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THE DIPHENYL RESIDUE IN CITRUS FRUIT WRAPPED  
IN DIPHENYL WRAPS

by

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סקירה מוקדמת

המחלקה לפירסומים

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# THE DIPHENYL RESIDUE IN CITRUS FRUIT WRAPPED IN DIPHENYL WRAPS\*

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## Introduction

Diphenyl is now the only product employed successfully against certain citrus fruit diseases which develop during storage and transport. Because of their diphenyl absorption, citrus fruits are classed in the category of foodstuffs containing an additive chemical, and their diphenyl content is limited by law.

What are the quantities of diphenyl generally found in citrus? Do they oscillate around fixed values, to be taken as absorption constants? There are little data on the subject and those which exist chiefly concern American citrus fruit preserved in diphenyl-impregnated cartons.

From the great number of determinations made in the U.S.A. by the Hazleton Laboratories (4) it was found that diphenyl quantities in whole fruits vary, in oranges, from 0 to 110 mg/kg, in lemons from 0 to 70 mg/kg, and in grapefruits from 0 to 30 mg/kg. The quantities found in citrus juices depend upon the way the juice is extracted and are generally higher when juice is extracted by pressure reaming, e.g. 0-4.36 mg of diphenyl per kg of orange juice, 1.5-11.2 mg/kg of lemon juice and 0.4-1 mg/kg of grapefruit juice when juice was obtained by pressure, against 0.05-2.5 mg/kg, 0.1-3.8 mg/kg, and 0.1-0.9 mg/kg respectively, when juices were obtained by reaming.

In Italy, Rogliani and Procassini (5) found, in lemons and oranges wrapped in diphenyl-impregnated wraps, 3.2-3.3 mg of diphenyl per one fruit peel and 0.3-0.6 mg of diphenyl per one fruit pulp. Tomkins and Isherwood (6) found, in Israeli oranges, up to 37 mg/kg of peel and none in the pulp. In Germany, Feuersenger (1) analyzed citrus of various origins and found in U.S.A. oranges 0.0-22.0 mg and in Italian lemons 0.5-16.0 mg/one fruit peel but none in the Israeli and Cyprus orange peels. With the exception of a few cases he found no diphenyl at all in the juice of analyzed citrus. Koether (2) found, in Italian lemons, 18-33 mg, in Brazilian oranges 3-22 mg, and in South African oranges 1-20 mg/kg of fruits but could find no evidence of the presence of diphenyl in the juice.

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Data concerning citrus pulp differ greatly. It seems, indeed, that the very presence of diphenyl in the citrus pulp - a very important fact from the standpoint of food hygiene - is not established. To know whether the diphenyl found in the pulp by some researchers comes from the pulp itself or from the losses incurred by the peel during juice preparation, diphenyl was determined separately in the peel and in the pulp, special care being taken when separating peel from pulp. Diphenyl was also determined in the albedo in order to discover to what extent albedo remnants can falsify determination results in the pulp.

Data concerning whole fruits also differ greatly. The diphenyl contents of whole fruits are so variable that they cannot be considered as absorption constants. These great differences constitute a disturbing factor and show that the maximal quantities permitted by law could be unintentionally surpassed. The cause of these differences is not clear. To determine if external agents, such as washing and waxing of fruit before wrapping, or if the diphenyl present in the atmosphere of the storage room influences the diphenyl absorption by diphenyl-wrapped fruit, a preliminary investigation was undertaken. Diphenyl was determined in both treated and non-treated diphenyl-wrapped fruit, and also in fruit wrapped in plain wraps, placed in a storage room together with fruits wrapped in diphenyl wraps.

#### Methods

Citrus fruit was taken at random, from a stock of fruit, and its diphenyl content determined.

#### Diphenyl Determination in Peel, Pulp and Albedo:

The fruit was carefully peeled, so as to avoid any contact between peel and pulp. To determine the diphenyl content in the albedo, the flavedo was very carefully removed from some whole fruits with the aid of a scalpel and the albedo separated from the pulp. Diphenyl was extracted and determined colorimetrically as described in (3). The results, corresponding to the average weight of 1 fruit, are expressed in ppm or by weight; in mg of diphenyl for whole fruit and peel, and in  $\mu$ g for pulp and albedo.



### Results and Conclusions

#### 1. Diphenyl residue in the various citrus fruits and its localization:

Diphenyl contents of various citrus fruits wrapped in diphenyl wraps is presented in Table 1. Peel and pulp of all citrus varieties were found to absorb diphenyl.

The quantities absorbed by whole fruits vary considerably. Nearly all the diphenyl is found in the peel; the albedo contains only traces. Thus the main amount of diphenyl concentrates in the flavedo which is rich in essences and coloring substances. In some cases, by simply rinsing the fruit with chloroform, it was possible to remove up to 30% from the total content of diphenyl in the peel; in other cases, only traces of diphenyl were removed by this treatment.

The edible portion generally shows very slight traces of diphenyl, and the quantities absorbed vary considerably. As no fixed correlations exist between the quantities absorbed by pulp and whole fruit, it is necessary to determine diphenyl separately each time in the peel and pulp.

#### 2. Effect of washing and waxing on diphenyl absorption:

The analysis of diphenyl-wrapped fruits with and without washing and waxing and stored under the same conditions, seems to show (Table 2) that treated fruits absorbed more diphenyl than non-treated ones. The treatment seems to have particularly influenced the rate of absorption by the peel. External agents can therefore act upon the absorptive power of the fruit and be one of the causes of the differences observed in the diphenyl contents of citrus.

Further studies on these agents should be made.

#### 3. Absorption of diphenyl by fruits wrapped in plain wraps and placed in a storage room together with fruits wrapped in diphenyl wraps:

As seen in Table 3 the fruit contains up to 2 mg diphenyl per fruit. Hence, the fruits can, independently of the wrapping, absorb non-negligible quantities of diphenyl from the atmosphere of the storage room. This could also be one of the causes of the differences observed in the quantities of diphenyl absorbed.



Table 1

Diphenyl Content in Citrus Fruits

Fruit Variety	D I P H E N Y L							
	Quantity per fruit					ppm		
	Whole fruit	Peel		Pulp		Whole fruit	Peel	Pulp
	mg	total mg	albedo mg	mg	% of total diphenyl			
Oranges:								
Shamouti	8.46	8.43	25.0	33	0.40	40	102	0.26
Valencia	12.60	12.57	98.0	31	0.25	70	247	0.24
Naval	7.85	7.80	30.0	55	0.70	38	152	0.35
Clementines	4.50	4.50	-	9	0.20	62	250	0.15
Grapefruit	0.73	0.73	-	7	1.0	32	15	0.04
Lemons	1.84	1.80	3.5	39	2.1	15	44	0.50

Table 2

Diphenyl Content of Treated and Non-Treated Fruit Wrapped in Diphenyl Wraps

Storage temperature: 17°C.

Treatment: Decasol, Citroshein Wax

Treatment	Duration of storage (days)	D I P H E N Y L						
		Quantity per 1 fruit					ppm	
		Whole fruit mg	Peel mg	Pulp mg	Whole fruit	Peel	Pulp	
		1. Shamouti						
-	31	3.61	3.56	48	13	33	0.28	
-	31	3.25	3.24	16	12	31	0.09	
+	36	8.46	8.43	33	40	102	0.26	
+	36	5.27	5.22	45	23	60	0.32	
		2. Valencia						
-	24	6.08	6.03	55	30	126	0.37	
+	24	8.70	8.62	77	45	192	0.55	
-	43	8.52	8.50	23	50	170	0.19	
+	43	12.61	12.58	31	70	247	0.24	



Table 3

Absorption of Diphenyl by Fruits Wrapped in Plain Wraps and Stored Together with  
Diphenyl-Preserved Fruits (Storage Temper. 17°C.)

Duration of Storage (days)	D I P H E N Y L					
	Quantity per 1 fruit			ppm		
	whole fruit mg.	peel mg.	pulp mg.	whole fruit	peel	pulp
	1. Non-treated fruit					
	Shamouti					
43	1.80	1.78	20	9	21	0-.16
43	2.23	2.22	11	11	26	0.07
	Valencia					
19	0.81	0.80	9	5	17	0.07
40	1.66	1.65	10	10	34	0.08
	2. Treated fruit					
	Shamouti					
30	1.27	1.24	23	6	14	0.18
30	2.03	2.01	17	9	24	0.13
	Valencia					
19	1.66	1.65	16	10	34	0.13
40	1.91	1.90	12	12	42	0.10

Summary

All the citrus fruits preserved by diphenyl wraps absorb diphenyl. Nearly all the diphenyl is to be found in the peel particularly in the flavedo; the pulp contains only slight traces. External agents can influence the rate of diphenyl absorption by the fruits and may be responsible for the differences observed in the diphenyl contents of the various fruits. A study of those agents is most necessary.

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