December, 1980] © 1980 The Chemical Society of Japan Bull. Chem. Soc. Jpn., 53, 3567—3572 (1980) 3567

Anodic Stripping Semidifferential Electroanalysis of Mercury(II) at Gold Disk Electrode and Its Application to Environmental Analysis

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(Received April 26, 1980)

Anodic stripping semidifferential electroanalysis using a gold disk working electrode was proposed for the trace determination of mercury(II) ion, theoretical predictions being verified. A gold electrode was steeped in aqua regia, activated electrochemically before use, and rotated during the course of pre-electrolysis. A hydroxylamine hydrochloride medium was selected as supporting electrolyte. The detection limit was found to be *ca.* 0.5 nM under the pre-electrolysis conditions of 10 min at 0 V *vs.* Ag/AgCl. The relative standard deviation was *ca.* ±4% for the measurement at 2 nM level. Removal of the oxygen dissolved in sample solutions was not necessary. Experiments were carried out to examine the feasibility of the technique to environmental analysis. The method consists of only the following procedure: sample acidification, standard mercury(II) ion addition, UV irradiation pretreatment, and medium exchange for stripping. It was successfully applied to river water and sea water to separately evaluate ionic mercury, mercury occluded in organic matter, and total mercury (excluding mercury occluded in inorganic matter).

Environmental scientists realize the increasing nead for reliable methods which can differentiate and determine the chemical forms of trace toxic metals such as mercury in natural waters.[1] Sensitive analytical techniques such as neutron activation analysis and atomic absorption spectrophotometry are not applicable to speciation studies since they deal only with total metal concentration. Anodic stripping voltammetry determines only the labile ionic metal and might be utilized in speciation studies.

Voltammetric methods reported so far for the determination of mercury(II) ion make use of carbon electrodes such as wax-impregnated graphite,[2,3] glassy carbon,[4,5] and carbon paste.[6] However, the methods provide no satisfactory sensitivity for the environmental analysis of mercury(II) ion because of a relatively small affinity of the carbon electrodes with metallic mercury. Recently gold electrodes have drawn renewed attention for trace mercury analysis because of the large solubility of mercury in gold.[7] Sipos *et al.*[8,9] successfully applied the voltammetric method using one or two gold electrodes for the simultaneous determination of total copper and mercury in environmental samples. All the voltammetric methods reported so far for mercury analysis make use of the conventional linear sweep voltammetry at a slow potential scan rate, potentiometry, or differential pulse voltammetry for stripping process. No report seems to have appeared on the systematic speciation analysis of mercury in natural waters.

Goto and Ishii[10] and Dalrymple-Alford *et al.*[11,12] developed a new voltammetric technique "*semidifferential electroanalysis.*" It was applied to anodic stripping analysis of heavy metal ions except mercury(II) ion at a hanging mercury drop[13] and a thin mercury film electrode.[14] Semidifferential electroanalysis has been applied to detection in anodic stripping process at a gold electrode, facilitating the analysis of trace mercury(II) ion. The method is recommended for simple speciation analysis of mercury in natural waters.

Theoretical

Consider a solution of reduced species in a thin layer of electrode. On oxidation the oxidized species diffuses into a solution of infinite thickness. The trace mercury deposited on a gold disk electrode from a mercury(II) ion solution may be regarded as a solution of mercury in a thin layer of gold. The electrode reaction is considered to be reversible without pre- or post-kinetics. In this case, the same approximate equation between the semiderivative of current, e, with respect to time, t, and the electrode potential, E, as that derived in anodic stripping process at thin mercury film electrode[14] holds under the conditions defined by the dimmentionless parameter, H, less than 1.6×10^{-3}. H is equal to $l^2\sigma/D'$ and $\sigma=nFv/RT$, l is the thickness of gold into which mercury diffuses, D' the diffusion coefficient of mercury in gold and v the potential scan rate, other symbols having their usual meanings. The theoretical e *vs.* E curve for anodic stripping process of mercury from a gold electrode has a maximum and a minimum (Fig. 1).[14] The height from the negative peak to the positive one is given by

$$e_{\mathrm{pp}} = \frac{0.429n^{2.5}F^{2.5}Av^{1.5}t_{\mathrm{p}}D^{2/3}N^{1/2}C}{\nu^{1/6}R^{1.5}T^{1.5}}, \tag{1}$$

the negative peak potential by

$$E_{\mathrm{np}} = E_{1/2} - \frac{0.656RT}{nF} + \frac{1.15RT}{nF}\log H, \tag{2}$$

and the width between the negative peak and the positive one in the e *vs.* E curve for re-dissolution process of mercury accumulated on a rotating gold disk electrode by pre-electrolyzing trace mercury(II) ion in sample solution by

$$W_{\mathrm{pp}} = \frac{2.52RT}{nF}, \tag{3}$$

where A is the electrode area, t_{p} the pre-electrolysis time, D the diffusion coefficient of mercury(II) ion in solution, N the revolution rate of electrode, C the concentration of mercury(II) ion, ν the kinematic viscosity, and $E_{1/2}$ the d.c. polarographic half-wave potential. We see that the height is proportional to the mercury(II) ion concentration in sample, the one half powers of the potential scan rate, the square root of the revolution rate of electrode during pre-electrolysis, and the pre-electrolysis time.

Experimental

Apparatus and Reagents. The same apparatus as that reported[14] was used. The working electrode, a gold disk of thickness 0.5 cm and diam. 0.3 cm, was sealed into glass tubing, length 15 cm and i. d. 0.3 cm. The exposed surface of the disk was metallographically polished with emery paper, then with Al_2O_3 powder (0.05 μm) on an acrylic resin plate. The surface had been dipped in aqua regia for 1 min, rinsed with distilled and deionized water, and then kept in 0.5% $HCl \cdot NH_2OH$ solution until use. A rotating electrode assembly (Yanagimoto Co., model P8-RE) was used to rotate the gold disk electrode. The Ag/AgCl (satd KCl) and platinum spiral were used as reference and counter electrodes, respectively, and separated from the sample solution by an agar salt bridge.

The stock solution of 100 mM** $Hg(NO_3)_2$ was prepared by dissolving mercury (99.9999%) in nitric acid. The solution or the mercury standard solution of 1000 ppm for atomic absorption spectrophotometry was used as the standard mercury(II) solution. The stock solutions of 5 M H_2SO_4 and 5% $HCl \cdot NH_2OH$ were prepared for one of supporting electrolytes. All the reagents were of special grade, the solutions being diluted with distilled and deionized water. The cell and all vessels used for sample preparation were filled with 1 M HNO_3 or 0.5 M H_2SO_4 for 1 d, rinsed out, and then filled with distilled and deionized water before use.

Procedure. *Standard Sample*: 25 ml of a standard mercury(II) solution with excess supporting electrolyte was poured into the cell, three electrodes being set. First, the working electrode was rotated, a potential of 0.8 V (*vs.* Ag/AgCl) applied for 30 s to clean the surface of the working electrode, and the required deposition potential of mercury(II) ion was applied for certain time. After this, the rotation was stopped, the potential set to 0 V, and the potential anodically swept at a definite scan rate following a rest period of 30 s. For the re-dissolution process of deposited mercury, the *e vs. E* curve was measured at room temperature (≈20 °C). Next, the working electrode was rotated, a potential of 0.8 V being applied for 30 s, after which the rotation was stopped, the potential of electrode set to 0 V, and the potential anodically swept again at the same scan rate following a rest period of 30 s, to measure the blank *e vs. E* curve. The net *e vs. E* curve was corrected by means of the blank curve.

Environmental Sample: Inland water and sea water were collected in polyethylene bottles and immediately acidified to pH 1 with concd. H_2SO_4. An aliquot of 25 ml of the sample was placed in an open weighing vessel, spiked 25 μl of 30% H_2O_2, and irradiated for a certain time with UV light from a 18 W lamp. The sample was then cooled and the volume adjusted to 25 ml by supplying water to compensate the loss caused by irradiation. The gold disk electrode was activated electrochemically in 0.25 M H_2SO_4 containing 0.5% $HCl \cdot NH_2OH$ by cyclic polarization in the range 0.8—1.6 V at a scan rate of 100 mV/s and again at 0.8 V for 1 min. The electrode was set in the sample solution and electrolysis of mercury(II) ion was carried out at 0 V for 10 min under rotation (rpm 1800) without deaeration of solution. The electrode was then transferred promptly to a medium of 0.25 M H_2SO_4 containing 0.5% $HCl \cdot NH_2OH$. The potential was anodically swept from 0 V to 0.8 V at a scan rate of 100 mV/s following quiet polarization for 30 s at 0 V . The blank *e vs. E* curve was constructed in the same way as in the case of standard sample, electrochemical activation of the electrode being carried out in the same medium. 10 μl of mercury(II) standard solution was then added to the sample solution, the above procedure being repeated to determine mercury.

Results and Discussion

e vs. E *Curve in Anodic Stripping Process.* Figure 1 shows a typical *e vs. E* curve of the re-dissolution process of mercury deposited on the gold disk electrode, the corresponding *i vs. E* curve being given for comparison. The dotted *e vs. E* and *i vs. E* curves indicate the blank curves measured after cleaning the gold electrode at 0.8 V for 30 s under rotation. As predicted theoretically, a maximum and a minimum were observed in the *e vs. E* curve for stripping process. We see that the *e vs. E* curve has the following advantages over the ordinary *i vs. E* curve. It is easy to estimate the height from the negative peak to the positive one rather than the peak height from the base line. From a comparison of both blank curves, the semidifferentiation of the *i vs. E* curve seems to reduce the interference of charging current.

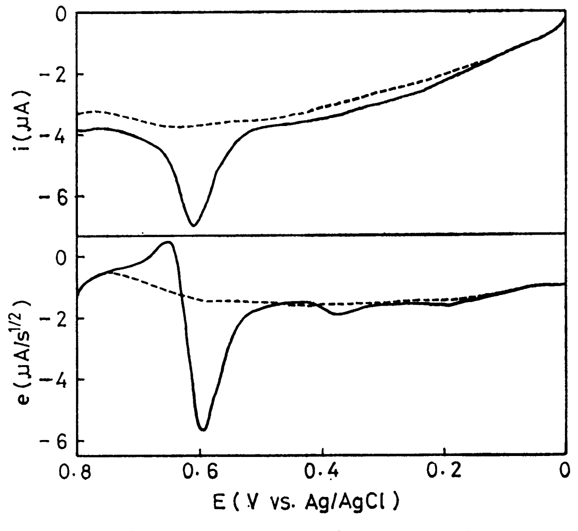

Fig. 1. Typical *e vs. E* curve for the re-dissolution of mercury deposited on the gold disk electrode. Sample: 98.4 nM Hg (II) in 0.25 M H_2SO_4 plus 0.5% $HCl \cdot NH_2OH$, potential scan rate: 100 mV/s, electrode area: 7.07 mm^2, pre-electrolysis potential: 0 V *vs.* Ag/AgCl, pre-electrolysis time: 5 min at rotating electrode and then 30 s at standing one, revolution rate of electrode: 1800 rpm. Dotted lines show the blank curves.

Effect of Supporting Electrolyte. Various supporting electrolytes including sulfuric acid, hydrochloric acid, potassium chloride, potassium thiocyanate, and hydroxylamine hydrochloride were investigated. In a sulfuric acid medium, the anodic stripping waves of mercury appeared at *ca.* 0.9 V, its reproducibility not being

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** 1 M = 1 mol dm^{-3}.

satisfactory. The oxidation of thiocyanate started from *ca.* 0.6 V in thiocyanate medium. A nice stripping wave of mercury was observed in all chloride media (Fig. 1). Since the hydroxylamine hydrochloride medium gave the most reproducible wave in the three chloride media, hydroxylamine hydrochloride was chosen as the supporting electrolyte in addition to 0.25 M H_2SO_4.

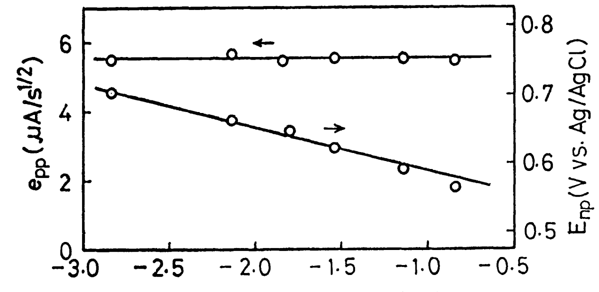

Fig. 2. Dependence of height from the negative peak to the positive one and negative peak potential of *e vs. E* curve for the re-dissolution process on concentration of $HCl \cdot NH_2OH$. Conditions except concentration of $HCl \cdot NH_2OH$ are the same as in Fig. 1.

Figure 2 shows the effect of $HCl \cdot NH_2OH$ concentration in the supporting electrolyte on e_{pp} and E_{np} in the *e vs. E* curve for anodic stripping process. e_{pp} was found to be independent of $HCl \cdot NH_2OH$ concentration. E_{np} shifted to negative potential side with increase in concentration, being linear with its logarithm. This indicates that mercurous ion forms complex with chloride ion:

$$Hg_2^{2+} + pCl^- \rightleftharpoons Hg_2Cl_p^{(2-p)-} \quad (4)$$

the electrode reaction being

$$2Hg + pCl^- \rightleftharpoons Hg_2Cl_p^{(2-p)-} + 2e \quad (5)$$

where p is the number of ligands involved in the complex. The d.c. polarographic half-wave potential for the electrode reaction of Eq. 5 is given by

$$E_{1/2} = E_s - \frac{RT}{2F}\ln\beta - \frac{pRT}{2F}\ln[Cl^-] \quad (6)$$

where E_s is the standard redox potential of $Hg_2^{2+}/2Hg$ couple and β the stability constant of the chemical reaction of Eq. 4. Thus E_{np} in the *e vs. E* curve should vary linearly with the logarithm of chloride concentration, since E_{np} is the simple function of $E_{1/2}$ (Eq. 2). The slope of the line for E_{np} *vs.* log $C_{HCl \cdot NH_2OH}$ was -62 mV, being comparable with the value -58 mV calculated from $-2.303\ RT/F$. The number of ligand involved in the complex is thus two.

For mercury analysis, the concentration of 0.5% $HCl \cdot NH_2OH$ seems to be most suitable, since the gold electrode itself starts to dissolve from about 0.8 V. A small unknown peak appears at *ca.* 0.4 V, the re-dissolution wave of mercury at this concentration appearing at *ca.* 0.6 V, not overlapping with the undesirable waves.

Pre-electrolysis Potential. The dependence of pre-electrolysis potential on e_{pp} in anodic stripping process was examined. e_{pp} was constant, independent of the pre-electrolysis potentials from 0.4 to -0.9 V. The pre-electrolysis potential of 0 V was chosen for mercury analysis. No influence of dissolved oxygen was observed, removal of oxygen in the solution thus being unnecessary.

Dependence of e_{pp} *on* t_p, v, N, *and* C. The relationship between e_{pp} and t_p at different concentrations was measured by keeping all other parameters constant (Fig. 3). e_{pp} was linear with pre-electrolysis time at rotating electrode up to 5 min at concentration of *ca.* 100 nM and 10 min at that of 10 nM. The positive intersepts with ordinate correspond to the pre-electrolysis for 30 s under standing electrode. Figure 4 shows the relationship between e_{pp} and v, and N. The line of e_{pp} *vs.* $v^{1.5}$ passed through the origin, e_{pp} being

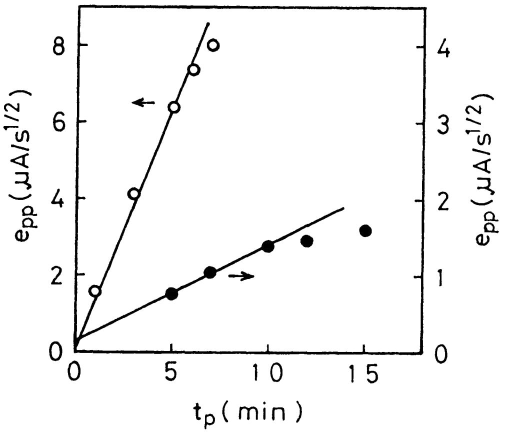

Fig. 3. Relationship between height from the negative peak to the positive one of *e vs. E* curve for the re-dissolution process and pre-electrolysis time at the rotating electrode. Concentration of Hg (II): ○ 98.4 nM, ● 9.8 nM, other conditions are the same as in Fig. 1.

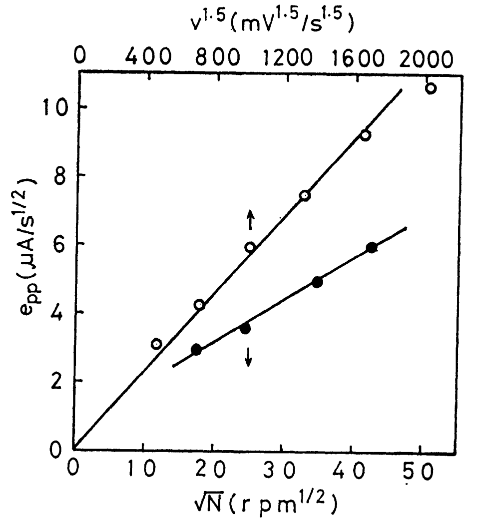

Fig. 4. Relationship between height from the negative peak to the positive one of *e vs. E* curve for the re-dissolution process and potential scan rate (○), and revolution rate of electrode (●). Conditions except potential scan rate (○), and revolution rate of electrode (●) are the same as in Fig. 1.

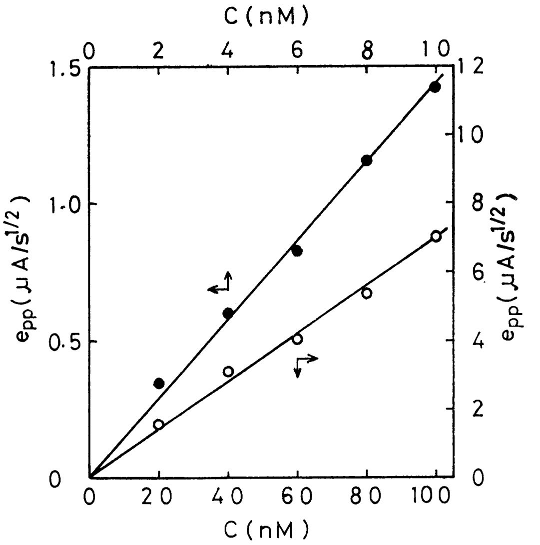

Fig. 5. Relationship between height from the negative peak to the positive one of *e vs. E* curve for the redissolution process and concentrations of Hg (II) present in original solution. Pre-electrolysis time: 5 min for (○), 10 min for (●) at rotating electrode and then 30 s at standing one, other conditions are the same as in Fig. 1.

linear with $N^{1/2}$. Figure 5 shows the relationship between e_{pp} and C at different concentration levels. The pre-electrolysis time employed was 5 min at 10 nM level and 10 min at 1 nM level, respectively. In both cases, e_{pp} was linear with C, the lines passing through the origin.

The results indicate that Eq. 1 applies in the anodic stripping process of mercury from the gold disk electrode.

Dependence of E_{np} *and* W_{pp} *on* C *and* v. The results are given in Table 1. E_{np} is independent of mercury(II) ion concentration to average 0.594 V at the scan rate of 100 mV/s, shifting towards the positive side with increasing scan rate as seen from Eq. 2. W_{pp} averaged *ca.* 52 mV, as compared with the theoretical value of 29 mV for two-electron transfer calculated by means of Eq. 3. This indicates that the electrode reaction (Eq. 5) is not completely reversible.

Table 1. Negative peak potential and width between negative and positive peak of *e vs. E* curve for redissolution process at different concentrations and potential scan rates[a)]

C(nM)	v(mV/s)	E_{np}(V *vs.* Ag/AgCl)	W_{pp}(mV)
19.7	100	0.590	49
39.4	100	0.593	50
59.0	100	0.596	51
78.7	100	0.592	51
98.4	100	0.597	52
98.4	60	0.592	52
98.4	80	0.596	52
98.4	120	0.600	53
98.4	140	0.603	59
98.4	160	0.600	57

a) Supporting electrolyte: 0.25 M H_2SO_4 plus 0.5% HCl·NH_2OH, electrode area: 7.07 mm^2, pre-electrolysis potential: 0 V *vs.* Ag/AgCl, pre-electrolysis time: 5 min at rotating electrode and then 30 s at standing one, revolution rate of electrode: 1800 rpm.

Table 2. Relationship between height from negative to positive peak of *e vs. E* curve for re-dissolution process and sweep number at different concentrations[a)]

Sweep number	e_{pp}(μA/s$^{1/2}$) (A)	e_{pp}(μA/s$^{1/2}$) (B)[b)]
1	1.43	0.33
2	1.46	0.32
3	1.41	0.33
4	1.41	0.30
5	1.50	0.32
6		0.33
7		0.30
8		0.33
av.	1.44	0.32
r.s.d.	±3%	±4%

a) Sample: (A) 9.8 nM, (B) 2.0 nM Hg (II), pre-electrolysis time: 10 min at rotating electrode and then 30 s at standing one, other conditions are the same as in Table 1. b) Working electrode was dipped in aqua regia for 1 min befor each measurement.

Analytical Aspects. Table 2 gives the reproducibility for successive measurements of e_{pp} on the same aliquot at different concentrations. The relative standard deviations were ±3% for 10 nM and ±4% for 2 nM. The detection limit of mercury(II) ion was 0.5 nM at the potential scan rate of 100 mV/s and pre-electrolysis time of 10 min.

Application to Environmental Analysis. In order to examine the applicability of the gold electrode for the determination of mercury(II) ion in natural waters, experiments were carried out to see whether the stripping process can be directly performed in natural water

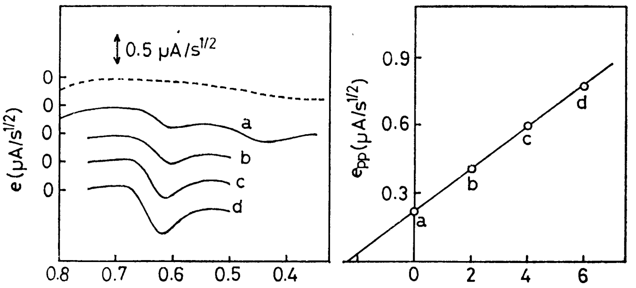

Fig. 6. Performance of anodic stripping semidifferential electroanalysis after medium exchange for 0.25 M H_2SO_4 plus 0.5% HCl·NH_2OH before stripping and regression line for mercury determination by the standard addition method in river water without UV irradiation pretreatment. a: unspiked, b: 2 nM spiked, c: 4 nM spiked, d: 6 nM spiked, pre-electrolysis time: 10 min at rotating electrode and 30 s at standing one after medium exchange, other conditions are the same as in Fig. 1.

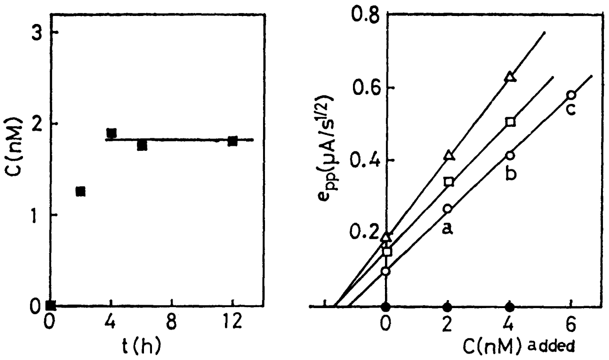

Fig. 7. Effect of pretreatment by UV irradiation on mercury determination in river water by anodic stripping semidifferential electroanalysis after medium exchange. UV irradiation time, *t*, (h): ● 0, ○ 2, □ 6, △ 12, other conditions are the same as in Fig. 6.

spiked with 0.25 M H_2SO_4 and 0.5% $HCl\cdot NH_2OH$. We were unable to establish the conditions including various electrode pretreatment to get a definite stripping peak of mercury. However, satisfactory results were obtained when the medium was exchanged for the supporting electrolyte solution of 0.25 M H_2SO_4 containing 0.5% $HCl\cdot NH_2OH$ after pre-electrolysis and the gold electrode was electrochemically activated before each run (see Experimental).[9] Figure 6 shows the *e vs. E* curves for the re-dissolution process of mercury after medium exchange following pre-electrolysis in the river water spiked with only 0.05 M H_2SO_4 at different standard mercury(II) ion additions, and the regression line for the determination of mercury in the river water with no pretreatment. The relationship between mercury(II) ion concentration added and e_{pp} remained linear. Samples from river waters frequently contain a large amount of dissolved, colloidal, and particulate organic matter. They must be subjected to UV irradiation to release trace mercury bound to, adsorbed on, and occuluded in components of organic matter by photolytic decomposition by addition of a small amount of H_2O_2. Figure 7 shows the effect of UV irradiation on e_{pp} for mercury analysis at different irradiation times. When the sample was not subjected to UV irradiation, no anodic stripping wave was observed even after standard mercury(II) ion addition. UV irradiation for 2 h with a 18 W lamp gave a regression line similar to that in Fig. 6, the slope increasing with increase in irradiation time. This indicates that the photolytic decomposition of organic matter progresses with irradiation time. The regression concentrations coincide with each other at irradiation times longer than 6 h (Fig. 7), the regression concentration indicating concentration at the intercept of regression line with abscissa.

Mercury Speciation in Natural Waters. The chemical forms of trace heavy metals in natural waters can be classified into the following groups with a slight modification of those given by Florence and Batley:[1] (1) simple hydrated metal ions, (2) labile metal-inorganic complexes, (3) labile metal-organic complexes, (4) stable metal-inorganic complexes, (5) stable metal-organic complexes, (6) metal adsorbed on inorganic colloids and particulate matter, (7) metal adsorbed on organic colloids and particulate matter, (8) metal occluded in inorganic colloids and particulate matter, (9) metal occluded in organic colloids and perticulate matter.

Anodic stripping voltammetry measures only the labile metal ions in (1)—(3) detecting no metal in other chemical form. UV irradiation with H_2O_2 oxidizes organic matter in solution to liberate metal in (5), (7), and (9). The metal-complexing and adsorbing capacity of natural water[15–18] can be determined by the standard addition method in anodic stripping voltammetry. When all metal-complexing and adsorbing material has been consumed, there will be a sudden increase in the slope of regression line. The break point represents the complexing and adsorbing capacity of the water. The slope before the break point would depend on the complexing and adsorbing ability of natural water, while the slope after it should agree with that in pure water.

In the chemical speciation analysis of mercury in river water and sea water by the standard addition method with and without UV irradiation pretreatment (Fig. 8), filtration of natural waters was not carried out in order to eliminate the contamination or loss of mercury. The total labile mercury(II) ions in (1)—(3) can be estimated from e_{pp} obtained with no standard addition and without UV irradiation and the slope of calibration curve in Fig. 5. The regression concentration without UV irradiation may represent the total mercury in (1)—(7), while that with UV irradiation the total mercury in (1)—(7) and (9). The coinciding

Table 3. Examples of mercury concentrations in some forms existing in natural waters

Species of mercury	Sample River water	 Sea water
Total ionic mercury	0.2 nM	1.0 nM
Mercury occluded in organic matter	0.9 nM	3.9 nM
Total mercury (excluding mercury occluded in inorganic matter)	1.2 nM	6.0 nM

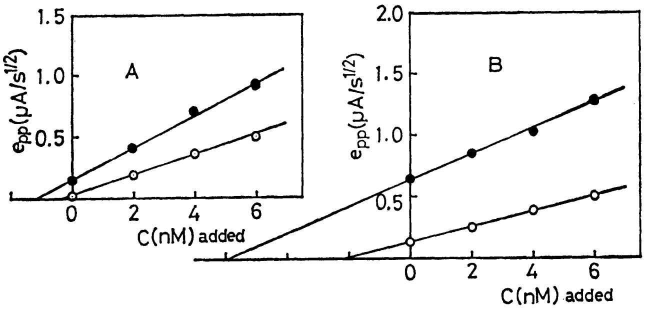

Fig. 8. Typical chemical speciation analysis of mercury in natural waters by the standard addition method with and without UV irradiation pretreatment. Sample: (A) river water, (B) sea water, UV pretreatment: ○ non, ● 12 h at 18 W lamp, other conditions are the same as in Fig. 6.

regression concentrations (Fig. 7) indicate that total mercury excluding (8) can be estimated so far as all mercury in (9) has been liberated, even though the decomposition of organic matter in solution is not complete. The difference between the regression concentrations with and without UV irradiation corresponds to the mercury in (9). When the decomposition of organic matter is complete, e_{pp} obtained with no standard addition and UV irradiation reflects roughly the total mercury in (1)—(3), (5), (7), and (9). Table 3 gives the analytical results of river water and sea water (Fig. 8). The proposed technique for chemical speciation of mercury is applicable to not only inland waters but also sea water.

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