Gakkaishi **1973**, *39*, 1315–1324.

Obatake, A.; Heya, H. "A Rapid Method to Measure Dark Muscle Content in Fish". *Nippon Suisan Gakkaishi* **1985**, *51*, 1001–1004.

Obatake, A.; Tsumiyama, S.; Yamamoto, Y. "Extractive Nitrogenous Constituents from the Dark Muscle of Fish". *Nippon Suisan Gakkaishi* **1985**, *51*, 1461–1468.

Raffin, J. P.; Leray, C. "Comparative Study on AMP Deaminase in Gill, Muscle and Blood of Fish". *Comp. Biochem. Physiol., B: Comp. Biochem.* **1980**, *67B*, 533–540.

Saito, T.; Arai, K.; Matsuyoshi, M. "A New Method for Estimation of the Freshness of Fish". *Nippon Suisan Gakkaishi* **1959**, *24*, 749–750.

Sakaguchi, M.; Murata, M. "Distribution of Free Amino Acids, Creatine, and Trimethylamine Oxide in Mackerel and Yellowtail". *Nippon Suisan Gakkaishi* **1986**, *52*, 685–689.

Sakaguchi, M.; Murata, M.; Kawai, A. "Changes in Free Amino Acids and Creatine Contents in Yellowtail (*Seriola quinqueradiat*) Muscle during Ice Storage". *J. Food. Sci.* **1982**, *47*, 1662–1666.

Sakaguchi, M.; Murata, M.; Kawai, A. "Changes in Free Amino Acid Contents in Juvenile Mackerel *Scomber Japonicus* Muscle during Ice Storage". *Nippon Suisan Gakkaishi* **1984**, *50*, 323–329.

Shewan, J. M. "The Bacteriology of Fresh and Spoiling Fish and Some Related Chemical Changes". *Rec. Adv. Food Sci.* **1962**, *1*, 167–193.

Simidu, U.; Terashima, H.; Simidu, W. "Studies on Muscle of Aquatic Animals 34: Changes in Muscle of Yellowtail (*Seriola quinqueradiata*) during Cold Storage". *Nippon Suisan Gakkaishi* **1958**, *24*, 586–590.

Uchiyama, H.; Ehira, S. "The Current Studies on the Freshness of Fish with Special Reference to Nucleic Acids and Their Related Compounds". *Nippon Suisan Gakkaishi* **1970**, *36*, 977–992.

Yamada, K.; Higashino, S.; Kawahara, T.; Ito, R. "Effect of Contamination of Dermis on the Estimation of K Value, an Index of Freshness, of Sardine Muscle". *Nippon Suisan Gakkaishi* **1981**, *47*, 631–636.

Yamaguchi, S. "Fundamental Properties of Umami in Human Taste Sensation" in *Umami: A Basic Taste*; Kawamura, Y., Kare, M. R., Eds.; Marcel Dekker: New York, 1987.

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Aluminum Levels in Food-Simulating Solvents and Various Foods Cooked in Aluminum Pans

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Release of aluminum from aluminum pans into food-simulating solvents with water, 4% acetic acid, and 0.5% sodium bicarbonate solution and into neutral, acidic, and alkaline foods was determined. Little aluminum was released into water with standing in nontreated and surface-treated aluminum pans at room temperature (20 ± 3 °C) for 24 h, boiling for 10 min, or boiling for 2 h, but the release of aluminum into acidic and alkaline solvents increased to 0.58–2.9 μg/mL with standing at room temperature for 24 h, 41–167 μg/mL with boiling for 10 min, and 161–501 μg/mL with boiling for 2 h. The aluminum concentrations of Chinese noodles (pH 8.9) and tomato juice (pH 4.2), whose aluminum contents were 1.1 ± 0.2 and 8.0 ± 0.8 μg/g, increased to 4.9 ± 0.2 and 11.6 ± 1.0 μg/g, respectively, with boiling in a nontreated aluminum pan for 10 min, whereas aluminum content of Japanese noodles (pH 5.5) remained at 1.1 ± 0.3 μg/g, showing no increase. The intake of one pack of Chinese noodles was estimated to cause ingestion of 3.3 mg of aluminum, including 2.6 mg of aluminum released from the aluminum pan.

Aluminum cooking utensils such as pans, pots, kettles, trays, and foil are widely used in homes, restaurants, cafeterias, and food industries. During recent decades, however, medical researchers have reportd that aluminum is suspected in etiology of osteomalacia (Bloom and Flinchum, 1960), dialysis encephalopathy (Parkinson et al., 1981; Alfrey et al. 1980), and Alzheimer's disease (Crapper et al., 1973; Perl and Brody, 1980). From a toxicological point of view, aluminum content in foods (Greger, 1985; Greger et al., 1985; Katsumura et al., 1973), environmental sources (Lione, 1983), and daily intake (Underwood, 1977; Greger, 1985) were studied, and the use of aluminum utensils for cooking has been discussed (Trapp et al., 1981; Lione, 1983).

We reported the release of copper and tin from copper or tin-plated copper utensils (Ishiwata et al., 1986) and lead and cadmium from experimentally produced enamelware glazed with lead- or cadmium-based color (Ishiwata et al., 1984). In the present study, the release of aluminum from nontreated and surface-treated aluminum pans with food-simulating solvents and with certain foods was examined.

EXPERIMENTAL SECTION

Materials. Nontreated aluminum pans whose inner diameter and depth were 15.0 and 9.0 cm and inner surface area and volume were 600 cm^2 and 1.6 L, respectively, and surface-treated aluminum pans whose inner diameter and depth were 15.5 and 5.9 cm and inner surface area and volume were 476 cm^2 and 1.1 L, respectively, were used. The pans were cleaned with a detergent and rinsed with water before use.

Equipment. Homogenizer: Waring-Gallon CB-6, Waring Products, New Hartford, CT 06057. Spectrophotometer: Shimadzu UV-240, Shimadzu Corp., Kyoto, Japan 604.

Migration Test. Water, 4% acetic acid, and 0.5% sodium bicarbonate solution as food-simulating solvents

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Table I. Recoveries of Aluminum from Food-Simulating Solvents and Foods

sample	spiked amt,[a] μg	rec, % (mean ± SD)
water	5	96 ± 9
4% acetic acid	500	99 ± 2
0.5% sodium bicarbonate	50	97 ± 1
Japanese noodles	50	97 ± 6
tomato juice	50	103 ± 18
Chinese noodles	50	90 ± 2

[a] Spiked to 200 mL of food-simulating solvents or 10 g of foods.

and both nontreated and surface-treated pans were used for the migration test of aluminum. The conditions of the test were maintenance at room temperature (20 ± 3 °C) for 24 h, boiling for 10 min, and boiling for 2 h. The solvent volumes were 1400 mL for nontreated and 800 mL for surface-treated pans for the room-temperature test, 1200 mL for the former and 700 mL for the latter pans for the boiling test for 10 min, and 1000 mL for the former and 700 mL for the latter pans for the boiling test for 2 h. After the experimental period, decreased volume was adjusted with water and the solution was transferred to a beaker.

To compare the difference in the concentration of aluminum released to food-simulating solvents and foods, Japanese noodles as a neutral food, Chinese noodles as an alkaline food, and tomato juice as an acidic food were used as foods. Chinese noodles are generally treated with sodium bicarbonate and sodium carbonate to improve wheat flour. Nontreated pans were used as cooking utensils for the migration test of aluminum with foods. One pack of dried Japanese noodles weighing 85 g or dried Chinese noodles weighing 80 g was put in 600 mL of boiling water without any of the packaged seasonings and boiled for 10 min. After boiling, noodles and broth were transferred into the blender and homogenized. In the experiment using tomato juice, 600 mL of the juice was heated in the nontreated pan and boiled for 10 min. The pot was covered with a lid of the same material during heating. As controls, the foods were cooked in 1-L beakers in the same manner as described above. All experiments were carried out in triplicate using new pans.

Determination of Aluminum. A 10-g portion of the homogenate of noodles and the cooked tomato juice was weighed, dried at 120 °C, charred, and ashed at 500 °C. The ash was dissolved in 4 mL of hydrochloric acid, dried on a water bath, and then dissolved again in 4 mL of 3.6% hydrochloric acid. The solution was brought to 200 mL with water. In the case of food-simulating solvents, the pH of 200 mL of the solution was adjusted to neutral and 4 mL of 3.6% hydrochloric acid was added. Aluminum in these solutions was determined by the standard method of the Japan Water Works Association (1985), which is based on colorimetry using 8-hydroxyquinoline. Recoveries of aluminum from food-simulating solvents and foods are shown in Table I.

RESULTS AND DISCUSSION

Release of aluminum was scarcely observed at the detection limit of 0.03 μg/mL under any conditions using water as a food-simulating solvent from either nontreated or surface-treated aluminum pans except for the release of 0.03 or 0.04 μg/mL from nontreated pans that occurred with boiling for 10 min or 2 h, respectively. When acidic or alkaline solvents stood at room temperature for 24 h, 0.58–2.90 μg/mL of aluminum was released from both types of pans. The release increased to 41–167 μg/mL with boiling of these solvents for 10 min and to 161–501 μg/mL with boiling for 2 h. Surface-treated pans were more resistant to the temperature and pH of the solvents in the release of aluminum than nontreated pans under all conditions except for the release of the same level of aluminum in 0.5% sodium bicarbonate solution allowed to stand at room temperature (20 ± 3 °C) for 24 h; it was also more resistant to the acidic than the alkaline solvent (Table II).

Table II. Release of Aluminum (μg/mL) to Food-Simulating Solvents from Aluminum Pans

solvent	material	rt, 24 h	boil, 10 min	boil, 2 h
water	nontreated	nd[a]	0.03 ± 0	0.04 ± 0
	surface-treated	nd	nd	nd
4% acetic	nontreated	2.90 ± 0.03	70 ± 3	501 ± 5
acid	surface-treated	0.58 ± 0.41	41 ± 2	161 ± 16
0.5% $NaHCO_3$	nontreated	1.34 ± 0.04	167 ± 16	323 ± 22
soln	surface-treated	1.35 ± 0.24	63 ± 5	263 ± 10

[a] Less than 0.03 μg/mL.

Table III. Aluminum Content (μg/g of Homogenate) in Foods Cooked in Beakers or Aluminum Pans[a]

food	pH	cookware	
		beaker	nontreated Al pan
Japanese noodles	5.5	1.1 ± 0.3	1.0 ± 0.2
tomato juice	4.2	8.0 ± 0.8	11.6 ± 1.0
Chinese noodles	8.9	1.1 ± 0.2	4.9 ± 0.2

[a] Cooking conditions: One pack of dried noodles was put in 600 mL of boiling water and boiled for 10 min; in the case of tomato juice, 600 mL was boiled for 10 min.

The aluminum concentration of the homogenate of Chinese noodles cooked for 10 min in a beaker was 1.1 ± 0.2 μg/g, but that cooked in the nontreated pan was 4.9 ± 0.2 μg/g. Tomato juice cooked in the nontreated pan also showed a higher concentration of aluminum, 11.6 ± 1.0 μg/g, than that cooked in a beaker, 8.0 ± 0.8 μg/g, but no difference was observed in the case of Japanese noodles, which contained 1.1 ± 0.3 μg/g of aluminum (Table III). The pHs of these foods after boiling for 10 min were 8.9, 4.2, and 5.5, respectively. Release of aluminum from aluminum pans with the cooking of alkaline and acidic foods was about 1/40 for alkaline and 1/20 for acidic solvents, and aluminum concentrations released into cooked foods were rather close to that treated with acidic or alkaline solvents at 20 ± 3 °C for 24 h. The aluminum concentration in the tomato juice cooked in the pan was almost the same as the results reported by Greger et al. (1985) that the release of aluminum from aluminum pans with the cooking of tomatoes for 10 min was 3.1 ± 1.4 μg/g. They reported that 57 μg/g (range 29.7–125 μg/g) of aluminum was released from aluminum pots with the boiling tomato sauce for 3 h. These concentrations corresponded to the results obtained under the conditions of boiling for 10 min with 4% acetic acid or 0.5% sodium bicarbonate solution in the present paper. The colorimetry for the determination of aluminum used in the present paper is known to be affected by the presence of a high concentration of calcium, but no effect was observed with the addition of up to 5 times the amount of calcium generally contained in tomato juice (Resources Council, Science and Technology Agency, 1983).

Daily intake of aluminum is estimated to be 3.5–51.6 mg (Underwood, 1977), 3–100 or 20–40 mg (Greger, 1985), and 20 mg (Lione, 1983), and it is estimated that 20% of the daily intake of aluminum comes from utensils (Lione, 1983). Our results indicated that at least 3.3 mg of aluminum, including 2.6 mg of aluminum released from an aluminum pan, will be ingested with one meal of Chinese noodles.

Registry No. $NaHCO_3$, 144-55-8; aluminum, 7429-90-5; acetic acid, 64-19-7.

LITERATURE CITED

Alfrey, A. C.; Hegg, A.; Craswell, P. "Metabolism and Toxicity of Aluminum in Renal Failure". *Am. J. Clin. Nutr.* **1980**, *33*, 1509–1516.

Bloom, W. L.; Flinchum, D. "Osteomalacia with Pseudofractures Caused by the Ingestion of Aluminum Hydroxide". *JAMA, J. Am. Med. Assoc.* **1960**, *174*, 1327–1330.

Crapper, D. R.; Krishnan, S. S.; Dalton, A. J. "Brian Aluminum Distribution in Alzheimer's Disease and Experimental Neurofibrillary Degenration". *Science (Washington, D.C.)* **1973**, *180*, 511–513.

Greger, J. L. "Aluminum Content of the American Diet". *Food Technol.* **1985**, *39*, 73–80.

Greger, J. L.; Goetz, W.; Sullivan, D. "Aluminum Levels in Foods Cooked and Stored in Aluminum Pans, Trays and Foil". *J. Food Prot.* **1985**, *48*, 772–777.

Ishiwata, H.; Inoue, T.; Yamada, T.; Tanimura, A. "Comparative Migration Tests of Lead and Cadmium from Tablewares". *J. Food Hyg. Soc. Jpn.* **1984**, *25*, 445–448.

Ishiwata, H.; Inoue, T.; Yoshihira, K. "Migration of Copper and Some Other Metals from Copper Tableware". *Bull. Environ. Contam. Toxicol.* **1986**, *37*, 638–642.

Japan Water Works Association *Standard Methods of Japan Water Works Association for Drinking Water*, 1st ed. The Japan Water Works Association: Tokyo, 1985; p 381.

Katsumura, K.; Ishizaki, M.; Sasamoto, T.; Ueno, S.; Hosogai, Y. "Determination of Aluminum in Food and Plant by Atomic Absorption Spectrometry". *J. Food Hyg. Soc. Jpn.* **1973**, *14*, 530–534.

Lione, A. "The Prophylactic Reduction of Aluminum Intake". *Food Chem. Toxicol.* **1983**, *21*, 103–109.

Parkinson, I. S.; Ward, M. K.; Kerr, D. N. S. "Dialysis Encephalopathy, Bone Disease and Anaemia: the Aluminum Intoxication Syndrome during Regular Haemodialysis". *J. Clin. Pathol.* **1981**, *34*, 1285–1294.

Perl, D. P.; Brody, A. R. "Alzheimer's Disease: X-ray Spectrometric Evidence of Aluminum Accumulation in Neurofibrillary Tangle-bearing Neurons". *Science (Washington, D.C.)* **1980**, *208*, 297–299.

Resources Council, Science and Technology Agency *Standard Tables of Food Composition in Japan*, 4th ed.; Kagawa Nutrition College: Tokyo, 1983; p 179.

Trapp, G. A.; Cannon, J. B.; Koning, J. H. "Aluminum Pots as a Source of Dietary Aluminum". *N. Engl. J. Med.* **1981**, *304*, 172–173.

Underwood, E. J. "Aluminum". In *Trace Elements in Human and Animal Nutrition*, 4th ed.; Academic: New York, 1977; p 430.

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Postirradiation Dosimetry of Meat by Electron Spin Resonance Spectroscopy of Bones

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Electron spin resonance (ESR) spectroscopy was used to measure the production of free radicals induced by ^{60}Co γ-rays in chicken bones. It was found that the radiation-induced ESR signal in bone could easily be distinguished from the endogenous ESR signal. Long-term (4 months) stability studies at 20 °C showed no decay of the radiation-induced ESR signal. A linear relationship was observed between the radiation-induced ESR signal intensity (peak-to-peak amplitude) and the absorbed dose (1–5 kGy). It was concluded that ESR measurements of bones can be used to determine whether the bone-containing meat has been irradiated and also at approximately what dose. The measurements indicate the feasibility of postirradiation dosimetry (PID) of meats when bones are present.

The treatment of natural and processed food products by ionizing radiation has been under consideration as a method of food preservation for the past 30 years (Goresline, 1983). The United States Food & Drug Administration has approved irradiation of foods up to an absorbed dose of 1 kGy (100 krads) (*Federal Register*, 1986), whereas the World Health Organization has set considerably higher limits at 10 kGy (1 Mrad).

A controversial aspect of food irradiation processing is the possible creation of radiolytic products unique to irradiated foods whose safety has not been assessed. Until these issues are resolved, an important aspect of food technology from the standpoint of safety and quality control is the development of convenient and reliable methods for determining whether a particular food item has been irradiated and at what dose. The measurement of dose after the fact by quantitative analysis of the specific radiation-induced changes in the specimen in question will be referred to here as postirradiation dosimetry (PID).

An important aspect of PID is the development of a highly specific marker that would serve as an internal dosimeter inherent to that particular food item. An ideal choice for this purpose would be a unique radiolytic product of the food item that changes linearly with the application of ionizing radiation over a required dose range and does not fade away during the lifetime of the food item. Initial studies into such techniques have already shown feasibility (Karam and Simic, 1986). The technique is based on generation of radiation-induced *o*-tyrosine (*o*-Tyr) from phenylalanine in meats such as chicken. The amount of *o*-Tyr, as determined by gas chromatography/mass spectrometry, in irradiated meat increases linearly with dose (0.5–5 kGy) (Karam and Simic, 1986) Though promising, the technique, or for that matter any one particular technique, cannot be expected to be applicable to all types of foods, e.g. all meats, fruits, spices, etc. Therefore, it is necessary to develop an array of techniques that can be used to maintain quality control

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