A New Procedure for the Analysis of Tung Fruit[1]

R. S. McKINNEY and NOAH J. HALBROOK, U. S. Tung Oil Laboratory, Gainesville, Fla.,* and G. WORTHEN AGEE, Barrow-Agee Laboratories Inc., Memphis, Tenn.

COLLABORATIVE analyses on samples of tung fruit were made during 1946 by members of a subcommittee on tung of the Seed and Meal Analysis Committee of the American Oil Chemists' Society, using procedures based upon methods of analysis developed in this Bureau (1). In the procedures employed on a first series consisting of five samples, 5- or 8-fruit subsamples were used for moisture determinations and 25-fruit subsamples were separated into components and the separated kernels used in the oil determination. In a second series consisting of the last three samples one carpel was taken from each of the 35 fruits in the sample and used for the oil determinations while another carpel was taken from each fruit for the moisture determinations. In the analysis of tung fruit by the carpel procedure it appeared advisable to use samples in regard to which information was available. Therefore the material used for sample 6 was drawn from the lot of tung fruit from which sample 3 had been drawn, sample 7 was drawn from the lot of fruit from which sample 1 had been drawn, while sample 8 was drawn from the lot of fruit from which sample 2 had been drawn. The results obtained by the collaborators using the two procedures are given in Table I.

Considerable variations are apparent in the oil and moisture contents of the first five samples, with standard deviations of 0.32%-0.90% oil and 0.42%-0.97% moisture, which could be attributed entirely to sampling errors on the basis of a recent study on the sampling of tung fruit (2). Considerably higher standard deviations in the oil content of the fruit samples occurred when carpels were used instead of the kernels of the fruit. This notable increase in the standard deviation was explainable by a wide variation in the oil content of individual carpels shown in a subsequent analysis of a sample of 25-tung fruit.

Tung producers have felt the need for increasing the accuracy of the analysis of tung fruit. It is obvious that neither the component nor the carpel procedure is entirely dependable. It appeared that accuracy could be improved only by the use of a much larger sample of tung fruit than that usually used in analysis by the component procedure. Increasing the size of the sample in the component procedure introduced the problem of the proper hulling and shelling of the large sample as the skilled workers required for the task objected to the tediousness involved. Therefore, in developing a new procedure, consideration was given to the possibility of grinding the whole sample of tung fruit and making the moisture and oil determinations on portions of the ground tung fruit. A difficulty in the development of such a procedure was that an appreciable amount of non-oil constituents, soluble in petroleum ether, occurs in the shell and hull portions of the tung fruit. In the component procedure the oil content of the fruit is calculated from the per cent kernels and the per cent oil in the kernels, the oil being located entirely in the kernels. The tung industry has used the component procedure for the analysis of tung fruit for a number of years, and the price paid for the fruit has been based upon the results obtained by this procedure. Therefore for the new procedure to be acceptable to the industry it had to yield results comparable to those obtained by the component procedure.

TABLE I

Analysis of Collaborative Samples by Component Procedure—1945-6

Sample	Collaborators 1	2	3	4	5	6	7	Average	S.D.
	Per Cent Oil in Tung Fruit								
1	19.5	21.0	19.8	19.8	19.1	20.4	19.5	19.8	0.63
2	19.6	19.9	19.2	19.7	20.5	19.1	18.9	19.6	0.55
3	19.9	19.9	20.2	19.8	20.4	19.5	19.0	19.8	0.48
4	22.4	22.7	22.6	23.0	22.1	22.7	23.0	22.6	0.32
5	22.7	23.2	23.6	23.0	21.0	22.7	23.7	22.8	0.90
6	19.4	21.8	20.6	19.2	18.9	20.9	19.4	20.0	1.10
7	21.8	21.8	20.5	19.7	18.3	22.0	20.8	20.7	1.35
8	20.7	20.5	18.7	20.2	20.5	21.3	20.1	20.3	0.80
	Per Cent Moisture in Tung Fruit								
1	15.1	16.4	16.3	15.6	16.0	15.0	14.7	15.6	0.67
2	15.0	14.8	16.8	15.3	14.3	15.3	14.5	15.1	0.82
3	11.9	11.6	12.1	11.9	12.9	11.8	12.1	12.0	0.42
4	13.0	12.6	10.4	13.4	12.5	12.4	12.8	12.4	0.97
5	11.8	12.5	9.6	11.7	11.2	11.3	11.1	11.3	0.89
6	11.3		9.4	10.6	10.5	10.5	10.2	10.4	0.69
7	11.5	10.7	10.0	10.7	10.8	10.5	10.5	10.7	0.42
8	10.1	10.5	9.8	10.8	10.0	10.1	10.6	10.3	0.35

The procedure finally developed for the determination of the moisture and oil content of tung fruit employs a sufficiently large sample to eliminate, to a considerable extent, the sampling errors which occur with the relatively small samples usually used in the component procedure. In this new procedure a sample of 200- to 250-fruit is ground in a Wiley mill[2] using a ¼-inch screen. After thoroughly mixing the Wiley-ground material by rolling on a large piece of paper or preferably in a large Maclellan mixer (30 quarts),[2] it is subdivided, either by quartering with a large spatula or with a riffle, into two portions of about 1-quart each. One portion is used in the moisture determination and the other in the oil determination. The portion to be used for the oil determination is reground either in a Raymond Laboratory mill[2] using a 1-millimeter screen or in a Bauer No. 148 Laboratory mill with No. 6912 plates at 3600 r.p.m., adjusted to produce a fine meal. The time of the initial grinding was shortened and loss of moisture was avoided by equipping the Wiley mill with the sieve having ¼-inch round holes, fitting an auxiliary hopper over the regular hopper to prevent material from being thrown out, and passing a tight fitting chute from the bottom of the mill through the cover of a large can into which the ground material is delivered without the possibility of spilling or drying.

Three methods may be used for determining the moisture content of the tung fruit in this new procedure. The first method consists of drying a 5-gram portion of the Wiley-ground sample for 4 hours at

* One of the Laboratories of the Bureau of Agricultural & Industrial Chemistry, Agricultural Research Administration, U. S. Department of Agriculture.

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[2] Identification of equipment, by giving name of manufacturer, should not be construed as an endorsement of such equipment by the U. S. Department of Agriculture.

101°C. in an oven at not more than 50 mm. of Hg. pressure. After cooling and weighing, the sample is redried under similar conditions for 1-hour periods until a loss of weight of not more than 2-mg. occurs. One redrying period is usually sufficient. In the second method a 5-gram sample of the Wiley-ground material is dried for 1 hour in a forced draft oven at 101°C., cooled and weighed, then redried for half-hour periods until the loss of weight is not more than 2 mg. Two redrying periods are usually required. The third method, that of Bidwell-Sterling (3) using either a 20- or 100-gram sample and run for 1¼-1½ hours, probably gives the most accurate estimate of the moisture content of tung fruit as some oxidation may occur in the two oven methods.

As some drying occurs in the preparation of the Wiley-ground material for the oil determination, it is necessary to make a moisture determination on the thoroughly mixed Raymond or Bauer-ground material using the same method employed on the Wiley-ground material. Redrying in the oven methods is usually not required. A 5-gram sample of the Raymond- or Bauer-ground material is extracted for 4 hours in a Butt-type extraction apparatus (1) and the oil content of the tung fruit is calculated to the original moisture basis.

TABLE II

Comparison of Oil and Moisture Content of Tung Fruit by Component Procedure and by New W-R Procedure

Sample	Component Procedure		New W-R Procedure	
	Moisture	Oil	Moisture[1]	Oil
	%	%	%	%
1	16.37	20.27	16.57	20.10
2	16.62	20.29	16.71	19.93
3	13.85	20.48	13.56	21.21
4	15.67	19.25	16.16	19.62
5	11.46	19.98	11.38	19.69
6	11.67	20.04	11.54	19.43
Average	14.27	20.05	14.32	20.00

[1] Using vacuum oven method.

To obtain the results reported in Table II, the new procedure was carried out as follows: six samples were drawn from commercial lots of tung fruit at a mill and each sample was thoroughly mixed and divided by quartering into two portions of about 100-fruit each. One portion of each sample was analyzed by the component procedure while the other portion was analyzed at the Gainesville Laboratory by the new procedure using the Wiley-Raymond grinding technique. The average results obtained with the component procedure and with the new procedure using the Wiley-Raymond grinding technique are in good agreement while the variations between the results for individual samples are about that to be expected from the previously mentioned sampling study (2) when 100-fruit samples are used.

In connection with the collaborative analyses on samples of tung fruit during 1947 it appeared desirable to compare the results obtainable with the new procedure using the Wiley-Bauer or the Wiley-Raymond grinding technique with those obtained by the component procedure. Five lots of tung fruit were thoroughly mixed and each lot was divided into three large samples of 200-250 fruit each. One of the large subsamples of each lot was then subdivided into six small subsamples which were analyzed by the members of the subcommittee on tung using the component procedure. The results obtained by the collaborators are given in Table III.

TABLE III

Analysis of Collaborative Samples by Component Procedure—1946-7

Sample	Per Cent Oil In Tung Fruit							
	Collaborators							S.D.
	1	2	3	4	5	6	Average	
1	20.4	21.3	20.8	19.9	20.1	19.3	20.3	0.70
2	19.4	19.3	19.5	(17.2)	18.5	19.3	19.2	0.40
3	18.5	19.0	21.0	18.9	19.1	19.8	19.4	0.90
4	20.8	20.5	20.6	21.0	20.9	20.4	20.7	0.24
5	19.3	19.3	20.2	18.4	19.4	20.6	19.5	0.84
	Per Cent Moisture in Tung Fruit							
1	14.1	13.6	14.5	14.0	13.5	14.6	13.9	0.48
2	10.5	10.7	12.2	10.2	11.7	11.3	11.1	0.80
3	11.2	11.8	12.5	11.2	11.7	11.9	11.7	0.49
4	14.2	14.7	14.8	14.2	14.9	14.1	14.5	0.37
5	14.1	15.6	15.6	14.2	14.8	15.5	15.0	0.70

The other two large subsamples of each lot of tung fruit were analyzed for moisture and oil content using the Wiley-Bauer and the Wiley Raymond grinding techniques. Because of the sampling errors in the component procedure with such small samples the average of the results of the collaborators were calculated and used for comparison. In Table IV

TABLE IV

Per Cent Oil and Moisture Content by Component and New Procedures

Sample	Per Cent Oil in Tung Fruit		
	Collaborators Component Procedure (Average Results)	Wiley & Raymond Ground Sample	Wiley & Bauer Ground Sample
1	20.30	20.74	20.80
2	19.20	19.57	20.30
3	19.38	19.47	19.80
4	20.70	20.42	20.25
5	19.50	18.85	19.80
Average	19.82	19.81	20.19
	Per Cent Moisture in Tung Fruit		
Sample	Collaborators Component Procedure (18-24 hrs. & 101°C.) (Average Results)	Wiley & Raymond Ground Sample (4 hrs. Vac. Oven)	Wiley & Bauer Ground Sample (B-S Method)
1	13.9	13.6	14.5
2	11.1	11.4	11.5
3	11.7	11.8	12.4
4	14.5	13.6	14.9
5	15.0	14.9	15.3
Average	13.2	13.1	13.7

are given the average results of the subcommittee members on the samples analyzed by the component procedure and by the new procedure using both Wiley-Raymond and Wiley-Bauer ground samples. The average of the results of the collaborators for oil content (19.82%) by the component procedure was found to be in good agreement with the average results obtained on the Wiley-Raymond ground samples (19.81%), but the average results obtained on the Wiley-Bauer ground material (20.19%) are appreciably higher than those obtained by the other procedures.

From these results it appears that a correction of 0.37% must be subtracted from the per cent oil obtained on samples ground in the Wiley-Bauer mills to obtain results comparable to those obtained by the component procedure. In Table V are given the average results of the subcommittee members for oil content on the samples analyzed by the component procedure, the results obtained on the Wiley-Raymond ground samples, and the corrected results on the Wiley-Bauer ground samples. From these data mean values were calculated for the oil content

of each collaborative sample and are listed in the last column of the table. Using the indicated correction, the oil percentages obtained by the new procedure, using the Wiley-Raymond and the Wiley-Bauer grinding techniques, were found to be in good agreement with the calculated means. The variations found in the oil content of the collaborative samples analyzed by the three procedures can be attributed to sampling errors, as even with samples of 200-tung fruit a standard deviation of 0.36% oil is to be expected (2).

TABLE V

Oil Values by Component and New Procedures with Corrected Values for Wiley-Bauer Ground Samples

Sample	Per Cent Oil In Tung Fruit			
	Collaborators Component Procedure (Average Results)	Wiley & Raymond Ground Sample (Uncorrected)	Wiley & Bauer Ground Sample (Corrected)	Average
1	20.30	20.74	20.43	20.49
2	19.20	19.57	19.93	19.57
3	19.38	19.47	19.43	19.43
4	20.70	20.42	19.88	20.33
5	19.50	18.85	19.43	19.26
Average	19.82	19.81	19.82	19.82

It was found that the Bidwell-Sterling method gave the highest results for moisture content of tung fruit; drying the whole tung fruit for 18-24 hours yielded moisture results which were slightly higher than those obtained by drying the Wiley-ground fruit in the vacuum oven. It appears to be difficult to remove the last traces of moisture from the whole fruit at atmospheric pressures and 101°C. in 18-24 hours, whereas some oxidation probably occurs in ground tung fruit at this temperature even at low pressures. The variations in the analysis of tung fruit by the new procedure using the three moisture methods should not have an appreciable effect upon the purchase of the fruit provided the same moisture method is employed with the Wiley-ground samples and with the Wiley-Raymond or Wiley-Bauer ground samples as the current prices of tung fruits (nuts) are based upon oil content.

In further tests to determine the amount of material extracted from the hulls and shell of tung fruit in the new procedure, so as to calculate the correction factors to be applied in the oil determination, the hulls and shell from several of the collaborative samples listed in Table IV were ground in the Wiley and Bauer mills and extracted for 4 hours with petroleum ether. The corresponding samples of tung fruit ground in the Wiley-Bauer mills were extracted for 4 hours with petroleum ether, then reground in a Sturtevant mill with alundum plates, and extracted for residual oil. Also, the hulls and shell from several collaborative samples of tung fruit were ground in the Wiley-Raymond mills and extracted for 4 hours with petroleum ether. Corresponding samples of tung fruit were ground in the Wiley-Raymond mills, extracted for 4 hours with petroleum ether, then reground with mortar and pestle using fine sand, and extracted for residual oil.

The results obtained in these tests, given in Table VI, have indicated that a correction of 0.40% should be subtracted from the percentage of ether-extractable material obtained from the Wiley-Bauer ground tung fruit in this procedure, and this correction factor is in good agreement with the one found in the collaborative work. The results have also indicated

TABLE VI

Extractable Material from Tung Hulls and Shell with Calculated Correction in Oil Determination

Wiley-Bauer Ground Fruit

Sample	Extract from		Hulls	Shell	Extract from Whole Fruit
	Hulls	Shell			
	%	%	%	%	%
2	1.48	0.46	49.3	20.0	0.81
3	0.88	0.60	47.8	20.7	0.54
4	1.04	0.43	45.7	21.0	0.58
5	0.94	0.24	46.7	20.5	0.49
Average	1.09	0.43	47.4	20.6	0.61

Extract after Second Grinding of Tung Fruit

Sample	2	3	4	5	Average
Per Cent	0.20	0.16	0.26	0.23	0.21

Correction = 0.61 − 0.21 = 0.40%

Wiley-Raymond Ground Fruit

Sample	Extract from		Hulls	Shell	Extract from Whole Fruit
	Hulls	Shell			
	%	%	%	%	
1	0.63	0.21	45.4	21.3	0.33
2	0.67	0.29	48.4	21.9	0.38
5	0.42	0.14	48.1	20.7	0.23
Average	0.57	0.21	47.3	21.3	0.31

Extract after Second Grinding of Tung Fruit

Sample	1	2	5	Average
Per Cent	0.11	0.16	0.09	0.12

Correction = 0.31 − 0.12 = 0.19%

a correction of 0.19% oil for the Wiley-Raymond ground tung fruit while the collaborative work has shown that no correction is necessary in this case. Further investigation may show that a correction should be used on the Wiley-Raymond ground material, but laboratory analyses have shown that it is practically impossible to thoroughly disintegrate a mixture of kernels, hulls, and shell by grinding with mortar and pestle because of the cushioning action of the hulls. It is believed that this factor accounts for the difference between the calculated and found corrections for the Wiley-Raymond ground material.

Summary

A new procedure for the analysis of tung fruit for oil and moisture content has been developed wherein it is possible to largely eliminate the sampling errors which occur in component procedure used in the past because of the relatively small samples used. Collaborative studies have indicated that to obtain results similar to those obtained in analysis by the component procedure a correction of 0.37% must be subtracted from the oil content obtained with the Wiley-Bauer ground fruit, but no correction appears to be necessary in the case of the Wiley-Raymond ground material.

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REFERENCES

1. R. S. McKinney and A. F. Freeman, Oil and Soap *16*, 151 (1939).
2. Raiford L. Holmes and Frank C. Pack, Proc. Am. Tung Oil Assoc. 1946, 40.
3. Methods of Analysis of the Association of Official Agricultural Chemists, Sixth Edition, 1945, 404.