retail handling of milk is recommended.

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LITERATURE CITED

Ames, S. R. "Methods for Evaluating Vitamin A Isomers". *J. Assoc. Off. Anal. Chem.* **1966**, *49*, 1071.
Bredeweg, R. A.; Rothman, L. D.; Pfieffer, C. D. "Chemical Reactivation of Silica Columns". *Anal. Chem.* **1979**, *51*, 2061.
Egberg, D. C.; Heroff, J. C.; Potter, R. H. "Determination of *all-trans-* and 13-*cis* Vitamin A in Food Products by High-Pressure Liquid Chromatography". *J. Agric. Food Chem.* **1977**, *25*, 1127.
Gaylord, A. M.; Warthesen, J. J.; Smith, D. E. "Effect of Fluorescent Light on the Isomerization of Retinyl Palmitate in Skim Milk". *J. Food Sci.* **1986**, *51*, 1456.
Landers, G. M.; Olson, J. A. "Absence of Isomerization of Retinyl Palmitate, Retinol, and Retinal in Chlorinated and Nonchlorinated Solvents under Gold Light". *J. Assoc. Off. Anal. Chem.* **1986**, *69*, 50.
Mulry, M. C. "Retinyl Palmitate Isomerization: Analysis and Quantization in Fortified Foods and Model Systems". Ph.D. Dissertation, University of Florida, Gainesville, FL, 1983.
Mulry, M. C.; Schmidt, R. H.; Kirk, J. R. "Isomerization of Retinyl Palmitate in Conventional Liquid Extraction Solvents". *J. Assoc. Off. Anal. Chem.* **1983**, *66*, 746.
SAS Institute Inc. *SAS Users Guide: Statistics*, version 5; SAS Institute: Cary, NC, 1985; 956 pp.
Slater, J. C.; Stone, H. A.; Palermo, B. T.; Duvall, R. N. "Reliability of Arrhenius Equation in Predicting Vitamin A Stability in Multivitamin Tablets". *J. Pharm. Sci.* **1979**, *68*, 49.
Stancher, B.; Zonta, F. "Comparison between Straight and Reversed Phases in High-Performance Liquid Chromatographic Fractionation of Retinol Isomers". *J. Chromatogr.* **1982a**, *234*, 244.
Stancher, B.; Zonta, F. "High-Performance Liquid Chromatographic Determination of Carotene and Vitamin A and Its Geometric Isomers in Foods: Application to Cheese Analysis". *J. Chromatogr.* **1982b**, *238*, 217.
Thompson, J. N.; Hatina, G.; Maxwell, W. B. "High Performance Liquid Chromatographic Determination of Vitamin A in Margarine, Milk, Partially Skimmed Milk and Skim Milk". *J. Assoc. Off. Anal. Chem.* **1980**, *63*, 894.
Wilkinson, S. A.; Earle, M. D.; Cleland, A. C. "Kinetics of Vitamin A Degradation in Beef Liver Puree on Heat Processing". *J. Food Sci.* **1981**, *46*, 32.
Wilkinson, S. A.; Earle, M. D.; Cleland, A. C. "Effects of Food Composition, pH and Copper on the Degradation of Vitamin A in Beef Liver Puree during Heat Processing". *J. Food Sci.* **1982**, *47*, 844.
Zahar, M.; Smith, D. E.; Warthesen, J. J. "Factors Related to the Light Stability of Vitamin A in Various Carriers". *J. Dairy Sci.* **1987**, *70*, 13.

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# Changes in Free Amino Acids and Adenine Nucleotides in Boiled Muscle Extracts of Yellowtail (*Seriola quinqueradiata*) Stored in Ice

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Free amino acids (FAA), adenine nucleotides, and their related compounds in boiled muscle extracts of yellowtail (*Seriola quinqueradiata*) were determined during ice storage. Little change in the amount of major FAA except alanine was detected in the white muscle during over 40 days of storage, while in the dark muscle almost all FAA, except taurine and histidine, increased significantly in the early stage of storage. Inosine 5′-monophosphate (IMP) content was abundant in the very fresh white muscle and then decreased gradually during storage. In the dark muscle, inosine (HxR) and IMP were predominant initially, followed by an extremely rapid decrease in IMP and a concomitant increase in HxR level within 1 day.

Fish muscle contains a variety of nonprotein nitrogenous compounds, some of which are important from a food chemical point of view (Konosu and Yamaguchi, 1982). The changes in content of adenine nucleotides and their related compounds during storage are closely associated with the quality of the fish (Uchiyama and Ehira, 1970; Connell and Shewan, 1980). Also the changes in amount of free amino acids (FAA) relate to autolysis and subsequent bacterial action (Shewan, 1962; Liston, 1980). FAA are known to evoke a meaty taste sensation together with adenine nucleotides (Maga, 1983). In our previous papers, we reported the changes in content of major FAA, adenine nucleotides, and their related compounds in yellowtail (*Seriola quinqueradiata*) muscle (Sakaguchi et al., 1982; Murata and Sakaguchi, 1986). Yellowtail is a dark-flesh fish and one of the most important species of cultured fish in Japan. Dark-fleshed fish have dark muscle along the lateral line in an amount of 10–20% of the whole musculature (Obatake and Heya, 1985). The rates and patterns of the changes in content of the above compounds during ice storage differ greatly between white and dark muscles. In the white muscle most FAA showed little change in content; in the dark muscle they increased markedly (Sakaguchi et al., 1982, 1984). Inosine 5′-monophosphate (IMP), among adenine nucleotides, accumulated in the former tissue in a certain early stage of storage, followed by the gradual decrease in level. IMP levels in the latter fall very rapidly, and inosine (HxR) rises concomitantly (Dyer et al., 1966; Murata and Sakaguchi, 1986). For extraction of nonprotein nitrogenous compounds from the muscle tissue, several deproteinizing reagents are commonly used. Such extracts obtained, however, are not applicable to sensory evaluation. In the present paper, we describe the changes in content of FAA, adenine nucleo-

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tides, and related compounds in boiled muscle extracts of yellowtail stored in ice. Comparison of the results was also made with those of our previous reports (Sakaguchi et al., 1982; Murata and Sakaguchi, 1986).

## MATERIALS AND METHODS

**Materials.** Five yellowtail (*S. quinqueradiata*) weighing about 1.5 kg each were obtained from a local market. They were killed by decapitation and filleted immediately. White muscle was taken from the dorsal part of the fillets, and dark muscle along the lateral line was separated. The muscles were put in a plastic vial (6.5-cm inner depth × 4.5-cm inner diameter) separately, and the vials were submerged in ice during storage. Approximately 0.5 g of the muscle from the anterior end of each piece was cut and discarded. Then, several slices of 0.5–0.7 g each were newly cut, pooled, and mixed well; 5 g was used for preparing boiled muscle extracts.

**Preparation of the Boiled Muscle Extracts.** Five grams of the muscle tissue was homogenized with 50 mL of ice-cold water, poured on a glass vessel, and heated gradually. After being boiled gently for 15 min with continuous stirring, the homogenates were centrifuged at 5000*g* for 30 min. The precipitate formed was stirred again with 10 mL of water, boiled for 5 min, and centrifuged again. The supernatants were combined, made up to 50 mL, and then stored frozen at −20 °C until analysis.

**Determination of Total Nitrogen (TN) and Nonprotein Nitrogen (NPN) in the Boiled Muscle Extract.** For determination of TN, 1 mL of the extracts was digested by the semimicro Kjeldahl method, and nitrogen contents were estimated colorimetrically by the method of Nimura (1973). To determine NPN in the muscle extract, 1 mL of 24% ice-cold trichloroacetic acid (TCA) was added to 3 mL of the extract. Precipitate formed was removed by centrifugation at 5000*g* for 30 min. The supernatant was digested, and nitrogen contents were determined in the same manner as TN.

**Determination of Major FAA and Adenine Nucleotides and Their Related Compounds.** An aliquot of the muscle extract was submitted to analysis of major FAA on an amino acid analyzer (Hitachi Model 835). For determination of adenine nucleotides and their related compounds, an aliquot of the muscle extracts was applied to column chromatography using an AG 1X4 ($Cl^-$) ion exchanger (Bio-Rad), as described by Kato et al. (1973) and Yamada et al. (1981).

**Calculation of *K* Value.** A freshness index *K* value (Saito et al., 1959) was expressed as a proportion (percent) of a content of inosine (HxR) plus hypoxanthine (Hx) to a total amount of adenine nucleotides, HxR and Hx.

## RESULTS AND DISCUSSION

**Total Nitrogen (TN) and Nonprotein Nitrogen (NPN).** Figure 1 shows changes in TN content in the white and dark muscles of yellowtail during ice storage. In the white muscle the TN content exhibited a 58 mg/100 g increase within the initial 4 days from 612 mg/100 g to 670 mg/100 g but changed little thereafter. The dark muscle showed a slow increase of TN for the initial 8 days from 412 mg/100 g to 432 mg/100 g and then was almost unchanged until the end of storage.

Figure 1 also shows changes in NPN levels in both the white and dark muscles during storage. The white muscle initially had an NPN content of 592 mg/100 g and then increased to 635 mg/100 g during the initial 4 days but thereafter was almost unchanged. The fresh dark muscle had 315 mg/100 g of NPN; the value agrees well with the data presented so far for dark-fleshed fish (Endo and Simidu, 1955; Lukton and Olcott, 1958; Arakaki and Suyama, 1966; Hashimoto et al., 1979; Sakaguchi, et al., 1982, 1984). The NPN level in the dark muscle increased gradually up to 374 mg/100 g on the 37th day, showing a 59 mg/100 g increase. The white muscle TN and NPN were almost in the same level at 0 day. The difference between the TN and NPN levels equals the nitrogen level of hot water soluble protein, which was precipitated by addition of TCA to the boiled muscle extracts. Such protein increased in content during the early but not later stage of storage. In the dark muscle, on the other hand, the content of the protein was significantly abundant compared with that in the white muscle through the entire storage period. The protein in both muscle extracts was thought to contribute to the taste sensation to some extent, although it might not function directly as a taste enhancer. Our preliminary sensory evaluation indicated that removal of the protein fraction by means of a membrane filter altered somewhat the taste quality of the muscle extracts, especially from the dark muscle. The characterization of the protein is now in progress.

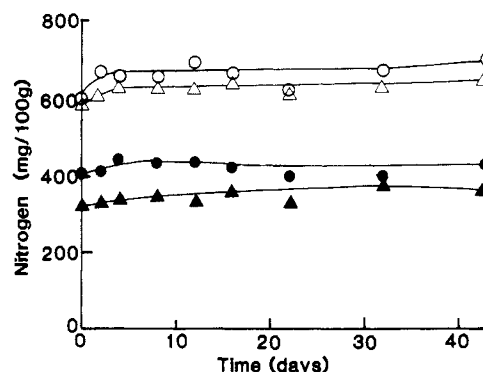


**Figure 1.** Changes in TN and NPN content in white and dark muscles during ice storage. Open circules and triangles represent nitrogen contents in the white muscle, and closed ones, those in the dark muscle: ○ and ●, TN; △ and ▲, NPN.

**Free Amino Acids (FAA).** Figures 2 and 3 show changes in major FAA levels in the white and dark muscles, respectively, during ice storage. Among FAA analyzed of the white muscle, histidine was extremely abundant, while alanine, lysine, taurine and glycine occurred in a range of 10–30 mg/100 g wet weight (Figure 2A,B). Glutamic acid known to provide a taste of "umami" was also less than 10 mg/100 g and began to decrease in the middle stage of storage. Alanine increased markedly in the early period, while arginine decreased after 22 days (Figure 2B). The others showed no measurable change.

In the dark muscle, taurine was present in an extremely high concentration (about 1000 mg/100 g) (Figure 3A). Generally, this compound is contained in high levels in the dark muscle of dark-fleshed fish and at low levels in the white muscle (Obatake et al., 1985; Sakaguchi and Murata, 1986). This evidence was in striking contrast to that of histidine, which was present in large amounts of the white muscle (Figure 2A) and in relatively small amounts in the dark muscle tissue (Figure 3C). FAA that exist at more than 10 mg/100 g in the initial stage were alanine, glutamic acid, and histidine, and others were relatively low (Figure 3A,B). Most of FAA analyzed in the dark muscle increased gradually; only histidine showed a decreasing trend (Figure 3A–C). Glutamic acid exhibited a complicated pattern of change, a rapid decrease initially and a gradual increase thereafter (Figure 3B). The drop of most FAA in the final stage of storage was thought to be due to bacterial spoilage, because after about 30 days the volatile base nitrogen level began to increase distinctly (Sakaguchi et al., 1982). From these results it can be concluded that contents of FAA in

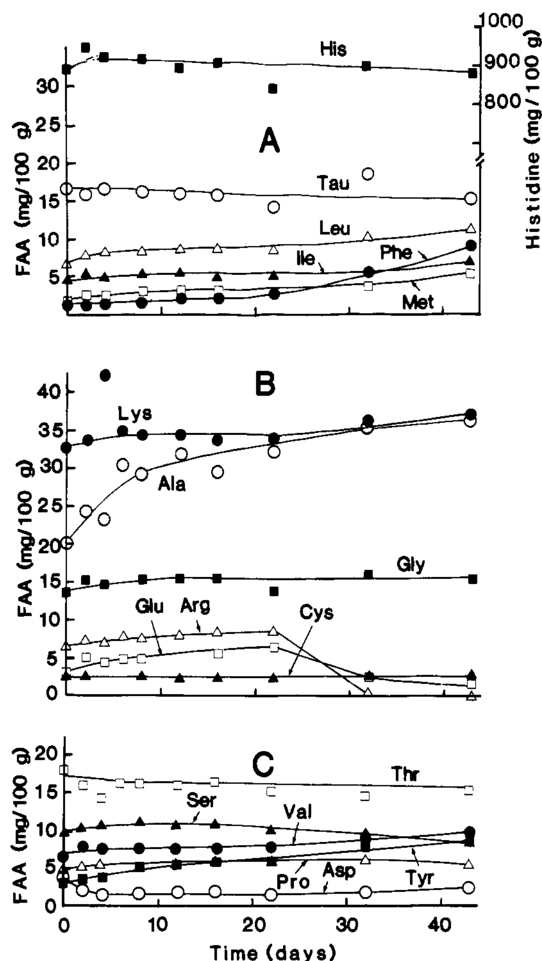


Figure 2. Changes in major FAA content in white muscle during storage in ice. A: ■, His; ○, taurine (Tau); △, Leu; ▲, Ile; ●, Phe; □, Met. B: ●, Lys; ○, Ala; ■, Gly; □, Glu; △, Arg; ▲, Cys. C: △, Pro; □, Thr; ▲, Ser; ●, Val; ■, Tyr; ○, Asp.

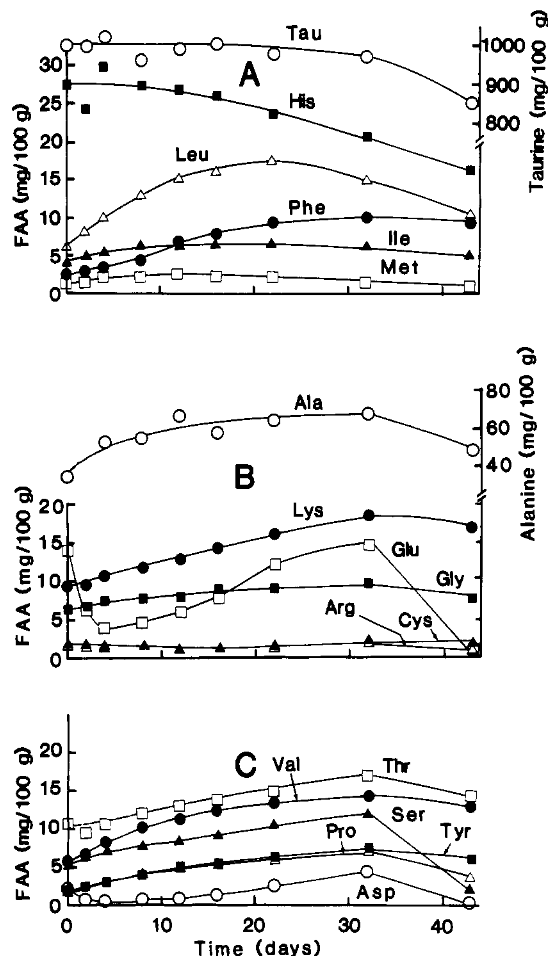


Figure 3. Changes in major FAA content in dark muscle during storage in ice. Symbols in A–C are the same as those in Figure 2.

the boiled muscle (white and dark) extracts and patterns of the change in FAA level during ice storage are similar to those observed previously for the TCA extracts (Sakaguchi et al., 1982).

**Nucleotides and Their Related Compounds.** Figures 4 and 5 illustrate changes in content of adenine nucleotides and related compounds in the white and dark muscles, respectively, during ice storage. No measurable ATP was present in the fresh white muscle (Figure 4). IMP instead was a major nucleotide (8.2 μmol/g wet weight) detected, followed by a gradual decrease until the end of storage. This pattern differed markedly from those reported previously (Murata and Sakaguchi, 1986). When the extraction was performed with perchloric acid (PCA), ATP was detected abundantly in the very fresh white muscle. An IMP content was low initially, increased rapidly to reach the highest level about 2 days after the beginning of storage, and then gradually decreased in level. Accompanied with the drop in IMP, a gradual buildup of HxR was observed. ADP and AMP levels were extremely low throughout the storage periods. A low content of Hx (0.06 μmol/g) was present initially, and the compound increased slowly. The boiled muscle extracts that were prepared from the very fresh muscle contained a high level of IMP, suggesting that ATP degraded rapidly with the concomitant accumulation of IMP during the homogenization and subsequent heating of the muscle. Concerning this phenomenon, it has been reported for many fish species that the white muscle has high AMP deaminase activity (Raffin and Leray, 1980; Fujisawa and Yoshino, 1987) but ex-

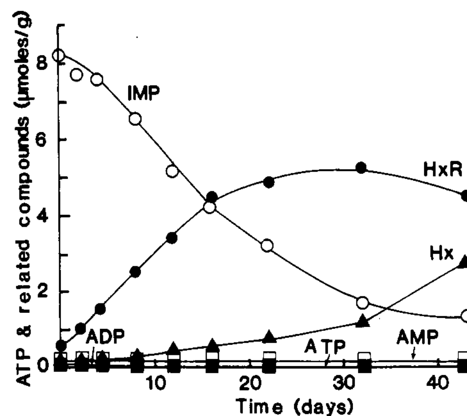


Figure 4. Changes in content of adenine nucleotides and related compounds in white muscle during storage in ice: ■, ATP; △, ADP; □, AMP; ○, IMP; ●, HxR; ▲, Hx.

tremely low 5′-nucleotidase activity (Fujisawa and Yoshino, 1987). IMP, therefore, accumulated for the short period of homogenization and heating. In addition, the enzyme systems (ATPase, myokinase, AMP deaminase) responsible for the degradation of ATP to IMP seemed to be heat-stable compared with an enzyme 5′-nucleotidase, which catalyzes the conversion of IMP to HxR. IMP is known to act synergistically with glutamic acid, resulting in lowered effective concentrations (Maga, 1983; Yamaguchi, 1987). Within the initial 3 weeks, glutamic acid changed little in level (Figure 2B). Therefore, IMP levels were thought to determine taste intensity of the fish muscle. Possibly, 8.2 μmol/g of the IMP level in the fresh

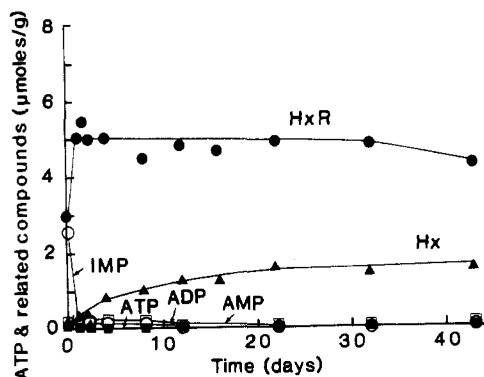


**Figure 5.** Changes in content of adenine nucleotides and related compounds in dark muscle during storage in ice. Symbols are the same as those in Figure 4.

muscle is enough to produce a maximum taste intensity. According to Simidu et al. (1958), the yellowtail muscle stored at 2 ± 2 °C is able to maintain the original taste rating until about 10 days and loses the rating sharply after this period. An IMP level attained on the 10th day was approximately 6 μmol/g (Figure 4). In the dark muscle, ATP, ADP, and AMP were hardly recognized when the tissue was very fresh (Figure 5). IMP existed at a level of 2.6 μmol/g wet weight first and disappeared within 1 day. HxR, which was relatively high at 0 day (3.0 μmol/g), rose at a high rate up to about 5 μmol/g by 1 day and fell very slowly thereafter, while Hx content continued to increase slowly. Generally, fish dark muscle has substantially high levels of ATP and undergoes the rapid degradation to IMP through ADP and AMP. The conversion of IMP to HxR proceeds also rapidly, but that of HxR to Hx is negligible. Consequently, HxR accumulates in high levels (Dyer et al., 1966; Murata and Sakaguchi, 1986). This series of reaction occurred within a short period of the homogenization and subsequent heating, resulting in the high production of HxR (Figure 5). Again, IMP existed at a considerable level first and disappeared rapidly within 1 day. It is generally believed that the dark muscle of dark-fleshed fish becomes tasteless soon during storage. Our result supports this, because both IMP and glutamic acid decreased in concentration at high rates. Our preliminary examination on addition of the amount of IMP corresponding to that of the 0-day sample back to the aged muscle extracts indicated the taste to be improved to a large extent. Most probably one of the reasons why the dark muscle becomes tasteless rapidly upon aging is loss of IMP.

***K* Value.** Figure 6 shows changes in *K* values during ice storage of the white and dark muscles. The value of the white muscle was very low (6.5%) initially, a little higher than that reported previously (Murata and Sakaguchi, 1986). The value continued to increase linearly until about the 16th day and slowly thereafter and reached approximately 80% at the end of storage (43 days). The value rose at a rate during storage similar to that observed for the PCA extracts, as reported previously (Murata and Sakaguchi, 1986). A suggested limit of the value is 20% for high-quality "sashimi" of dark-fleshed fish (Uchiyama and Ehira, 1970), indicating that a shelf life for yellowtail "sashimi" is 5–6 days in ice. Further storage for 5 days raised the *K* value to a level of about 35% (Figure 6). In this period of storage the original quality grade does not drop significantly, because Simidu et al. (1958) reported that the overall score including surface appearance, texture, odor, and taste falls in very low rates about 10 days after the beginning of storage, when yellowtail are stored at 2 ± 2 °C. The pattern of change in the *K* value of the dark muscle differed greatly from that of the white muscle (Figure 6). Even in the extremely fresh muscle, the value was 62% and reached about 90% as early as 1 day after the beginning of storage. The value at the initial period was much higher than that of the PCA extract (14.5%) (Murata and Sakaguchi, 1986). This was attributable to the rapid accumulation of HxR, possibly because of the accelerated breakdown of IMP to HxR by the homogenization and subsequent heating. In the case of the white muscle, however, this breakdown was not stimulted so much as that of the dark muscle; thus, the initial *K* value remained low.

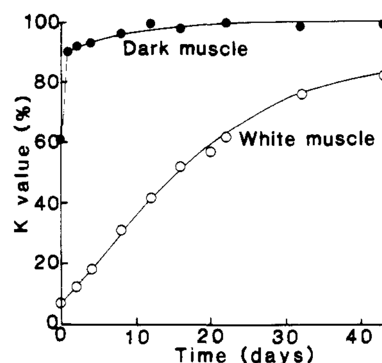


**Figure 6.** Changes in *K* value in white and dark muscles during storage in ice: ○, white muscle; ●, dark muscle.

LITERATURE CITED


Arakaki, J.; Suyama, M. "Free and Conjugated Amino Acids in the Extractives of Anchovy". *Nippon Suisan Gakkaishi* **1966**, *32*, 74–79.

Connell, J. J.; Shewan, J. M. "Past, Present and Future of Fish Science". In *Advances in Fish Science and Technology*; Connell, J. J., Ed.; Fishing News Books: Surrey, England, 1980.

Dyer, W. J.; Fraser, D. I.; Lohnes, D. P. "Nucleotide Degradation and Quality in Ordinary and Red Muscle of Iced and Frozen Swordfish (*Xiphias gladius*)". *J. Fish. Res. Board Can.* **1966**, *23*, 1821–1833.

Endo, K.; Simidu, W. "Studies on Muscle of Aquatic Animals X111: Distribution of Extractive Nitrogens in Bloody Muscle". *Nippon Suisan Gakkaishi* **1955**, *21*, 127–129.

Fujisawa, K.; Yoshino, M. "Activities of Adenylate-Degrading Enzymes in Muscle from Vertebrates and Invertebrates". *Comp. Biochem. Physiol., B: Comp. Biochem.* **1987**, *86B*, 109–112.

Hashimoto, K.; Watabe, S.; Kono, M.; Shiro, K. "Muscle Protein Composition of Sardine and Mackerel". *Nippon Suisan Gakkaishi* **1979**, *45*, 1435–1441.

Kato, N.; Uchiyama, H.; Uda, F. "A Rapid Method for Determination of Inosine, Hypoxanthine, Uric Acid, and Nucleotides in Fish Muscle by Continuous Gradient Column Chromatography". *Nippon Suisan Gakkaishi* **1973**, *39*, 1039–1044.

Konosu, S.; Yamaguchi, K. "The Flavor Component in Fish and Shellfish". In *Chemistry & Biochemistry of Marine Food Products*; Martin, R. E., Flick, G. J., Hebard, C. E., Ward, D. R., Eds.; AVI: Westport, CT, 1982.

Liston, J. "Microbiology in Fishery Science". In *Advances in Fish Science and Technology*; Connell, J. J., Ed.; Fishing New Books: Surrey, England, 1980.

Lukton, A.; Olcott, H. S. "Content of Free Imidazole Compounds in the Muscle Tissue of Aquatic Animals". *Food Res.* **1958**, *23*, 611–618.

Maga, J. A. "Flavor Potentiators". *CRC Crit. Rev. Food Sci. Nutr.* **1983**, *18*, 231–312.

Murata, M.; Sakaguchi, M. "Storage of Yellowtail (*Seriola quinqueradiata*) White and Dark Muscle in Ice: Changes in content of Adenine Nucleotides and Related Compounds". *J. Food Sci.* **1986**, *51*, 321–326.

Nimura, Y. "A Direct Estimation of Microgram Amounts of Ammonia in Water without Salt-error". *Nippon Suisan*

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

Takiko Inoue,* Hajimu Ishiwata, and Kunitoshi Yoshihira

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.

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## 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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