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Received for review August 29, 1953. Accepted December 28, 1953. Presented before the Division of Agricultural and Food Chemistry, Symposium on Problems of Deterioration in Food Fats, at the 124th Meeting of the AMERICAN CHEMICAL SOCIETY, *Chicago, Ill.*

SUGAR SOLUBILITY

Sugar Mixtures in Aqueous Glycerol

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Sucrose, dextrose, and invert sugar are used in foods, candies, sirups, and pharmaceutical preparations and, at times, glycerol is included as a softener or a humectant, or for its effect upon texture. Consequently, it is desirable to know the amount of sugar that can be dissolved in aqueous glycerol, so that maximum sugar concentrations can be estimated. Higher concentrations of dissolved sugar can be attained with mixtures of sucrose and dextrose than with either sugar alone. Invert sugar is also a mixture but of constant composition, its equimolar dextrose-levulose composition fixed by its origin. Used by itself, it shows the same type of solubility curve as do the single sugars—i.e., solubility decreases with decreasing temperature and increasing glycerol concentration. Its solubility is limited by its dextrose component.

MANY FOODS, CONFECTIONS, AND PHARMACEUTICAL PREPARATIONS are improved by the addition of glycerol. As a sugar will also be an ingredient in most of such uses, it is desirable to know the solubility of the sugar or sugars in the water phase—i.e., in aqueous glycerol. The solubilities of single sugars in aqueous glycerol at 15°, 25°, and 35° C. have been previously reported (*8*).

If a mixture of sucrose and dextrose is used, the presence of one will lessen the solubility of the other, but their combined solubilities will exceed that of either. This affords a way of introducing the maximum amount of sugar into a solution. Information is presented here on the solubility of sucrose-dextrose mixtures and of invert sugar in aqueous glycerol at 25° and 35° C.

Materials

Glycerol, U.S.P., 95% concentration. Armour and Co., Chicago.

Sucrose, C.P., 99.9+%. Specific rotation 66.5°. Pfanstiehl Chemical Co., Waukegan, Ill.

Dextrose, C.P., anhydrous, 99.9+%. Special sample. Corn Products Refining Co., Argo, Ill.

d(−) Levulose, C.P., special. Specific rotation −92°. Pfanstiehl Chemical Co., Waukegan, Ill.

Water, distilled.

Apparatus and Procedure

Apparatus and procedure have been described (*8*). In brief, the solutions were prepared in 100-ml. serum bottles closed with sleeve stoppers and immersed in a water bath kept within ±0.05° C. of the desired temperature. The specific gravity of the solutions was measured at 25° C. with 25-ml. pycnometers and refractive index at 25° C. with an Abbe

Table I. Solubility of Dextrose in Aqueous Glycerol Containing a Fixed Amount of Sucrose

Solvent, Glyc.-Water Ratio	At 25° C. Solution		At 25° C. Solution, % Composition			At 35° C. Solution		At 35° C. Solution, % Composition		
	Sp.gr., 25/25	n_D^{25}	Dextrose	Sucrose	Solvent	Sp.gr. 25/25	n_D^{25}	Dextrose	Sucrose	Solvent
25:75	1.2481	1.4323	46.5	0.0	53.5	1.2836	1.4463	54.5	0.0	45.5
	1.2662	1.4381	37.8	9.5	52.6	1.3010	1.4509	49.1	8.1	42.8
	1.3008	1.4511	33.9	23.3	42.8	1.3181	1.4569	42.3	19.1	38.6
	1.3338	1.4640	27.0	36.2	36.8	1.3437	1.4689	35.3	30.4	34.3
	1.3347	1.4630	10.3	52.1	37.6	1.3482	1.4688	9.9	55.6	34.5
50:50	1.2585	1.4430	38.1	0.0	61.9	1.2915	1.4541	46.6	0.0	53.4
	1.2732	1.4478	33.9	6.1	60.0	1.2944	1.4569	41.6	6.8	51.6
	1.2974	1.4565	28.9	17.3	53.8	1.3182	1.4640	37.2	15.3	47.5
	1.3249	1.4660	27.4	26.3	46.3	1.3442	1.4730	33.3	23.4	43.3
	1.3286	1.4672	16.3	37.8	45.9	1.3364	1.4695	13.8	41.1	44.6
62.5:37.5	1.2647	1.4503	33.0	0.0	67.0	1.2918	1.4587	40.9	0.0	59.1
	1.2745	1.4531	27.8	5.4	66.8	1.3056	1.4631	38.1	6.2	55.7
	1.2989	1.4610	26.2	13.5	60.3	1.3200	1.4682	34.8	13.0	52.2
	1.3156	1.4673	25.0	21.6	53.4	1.3366	1.4734	31.3	21.3	47.4
	1.3229	1.4696	17.6	30.3	52.7	1.3531	1.4795	24.9	28.1	47.0
75:25	1.2673	1.4570	25.6	0.0	74.4	1.2869	1.4636	32.5	0.0	67.5
	1.2862	1.4630	26.4	4.3	69.3	1.3025	1.4674	31.1	5.3	63.6
	1.2965	1.4660	25.1	7.9	67.0	1.3142	1.4715	30.8	9.9	59.3
	1.3111	1.4703	22.2	16.1	61.7	1.3257	1.4747	27.6	15.8	56.6
	1.3255	1.4748	21.7	21.3	57.0	1.3397	1.4794	24.3	23.7	52.0
82.5:17.5	1.2690	1.4620	21.0	0.0	79.0	1.2862	1.4670	27.1	0.0	72.9
	1.2879	1.4672	20.7	5.0	74.3	1.2995	1.4708	25.6	4.0	70.4
	1.2954	1.4700	19.5	8.5	72.0	1.3099	1.4736	24.9	8.7	66.4
	1.3058	1.4725	19.1	12.3	68.6	1.3202	1.4764	23.4	13.8	62.8
	1.3156	1.4753	18.8	16.2	65.0	1.3089	1.4732	12.4	20.2	67.4
95:5	1.2723	1.4711	14.3	0.0	85.7	1.2833	1.4740	17.3	0.0	82.7
	1.2897	1.4761	11.7	4.3	84.0	1.2996	1.4775	17.3	3.1	79.6
	1.2922	1.4766	11.2	5.6	83.2	1.3073	1.4808	17.4	6.9	75.7
	1.2931	1.4772	9.8	7.8	82.4	1.3113	1.4813	16.0	8.4	75.6
	1.2782	1.4731	8.9	8.0	83.1	1.3156	1.4828	12.8	10.2	77.0

refractometer. Sugar concentrations of the sucrose-dextrose mixtures were determined by the Clerget method. Concentrations of invert sugar were calculated from simple saccharimeter readings.

Experimental

Glycerol-water solutions containing 25.0, 50.0, 62.5, 75.0, 82.5, and 95.0 ± 0.10% by weight of glycerol were prepared, their concentrations being determined from their specific gravity by reference to the table of Bosart and Snoddy (*5*).

Sugar solutions in the glycerol-water solvents were prepared in the 100-ml. serum bottles which were submerged and rotated in the constant-temperature water bath. Saturation was judged to be complete when about 0.5 gram of sugar crystals remained undissolved and the refractive index of the solution did not change in 4 days. After saturation was complete, the bottles of solution were stored in the constant-temperature bath. Solutions were prepared at 25° and 35° C.

To prepare the solutions of mixed sugars, one sugar was added in fixed amounts sufficient for $^1/_4$, $^1/_2$, $^3/_4$, and complete saturation of the glycerol-water solvent [see (*8*) for solubility data]. The second sugar was then added to saturation. A second set of solutions was prepared with the roles of the sugars reversed. The second sugar was added to the solution, in larger amounts at first, then in 1-gram portions as saturation was approached, until crystals remained after 4 days of continuous agitation and the refractive index of the solution remained constant. The solution was then considered to be saturated, whether the crystals were of the sugar first added or of the second sugar. Crystals of the first sugar, when present, were formed as a result of displacement by the second sugar. The sugar crystals were identified by microscopic examination.

The sucrose-dextrose solutions were analyzed by the Clerget method (*2*), in which the optical rotation of the solutions is measured before and after inversion of the sucrose. The following form of the method was used:

Direct Polarization. Weigh accurately 10 ml. of the saturated sugar solution in a 50-ml. volumetric flask. Add one drop of ammonia to assure rapid mutarotation of the dextrose and dilute to 50 ml. Read the optical rotation of the solution at 20° C.

Invert Polarization. Weigh accurately 10 ml. of the saturated sugar solution in a 50-ml. volumetric flask. Add 25 ml. of water and 10 ml. of 1 to 1 diluted hydrochloric acid. Mix and let stand overnight at 20° to 25° C. Dilute the solution to 50 ml. and measure the optical rotation at 20° C.

The concentration of sucrose was calculated with the formula given by the Association of Official Agricultural Chemists (*1*) and the concentration of dextrose was calculated with the formula given by Browne and Zerban (*6*).

The specific gravity and the refractive index of the solutions were measured at 25° C.

Data for the sucrose-dextrose solutions are given in Tables I and II. The solubilities of the mixed sugars in water were determined by Jackson and Silsbee (*7*). Interpolations of their data are given in Table III. The solubility curves of the solutions are given in Figures 1 and 2. The dextrose is calculated as anhydrous dextrose.

The accuracy of the analyses by the Clerget method was confirmed by determination of the sugar concentrations of some of the solutions by the copper reduction method of Hammond (*3*). The results are given in Table IV.

For the purpose of this work, invert sugar is considered as a mixed sugar because it is composed of dextrose and levulose in equimolar amounts. Because this ratio is always the same, no solubility measurements were made with other mixtures of the two sugars. To avoid the uncertainties of purity and completeness of inversion that would occur if sucrose were inverted, an artificial invert sugar was made by mixing equimolar amounts of anhydrous dextrose and levulose. This mixture was then added to the several glycerol-water solvents until they were saturated with

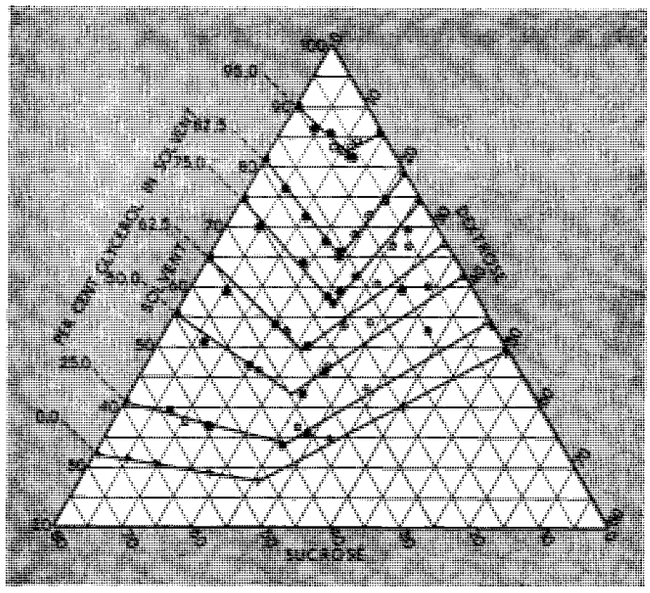


Figure 1. Solubility of sucrose and dextrose in aqueous glycerol at 25° C.

● Dextrose fixed, sucrose added to saturation
○ Sucrose fixed, dextrose added to saturation
+ Jackson and Silsbee data

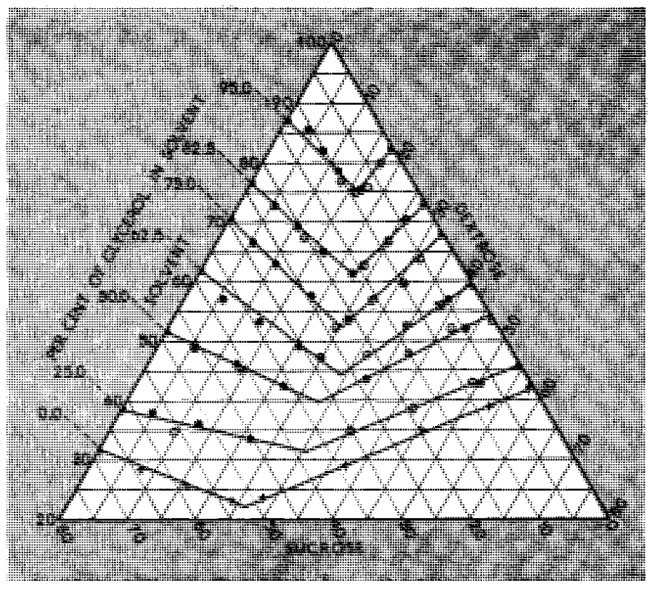


Figure 2. Solubility of sucrose and dextrose in aqueous glycerol at 35° C.

● Dextrose fixed, sucrose added to saturation
○ Sucrose fixed, dextrose added to saturation
+ Jackson and Silsbee data

the sugar. Levulose is more soluble than dextrose and therefore the solubility of the mixture is limited by the dextrose solubility. Solutions were prepared at 25° and 35° C. and the same criteria of saturation were used as before. The data for these solutions are given in Table V and Figure 3.

Discussion

An inspection of Figures 1 and 2 shows that the presence of one sugar in aqueous glycerol will diminish the solubility of a second sugar, but the total amount of the two sugars will, in many cases, be greater than either alone. As is expected from the solubility of single sugars, the solubility of sugar mixtures is diminished by

Table II. Solubility of Sucrose in Aqueous Glycerol Containing a Fixed Amount of Dextrose

Solvent, Glyc.-Water Ratio	At 25° C.					At 35° C.				
	Solution		Solution, % Composition			Solution		Solution, % Composition		
	Sp.gr. 25/25	n_D^{25}	Sucrose	Dextrose	Solvent	Sp.gr. 25/25	n_D^{35}	Sucrose	Dextrose	Solvent
25:75	1.3175	1.4571	59.4	0.0	40.6	1.3286	1.4610	61.9	0.0	38.1
	1.3249	1.4600	53.5	7.4	39.1	1.3369	1.4650	57.9	4.9	37.2
	1.3365	1.4639	49.7	13.8	36.5	1.3464	1.4689	51.9	12.5	35.6
	1.3527	1.4704	40.2	26.1	33.7	1.3588	1.4731	45.4	21.5	33.1
	1.3349	1.4635	35.9	28.6	35.5	1.3041	1.4523	7.3	50.2	42.5
50:50	1.2963	1.4564	44.8	0.0	55.2	1.3077	1.4599	48.7	0.0	51.3
	1.3094	1.4603	43.0	6.4	50.6	1.3203	1.4650	45.7	5.5	48.8
	1.3224	1.4650	38.8	14.6	46.4	1.3319	1.4695	41.3	13.0	45.7
	1.3388	1.4704	33.1	24.8	42.1	1.3456	1.4740	36.1	21.7	42.2
	1.3264	1.4665	28.0	26.2	45.8	1.3029	1.4586	4.7	43.6	51.7
62.5:37.5	1.2830	1.4570	35.3	0.0	64.7	1.2988	1.4610	38.4	0.0	61.6
	1.2991	1.4613	35.7	5.4	58.9	1.3096	1.4660	37.6	5.7	56.7
	1.3157	1.4665	31.1	15.2	53.7	1.3225	1.4699	34.1	12.9	53.0
	1.3280	1.4704	28.7	21.3	50.0	1.3371	1.4751	30.4	20.9	48.7
	1.2944	1.4592	10.2	30.3	59.5	1.3039	1.4633	4.7	38.9	56.4
75:25	1.2733	1.4597	25.4	0.0	74.6	1.2894	1.4639	29.5	0.0	70.5
	1.2898	1.4634	25.6	4.9	69.5	1.2988	1.4678	28.3	5.3	66.4
	1.3033	1.4671	22.3	13.7	64.0	1.3109	1.4710	26.8	10.5	62.7
	1.3214	1.4731	20.5	21.1	58.4	1.3248	1.4750	24.3	18.3	57.4
	1.3189	1.4722	19.3	21.6	59.1	1.3391	1.4790	21.0	25.7	53.3
82.5:17.5	1.2610	1.4640	19.0	0.0	81.0	1.2835	1.4666	23.3	0.0	76.7
	1.2843	1.4649	18.5	5.2	76.3	1.2930	1.4688	22.2	5.1	72.7
	1.2949	1.4689	17.6	10.5	71.9	1.3048	1.4728	20.6	10.4	69.0
	1.3060	1.4724	17.2	15.5	67.3	1.3168	1.4760	19.0	16.2	64.8
	1.3127	1.4740	16.3	18.2	65.5	1.3270	1.4789	16.3	22.4	61.3
95:5	1.2753	1.4723	9.9	0.0	90.1	1.2812	1.4743	13.0	0.0	87.0
	1.2827	1.4737	9.1	4.3	86.6	1.2856	1.4750	10.6	3.9	85.5
	1.2889	1.4750	7.6	7.1	85.3	1.2946	1.4774	10.4	7.6	82.0
	1.2953	1.4772	6.4	11.8	81.8	1.3022	1.4796	9.8	11.5	78.7
	1.2916	1.4764	6.4	12.2	81.4	1.3114	1.4816	8.7	16.2	75.1

Table III. Solubility of Dextrose and Sucrose in Water

(From Jackson and Silsbee)

Composition of Solution, % Weight		
Dextrose	Sucrose	Water
	At 25° C.	
0.0	68.0	32.0
4.8	64.0	31.2
9.4	60.2	30.4
18.0	53.0	29.0
25.7	46.9	27.4
32.7	32.6	34.7
40.5	19.4	40.1
50.8	0.0	49.2
	At 35° C.	
0.0	68.1	31.9
7.6	64.2	28.2
15.5	60.6	23.9
23.4	53.4	23.2
28.0	48.8	23.2
37.1	34.0	28.9
45.8	19.8	34.4
53.6	7.5	38.9
58.0	0.0	42.0

Table IV. Comparison of Sugar Determinations by Clerget and Hammond Methods

% Glycerol in Solvent	Clerget Method		Hammond Method	
	% dextrose	% sucrose	% dextrose	% sucrose
25	38	10	38	10
50	34	6	33	6
62.5	28	5	27	6
75	26	4	27	4
82.5	21	5	20	5
95	12	4	13	5
25	7	54	6	53
50	6	43	7	44
62.5	5	36	6	36
75	5	26	4	27
82.5	5	19	5	18
95	4	9	5	10

Table V. Solubility, Specific Gravity, and Refractive Index of Aqueous Glycerol Saturated with Invert Sugar, at 25° and 35° C.

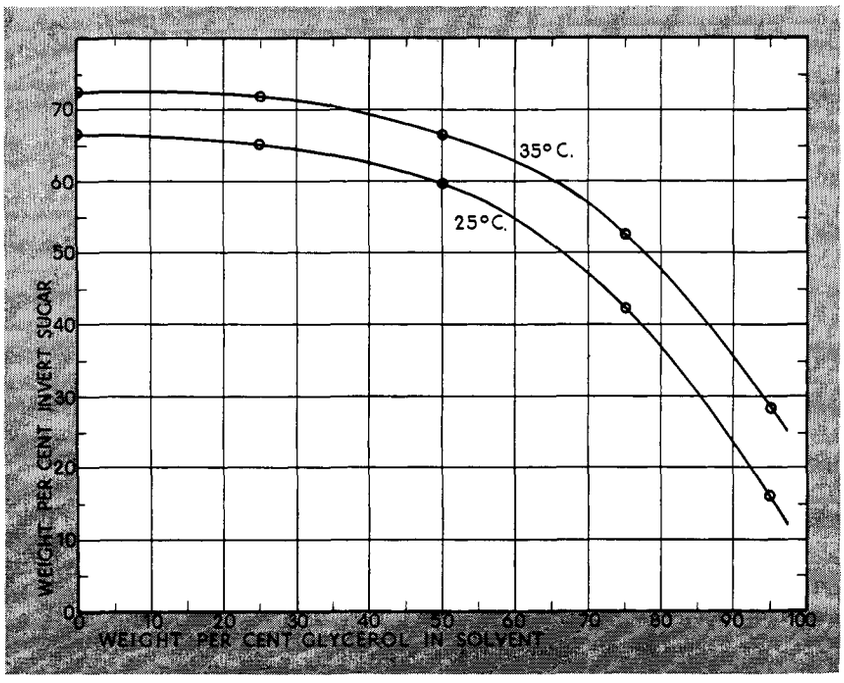
Solvent, Glyc.-Water Ratio	Solution Saturated at 25° C.				Solution Saturated at 35° C.			
	Sugar, % wt.	Sugar, g./100 ml.	Sp.gr. 25/25	n_D^{25}	Sugar, % wt.	Sugar, g./100 ml.	Sp.gr. 25/25	n_D^{25}
0:100	66.2(4)	. .	. .	. .	72.2	. .	. .	. .
25:75	65.0	86.7	1.3332	1.4621	71.6	98.6	1.3765	1.4781
50:50	59.7	79.6	1.3327	1.4686	66.2	90.3	1.3643	1.4785
75:25	42.4	55.9	1.3174	1.4710	52.5	70.8	1.3482	1.4804
95:5	16.0	20.6	1.2874	1.4750	28.7	37.9	1.3190	1.4829

lower temperatures and higher glycerol concentrations.

In sucrose-dextrose solutions the sugar which will separate as the solid phase depends upon the amount of each sugar present. It may be either the second sugar being added, or the first sugar being displaced by the second sugar going into solution. When dextrose is the solid phase, it may be either the monohydrate or the anhydrous form. The monohydrate is stable in solutions containing 62.5% or less of glycerol in the solvent, excepting 62.5% glycerol at 35° C., and anhydrous dextrose is stable in solutions containing 75% or more of glycerol in the solvent, and in 62.5% glycerol at 35° C. The range of glycerol concentration from 62.5% to 75% has not been explored.

Invert sugar, although a mixture, was treated essentially as a single sugar because the proportions of dextrose and levulose in it are constant and its solubility is limited by the dextrose. Therefore, its solubility as depicted in Table V and Figure 3 is analogous to the solubilities of single sugars as previously reported (*6*).

Figure 3. Solubility of invert sugar in aqueous glycerol at 25° and 35° C.



Acknowledgment

The pure dextrose used in this work was generously supplied by George R. Dean, Corn Products Refining Co.

The work was sponsored by the Glycerine Division, Association of American Soap and Glycerine Producers, New York, N. Y.

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Received for review October 16, 1953. Accepted December 19, 1953.