

R is the gas constant = 8.314 J/mole K

T is the absolute temperature

The energy of activation and the frequency factor can be found from an Arrhenius plot of Log corrosion rate against 1/T as shown in Figure (9). The activation energy for the corrosion rate (leaching process) is equal to 26.407 KJ /mole. This can be concluded from the following explanation.



Figure 9. Activation energy measurement by applying Arrenhius equation.

The effect of temperature on the Indian aluminum sample is studied using drinking water after two hours of exposure (Figure (10)). The corrosion rates at temperatures below 100 $^{\circ}$ C are less than that in drinking water, while at 100 $^{\circ}$ C the corrosion rate is three times more than that in drinking water.

The aluminum intake (mg/person) is recorded for temperatures (°C) of 23, 70, 90 and 100 to be 20.15, 20.15, 40.3 and 301.93, respectively.



Figure 10. Effect of temperature on the corrosion rate of the Indian sample after two hours exposure to tap water.

The temperature dependence of the corrosion rate (C.R.) using Arrhenius equation correlation showed that the activation energy is approximately equal to 86.296 KJ/mole which is higher than the value of drinking water by a factor of 3.3. This factor is also approximately equated by considering the aluminum intake values.

From this equation, it can be seen that increasing the temperature from 70 $^{\circ}$ C to 100 $^{\circ}$ C caused an increase in corrosion rate by more than ten times.

3.3. Effect of pH

The effect of the pH on the leaching behavior of the Egyptian cookware in salt solution (3.5 weight percent of NaCl in tap water) at room temperature, after six days of exposure, is reported in Table (3).

Table (3) shows that the corrosion rate is high at low pH, it decreases after to a minimum pH of 6.4 then increases sharply to very high values at pH of 8 and 10. Similar behavior is reported by Wong et al [16]. It is also reported that aluminum is very sensitive to high pH and shows a corrosion increase

in alkaline environments. This result is almost similar to previous work [14]; also, Shuping Bi indicated that leaching enhanced dramatically in the ranges of pH < 4 or pH > 8 [7].

Table 3. Effect of pH value on the corrosion rate of the Egyptian sample after six days of exposure to3.5% NaCl solution in tap water at room temperature

Initial pH Value	Final pH Value	Weight Loss (mg)	Corrosion rate (mg.cm ⁻² .hr ⁻¹)
1.2	1.7	20	0.0580
2.8	2.9	9.6	0.028
6.4 (native)	7.8	5	0.0146
8.2	7.6	37	0.1078
10	8.5	60	0.174

Figures (11) and (12) show localized corrosion with some uniform corrosion occurring at low pH of 1.2 and 2.8, respectively. Figure (13) shows that almost uniform corrosion is occurring with little localized corrosion at high pH of 10.



Figure 11. ESEM of the Egyptian sample after six days of exposure to 3.5% NaCl solution in tap water (pH 1.2) at room temperature (20 mg).



Figure 12. ESEM of the Egyptian sample after six days of exposure to 3.5% NaCl solution in tap water (pH 2.8) at room temperature (9.6 mg).



Figure 13. ESEM of the Egyptian sample after six days of exposure to 3.5% NaCl solution in tap water (pH 10) at room temperature (60 mg).

Figure (14) resembles the corrosion rate of Egyptian samples in 3.5 wt. % of NaCl in drinking water at room temperature after two weeks exposure, but with huge reduction in the corrosion rate compared to the first case. At pH of 2, 2.4, and 3.1 the localized attack is clearly noticed in Figures (15), (16), and (17), respectively. While at pH of 6.7, there is very low uniform corrosion rate (Figure (18)). Increasing the pH to 10 shows very severe local attack (Figure (19)); pitting is filling the whole surface of the sample. It is previously reported that aluminum usually develops a protective surface of oxide film upon exposure to the atmosphere or to aqueous solutions [17]. This film is responsible for the corrosion resistance of aluminum in most environments when aluminum is exposed to high concentration of acids or bases. This solution causes pitting corrosion to the aluminum in the presence of chloride ions.



Figure 14. Effect of pH on the corrosion rate of the Egyptian sample after two weeks of exposure to 3.5% NaCl solution in drinking water at room temperature (solid line for mg/cm².hr x 10⁻², and dotted line for mm/yr).



Figures 15. ESEM of the Egyptian sample after 14 days of exposure to 3.5% NaCl solution in drinking water (pH 2) at room temperature (8.4 mg).



Figure 16. ESEM of the Egyptian sample after 14 days of exposure to 3.5% NaCl solution in drinking water (pH 2.4) at room temperature (7 mg).



Figure 17. ESEM of the Egyptian sample after 14 days of exposure to 3.5% NaCl solution in drinking water (pH 3.1) at room temperature (4.4 mg).



Figure 18. ESEM of the Egyptian sample after 14 days of exposure to 3.5% NaCl solution in drinking water (pH 6.7) at room temperature (0.2 mg).



Figures 19. ESEM of the Egyptian sample after 14 days of exposure to 3.5% NaCl solution in drinking water (pH 10) at room temperature (0.3 mg).



Figure 20. ESEM of the Egyptian sample after 14 days of exposure to 3.5% NaCl solution in drinking water (pH 11) at room temperature (10 mg).

The leaching in the same solution but at pH of 11 is higher than at any other pH. The mode of reaction is a uniform corrosion; the grains and the grain boundaries are clearly shown in Figure (24). The leaching is removing all the oxides from the surface. The above finding is in agreement with previous works [7, 12-13].

3.4. Effect of salinity

Sea water is usually more corrosive than fresh water; this is related to the conductivity and the penetrating power of the chloride ions through the surface. Egyptian cookware sample in various NaCl solutions using tap water is studied for six days. This shows that the corrosion rate is increasing with the concentration of NaCl; maximum value is at 2 wt. % of NaCl. After that, the corrosion rate decreases to a constant value in 2.5% to 3.5%. The same behavior is reported for steel in various NaCl solutions, but with a maximum corrosion rate in 2.7 wt. % of NaCl [13]. This is attributed to the combination of high conductivity and the oxygen solubility to be at a maximum at this point. Solubility of oxygen is reduced with increasing salt concentration; this is why the corrosion reduces as the concentration increases.

Figure (21) shows the same behavior of the Egyptian samples in various NaCl solutions using drinking water after 14 days exposure with the peak at 1.5 wt. % of NaCl.



Figure 21. Effect of NaCl concentration on corrosion rate of the Egyptian sample (solid line for mg/cm².hr x 10⁻², and dotted line for mm/yr).

Figure (22A) indicates that the corrosion mode in 1.5 wt. % of NaCl is uniform, the grains and the grains boundaries are clearly shown in the ESEM micrograph. Also the big weight loss of 20 mg explains this behavior, while it is only 0.2 mg in the 2.5 wt. % of NaCl solution. Figure (22B) clearly shows the pitting on the surface after the exposure to 2.5 wt. % of NaCl solution.





Figure 22. ESEM of the Egyptian sample after 14 days of exposure to 1.5% NaCl (20 mg) (A) and to 2.5% NaCl (B) solution in drinking water at room temperature (0.2 mg).

4. CONCLUSIONS

The amount of aluminum leaching depends on pH, salinity, temperature and time of exposure as well as present ions in the medium. Increased ion concentration significantly increases aluminum leaching since the complexing effect plays an important role which is studied by using tap water and an ion-free medium, drinking water. Increasing salinity of the medium increases aluminum leaching for both Indian and Egyptian samples under investigation with up to an effect of 6 times more than at salt-free conditions. The aluminum intake per person increases significantly at higher cooking temperatures. The obtained results indicate the necessity of aluminum leaching societal awareness and call for undertaking necessary precautions either by aluminum cookware replacement or by the cheaper option of aluminum passivation.

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