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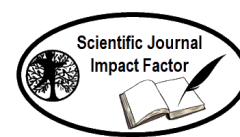
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Study of Elements Released from Various Cooking Utensil After Heating on Cooking Utensil of Aluminum, Stainless Steel, Titanium-coated Stainless Steel and Teflon and Their Potential Health Hazards

Manogari Sianturi^{1*}, Fajar L. Gultom¹, Faradiba^{3*}, Patricya V. Heumasse⁴, Faris Febriza⁵

Universitas Kristen Indonesia



Abstract – Alloy products is widely used as raw material in daily life, one of which used as material for cooking utensils. High thermal conductivity, very high resistance to corrosion, high stability, biocompatibility, low specific weight, very low toxicity, heat resistance and affordable prices are public references in choosing raw materials for cooking utensils. The migration of metal elements from cooking utensil materials into food can cause potential health hazards to humans. This study investigated metal release from the cooking utensils material into a solution of water and sodium bicarbonate by cooking the solution in four different types of cooking pots. Before determining the metal elements that were released from the alloys of the cooking utensils material, the composing of the cooking utensils were examined quantitatively by X-Ray Fluorescence (XRF) and Scanning Electron Microscopy- Energy Dispersive X-ray (SEM-EDX). The elements released from the material in contact with the solution detected by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES) were Al, Si, Ca, Fe, Cr, Ni, Ti, Mn, Mg and Na elements. The highest to smallest elemental concentrations detected in the solution of the cooking utensil material studied were elements of Na, Si, Mg, Al, Ca, Fe, Ni, Mn, Ti and Cr with concentrations of 2400.4900, 52.02610, 4.90241, 1.64646, 1.57894, 0.106696, 0.02521, 0.02146, 0.008743 and 0.00635 levels/ppm of each element from the various cooking utensil materials studied.

Keywords – Alloy; cooking utensils; health hazards; XRF; SEM-EDX; ICP-OES.

I. INTRODUCTION

Alloys are widely used for the production of food contact materials for cooking at high temperatures, because of their high thermal conductivity and thermostability (1). The majority of metal kitchen utensils consist of an alloy and not a single material. A typical example of an alloy is stainless steel, which consists of an alloy of iron (Fe) with nickel (Ni) and chrome (Cr). Stainless steel does not rust and exhibits good strength and thermal conductivity. The most widely used metals include aluminum (Al), iron (Fe), tin (Sn), copper (Cu), nickel (Ni), Manganese (Mn), and chromium (Cr).

Apart from these elements, other groups of toxic elements, including arsenic (As), lead (Pb), and cadmium (Cd), that can be included as contaminants or impurities in the manufacturing process (2). Brass and bronze generally use a copper (Cu) alloy consisting of an alloy of Cu with zinc (Zn) and tin (Sn). Al elements can be combined with various elements such as Mg, Cu, and Zn. Al alloys are commonly used for the production of cans and various types of food and beverage packaging. Other kitchen utensils that are often used for cooking are non-stick coatings or Teflon.

Metals in food have a dual function because they are essential nutritional elements but can also be toxic when present at high concentrations. The majority of metals have a significant effect on human health. Various metals such as copper, iron, manganese, zinc, calcium, magnesium, potassium and sodium are essential elements. Although small levels of some metals, such as Cr, are very important for vital functions in the human body, excessive intake of these metals can cause health problems (3). Therefore, the release of toxic metals such as Ni, Cd, and Pb from food contact materials into food is a major public health problem. Recent studies have reported that the toxic metals Al, Fe, and Pb are still found in fufu foods and fried rice (4). In addition, Ni predominantly affects the kidneys through oral exposure (5). Such metals can migrate into food from metal kitchen utensils used to package, cook or store food (6). Migrating elements Al, boron (B), barium (Ba), Co, Cr, Cu, Fe, Li, Mg, Mn, Ni, antimony (Sb), tin (Sn), strontium (Sr), titanium (Ti), vanadium (V), zinc (Zn) and zirconium (Zr) from ceramic utensils into food. According to the classification proposed by the International Agency for Research on Cancer (IARC), these elements are cause organ toxicity and also carcinogenic (7).

Cooking utensils can be the main source of total daily Al intake depending on temperature, time and pH of food (8). The release of the Cr element from the cooking utensil material into the solution depends on the pH of the solution used (9). Previous research has mainly focused on the release of alloying elements from stainless steel or Al products (10), different Cu and cast iron alloys (11), into foods with the influence of PH, acidic properties and solution temperature. However, very few studies have focused on the release of metals from food contact materials with various types of solution-based cookware that are actually used in everyday cooking, in this study a solution of gallon water and sodium bicarbonate salt was used. The Cr, Ni, and Mn release studies of six types of stainless steel reported that there are differences in the amount of metal released based on the type of stainless steel (12).

The food safety authority of a country has regulated the release of metals from cookware contact material into food because the excessive release of these metals can threaten human health. According to Korean regulations administered by the Ministry of Food and Drug Safety, Ministry of Food and Drug Safety (MFDS), the release levels of both Ni and Cr (VI) from food contact material into food must be less than 0.1 mg / L (level / ppm), each of these metals. In Europe, specific release limits for various metals and metalloids were

adopted from the Council of Europe (CoE) in 2013, these limits stipulate that the release rates of both Cr and Ni metals must be less than 0.25 and 0.14 mg / kg respectively.

II. OBJECTIVE

The aim of this research is to study the type of release of elements and their levels from alloy cookware, aluminum, stainless steel, titanium-coated stainless steel and non-stick coating or Teflon into the solution by heating sodium bicarbonate solution in four different types of cooking utensil materials. and its potential harm to human health.

III. MATERIALS AND METHODS

The test for the release of elements from the four types of cooking utensils studied was the cooking utensils commonly used by the Indonesian people were made of alloy, aluminum, stainless steel, titanium coated stainless steel and Teflon. The salt solution used in this study was a mixture of 250 ml of gallon water and 3 grams of NaHCO₃ in crystal form, as a substitute for table salt because it dissolves easily in water, used as an acid neutralizer and often used as a physiological pH buffer. The mixture of 250 ml gallon water and 3 grams of NaHCO₃ was stirred so that it becomes a homogeneous solution in four 500 ml beaker glasses, then this solution was put into four different types of cooking pots. Before the solution was boiled, 4 empty bottles were prepared and cleaned with detergent and then rinsed with clean water from tap water, then dried with a hair drier so that the bottles were clean and do not contain any impurities. The salt solution was heated using 4 similar cooking stoves which are considered to have the same heat intensity so that the solution boils (100 °C) in 5 minutes 45 seconds. The salt solution was heated using 4 similar cooking stoves which are considered to have the same heat intensity so that the solution boils (100 °C) in 5 minutes 45 seconds for each cooking utensil. After the solution boils, the stove was immediately turned off then the solution was put into a bottle that had been prepared beforehand, then cooled until the solution temperature reaches room temperature (27 °C), closed and sealed. Bottles were marked A, B, C and D on each bottle containing a solution from alloy of aluminum, stainless steel, titanium-coated stainless steel and Teflon. Finally, testing was done to the elements contained in each cooking utensil solution with begins testing to the elements forming of each raw material cooking utensil. Initial testing was required of all the elements forming of each raw material cooking utensil so that it could be predicted which elements will be released from each the material cooking utensil into the solution.

IV. INSTRUMENT

Quantitative examination of the raw material composition of each cooking utensil by using X-Ray Fluorescence (XRF). In addition to using XRF, further examination of the atomic composition of the raw material for cooking utensils was carried out with Energy Dispersive X-ray (EDX) and the surface morphology of the samples was observed by Scanning Electron Microscopy (SEM). Elements and concentrations released from each cooking utensil were measured by Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES).

In addition to using XRF, further examination of the atomic composition of the raw material for cooking utensils was carried out with Energy Dispersive X-ray (EDX). and the surface morphology of the samples was observed by

Scanning Electron Microscopy (SEM). Element and concentration of elements released from cookware materials (Al, Si, Ca, Fe, Cr, Ni, Ti, Mn, Mg and Na) were measured using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES).

V. RESULTS

Table 1 shows the test results of the concentration (%) of the elements of raw material for cooking tools sampled in this study using XRF, where XRF itself is a non-destructive analysis technique used to identify the elements and their concentrations that form a raw material in form of solid, powder, or liquid preparations.

Table 1. Quantitative examination results for composition of raw material cooking utensils using XRF

Element	Cooking utensils			
	A	B	C	D
	Concentration (%)	Concentration (%)	Concentration (%)	Concentration (%)
Al	98.96	0.74	0.98	28.15
Si	0.41	1.01	0.97	21.79
S	ND	ND	ND	9.07
Cl	ND	ND	ND	10.38
Ca	ND	ND	ND	1.41
Fe	0.33	85.37	67.99	4.94
Cr	0.19	12.89	17.13	ND
Ni	ND	ND	10.18	ND
Ti	ND	ND	ND	18.09
Mn	0.11	ND	1.03	6.17
Mo	ND	ND	1.73	ND
Total	100	100	100	100

ND : means below the detection limit, bold means the highest element concentration

Furthermore, Table 2 and Figure 1 show the results of quantitative examination of the concentration (%) and the spectrum of the atoms forming the cooking utensil material using SEM-EDX. Figure 2 shows the results of the microstructure examination of the inner surface of each

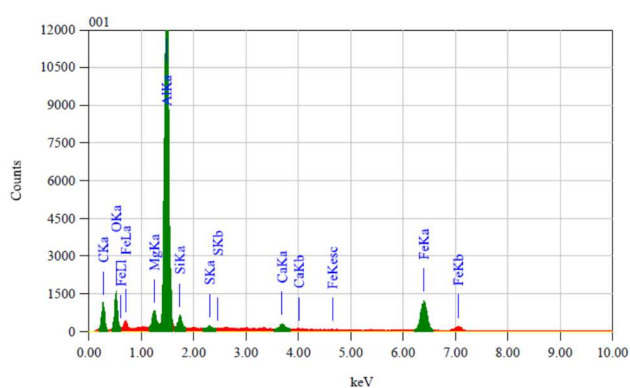
cooking utensils using SEM. Table 3 shows the results of examining the metal elements released from each raw material of the cooking utensils into a bicarbonate salt solution that has been heated to 100 °C using ICP-OES.

Table 2. Quantitative examination results of the raw material cooking utensils composition using SEM-EDX

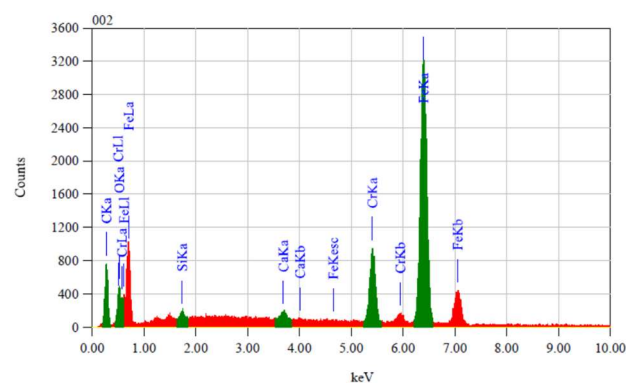
Element	Cooking utensils pots			
	A	B	C	D
	Atom (%)	Atom (%)	Atom (%)	Atom (%)
Al	28.47	ND	2.16	ND
Si	0.87	0.52	ND	2.10
S	0.19	ND	ND	ND

Cl	ND	ND	ND	ND
Ca	0.37	0.57	ND	ND
Fe	4.57	35.65	63.06	ND
Cr	ND	5.94	14.11	ND
Ni	ND	ND	ND	ND
Mg	12.25	ND	ND	2.15
C	42.52	47.08	6.6	31.24
O	20.76	10.24	ND	16.21
F	ND	ND	13.28	48.29
Ta	ND	ND	0.79	ND
Total	100	100	100	100

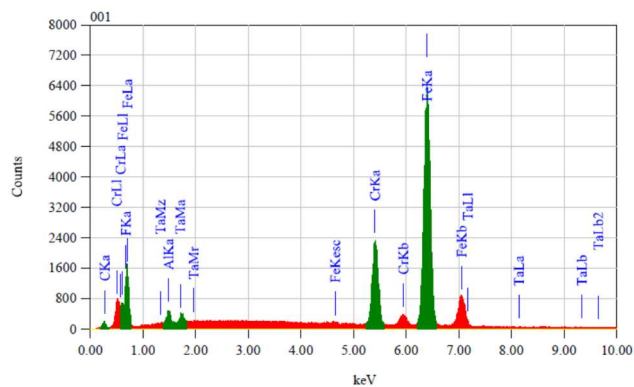
ND : means below the detection limit, bold means the highest element concentration



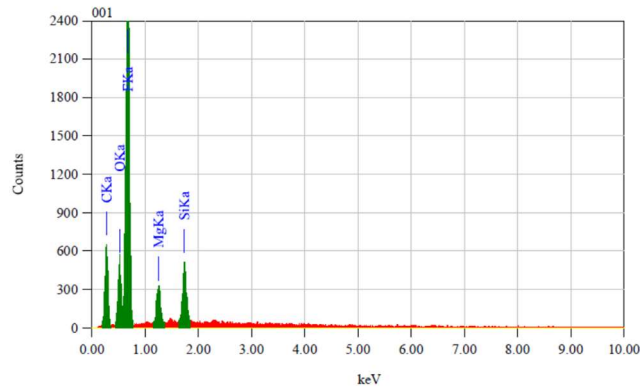
A



B



C



D

Figure 1. Spectrum graph of the composition atoms of the raw materials alloy for various cooking utensils A. Aluminum, B. Stainless steel, C. Titanium coated stainless steel and D. Teflon.

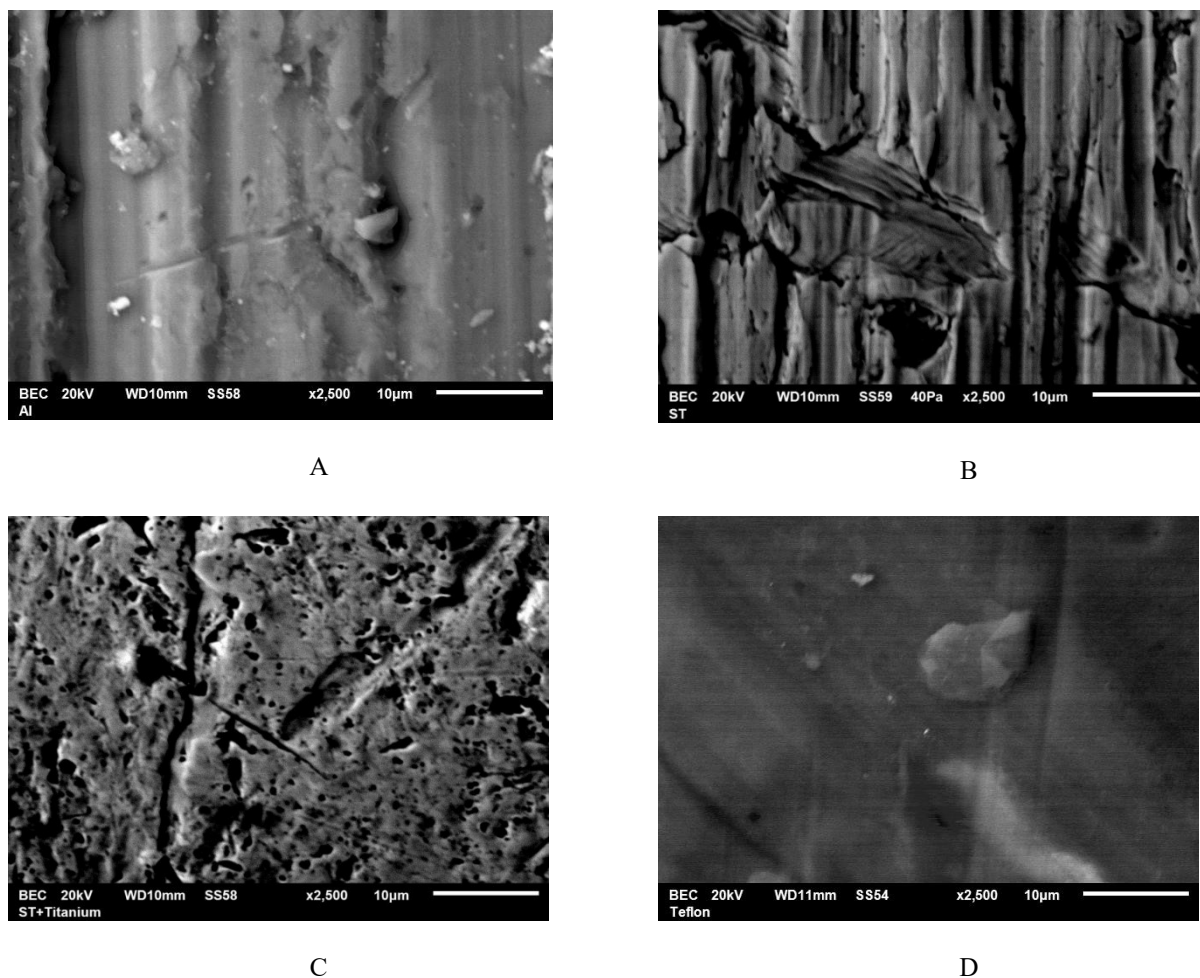


Figure 2. SEM image of the surface microstructure inner of various cooking utensils A. Aluminum, B. Stainless steel, C. Titanium coated stainless steel and D. Teflon.

Table 3. Quantitative examination results of the raw material cooking utensils composition using ICP-OES

Element	Cooking utensils			
	A	B	C	D
	Levels/ppm	Levels/ppm	Levels/ppm	Levels/ppm
Al	1.64646	0.034288	0.068818	0.010112
Si	48.1776	37.5952	41.4391	52.0261
Ca	1.11818	1.39875	1.5509	1.57894
Fe	0.088518	0.057327	0.106696	0.071594
Cr	0.000813	0.00095	0.00635	0.002507
Ni	0.02521	0.021318	0.0015	0.020469
Ti	0.008743	0.002485	0.006466	0.004447
Mn	0.02146	0.003032	0.005795	0.006559
Mg	4.22183	3.68106	3.44527	4.90241
Na	2064.88	1616.71	1823.24	2400.49

Bold means the highest element concentration

VI. DISCUSSION

Tables 1 and 2 show that the alloy elements composition of the cooking utensil studied consists of the elements Al, Si, S, Cl, Ca, Fe, Cr, Ni, Ti, Mn, and Mo. The results of this examination are almost the same as the information from AISI 304 (UNS S30400, EN 1.4301), AISI: American Iron and Steel Institute; UNS: Unified Numbering System; EN: European Standard, that generally the alloy elements composition of cooking utensils often used consist of Fe, Cr, Ni, Mo, Mn, C, S, Si, Cu, Co, P and N (9).

The examining ICP-OES results to the salt solution show that the elements found of salt solution from each cooking utensils were Al, Si, Ca, Fe, Cr, Ni, Ti, Mn, Mg and Na as shown in table 3. The order of the elements from the highest to the smallest elemental concentration in the salt solution

was Na, Si, Mg, Al, Ca, Fe, Ni, Mn, Ti and Cr as shown in table 4. The study revealed that the solution from Teflon and aluminum cooking utensils found to release the most types of elements with the highest concentration of all elements found in the solution among the cooking utensils studied by examination of ICP-OES. Elements of Na, Si, Mg and Ca with level concentrations of 2400.49, 52.0261, 4.90241 and 1.57894 (levels/ppm) that found from Teflon pot salt solution and elements Al, Ni, Mn and Ti with level concentration of 1.64646, 0.02521, 0.02146 and 0.008743 (levels/ppm) that found from aluminum pot salt solution, element respectively with the highest concentration. Titanium coated stainless steel was a cooking utensils that releases Fe and Cr elements with the highest concentration in the salt solution of 0.106696 and 0.00635 (levels/ppm) element respectively.

Table 4. The order of the elements based on the concentration level of examining results on the salt solution using ICP-OES

Element	Cooking utensils	Concentration (level/ ppm)
Na	D	2400.4900
Si	D	52.02610
Mg	D	4,90241
Al	A	1.64646
Ca	D	1.57894
Fe	C	0.106696
Ni	A	0.02521
Mn	A	0.02146
Ti	A	0.008743
Cr	C	0.00635

The element Na is the element with the highest concentration and is much higher than other elements found in all salt solutions that come from cooking utensils studied, this happens because 3 grams of bicarbonate (NaHCO_3) were added to the solution. All salt solution from the cooking utensils that studied, it was found that the highest concentration of Na elements came from salt solution of the alloy Teflon cooking utensil. This shows that the element Na was the most unstable in the alloy Teflon cooking utensil compared to another cooking utensil when it in contact with a heated salt solution.

Based on the examining results of the composition of the elements forming the Teflon cooking utensils showed that the Si element had a concentration of 21.79% and 2.10 atoms (%) by examination using XRF and using SEM-EDX respectively as shown in Tables 1 and 2. Si elements were found in the highest levels as an element for the formation of Teflon material among the cooking utensil materials studied

using both XRF and SEM-EDX examinations. Therefore, when the Si element was found in the salt solution from the Teflon material in the highest levels, this shows that it released from the material alloy Teflon into the salt solution in the highest levels when the cooking utensils material was heated up to 100 °C. This shows that the Si element in material alloy Teflon more easily released compared to other material alloy cooking utensils.

Based on the results of examining SEM-EDX, shows that the Mg element was found as the composition of the elements forming the alloy material of aluminum and Teflon cooking utensil with atomic percentages of 12.25 and 2.15 atoms (%), respectively, in while in cooking utensils other, Mg element not detected as the composition of the forming elements in the raw material cooking utensil, as shown in Table 2. Based on the results of the ICP-OES examination, it showed that the Mg element was found in the salt solution from material alloy of aluminum and Teflon cooking utensil, with levels of

4.22183 and 4.90241(levels/ppm) respectively, as shown in Table 3. The highest levels of Mg elements were found from the salt solution that came from the Teflon cooking utensil, on the other hand, the highest Mg atomic content was found as the element composition forming of the alloy raw material aluminum whose levels were nearly 6 times level of in the Teflon cooking utensil alloy forming material. This phenomenon indicates that the Mg element was more reactive and migrates effortlessly from the Teflon alloy material into the salt solution compared to the material alloy aluminum when cooking utensil heated. Another phenomenon shows that solution from stainless steel and titanium coated stainless steel, Mg elements were found with a concentration of 3.68106 and 3.44527 (levels/ppm) even though these elements were not detected as elements composition of alloys of stainless steel and titanium coated stainless steel as shown in tables 1 and 2. This phenomenon shows that it was likely that the water used in the salt solution contains element Mg. This result appropriate with previous studies that water originating from Indonesia, especially Jakarta, contains element Mg (13).

Al element was found as an element composition of materials alloy aluminum, stainless steel, titanium coated stainless steel and Teflon cooking utensil with concentrations of 98.96, 0.74, 0.98 and 28.15 (%) for each cooking utensil by using XRF as show in Table1. Based on the examination of ICP-OES that Al element released of cooking utensil into salt solution of each cooking utensil material, it was found that the highest concentration of Al elements was found in the solution from the aluminum cooking utensil of 1.64646 (levels/ppm) as shown in Table 3. Based on results of the examination reveal that the Al element content was found less from the salt solution of the Teflon cooking utensil than from salt solution both of stainless steel and titanium-coated stainless steel cooking utensil, although the Al elemental composition content of the Teflon cooking utensil has a higher Al elemental composition in the stainless steel and titanium coated stainless steel cooking utensil. This phenomenon proves that the Al element was more stable or inert in material Teflon cooking utensil compared to materials stainless steel and titanium coated stainless steel cooking utensil when material cooking utensil was heated. The Al element found in all the solution which came from all the alloy materials of the cooking utensils studied proves that the Al element in the cookware pans produced locally has dissolved into a heated salt solution, the result of this study is appropriate with previous research (14).

Through examination using XRF, shows that the Ca element was found as element composition of material alloy Teflon cooking utensil only, with a concentration of 1.41 (%) as shown in Table 1. While examination using SEM-EDS, Ca element was found in both materials alloy aluminum and stainless-steel cooking utensil with concentrations of 0.37 and 0.57 atoms (%) respectively. Based on the examination of the salt solution using ICP-OES, it shows that the highest amount of Ca was found in the solution that came from material alloy Teflon cooking utensil of 1.57894 (levels/ppm). Therefore, of Ca element was more easily released of the material alloy Teflon cooking utensil compared to other material alloy cooking utensils.

Fe element was found as an element composition of all the raw materials of cooking utensils studied. This element was found with the first highest levels was in raw material alloy stainless steel and second one in raw material alloy titanium-coated stainless steel of 85, 37 and 67.99% respectively by examination using XRF as shown in Table 1. The highest levels Fe element released from material alloy titanium coated stainless steel cooking utensil of 0.10670 (levels/ ppm) by examination using ICP-OES. The results of examination revealed that the Fe element was more easily released from material alloy titanium-coated stainless steel into salt solution than material alloy stainless steel cooking utensil when those cooking utensil heated.

Based on the results of the examination using ICP-OES shows that the types of elements Ni, Mn and Ti with the highest concentrations found in the salt solution came from alloy aluminum cooking utensils as shown in Table 4. Although each elements of Ni and Ti could only be detected in both material alloy titanium coated stainless steel and Teflon cooking utensil, but both the elements were detected in the highest concentration in the salt solution from aluminum cookware of 0.02521 and 0.008743 levels/ppm of element respectively. This phenomenon reveals that it was likely that Ni and Ti elements were actually present in metal and aluminum alloy materials, but their levels are below detection limit of XRF and SEM EDS used. Both Ni and Ti elements in material alloy aluminum cooking utensil easily release from this raw material and enter into salt solution when cooking utensil heated, on the other hand, both Ni and Ti elements are considered very inert in both material alloy titanium coated stainless steel and Teflon cooking utensil. The Mn element was found as an forming element composition of all the raw materials of cooking utensils except for stainless steel which is this element was not found either through XRF or SEM-EDX examinations. The

concentration of the element Mn was found to be the highest as the composition of the elements forming of material alloy Teflon cooking utensil, but this element was found in the highest levels in the salt solution came from material alloy aluminum cooking utensil of 0.02146 levels/ppm, this reveals that the Mn element was very easily migrate from material alloy aluminum cooking utensil and enter into salt solution, otherwise this element was very inert in the material alloy Teflon cooking utensil.

Based on the results of XRF and SEM-EDX examinations of all material alloy cooking utensil that studied, it found that it in the highest levels of Cr element as the composition of the forming elements in material alloy titanium coated stainless steel cooking utensil compared to other cooking utensil of 17.13% by using XRF. This result was in line with the results of the ICP-OES examination, that the highest levels of Cr element found came from the salt solution from material alloy titanium coated stainless steel cooking utensil of 0.00635 levels/ppm. This phenomenon proves that the Cr element was most easily released from material alloy titanium coated stainless steel cooking utensil and enter into salt solution when this cooking utensil was heated.

Potential Health Effects of Release of Elements Raw Material Alloy Cooking Utensil

Various studies have been conducted to determine the effects of exposure of metallic elements on human target cells. The impact of chromate and nickel elements on human target cells has been widely carried out, one of which is through human B lymphoblastoid cells exposed using potassium dichromate ($K_2Cr_2O_7$) and nickel chloride ($NiCl_2$). The effect of occurs when B lymphoblastoid cells are exposed to $K_2Cr_2O_7$ and $NiCl_2$ results in a cytotoxic effect of human B lymphoblastoid cells in the form of decreased cell viability that depends on concentration and exposure time of the compounds (Jianlin Lou et al., 2013). In addition, cytotoxic effects can also occur as a result of the synergistic combination of the two compounds. The combined effect of chromium and nickel on cell viability is complex, and the most likely mechanism is the activation of HSPs (especially HSP27 and HSP70) and NF κ B which suppress signaling suppression and lead to cell survival. Meanwhile, synergistic cytotoxic effects are characterized by the production of oxidative stress and inhibition of HSPs and NF κ B, PARP-1 expression during activation and ATP depletion. The attenuating effect of nickel on the cytotoxicity of $K_2Cr_2O_7$ allows damaged cells to survive, but leads to abnormal cell accumulation, a similar result has also been reported (15), and occurs in human liver carcinoma cells (HepG2) (16).

Although the mutagenic potential of nickel compounds assessed in many mutation systems, single-strand DNA damage caused by Ni compounds, including nickel chloride, has been reported in human lymphocytes (17,18). Other researchers have also revealed that heavy metal contamination often occurs together with several metal ions. The results indicated that significant DNA oxidative damage is observed in workers exposed to chromate. Collectively, the results show workers who have worked in chromate factories simultaneously exposed to chromate have an interactive effect on workers' DNA damage (19). The human health effects associated with exposure to silicones in the form of silica, particularly crystalline silica (0.5-10 μ m), have been extensively studied. Exposure to crystalline silica to humans induces silicosis (fibrotic lung disease) and is also associated with lung cancer, emphysema, and pulmonary tuberculosis (20). The health effects on the human body caused by magnesium levels exceeding normal limits, can be said to be a state of hypermagnesia. Hypermagnesia itself can cause several disorders such as abnormalities of muscular contraction, sugar and insulin metabolism (21).

Excess intake of the metallic element Fe plays a major role in pathological events such as iron oxide deposition in Parkinson's disease (22). Apart from aiding neurological deposition, these active redox metals promote premature aging and cause oxidative damage, a key component of chronic inflammatory disease and cancer triggers (23). Heavy metals such as the cause of mental retardation, kidney failure, liver damage, hearing loss and shortened pregnancy in humans (24).

VII. CONCLUSION

The composition of the element raw material alloy of cooking utensils studied consists of elements Al, Si, Ca, Fe, Cr, Ni, Ti, Mn, S, Cl and Mo. The elements released from the raw material of the cooking utensil and enter into the salt solution which can be detected consist of the elements Al, Si, Ca, Fe, Cr, Ni, Ti, Mn, Mg and Na. The aluminum cooking utensil released the highest levels of Al, Ni, Mn and Ti elements among the cookware studied. The Fe and Cr elements released with the highest levels occurred in raw material alloy titanium coated stainless steel cooking utensil. Previous studies revealed that exposure of metals such as Fe, Si, Ni and Cr to human target cells had the potential to cause health hazards in humans.

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