

Analysis of Sulfites in Foods by Ion Exclusion Chromatography with Electrochemical Detection

New method provides faster, more sensitive, and interference-free analysis of sulfites than current methods

H.-J. Kim, G.Y. Park, and Y.-K. Kim

□ ON JULY 9, 1986, the Food and Drug Administration published two regulations regarding sulfites in foods. In the first regulation, FDA required that the presence of sulfiting agents in foods at levels of 10 ppm or higher be declared, effective January 9, 1987, whether they have been added directly or indirectly via the ingredients of the food (FDA, 1986a). In the second regulation, FDA revoked the Generally Recognized as Safe (GRAS) status of sulfites for use on fruits and vegetables intended to be served or sold raw to consumers, effective August 8, 1986 (FDA, 1986b).

Sulfiting agents are used in foods for various reasons (Wedzicha, 1984; IFT, 1986). Two important roles are the prevention of enzymatic browning in fresh fruits and vegetables and the control of non-enzymatic browning in processed foods. The FDA regulation revoking the GRAS status of sulfite on fresh produce appears appropriate, because sulfited fresh vegetables such as lettuce represent a major portion of the sulfite consumed (IFT, 1986). Also, control of enzymatic browning is relatively easy, and several sulfite substitute formulas are commercially available. On the other hand, control of non-enzymatic browning is much more difficult, and no substitute seems to be at hand.

In the absence of an alternative, it would be desirable to minimize the amount of sulfite in processed foods while maintaining the quality and shelf life of the foods. The current sulfite level observed in some samples of foods (5,800 ppm in freeze-dried green bell pepper dice and 1,600 ppm in golden raisin) may be well above the mini-

mum requirement for each product. To determine the minimum level of sulfite in different foods, a reliable analytical technique for sulfite is needed. Preferably, the technique should be able to distinguish between free and bound sulfite, because the hypersensitive reaction might be to free, but not to bound, sulfite.

More importantly, for enforcement of and compliance with the labeling regulation, an analytical technique reliable at the 10 ppm level is needed. The widely accepted method—the modified Monier-Williams method (AOAC, 1984)—is time-consuming and subject to interference at low sulfite levels. FDA suggested several modifications to overcome some of the problems with the method (FDA, 1986a). Still, the modified Monier-Williams method requires distillation for 1.75 hr. In response to comments about the inappropriateness of the procedure for quality control use, FDA stated that “processors are under no obligation to use the designated analytical procedure for quality control or for any other purpose” (FDA, 1986a). Therefore, it would be desirable to develop an alternative technique that is not only sensitive and interference-free but also fast and easy-to-use for quality control and compliance test purposes.

Several alternative techniques for the measurement of sulfites in

foods are summarized in Table 1. In this article, we present several key features of a new ion exclusion chromatographic technique that make it an attractive alternative to the standard AOAC methods.

New Method Strategy

In the Monier-Williams method, sulfite is removed from the food matrix by distillation in strong acid and detected by titration with alkali after oxidation of the sulfur dioxide to sulfuric acid by H_2O_2 . Some volatile materials such as acetic acid and other sulfur compounds tend to codistill with sulfur dioxide. The detection method also lacks specificity because titration with alkali measures the total acidity in the trapping solution but not necessarily the sulfuric acid derived from sulfite.

Recently, ion chromatographic methods have received much attention. In most approaches, ion exchange chromatography is used to separate sulfite from other interfering compounds in the distillate and to improve the sensitivity of detection (Sullivan and Smith, 1985; Anderson et al., 1986; Cooper et al., 1986). Our strategy was to replace the time-consuming distillation with a rapid extraction and to achieve the needed specificity by a selective chromatographic separation and detection (Kim and Kim, 1986).

The basic procedure for our ion exclusion chromatographic technique for sulfite analysis has been published by Kim and Kim (1986). Two modifications have been made to reduce the analysis time and to improve the separation. The revised procedure is outlined in Figure 1. A Brownlee Polypore H high-

Authors H.-J. Kim and Park are Senior Chemist and Physical Science Technician, respectively, with the Food Engineering Directorate, and author Y.-K. Kim is Research Chemist with the Science and Advanced Technology Directorate, U.S. Army Natick Research, Development and Engineering Center, Natick, MA 01760

speed anion exclusion column (sulfonated polystyrene/divinylbenzene, 4.6 × 100 mm) is used. A 6 mM, instead of 5 mM, H₂SO₄ solution is used as eluant, because a better separation of sulfite from ascorbic acid is achieved. Under these conditions, sulfite is eluted from the column about 2 min after injection.

New Method Features

The new method has several advantages over the previous methods:

1. **It Is Fast.** The most time-consuming step in the Monier-Williams method and recently published ion chromatographic methods is the distillation (15–105 min). Our method uses a 1-min extraction with Polytron instead of distillation. The separation achieved by distillation in the previous methods is now achieved by chromatography on a high-speed anion exclusion column within several minutes. The detection of sulfite that is done by titration with alkali in the Monier-

Williams method and by other methods such as titration with iodine (DeVries et al., 1986) or colorimetry (Jennings et al., 1978; Ogawa et al., 1979; Madison and Wharton, 1986) is done amperometrically in line with the separation. The printer/integrator can calculate the sulfite concentration in the sample in line with the electrochemical detector. The whole analysis, including weighing, extraction, filtration, separation, detection, and data reduction, can be carried out within 10 min, which compares very favorably with any other method for sulfite analysis.

2. **It Is Sensitive.** To comply with the FDA labeling regulation, it is important to be able to measure sulfite at the 10 ppm level in foods. If a food containing 10 ppm sulfite is extracted with a 100 times excess of the extracting solution, the sulfite concentration in the extract will be 0.1 ppm. We have demonstrated that using an electrochemical detector under our experimental conditions, one can detect 0.1 ppm

SO₂ in solution with a signal-to-noise (S/N) ratio of approximately 20. Moreover, the detector response is linear in this concentration range. Therefore, sulfite present at 10 ppm in the foods can be determined easily by comparing the signal intensity of the sample with that of the standard solution containing a known amount of sulfite. Sulfite present in foods at far below the 10 ppm level can also be determined by increasing the sample-to-extractant ratio and centrifuging the extract before filtration if necessary. Sulfite present at higher levels can be determined similarly, because the detector response is linear up to 20 ppm SO₂ in the extract if a 20-μL sample injection loop is used. Therefore, if sulfite is diluted 100-fold during extraction, sulfite present at anywhere between 2 and 2,000 ppm can be determined by the same procedure.

The chromatograms for flaked coconut in Figure 2 demonstrate the sensitivity of the present method. One gram of coconut sample

Table 1—Alternative Techniques for measurement of free and total sulfite in foods

Sulfite	Treatment	Separation	Detection	Reference		
Free	Acid	None	Titration with iodine	AOAC (1984)		
		Gas chromatography	Flame photometric detection	Hamano et al. (1979)		
	None	Cold N ₂ bubbling	H ₂ O ₂ , titration with NaOH	Fujita et al. (1979)		
		Cold N ₂ bubbling	Color with p-rosaniline	Ogawa et al. (1979)		
Acid	None	Polarography	Bruno et al. (1979)			
	Ion exclusion chromatography	Electrochemical detection	Kim and Kim (1986)			
Total	Acid	Distillation	H ₂ O ₂ , titration with NaOH	AOAC (1984)		
		Distillation	Color with p-rosaniline	Jennings et al. (1978)		
		Distillation	Color with p-rosaniline	Ogawa et al. (1979)		
		Distillation/ion exchange chromatography	Conductivity detection	Sullivan and Smith (1985)		
		Distillation/ion exchange chromatography	Conductivity/electrochemical detection	Anderson et al. (1986)		
		Distillation/ion exchange chromatography	Conductivity detection	Cooper et al. (1986)		
		Distillation	Color with Eilman's reagent	Madison and Wharton (1986)		
		Distillation	Titration with iodine	DeVries et al. (1986)		
		Total	Alkali	None	Color with p-rosaniline	AOAC (1984)
				None	Titration with iodine	Ponting and Johnson (1945)
Gas chromatography	Flame photometric detection			Hamano et al. (1979)		
None	Polarography			Bruno et al. (1979)		
None	Reaction with sulfite oxidase			Beutler (1984)		
Flow-injection analysis	Reaction with malachite green			Sullivan et al. (1986)		
Total	Alkali	Ion exchange chromatography	Conductivity detection	Cooper et al. (1986)		
		Ion exclusion chromatography	Electrochemical detection	Kim and Kim (1986)		

was homogenized with 50 mL of pH 8.9 extraction buffer. The area under the sulfite peak at around 2 min corresponds to 0.196 ppm SO_2 , which translates to 9.8 ppm of total SO_2 in the coconut. The area under

the peak for free sulfite shows less than 0.01 ppm SO_2 ; therefore the amount of free sulfite in the coconut is less than 0.5 ppm.

3. It Is Selective. In the Monier-Williams method, the sum of the

free and reversibly bound sulfite is separated from the rest of the food by distillation with strong acid. Even though the results thus obtained are reliable in most cases, the method is subject to interfer-

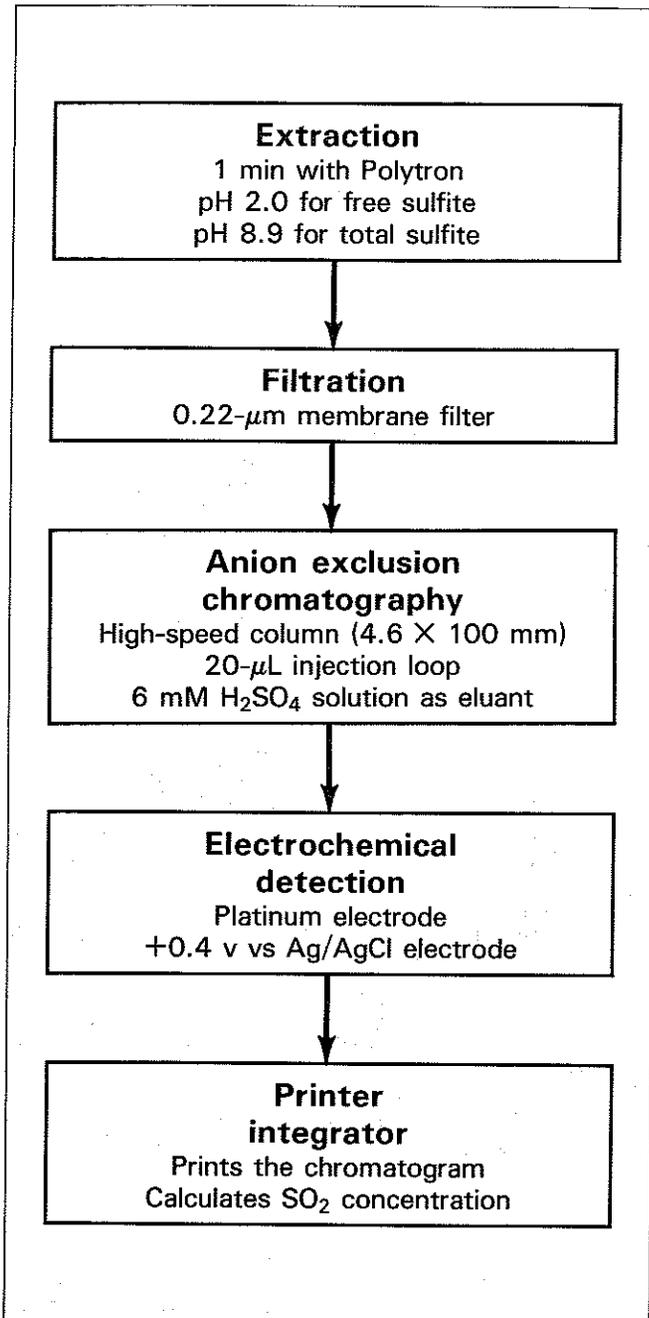


Fig. 1 (above)—Outline of Experimental Procedure for ion exclusion chromatographic analysis of sulfites in foods

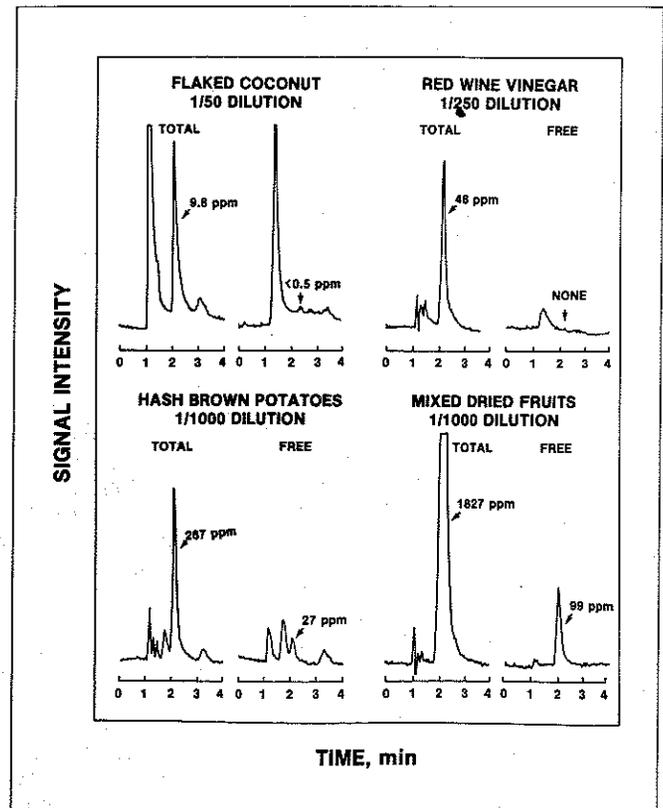


Fig. 2 (above)—Chromatograms Showing Total and Free Sulfite in four different foods with different levels of sulfite; numbers represent SO_2 concentration in ppm

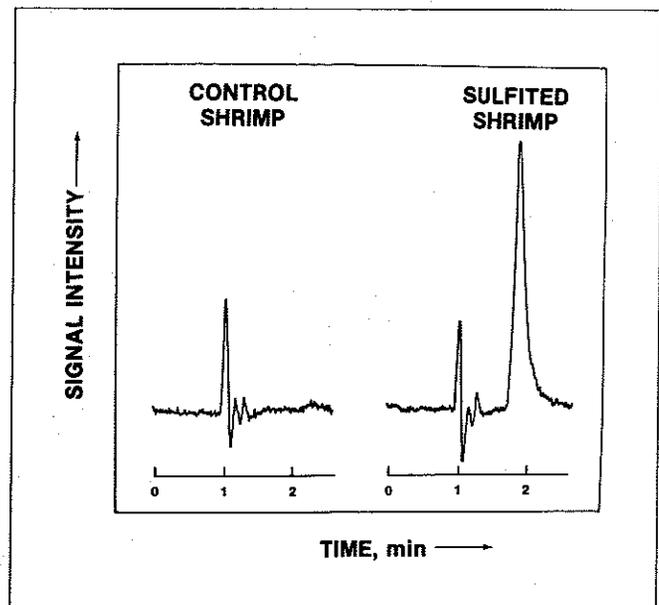


Fig. 3 (at right)—Chromatograms Obtained from Control Shrimp and Shrimp Dip-Treated in 1.25% sodium metabisulfite solution for 1 min. The sulfite peak with 2 min retention time on the right corresponds to 268 ppm SO_2 in the shrimp

ence when volatile compounds are present in the food in significant amounts.

In the new method, the selectivity is introduced in two steps, i.e., in the column and in the detector. Separation on the anion exclusion column is achieved by a combination of Donnan exclusion, partitioning, and size exclusion (Jupille et al., 1981; Fritz et al., 1982). Of the various water-soluble components in the food extract, strong anions are repelled by the negatively charged groups of the resin, and the cations are retained on the column. Organic acids and alcohols are the primary compounds in foods that show reasonable retention time on the anion exclusion column. The pK_1 of H_2SO_3 is 1.81, and the pH of the eluant is about 2. Therefore, the property of H_2SO_3 as a weak acid is fully utilized on the anion exclusion column with the pH 2 eluant. On the other hand, at the typical operating pH of an anion exchange column (pH 8–10), both weak and strong acids are fully ionized, and the coelution of such divalent anions as HPO_4^{-2} with SO_3^{-2} is a potential problem. Further selectivity is achieved by the electrochemical detector, which only detects the compounds oxidizable at the applied voltage at the acidic pH.

The chromatograms for shrimp in Figure 3 demonstrate the combined selectivity of the anion exclusion chromatographic separation and the electrochemical detection. From the control shrimp, no signal was observed other than the system peak at around 1 min. From sulfited shrimp (shrimp dipped in a 1.25% sodium metabisulfite solution for 1 min), a single sulfite peak was observed at around 2 min, corresponding to 268 ppm SO_2 . Cooper et al. (1986) observed more than five peaks from sulfited shrimp after alkaline/formaldehyde extraction and separation on an anion exchange column with conductivity detection. They also observed an interference corresponding to 37 ppm SO_2 coeluting with sulfite.

The chromatograms in Figure 4 demonstrate the selectivity of the electrochemical detection compared with the spectrophotometric detection. The UV detection at 210 nm shows at least five overlapping peak for peppers in vinegar. Of the coeluting compounds at around 2

Table 2—Comparison of Total Sulfite in Foods measured by the Monier-Williams method (AOAC, 1984) and the Kim and Kim (1986) method

Sample	Total SO_2 (ppm) ^a	
	Monier-Williams method	Kim and Kim method
Golden raisin	1,601	1,555
Instant mashed potatoes	390	488
Lemon juice	255	278
Dip-treated shrimp ^b	231	268
Wine vinegar	116 ^c , 49 ^d	48
Instant vegetable soup	41	43
Flaked coconut	11.1	9.8
Beer	0.9	<0.5

^aAverage of duplicate measurements
^bDipped in 1.25% sodium metabisulfite solution for 1 min
^cUncorrected for acetic acid
^dCorrected for acetic acid (see text)

min, the electrochemical detector selectively detects the sulfite.

The selectivity, i.e., the lack of interference, of the new method is best demonstrated by the results for wine vinegar shown in Table 2. The results for wine vinegar were 116 ppm SO_2 by the Monier-Williams method and 48 ppm SO_2 by the new method. We determined the acetic acid concentration in the wine vinegar to be 4.6% by separating the acetic acid on the same anion exclusion column and detecting it at 210 nm. When a Monier-Williams run was made with a 4.6% acetic acid solution in water, a blank reading of 67 ppm was obtained. After the correction for acetic acid, the result was 49 ppm SO_2 in the wine vinegar, which was in excellent agreement with the new method. Clearly, sulfite is separated and detected selectively from various food components in the extract by the new method.

4. It Is Reliable. However fast, sensitive, and selective it may be, the new method would be of little value if the results obtained were unreliable. We checked the accuracy of the method in two ways. First, the recovery of added sulfite was investigated. When diluted lemon juice containing 11 ppm SO_2 was spiked with 10, 30, and 50 ppm SO_2 and total sulfite was determined by the new method, 11, 31, and 55 ppm SO_2 were recovered from the juice sample, respectively. Second, the results obtained by the Monier-Williams method and the new method were compared for samples containing sulfite at a wide range of levels. As shown in Table 2, a good

agreement was observed from a variety of foods containing sulfite at less than 10 ppm to 1,600 ppm. The only major discrepancy was observed in wine vinegar, which contains 4.6% acetic acid. As discussed above, an excellent agreement was obtained after correction for acetic acid (Table 2), which demonstrates that the new method is more accurate than the Monier-Williams method when a significant amount of volatile, interfering compounds is present in the food. Other such examples may include dehydrated onions and cabbage.

In some cases, such as instant mashed potatoes, the amount of total sulfite determined by the new method was 10–20% higher than that by the Monier-Williams method. This difference might be due to a more complete release of the bound sulfite at the alkaline pH used in the new method, because the dissociation of the bound sulfite is favored at higher pH on both equilibrium constant and rate considerations (Wedzicha, 1984).

An important factor influencing the precision of the method is the loss of sulfite during the analysis. By adding D-mannitol to the extractant and minimizing the analysis time, the precision of measurements corresponding to a coefficient of variation of approximately 4% was achieved. Thus, the new method is extremely reliable in terms of both accuracy and precision.

5. It Is Versatile. Currently, for quantitative analysis of total sulfite, AOAC (1984) recommends the modified Monier-Williams method

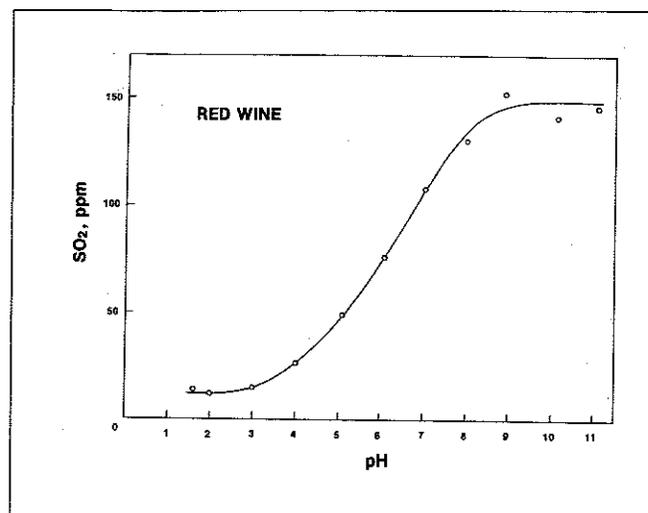
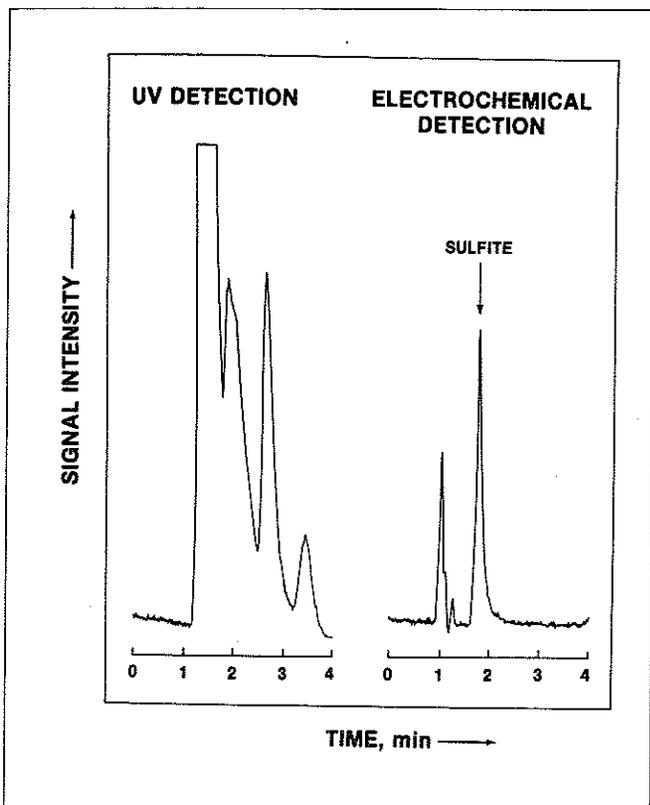


Fig. 5 (above)—pH Dependence of the Amount of Released Sulfite from red wine measured by the new method. At pH 2, only free sulfite is measured; at pH >8, total sulfite is measured

Fig. 4 (at left)—Chromatograms obtained from Peppers in Vinegar with UV detection (left) and electrochemical detection (right), showing the selectivity of the electrochemical detection

for most foods, including meat and wine but excluding dried onions, leeks, and cabbage, and the colorimetric method for dried fruits and beer. It would be desirable to have a technique which is applicable to all sulfite-containing foods and can be used to measure free sulfite as well as total sulfite.

Because the new technique is highly selective and interference-free, it can be used with most known foods containing sulfite, as shown in Table 3. So far, we have used the technique to measure sulfite from dehydrated fruits and vegetables, instant vegetable soups, dehydrated potatoes, bottled vegetables, juices, wine vinegar, wine, beer, meat, and dip-treated lettuce and shrimp containing sulfite at <10 ppm to >5,000 ppm. No significant interference was observed with these foods. The only major food component that behaves like sulfite on our system is ascorbic acid, which precedes the sulfite under our experimental conditions. A baseline separation of ascorbic acid and sulfite is achieved with 6 mM H₂SO₄ solution as eluant.

The new method is also versatile in the sense that it enables one to measure free and total sulfite sepa-

rately. It is well known that the carbonyl-sulfite adducts show maximum stability at around pH 2 and that dissociation of the adducts is favored at pH > 6 (Wedzicha, 1984). Therefore, for analysis of free sulfite, the food sample is treated with acid without heat, and the resulting SO₂ is analyzed by various techniques, as summarized in Table 1. For analysis of total sulfite, the sulfite is released by distillation in strong acid with nitrogen bubbling or by treatment with alkali and analyzed by various techniques (Table 1).

Figure 5 shows the pH profile of sulfite released from red wine with buffers at different pH values and analyzed by the new method. Maximum and minimum amounts of sulfite are obtained at pH values of about 9 and 2, respectively. This observation is consistent with the idea that one can measure the total and free sulfite by alkali and acid extraction, respectively. In the new method, free sulfite is analyzed by extracting the sulfite with 5 mM H₂SO₄, pH 2, separating the sulfite by anion exclusion chromatography, and detecting it with an electrochemical detector. Total sulfite is analyzed similarly, except that 20

mM sodium phosphate buffer, pH 8.9, is used for extraction. The extract is injected into the chromatograph without adjustment of the pH. Yet, because the sample size is small (20- μ L injection loop used), the pH adjustment by the eluant (6 mM H₂SO₄) takes place within the chromatograph, and the sulfite in the extract is eluted with the same retention time as the authentic sulfite dissolved in 6 mM H₂SO₄.

Figure 2 shows typical chromatograms for free and total sulfite obtained from four different foods containing sulfite at widely different levels. Results for free and total sulfite in a variety of foods are summarized in Table 3. A wide range of values in the free sulfite/total sulfite ratio as well as in the absolute sulfite level are observed. The changes in the free and total sulfite in foods during processing and storage can be measured easily by the new method. Correlation of such changes with the changes in product quality will be very useful in determining the optimal sulfite level in the foods.

6. It Is Convenient. From a practical standpoint, one of the important features of any analytical

method is convenience. It is especially true for routine analysis of multiple samples for quality control. A major disadvantage of the methods listed in Table 1 that involve distillation is inconvenience. Distillation is time-consuming, and the distillation apparatus and the trap have to be disassembled, washed, and reassembled before the next run is made.

In the new method, distillation is replaced by a simple extraction with Polytron and chromatographic separation. After an extraction, the Polytron is washed by running with water for several seconds and is ready for the next extraction. The extract can be injected directly into the chromatograph through a membrane filter, and the separation, detection, and data manipulation take place in line within several minutes. Since isocratic elution is used, there is no need to reequilibrate the column after each run, as in the gradient elution. For multiple sample analysis, a second sample can be weighed and extracted while the first sample is being analyzed on the chromatograph. An experienced person can analyze 5-10 samples an hour.

Formulation and Labeling Considerations

Americans spent nearly \$400 billion on food in 1985, and 6,694 new food products were introduced to the market that year alone (Anonymous, 1986a). FDA requires that the presence of sulfite at or above 10 ppm in the final food product be declared on the label, whether the sulfite has been added directly or indirectly. An example of a common source of an indirect addition of sulfite is the use of dehydrated components as ingredients in other products. Sulfite is either added to or naturally occurring in a number of dehydrated foods. Considering the popularity of instant foods and dehydrated food ingredients, it is suspected that a substantial fraction of the food products introduced each year might contain sulfited ingredients such as dehydrated potatoes, fruits, and vegetables. Therefore, the sulfite level in such foods as instant soup, sauce and gravy mix, guacamole mix, breakfast cereals, salad dressing, etc., needs to be determined and declared on the label if it is at or exceeds 10 ppm. It would be advan-

Table 3—Free and Total Sulfite in various foods measured by the Kim and Kim (1986) method

Sample	SO ₂ (ppm) ^a		Free SO ₂ /total SO ₂ ratio
	Free	Total	
Freeze-dried green bell pepper dice	1,747	5,819	0.30
Freeze-dried chopped celery	320	517	0.62
Freeze-dried peas	47	345	0.14
Freeze-dried cauliflower	0	0	—
Mixed dried fruits	99	1,827	0.05
Golden raisin	95	1,555	0.06
Instant mashed potatoes	219	488	0.45
Hash brown potatoes	61	347	0.18
Dip-treated lettuce ^b	380	536	0.71
Pepper in vinegar	184	307	0.60
Cocktail onion	8.7	64.1	0.14
Lemon juice	174	278	0.63
Lime juice	135	218	0.62
Wine vinegar	3.3	48.4	0.07
Red wine A	32	163	0.20
Red wine B	10	156	0.06
Beer	<0.5	<0.5	—
Flaked coconut	<0.5	9.8	<0.05

^aAverage of duplicate measurements
^bCut ½ in X ½ in, dipped in ½ oz/gal sodium metabisulfite solution for 1 min, and drained

tageous for the food industry to bring the sulfite level in the final product to below 10 ppm if at all possible. To do so, the food developer could attempt to lower the sulfite level in the ingredients to the acceptable minimum or decrease the amount of the ingredients containing high levels of sulfite. The new method of analysis described in this article would be useful in accurately determining the sulfite level in the ingredients as well as in the final products.

The advantage of the new method to the food developer or manufacturer with regard to sulfite labeling would be most critical where the Monier-Williams method gives "false-positive" response. For example, the Monier-Williams analysis of Ralston Purina Co.'s isolated soy protein produced without addition of sulfite showed a false-positive response up to 39 ppm (Anonymous, 1986b). Analysis of the same isolated soy protein sample by our method showed only 4 ppm SO₂ (Kim, unpublished data). Other such examples include dehydrated onion and cabbage. Since these are common ingredients in such foods as instant soups, gravy mix, salad dressing, etc., false-positive response from these foods is a poten-

tial problem. Such problems could be avoided by the new method, and the labeling need obviated in many cases.

Following the FDA regulation, the Bureau of Alcohol, Tobacco and Firearms also required labeling of alcoholic beverages for the presence of total sulfite at or above 10 ppm (BATF, 1986). Imported wine and foods are not exempt from the labeling requirement. We determined, by the new method, the sulfite levels in imported red wine, instant vegetable soup, and seasoning mix to be 163, 43, and 8 ppm, respectively. Red wine and instant vegetable soup clearly need to have the label indicate the presence of sulfite, whereas the seasoning mix containing less than 10 ppm SO₂ does not.

The IFT Expert Panel on Food Safety & Nutrition stated that "future research is needed on the reactivity of sensitive individuals to sulfited foods and improved methods for the accurate detection of sulfite residues in foods" (IFT, 1986). Especially at issue is whether free or total sulfite is responsible for the adverse reactions to sulfite. The new technique can distinguish between free and bound sulfite and will be useful in investigating the

role of free vs bound sulfite in the hypersensitive reactions of individuals to foods containing sulfite.

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