Pak. j. sci. ind. res., vol. 35, no. 3, March 1992

IMPROVEMENT IN EMULSIFYING NATURE OF A PLANT GUM BY CHEMICAL DERIVATIZATION. *Part -I*

R. K. Agrawal, Ashok K. Shakya and Pradeep Mishra
Department of Pharmaceutical Sciences, Doctor Harisingh Gour Vishwavidyalaya, Sagar (M.P.), 470 003, India

(Received April 8, 1991)

Anogeissus latifolia gum was chemically derivatised to obtain acetyl and methyl derivatives with different percentages of group content. They were quantitatively analysed and evaluated as o/w emulsifying agent. Methylated gum with maximum methyl content showed all the qualities desirable in a good emulsion formulation. It had the advantage over normally used emulsifiers like Span: Tween of imparting viscosity to the emulsion and allowing formulations of medicaments in it.

Key words: Plant gum, *Anogeissus latifolia*, Emulsifier.

Introduction

Natural plant gum obtained from *Anogeissus latifolia* Wall (Family Combretacae) is widely available in the central part of India. In admixture with gum Acacia it is sold as Ghatti gum in European markets [1]. Being non ionic in nature, plant gums stabilize the emulsions by forming multimolecular films at the interphase and also impart viscosity to the continuous phase [2].

These plant gums which have the inherent characteristics of viscosity builder, also provide us ample opportunity to introduce certain groups to get the desired emulsifier out of them.

Various ether and ester derivatives of cellulose, depending on kind and percentage of ether or ester linkage content, show varied hydrophilic and hydrophobic behaviour. They also exhibit different rheological characteristics [3].

Based on the above facts about derivatives of cellulose, methyl ether and acetate esters of the gum were prepared with varied percentage of group content. They were evaluated for emulsifying properties.

Experimental

Acetyl derivatives of gum. These derivatives with different acetyl contents were prepared by Wilson and Hughes method [4] using acetic anhydride in pyridine as acetylating agent. The reaction conditions were standardized by three repeat experiments preparing acetyl derivatives with different acetyl contents under different reaction conditions (Table 1).

Methoxyl derivatives of gum. The methoxyl derivatives with lower methoxyl content were prepared by the direct methylation method of Wadman *et al.* [5]. The higher methoxyl content derivatives were prepared by indirect methylation method of Howarth *et al.* [6]. which involved deacetylation and methylation of acetylated gum. A number of methoxyl derivatives were prepared under different reaction conditions out of which three methoxyl derivatives were selected for final preparation by the direct methylation method. The reaction conditions for their preparation were standardized by three repeat experiments. Two methoxyl derivatives were prepared by the de-acetylation methylation method with higher methoxyl content (Table 2).

Table 1. Preparation of Acetyl Dreivatives of Gum.

S. No.	Code name of acetylated gum	Acetylation mixture	Reaction time (hrs.)	Precipitating solvent	Yield (gm)	Acetyl content (%)
1.	AcG1	50ml acetic anhydride +100 ml pyridine	6	Methanol	53	11.1
2.	AcG2	50ml acetic anhydride +100 ml pyridin	12	Methanol	67	17.7
3.	AcG3	100 ml acetic anhydride +200 ml pyridine	6	Distilled water	76	29.5

- In each case 50 g gum was swollen with 600 ml of dimethylformamide.
- In each case the reaction was carried out at 80 + 5°.

Table 2. Preparation of Methoxy Derivatives of Gum.

S No.	Code name of methyled gum	Swelling agent (Aq. sodium hydroxide) (w/v)	Reaction time (hrs)	Volume dimethyl sulphate (ml)	Yield (gm)	%OCH_3 content
1.	MG1	20% 200 ml	3	100	42	6.54
2.	MG2	30% 200 ml	3	200	48	13.03
3.	MG3	40% 300 ml	3	300	52	19.62
4.	MG4*	40% 750 ml	6	400	48	32.65
5.	MG5*	40% 1000 ml	6	600	53	36.39

- In each case 50 g gum was used. The reaction was carried out at 70± 5°.
- Fully acetylated gum was taken in 500 ml Dioxane and reaction was carried out in nitrogen.

Quantitative analysis. The acetate derivatives were analysed by the USP. method [7]. The methyl derivatives were analysed by the zeissel method [8] (Table 1 and 2).

EVALUATION OF EMULSIFYING PROPERTY

Preparation of liquid paraffin emulsions. Light liquid paraffin I. P. emulsions were prepared using the prepared emulsifying agents. For comparative study, Tween 80 and Span 20, Acacia and the original gum were also used as emulsifying agents. For preparation of emulsion using Acacia and the gum under study, the dry gum method was employed [9]. The resultant emulsion was homogenized twice by passing through a hand homogenizer.

In the case of emulsions with prepared derivatives, the emlusifying agent was first triturated with a little chloroform water and liquid paraffin was added all at once. With continuous trituration, aqueous phase was added slowly and homogenized. For emulsions using Span 20 and Tween 80; Span 20 was dissolved in liquid paraffin and Tween 80 in chloroform water, both were mixed, shaken and homogenized.

Evaluation of emulsions. The emulsions were evaluated for the following four parameters of stability under normal storage conditions for four weeks.

(1) Physical characteristics. All emulsions were allowed to equilibrate for 2 hrs. Their visual appearance regarding colour was noted. The redispersibility and pourability studies were carried out after keeping for four weeks (Table 3).

(2) Creaming and phase separation. The creaming was studied by recording the creaming volume after 1,7,15 and 30 days (Table 4).

(3) Viscosity. The viscosity was determined after 1,7,15 and 30 days using Aimil-Emila Viscometer (Table 5).

(4) Globule size distribution. A study of globule size distribution with time was carried out using the microscopical method. The dispersed globules were counted with the help of a calibrated occular micrometer after suitable dilutions after 1 and 28 days (Table 6). The emulsions prepared with gum and its derivatives were compared with emulsions prepared with Acacia and combinations of Span 20 and Tween 80 for globule size distribution. The ratio of Span 20 and Tween 80 were calculated, based on the HLB value of liquid paraffin 12.

TABLE 3. FORMULAE AND PHYSICAL CHARACTERISTICS OF LIQUID PARAFFIN EMULSIONS PREPARED WITH THE GUM AND ITS DERIVATIVES.

S. No.	Emulsifying agent	Wt. of emulsifier (g)	Polar phase chloroform water (ml q.s.)	Colour	Redispersibility after 4 week (sec.)	Permeability after 4 week
1.	Acacia	6.25	100	White	13	++
2.	Gum	6.25	100	White	17	++
3.	MG1	6.25	100	Creamy	6	+++
4.	MG2	6.25	100	Creamy	8	+++
5.	MG3	6.25	100	Creamy	8	+++
6.	MG4	6.25	100	Creamy	7	+++
7.	MG5	6.25	100	Creamy	9	+++
8.	AcG1	6.25	100	Creamy	11	+
9.	AcG2	6.25	100	Creamy	8	+
10.	AcG3	6.25	100	Creamy	10	++
11.	Span 20 I	2.90	100	White	7	+++
	Tween 80	3.30				
12.	Span 20 II	1.50	100	White	6	+++
	Tween 80	1.70				

– In each case 25% w/v emulsion was prepared using liquid paraffin (density - 0.8358/cc) in chloroform water I.P. -Fair (+); good (++) and very good (+++).

TABLE 4. EFFECT OF AGING ON CREAMING OF EMULSIONS PREPARED WITH THE GUM, ITS DERIVATIVES AND OTHER EMULSIFYING AGENTS.

S. No.	Emulsifying agent	Orignal volume (ml)	Ultimate volume (Vu/ml) after days				Creaming ratio (Vu/Vo) after days			
			1	7	15	30	1	5	15	30
1.	Acacia	20	6.0	5.8	5.7	5.6	0.300	0.290	0.285	0.280
2.	Gum	20	5.3	5.4	5.5	5.5	0.265	0.270	0.275	0.275
3.	MG1	20	5.9	5.9	6.0	6.0	0.295	0.295	0.300	0.300
4.	MG2	20	7.9	7.9	7.5	6.4	0.395	0.395	0.375	0.375
5.	MG3	20	6.7	6.3	5.8	5.6	0.335	0.315	0.290	0.280
6.	MG4	20	7.7	7.4	6.6	6.0	0.385	0.370	0.330	0.300
7.	MG5	20	9.5	9.4	9.2	9.2	0.475	0.470	0.460	0.460
8.	AcG1	20	11.5	11.2	11.0	10.7	0.575	0.560	0.550	0.535
9.	AcG2	20	18.8	18.4	17.2	16.6	0.940	0.920	0.850	0.830
10.	AcG3	20	9.4	9.5	10.0	10.1	0.470	0.475	0.500	0.505
11.	Span 20 I Tween 80	20	6.9	6.3	5.9	5.5	0.345	0.315	0.295	0.275
12.	Span 20 II Tween 80	20	6.2	5.8	5.5	5.4	0.310	0.290	0.275	0.270

Discussion

The pourability and redispersibility studies were done after 4 weeks of aging. Pourability was poor with acetyl derivatives of gum which formed redispersible flocculates. The methoxy derivatives of gum and span: Tween combination emulsions showed very good pouring characteristics. Redispersibility was quite satisfactory with methoxy derivatives of gum and Span: Tween combination. Acacia, gum and acetyl derivatives (AcGl and AcG2) gave poor redispersible emulsions. Poor pourability and redispersi bility in such emulsions may be attributed to the swelling nature of them.

The effect of aging on viscosity after 7, 15 and 30 days was studied. Emulsions made with gum had higher viscosity than that made with Acacia. All the emulsions with methoxy and acetyl derivatives showed lower viscosity than emulsions with gum and it changed very slightly on deeping for one month. It was a good sign for better emulsifying nature. Span: Tween emulsions had quite low viscosity. MG5 with highest methoxy content, gave emulsion, viscosity which remained unchanged under the period of study. It imparted good nonpolar nature to the gum and retained or even improved other characteristics of gum as emulsifying agent, like pourability and re-dispersibility.

It was observed that with increase in methoxy content, the methoxy derivatives showed successively higher percentage of reducing size of oil globules. On storage, neither the globule size distributions nor viscosities were affected appreciably.

The acetyl derivaties showed varied effects. As acetyl content increased the nonpolar nature increased with reduction in polar nature. Hence AcG2 which had intermediate polarity could provide better emulsions. It had the highest creaming ratio. So far globule size distribution was concerned, it could not compete with MG5. All these emulsifying agents proved overall better than Acacia and the gum under study. The results are comparable with Span: Tween emulsions. These emulsions have added quality of higher viscosity which may be helpful in compounding medicinal agents in them.

TABLE 5. EFFECT OF AGING ON VISCOSITIES OF EMULSIONS PREPARED WITH THE GUM, ITS DERIVATIVES AND OTHER EMULSIFYING AGENTS.

S.No.	Emulsifying agents	Viscosities in centipoises at room temp. after days			
		1	7	15	30
1.	Acacia	3.5	3.5	4.0	4.0
2.	Gum	7.5	7.0	6.5	6.5
3.	MG1	2.5	2.5	2.0	2.0
4.	MG2	8.5	8.0	8.0	7.5
5.	MG3	4.5	4.5	4.0	3.5
6.	MG4	3.5	3.0	2.5	2.5
7.	MG5	4.5	4.5	4.5	4.5
8.	AcG1	3.0	3.0	2.5	2.5
9.	AcG2	2.0	2.5	2.5	2.5
10.	AcG3	2.0	2.5	2.5	2.5
11.	Span 20 I Tween 80	1.5	2.0	2.0	2.0
12.	Span 20 II Tween 80	1.5	1.5	1.5	1.5

TABLE 6. EFFECT OF AGING ON GLOBULE SIZE DISTRIBUTION OF EMULSIONS PREPARED WITH THE GUM, ITS DERIVATIVES AND OTHER EMULSIFYING AGENTS.

S. No.	Emulsifying agent	Globule size in microns											
		0.5 – 1		1 – 2		2 – 3		3 – 4		4 – 5		5 – 8	
		I*	II*	I	II	I	II	I	II	I	II	I	II
1.	Acacia	87	60	7.0	30	3.0	7.0	2.0	2.0	0.6	0.6	0.4	0.4
2.	Gum	75	70	16.0	14	6.0	11.0	2.8	4.0	0.2	0.7	0.0	0.0
3.	MG1	79	72	19.0	15	1.5	8.0	0.5	3.0	0.0	1.5	0.0	0.5
4.	MG2	88	78	10.0	15	1.5	4.5	0.3	1.5	0.2	0.7	0.0	0.3
5.	MG3	89	82	6.0	12	4.0	4.5	0.5	1.0	0.3	0.4	0.2	0.1
6.	MG4	91	85	6.0	13	2.5	1.5	0.3	0.4	0.2	0.1	0.0	0.0
7.	MG5	97	94	2.0	4	0.6	1.0	0.3	0.4	0.1	0.4	0.0	0.2
8.	AcG1	77	72	21.0	11	1.5	10.0	0.5	6.0	0.0	0.4	0.0	0.6
9.	AcG2	80	73	17.0	22	2.0	4.0	1.0	0.6	0.0	0.4	–	–
10.	AcG3	58	45	28.0	20	10.0	28.0	2.0	3.0	1.0	2.0	1.0	2.0
11.	Span 20 I Tween 80	98	88	1.5	10	0.5	1.0	0.0	0.5	0.0	0.5	0.0	0.0
12.	Span 20 II	92	83	5.0	14	2.0	2.0	0.4	0.5	0.4	0.5	0.2	0.0

I – Initial; II – After 28 days.

Acknowledgements. The authors are thankful to Prof. N. K. Jain, Head, Dept. of Pharm. Sci., Dr. H. S. Gour Vishwavidyalaya, Sagar (M.P.) for providing necessary facilities.

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