# Preferential Inclusion of Food Colours via Formation of their Inclusion Complexes with $\beta$ -Cyclodextrin ( $\beta$ -CD)

#### BHASKAR HOSANGADI\* and SHASHIKANT PALEKAR

Department of Chemistry, University of Bombay, Vidyanagari, Bombay-400 098, India

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Abstract. The encapsulation of commercially utilized food colours with  $\beta$ -cyclodextrin is reported. Thus, inclusion complexes of Sunset Yellow, Amaranth, Ponceau 4R, Carmoisine, Fast Red E, Tartrazine and Erythrosine with  $\beta$ -CD were prepared. The formation of inclusion complexes was established by, among other methods, UV, reflectance and X-ray diffraction techniques. The host-to-guest ratio was determined by a UV spectral method. The effect of inclusion with  $\beta$ -CD on binary mixtures such as Raspberry Red, Tomato Red and Orange Red was also studied.

Key words.  $\beta$ -Cyclodextrin ( $\beta$ -CD), food colours, spectrophotometry, reflectance spectra, X-ray line diagrams.

## 1. Introduction

In recent years cyclodextrins [1] have been the subject of wide-ranging studies, including crystallographic studies [2], inclusion complexes [3, 4] and their role in catalysis as a model for enzyme action [5]. Chromatographic separations of a few inclusion complexes of cyclodextrins have been reported [6]. Inclusion complexes of some azo dyes have been reported [7–11]. However, food colours, which are marketed as aqueous solutions of azo dyes, have not been subjected to inclusion studies. Inclusion is expected to lend stability to such aqueous solutions. In continuation of our study on encapsulation of organic molecules with  $\beta$ -CD [12], experimental work on food colours was undertaken, the results of which are reported in this paper.

## 2. Experimental

 $\beta$ -CD (Fluka) was used for this study. Food colours and binary mixtures were obtained commercially. Food colours were further purified by repeated crystallisations from water. The purity of the samples thus obtained was checked by TLC [13]. A Beckman DU-2 spectrophotometer was used for spectrophotometric studies. Reflectance spectra were measured on a Carl-Zeiss VSU-2P spectrophotometer using  $\beta$ -CD as the reference standard. X-ray powder diffractograms were recorded on a Rigaku Geigerflex D/max-B series X-ray diffractometer.

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## 2.1. PREPARATION OF INCLUSION COMPLEXES

 $\beta$ -CD was dissolved in the minimum amount of distilled water (slight heating was necessary) and to this solution were added the food colours shown in Chart 1, such as Sunset Yellow (I), Amaranth (II), Ponceau 4R (III), Carmoisine (IV), Fast Red E (V), Tartrazine (VI) and Erythrosine (VII) in 1 : 1 mole proportions, and the solution was intermittently shaken in a water bath at 80°C for 45 minutes. It was then kept overnight at room temperature. The inclusion complex so formed was filtered, washed with ethanol to remove the untrapped feed and was dried in an oven at 100°C.

The inclusion complexes of binary mixtures such as Raspberry Red, Orange Red and Tomato Red were prepared in the same way.



Chart 1.

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# INCLUSION OF FOOD COLOURS WITH $\beta$ -CYCLODEXTRIN

Food Colours	Technique	For food colour	Inclusion complex of the food colour	Physical mixture of the food colour with $\beta$ -CD
Sunset	m.p.		257–8°C	2468°C
renow	maxima (nm)	235, 315	240, 310	235, 313
	maxima (nm)		360	370
	of inclusion		4.96%	
Amaranth	m.p.		260–1°C	240–1°C
	UV absorption maxima (nm) reflectance	330	245	325
	maxima (nm) percentage		390, 440	390, 430, 480
	of inclusion		3.97%	
Carmoisine	m.p.		254–5°C	2378°C
	UV absorption maxima (nm) reflectance	290, 325	280, 320	283. 320
	maxima (nm)		360, 400	370, 400, 530
	of inclusion		5.60%	
Ponceau 4R	m.p.		251–2°C	2423°C
	UV absorption maxima (nm) reflectance	245, 335	240, 325	245, 335
	maxima (nm) percentage		370, 520	370, 470
	of inclusion		4.29%	
Fast Red E	m.p. UV absorption		252–3°C	2456°C
	maxima (nm) reflectance	280, 317	265. 320	280, 315
	maxima (nm) percentage		380	370
	of inclusion		3.09%	
Tartrazine	m.p.		265–6°C	250–2°C
	maxima (nm) reflectance	255	260	255
	maxima (nm) percentage		380	390
	of inclusion		5.02%	
Erythrosine	m.p. UV absorption		234–5°C	220–2°C
	maxima (nm) reflectance	260, 310	265, 310	260, 313
	maxima (nm) percentage		360	370
	of inclusion		8.66%	

Table I. Analytical data of the inclusion complexes of food colours with  $\beta$ -CD

<sup>1</sup> Food colours did not show characteristic melting points. <sup>2</sup> Food colours being too dark in colour, it was not possible to record their reflectance. <sup>3</sup> Percentage of inclusion denotes percentage of food colour in  $\beta$ -CD by weight.



Fig. 1. X-ray powder diffraction pattern (line diagram).

Complex formation was confirmed by melting point, UV spectra, reflectance spectra and X-ray diffraction techniques. The percentage of inclusion of the guest in the complex was determined by using the UV spectral method.

A typical diffraction pattern line diagram of  $\beta$ -CD and its inclusion compound with Erythrosine illustrates the changes in the pattern after inclusion (Figure 1).

Before undertaking studies on preferential inclusion it was thought necessary to determine the percentage composition of individual food colours in the binary mixtures. For this purpose the following two methods were used.

- (1) The Graphical Absorbance-Ratio Method [14]
- (2) The Simultaneous Equations method [15]

Binary mixture	Constituents	% composition before inclusion	% composition after inclusion	
Orange Red	Carmoisine : Sunset yellow	8.87 : 91.13	15.06 : 84.74	
Raspberry Red	Carmoisine : Amaranth	89.28 : 10.72	84.83 : 15.17	
Tomato Red	Carmoisine : Ponceau 4R	3.08 : 96.92	48.19 : 52.81	

Table II. Percentage composition of binary mixtures before and after inclusion

#### 3. Results

The above two methods constitute useful procedures for determing the percentage composition of food colours. By using the above two UV spectral methods, studies were also carried out to determine the preferential inclusion of complexes of binary mixtures. However, the graphical absorbance-ratio method did not work satisfactorily in this case. Results using the simultaneous equations method are recorded in Table II.

The above study indicates that  $\beta$ -CD shows preferential inclusion for carmoisine present in binary mixtures of food colours, e.g. Orange Red and Tomato Red.

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## References

- 1. M. L. Bender and M. Komiyama: Cyclodextrin Chemistry, Springer-Verlag (1978).
- 2. W. Saenger, M. Noltemeyer, P. C. Manor, B. Hingerty and B. Klar: Bioorg. Chem. 5, 187 (1976).
- 3. S. Harata: Bull. Chem. Soc. Jpn. 48, 2409 (1975).
- 4. J. Szejtli: Cyclodextrins and Their Inclusion Complexes, Akademiai Kiado, Budapest (1982).
- 5. I. Tabushi: Acc. Chem. Res. 15, 66 (1982).
- 6. W. L. Hinze: Separation and Purification Methods, Vol. 10, p. 159, Marcel Dekker (1981).
- 7. J. Szejtli, Z. Budai and M. Kajtar: Magy. Kem. Foly. 84, 68 (1978).
- 8. R. P. Rohrbach and J. F. Wojcik: Carbohydr. Res. 92, 177 (1981).
- 9. H. Hirai, N. Toshima, and S. Uenoyama: Polym. J. (Tokyo) 13, 607 (1981).
- 10. W. V. Gerasimowicz and J. F. Wojcik: Bioorg. Chem. 11, 420 (1982).
- 11. A. Orstran and J. F. Wojcik: Carbohydr. Res. 143, 43 (1985).
- 12. B. D. Hosangadi and R. D. Prabhukhanolkar: J. Incl. Phenom. 3, 151 (1985).
- 13. CRC Handbook of Chromatography (Ed. Gunter Zweig and Joseph Sherma), CRC Press, Cleveland, Ohio, third edn. (1976), Vol. I, p. 490.
- 14. R. C. Hirt, F. T. King and R. G. Schmitt: Anal. Chem. 26, 1270 (1954).
- 15. A. C. Hardy and F. M. Young: J. Opt. Soc. Am., 38, 854 (1948).