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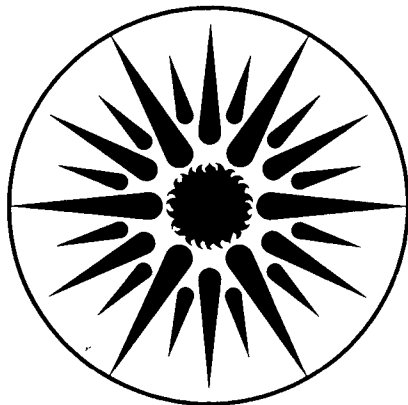
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W.H. Benner

April 1984

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THE DETERMINATION OF PYROLYZABLE OXYGEN IN AMBIENT PARTICULATE MATTER*

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Abstract

A technique is described for the determination of tens of micrograms of oxygen that can be pyrolyzed at $T < 1135^{\circ}\text{C}$. Standard oxygen-containing compounds and filter samples of ambient particulate matter are pyrolyzed in purified N_2 , and oxygen in the pyrolysis products is quantitatively converted to CO by passage over carbon at 1135°C . The evolved CO is detected by NDIR spectrometry or by coulometry after conversion to CO_2 . The application of the technique for the analysis of ambient particulate matter is shown.

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Oxygen is a very common component of organic and inorganic substances and by inference should be a common component of ambient particulate matter. The determination of oxygen in ambient particulate matter would supply insights into the chemistry of haze formation, but little work has been reported in this area of analysis. Clemenson (1) used a cyclotron technique to determine the oxygen content of a few atmospheric aerosol samples. His technique is unique in that total oxygen is determined, as compared with a variety of other techniques (2-10) that, if used to analyze an atmospheric aerosol sample, would only determine a fraction of the oxygen in the sample. For example, a gas-chromatographic technique (2) requires that the substance of interest elutes from a column — a requirement not suitable for nonvolatile substances — and ESCA (4) or Auger (5) spectroscopy is a surface sensitive technique.

We have modified and combined several analytical methods to quantitate pyrolyzable oxygen in ambient particulate matter. The method first reported by Schütze (7) and later modified by Unterzaucher (7) and others (8-10) consists of thermally decomposing the sample and then reacting the oxygen-containing decomposition products with hot carbon to produce CO. The CO is then detected either directly by nondispersed infrared absorption (NDIR) or coulometrically after it is converted to CO₂. With a focus on oxygen-containing molecules, ambient particulate matter in general contains organic oxygen, carbonates, silicates, sulfates, nitrates, metal oxides, water, and adsorbed gases like O₂ and CO₂. The technique reported here detects oxygen in all species that decompose at T < 1135°C — this excludes refractory metal oxides like Al₂O₃. SiO₂ does not decompose at 1135°C but -OH functional groups on the SiO₂ particles can decompose.

Experimental Section

Reagents

Prepurified compressed N₂ was further purified by passage through columns

of $\text{Mg}(\text{ClO}_4)_2$ and P_2O_5 -coated glass beads to remove H_2O vapor and over Cu metal turnings (at 325°C) to remove molecular oxygen. The granular carbon packing and Schütze reagent (acidic I_2O_5) were purchased from Coulometrics, Inc. (Wheat Ridge, Colorado). Oxygen-containing standards were at least 98% pure and were not further purified. The benzoic acid was primary standard grade. All standards were desiccated over P_2O_5 for several days before use. CO_2 was obtained from a cylinder of liquified CO_2 (99.9%). CO calibration gas (primary standard, $40 \pm .4$ ppm CO in N_2) was from Matheson. Ambient particulate matter was collected by filtration in Berkeley, California, onto quartz fiber filters (tissue quartz, Pallflex Products Corp., Putnam, Connecticut, pre-fired at 800°C). The pyrolysis tube was fabricated from quartz tubing.

Apparatus

The quartz pyrolysis tube (2 cm o.d., 0.25 cm wall, 65 cm long; see Fig. 1) was packed with a 4-cm length of granular carbon, and the packing was held in place with 3-cm long quartz wool plugs and was heated in a tube furnace (Lindberg, Watertown, Wisconsin). The scrubbers for purifying the N_2 consisted of vertically mounted columns (1.2 cm dia. x 30 cm) of $\text{Mg}(\text{ClO}_4)_2$ and P_2O_5 -coated glass beads (0.3 cm dia. beads). The Cu metal turnings were packed into a Pyrex tube (2.5 cm dia. x 35 cm) and heated in another tube furnace to 325°C .

Two different detectors were optionally used in this study. The first was a CO_2 coulometer (Coulometrics, Inc.); and when this detector was used, the CO exiting from the pyrolysis tube was oxidized to CO_2 by passage through a packed column (1.2 cm dia. x 15 cm) containing 5 cm of $\text{Mg}(\text{ClO}_4)_2$, 5 cm of Schütze reagent, and 3 cm of $\text{Na}_2\text{S}_2\text{O}_8$. Each packing was isolated with small quartz wool plugs. The $\text{Mg}(\text{ClO}_4)_2$ protected the I_2O_5 from moisture when the system was opened, the I_2O_5 oxidized CO to CO_2 , and the $\text{Na}_2\text{S}_2\text{O}_8$ absorbed liberated I_2 . With this detector, the flowstream of N_2 from the pyrolysis tube was forced through the CO_2 coulometer without any added diluent N_2 . This detector integrates the

[CO₂] over a selectable time period and is not flowrate sensitive below several hundred ml/min. The difference between an operational blank and a sample was the net oxygen (detected as CO₂).

An NDIR CO detector (0-100 ppm, Mine Safety Appliance Co., Pittsburgh, Pennsylvania) was the second detector used in this study. The signal from this detector was fed through an 11-bit analog-to-digital converter to a small computer (Commodore 2001 Microcomputer). A plotter (Commodore Series 2022) was used to observe the data. The computer software permitted baseline subtraction and peak area integration. The flow system in Fig. 1, into which this detector was plumbed, was designed so that the flow of N₂ through the pyrolysis tube was diluted during entrance into the detector. This permitted the background signal to be low (~ 4 ppm CO) and yet provided good sensitivity. The design of the flow system is not sensitive to small N₂ flowrate changes through the pyrolysis tube for two reasons: 1) The background level of CO is not flowrate dependent because all major oxygen impurities in the N₂ were removed, and 2) all the effluent from the pyrolysis tube entered the detector (N₂ was added upstream of the NDIR detector to supply 1.98 l/min of total flow through the detector.). In Fig. 1 the only flowrate that needs to be accurately and precisely controlled is the flowrate through the NDIR detector. The optimal flowrate of N₂ through the carbon packing is between 10 and 50 ml/min. Below 10 ml/min it becomes difficult to adjust the flowrate with a rotameter, and above 50 ml/min the decomposition products don't have time to react completely with the hot carbon.

Gas samples were injected into the pyrolysis tube via a septum. A multiport valve (Condyne, La Canada, California) was used to change the direction of flow through the pyrolysis tube. Solid samples and filter samples were loaded into a small platinum boat (formed from 5 mil Pt foil) and placed into the cool zone of the pyrolysis tube while backflushing with N₂ so that room air did not contact

the hot carbon. After a sample was loaded, the pyrolysis tube was resealed (at the O-ring seal) and any entrained air was backflushed from the system (~ 4 min). Then the multiport valve was switched; and finally after the CO detector stabilized, the sample boat was slid into the hot zone with a magnetically moved plunger.

Results and Discussion

The digitized analog NDIR detector signal deviated less than 0.5% from the analog signal in the 1.0-99.0 ppm CO range. This good digitization was, in part, due to the use of a 12-bit A/D converter rather than the more commonly used 8-bit A/D. Table 1 shows the results of the determination of oxygen in a variety of oxygenated organic compounds, $(\text{NH}_4)_2\text{SO}_4$, and NH_4NO_3 . These chemicals were either weighed into an empty boat or solutions containing these chemicals were evaporated onto 1-cm^2 discs of quartz fiber filter material. Also, several microliter volumes of CO_2 were analyzed and served as a convenient way to check out the system.

Direct injections of CO_2 into the coulometer (footnote f, Table 1) shows good recovery (100.4%) with a standard deviation (σ) of 2.3%. Passage of CO_2 through the hot carbon and subsequent conversion of the evolved CO back to CO_2 did not affect the accuracy (footnote g, Table 1). The injection of CO_2 upstream of the hot carbon with NDIR detection of the evolved CO also gave good recovery ($100.0 \pm 0.4\%$). Each detector offers certain advantages over the other one. The coulometer enables larger samples to be analyzed: 55-400 $\mu\text{g O}$ is a good operating range. Below 55 $\mu\text{g O}$, one is operating below the limit of quantitation (LOQ) (See Table 2) for the coulometer; and above $\sim 400 \mu\text{g O}$, complete elution of CO_2 from the I_2O_5 column is delayed. An increase in analysis time would solve the elution problem. The coulometer offers the simplest operation in that, aside from the required I_2O_5 column, it is self contained. The NDIR detector requires

a small computer for signal processing, particularly for peak area determination; but because the detector signal was tracked graphically with a computer-driven plotter, much better baseline subtraction can be obtained with the computer system. NDIR detection offers the best sensitivity with an LOQ of 10.9 $\mu\text{g O}$ for the NDIR system.

A number of compounds were analyzed to determine the appropriateness of this technique for analyzing samples of ambient particulate matter. The compounds presented in Table 1 have a variety of oxygen functional groups, some of which have been observed in ambient air and smog chamber samples (11,12). The recovery of oxygen for each of these compounds is shown in Table 2, and the overall results (percent recovery, average deviation, etc.) are summarized in Table 2. The recovery for all standards was $101.4 \pm 4.1\%$ ($n = 65$). For standards analyzed with the coulometer, recovery was $101.2 \pm 3.9\%$ ($n = 43$), and recovery was $101.8 \pm 4.5\%$ ($n = 22$) for standards analyzed with NDIR detection. The analytical limits of detection (LOD) and LOQ were calculated according to American Chemical Society recommendations (14), as shown in Table 2. Either of the detectors is suitable for analyzing oxygen in ambient particulate matter, but the NDIR system is probably more appropriate because of the low concentrations of oxygen expected on an air filter sample.

A few comments should be made about preparation of low-concentration oxygen standards and their analysis. Benzoic acid was suggested as a reliable standard for this type of analysis (13) because it's available commercially in a primary standard grade. Our experience has been that only fresh primary standard benzoic acid gave results 100% of the expected value. For example, primary standard benzoic acid that sat on a laboratory shelf since 1953 gave results about 25% high, while fresh primary standard benzoic acid gave results as presented in Table 1; however, the fresh benzoic acid decomposed to give 110-115% values

around 1 year after purchase. Preparation of standards containing a few micrograms O from benzoic acid is complicated because of its tendency to sublime. Gentle heating or evacuation caused losses of benzoic acid. Primary standard KHP or Na_2CO_3 is also a good standard because samples, if prepared from aqueous solutions, can be baked dry. But salts like KHP and Na_2CO_3 volatilize when placed in the pyrolysis tube, and the K or Na soon devitrifies the hot quartz so that it becomes translucent and breaks. Therefore, Na- and K-salts are not standards of choice for this pyrolysis tube technique. Ammonium sulfate is available as a primary standard for N analyses and appears to be a good choice for an oxygen standard, too. Solutions of $(\text{NH}_4)_2\text{SO}_4$ can be evaporated onto discs of quartz fiber filter material at 110°C without decomposition. Heating circumvents the need for overnight desiccation that is required to dry aqueous solutions of organic oxygen compounds. Organic solvents could be used to prepare standards of organic oxygen compounds, but several organic acids we tried were not soluble in nonoxygenated solvents. Evaporation of oxygenated solvents on quartz fiber filters always increased the blank value either due to incomplete evaporation or the presence of contaminants. Some variability in analysis was introduced during preparation of low-level oxygen standards because microliter injections of CO_2 (Table 1, NDIR detector) would be analyzed with $\sigma = \pm 0.4\%$, while evaporated solutions of $(\text{NH}_4)_2\text{SO}_4$, KHP, and glutaric acid showed larger deviations ($\sigma = \pm 3.8\%, \pm 4.0\%, \pm 5.7\%$, respectively).

Figures 2a and 2b show the linearity and the scatter than can be expected for the analysis of standards prepared by the evaporation of solutions onto 1-cm^2 quartz fiber discs. Each datum point is the determination of oxygen on a 1-cm^2 disc.

Replicate analyses of two ambient air filter samples are presented in Table 3 to show the applicability of this technique. A high-volume filter

sampler (~ 1100 lpm of ambient air was drawn through 400-cm^2 prefired quartz fiber filters for several hours.) was used to collect samples in Berkeley, California. Multiple 1-cm^2 discs of these filters were dessicated for 12 hrs over P_2O_5 and then analyzed for oxygen. A separate technique (15) was used to analyze for particulate carbon. The results indicate that the volumetric flowrates frequently used to sample ambient particulate matter will collect enough oxygenated particulate matter so that 1 cm^2 of filter will give a signal above the LOQ in the NDIR analysis scheme. Comparison of the carbon loadings in Table 3 to Table 2 in Novakov (16) indicates that these loadings are typical for many United States sampling sites.

Conclusions

A technique is described by which microgram quantities of oxygen can be determined in ambient particulate matter. The limitation is that the oxygen-containing species must decompose at $T < 1135^\circ\text{C}$ to be analyzed. In ambient particulate matter, this excludes silica and metal oxides like Al_2O_3 . A sample is pyrolyzed at 1135°C under dry N_2 , and oxygen in the pyrolysis products is converted quantitatively to CO by passage over carbon at 1135°C . The CO is then detected either by NDIR absorption or by CO_2 coulometry after it's converted to CO_2 . For the NDIR detector, $\text{LOD} = 8\text{ }\mu\text{g O}$ and $\text{LOQ} = 11\text{ }\mu\text{g O}$, while $\text{LOD} = 35\text{ }\mu\text{g O}$ and $\text{LOQ} = 55\text{ }\mu\text{g O}$ for the coulometric detector.

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Table 1. Oxygen in standard compounds.

Sample	Sample Type ^a	Detector ^d	Sample wt (μgO)	μgO Recovered (blank corrected) ^b	% Recovery $\bar{x} \pm s$ (n)	Deviation ^c (μgO)	Ave Deviation $\bar{x} \pm s$
Ammonium nitrate	S	C	612.6	605.0	99.0 ± 0.3 (2)	7.6	
			406.0	402.8		3.2	
Ammonium sulfate	W	N	7.1	6.8	99.8 ± 3.8 ^e (7)	0.3	0.7 ± 0.5
			14.3	15.1		0.8	
			14.3	14.6		0.3	
			28.6	29.0		0.4	
			35.7	34.9		0.8	
			35.7	33.9		1.8	
			35.7	36.1		0.4	
Anthracene	S	C	1216.	7.2	0.6 ± 0.1 (2)	—	
			1517.	10.4		—	
Benzaldehyde 2,4-dihydroxy	S	C	318.0	343.4	108.1 ± 0.1 (2)	25.4	25.6 ± 0.4
			318.7	344.6		25.9	
Benzene, p-deithoxy	S	C	304.8	305.1	101.9 ± 2.5 (2)	0.3	5.1 ± 6.7
			268.4	278.2		9.8	
Benzoic acid	S	C	413.3	427.5	103.0 ± 3.7 (12)	14.2	10.7 ± 7.9
			406.4	424.5		18.1	
			319.9	330.9		11.0	
			322.7	331.1		8.4	
			198.9	196.4		2.5	
			140.9	150.3		9.4	
			316.8	325.8		9.0	
			155.8	160.6		4.8	
			240.5	255.8		15.3	
			322.8	352.4		29.6	
			103.5	98.5		5.0	
			177.5	178.5		1.0	
Benzoic acid, m-hydroxy	S	C	316.7	319.1	100.8 ± 0.1 (2)	2.4	2.3 ± 0.1
			252.7	254.9		2.2	
Blank boat (for weighed standards)		C	0	31.4	26.1 ± 2.9 (10)		
			0	23.0			
			0	28.3			
			0	22.6			
			0	27.4			
			0	22.6			
			0	26.2			
			0	25.4			
			0	28.2			
			0	25.5			
Blank boat + 1 cm ² disc of quartz filter blanks		N	0	6.2	6.9 ± 0.4 (7)		
			0	7.4			
			0	7.1			
			0	6.4			
			0	7.2			
			0	6.9			
			0	7.0			

Table 1. Continued.

Sample	Sample Type ^a	Detector ^d	Sample wt (μgO)	μgO Recovered (blank corrected) ^b	% Recovery $\bar{x} \pm s$ (n)	Deviation ^c (μgO)	Ave Deviation $\bar{x} \pm s$
CO ₂ ^f	G	C	653.1	644.8	100.4 ± 2.3 (10)	8.3	13.1 ± 7.0
				651.7		1.4	
				678.1		25.0	
				672.5		19.4	
				671.2		18.1	
				668.8		15.7	
				638.9		14.2	
				644.0		9.1	
				646.9		6.2	
639.2	13.9						
CO ₂ ^g	G	C	653.1	648.5 657.5	100.0 ± 1.0 (2)	4.6 4.4	4.5 ± 0.1
CO ₂	G	N	26.1	26.1	100.0 ± 0.4 (5)	0.0	0.3 ± 0.2
				26.5		0.4	
				25.8		0.3	
				25.6		0.5	
				26.4		0.3	
Glutaric acid	S	C	365.6	358.3	101.3 ± 3.0 (4)	7.3	8.7 ± 8.2
				385.9		20.7	
				295.3		3.2	
				431.0		3.7	
	W	N		8.5	104.0 ± 5.7 ^h (8)	0.3	1.4 ± 0.9
				8.5		0.2	
				17.0		2.4	
				17.0		1.4	
				34.1		2.6	
				45.6		1.1	
				45.6		1.3	
				45.6		2.3	
				119.7		4.0	
				229.5		5.8	
250.3	7.6						
188.8	1.7						
Myristic acid	S	C	119.7	123.7	102.5 ± 1.1 (4)	4.0	4.8 ± 2.5
				229.5		5.8	
				250.3		7.6	
				188.8		1.7	
Phthalic acid	S	C	215.7	199.5	92.2 ± 1.5 (3)	16.2	18.3 ± 1.9
				290.2		18.6	
				212.1		20.0	
Phthalic acid, monopotassium salt (KHP)	W	N	17.2	103.0 ± 4.0 (2)	0.0	0.4 ± 0.6	
			17.2		0.9		

^aG ≡ gas injection; S ≡ solid, weighed into boat; W ≡ water solution evaporated onto 1 cm² disc of quartz fiber filter.

^bExcept blank boats.

^crecovered O - expected O

^dC ≡ CO₂ coulometer, N ≡ NDIR CO detector

^eSee Figure

^fCO₂ injected after I₂O₅ column.

^gCO₂ injected upstream of hot carbon (CO₂ → 2CO → 2CO₂).

^hSee Figure 2a.

Table 2. Summary of analyses of oxygen standards.

Detector	Overall recovery		Ave. deviation ^a	Sample size range
	% recovery ± σ	(n)		
Coulometer	101.2 ± 3.9	(43)	10.7 μg O	101-678 μg O
NDIR	101.8 ± 4.5	(22)	0.9 μg O	7.1-45.6 μg O
Overall	101.4 ± 4.1	(65)		

$$\text{Limits of detection} = \text{blank} + 3 \sigma_{\text{blank}}$$

$$\text{Coulometer: } 26.1 + 3(2.9) = 34.8 \mu\text{g O}$$

$$\text{NDIR: } 6.9 + 3(0.4) = 8.1 \mu\text{g O}$$

$$\text{Limits of quantitation} = \text{blank} + 10 \sigma_{\text{blank}}$$

$$\text{Coulometer: } 26.1 + 10(2.9) = 55.1 \mu\text{g O}$$

$$\text{NDIR: } 6.9 + 10(0.4) = 10.9 \mu\text{g O}$$

$$^a \text{Ave. dev.} = \frac{1}{n} \sum_{i=1}^n \frac{|\text{expected-recovered}|}{\text{expected}}$$

Table 3. Analysis of filtered ambient particulate matter.

Sample			$\mu\text{g O/cm}^2$ ^a	$\mu\text{g O/m}^3$	$\mu\text{g C/cm}^2$	$\mu\text{g C/m}^3$
Berkeley, CA	1000-1715 hrs	11 Mar 81	20.5	16.3	13.7	11.0
"	"	20.3				
"	"	20.2				
"	"	20.2				
Berkeley, CA	1800-0600 hrs	11-12 Mar 81	21.0	10.1	12.9	6.3
"	"	20.5				

^aBlank corrected replicates

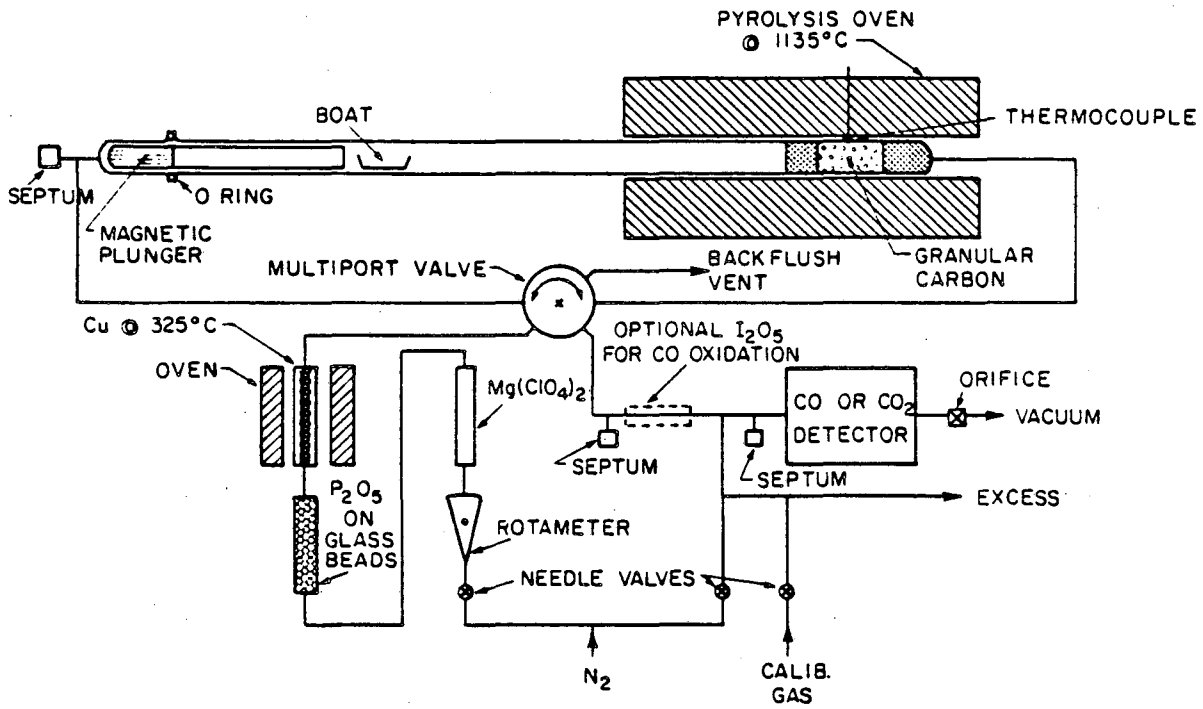
Figure Captions

Figure 1. Schematic drawing of oxygen analysis system. (XBL 833-127)

Figure 2a (XBL 833-128)

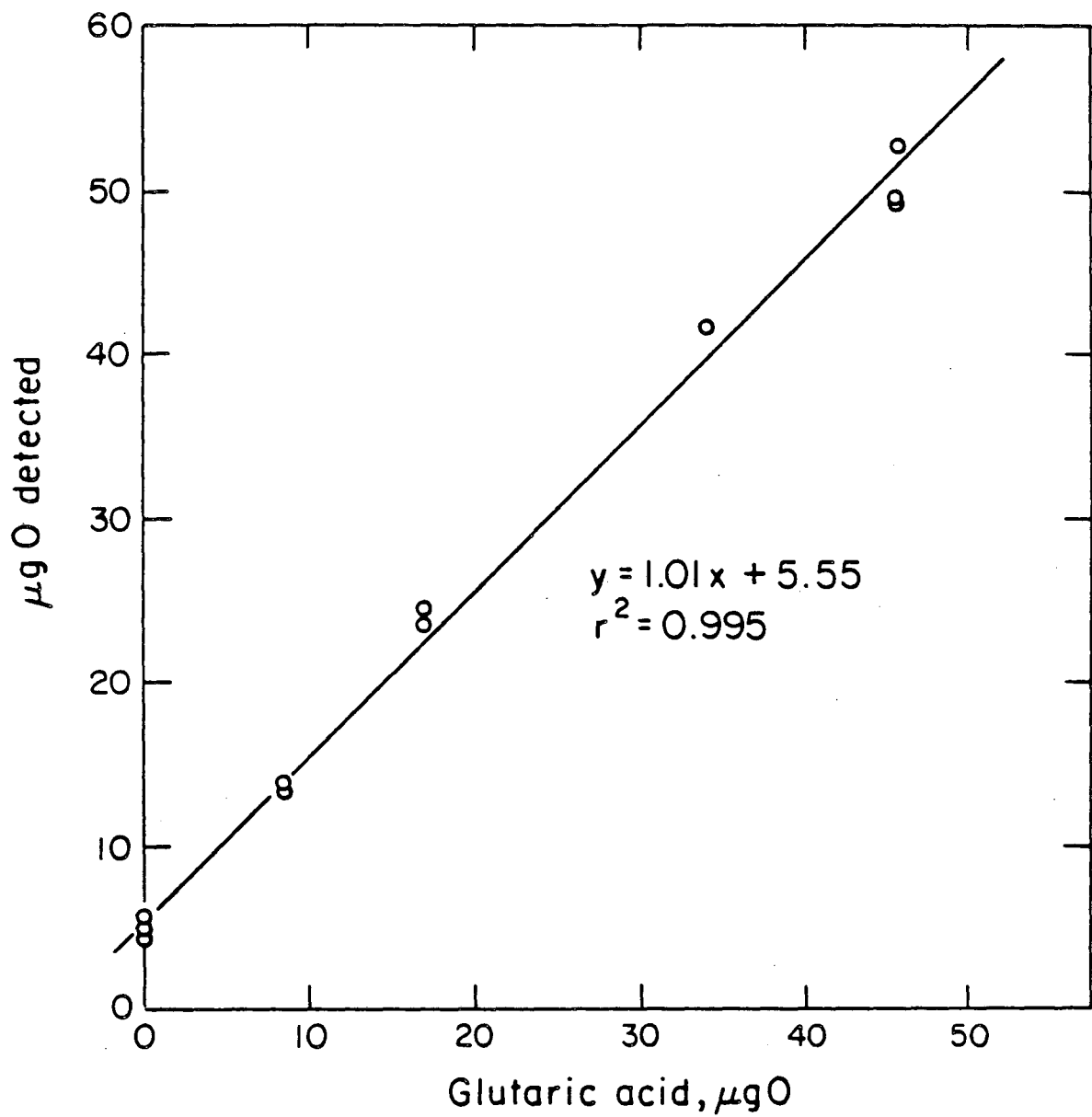
Recovery of oxygen in standards.

2b (XBL 833-129)



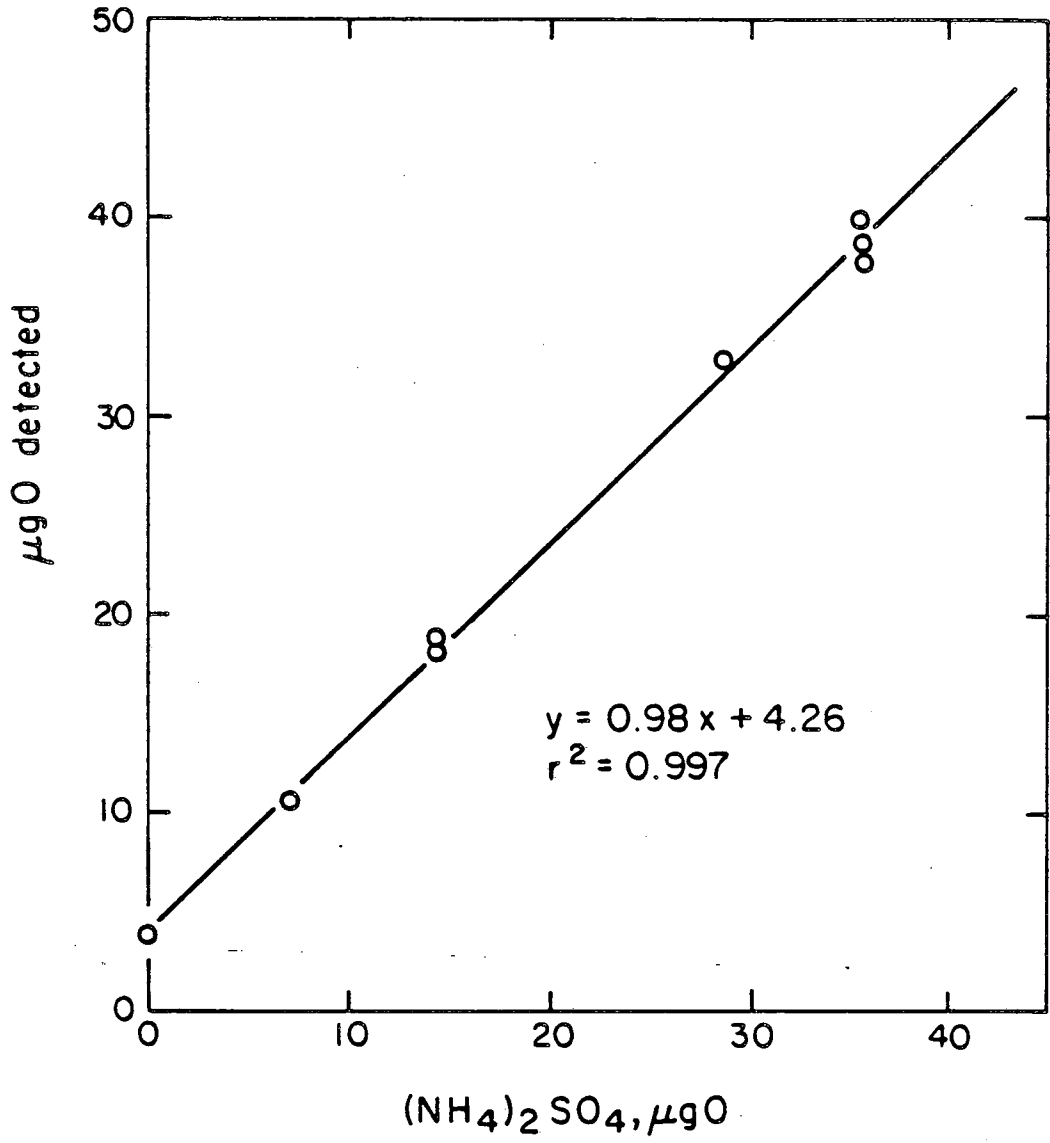
XBL 833-127

Figure 1



XBL 833-128

Figure 2a



XBL 833-129

Figure 2b

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