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Comparison of the viscosity of trehalose and sucrose solutions at various temperatures: Effect of guar gum addition

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ABSTRACT

The viscosity of trehalose and sucrose solutions was measured and compared as well as the effect of adding guar gum. Viscosity measurements of trehalose and sucrose solutions were performed in the range 20–45 g/100 g; 0.1 g guar gum/100 g was added in mixed systems. Viscosity of trehalose or sucrose solutions was measured at 20, 27 and 34 °C. Trehalose solutions had always a higher viscosity than sucrose, with the biggest difference ($p < 0.001$) in the range 30–45 g/100 g. Addition of guar gum revealed a synergistic effect with both disaccharides, particularly for trehalose. Viscosity dependence on temperature was also analyzed by calculating activation energies for all systems studied.

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1. Introduction

Trehalose (α -D-glucopyranosyl- α -D-glucopyranoside) is a naturally occurring, non toxic disaccharide with the same chemical formulae as sucrose but slightly different structure, consisting of two glucose molecules linked by a 1,1 α , α -glycosidic bond which is not cleaved by α -glucosidase. It is increasingly available for food application, with considerable potential for the food industry, being used to improve existing products or to create innovative new ones. Among its properties, trehalose is almost half as sweet as sucrose while having a similar sweetness dynamic profile (Galmarini, Zamora, & Chirife, 2009), it is non-reducing thus not reacting with amino acids or proteins according to Maillard browning reaction, and it is also stable under low pH conditions where other disaccharides would hydrolyze into their component monosaccharides (Komes, Lovric, Ganic, & Gracin, 2003; Komes et al., 2005). Trehalose has also a low cariogenic potential when compared to sucrose and lower glycaemic and insulinemic response, particularly in obese men according to Maki, Kanter, Rains, Hess, and Geohas (2009) and depresses water activity in the same way as sucrose (Galmarini, Chirife, Zamora, & Perez,

2008). For these reasons in the past few years trehalose has been used (or proposed to be used) alone or in combination with sucrose in a variety of beverages and food products since it could improve physico chemical characteristics, while optimizing sweetness and maintaining product shelf life.

Besides sugars, hydrocolloids are also commonly used when formulating beverages as stabilizers and/or thickening agents. Therefore it is important to study mixed systems (sugar + hydrocolloids) since there could be different interactions which could affect their rheological behavior. Guar gum is a well known thickener usually employed in the food industry (Maier, Anderson, Karl, & Magnuson, 1993) where rheological properties play an important role especially in food process design, being required for calculation in any process involving fluid flow.

The aim of the present work was to measure and compare the viscosity of trehalose and sucrose solutions in a concentration and temperature range of practical interest and to evaluate the effect of the addition of guar gum on the viscosity of the mentioned solutions.

2. Materials and methods

2.1. Preparation of solutions

Sucrose (S) was obtained from Laboratorio Anedra, Buenos Aires, Argentina; crystalline trehalose dihydrate (T) was provided by Cargill Inc., Wayzata MN, U.S.A and guar gum (GG) was from

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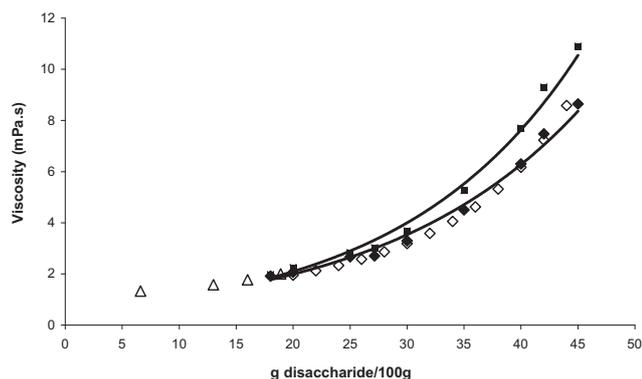


Fig. 1. Comparison of viscosity of trehalose and sucrose solutions (present work and literature data) at 20 °C. ◆ Sucrose (this work); ■ trehalose (this work); ◇ sucrose (Bubnik et al., 1995); △ trehalose (Sampedro et al., 2002).

Gelfix S.A., Argentina. Distilled water was used to prepare all solutions.

Disaccharide solutions were prepared by adding the sugar to a weighed amount of distilled water with constant stirring. Selected concentrations were: 18; 20; 25; 27; 30; 35; 40; 42 and 45 g/100 g. It is to be noted that a 45 g/100 g trehalose solution is near to its maximum solubility at 20 °C (Lammert, Schmidt, & Day, 1998). Final weight was controlled after stirring the solutions in order to compensate any water loss by evaporation.

Disaccharide systems containing guar gum at 0.1 g/100 g were prepared by first mixing the solids which were then dissolved in a weighed amount of distilled water with constant stirring. For the purpose of comparison, solutions consisting of only guar gum (in the range 0.1 to 0.3 g/100 g) were prepared and measured under the same conditions above mentioned.

2.2. Viscosity determination

Viscosity was measured by means of a rotating viscometer (Brookfield DV-LVT; Brookfield Engineering Laboratories, Inc., Middleboro, U.S.A) using the UL/Y adapter with UL spindle. The sample chamber was placed in a water jacket connected to a constant temperature bath in order to determine viscosities at 20, 27 and 34 °C in order to cover a “practical” range of interest for beverage storage. Measurements were made at several rotational speeds corresponding to a percentage torque ranging from 10 to 100. All determinations were done in duplicate at increasing shear rates, discarding values corresponding to a percentage torque

smaller than 10% due to the amplified errors in the readings. Reproducibility was calculated measuring seven replicates of trehalose solutions at 20 and 42 g/100 g at 20 °C obtaining an error (considered as twice the standard deviation) below 2% in both cases.

2.3. Statistical analysis

Analysis of variance (ANOVA) was carried out to assess if viscosity was significantly different between trehalose and sucrose using the general linear model command in SPSS version 13.0 (SPSS, Inc., Chicago, IL). The variability of the viscosity was studied using a model where concentration and disaccharide were considered as fixed factors. Multiple means comparisons were carried out by Tukey's honestly significantly different test at $p < 0.05$. A more conservative test such as Tukey was used to reduce the probability of error (finding a significant difference when there is none).

The effect of temperature on viscosity for disaccharide solutions (in the range 20–45 g/100 g) was analyzed by a One Way ANOVA. Measured and predicted viscosities of mixed systems (guar gum + saccharide) at different concentrations were compared by t- test for two paired samples also using SPSS v. 13.0 (Inc. Chicago, IL).

3. Results and discussion

3.1. Comparison of the viscosity of trehalose and sucrose solutions

Sucrose and trehalose showed a Newtonian behaviour, being their viscosity independent from the shear rate. Viscosity values here reported were obtained from the slope of the straight line resulting from plotting shear stress as a function of shear rate (data not shown). Fig. 1 shows measured viscosities at 20 °C of trehalose and sucrose solutions. The data from Bubnik, Kadlec, Urban, and Bruhns (1995) (for sucrose solutions) and from Sampedro, Muñoz-Clares, and Uribe (2002) (for trehalose solutions) where also plotted in Fig. 1 for the purpose of comparison; it is to be noted that the data from Sampedro et al. (2002) corresponds to a lower range of trehalose concentration following the same tendency as present data. Although data for viscosity of sucrose solutions are available from literature, they were also measured in order to check the reliability of our determinations.

Viscosity of sucrose and trehalose solutions increased in an exponential way as concentration increased; being the viscosity between the two sugars different ($p < 0.001$) in the range 35–45 g/100 g.

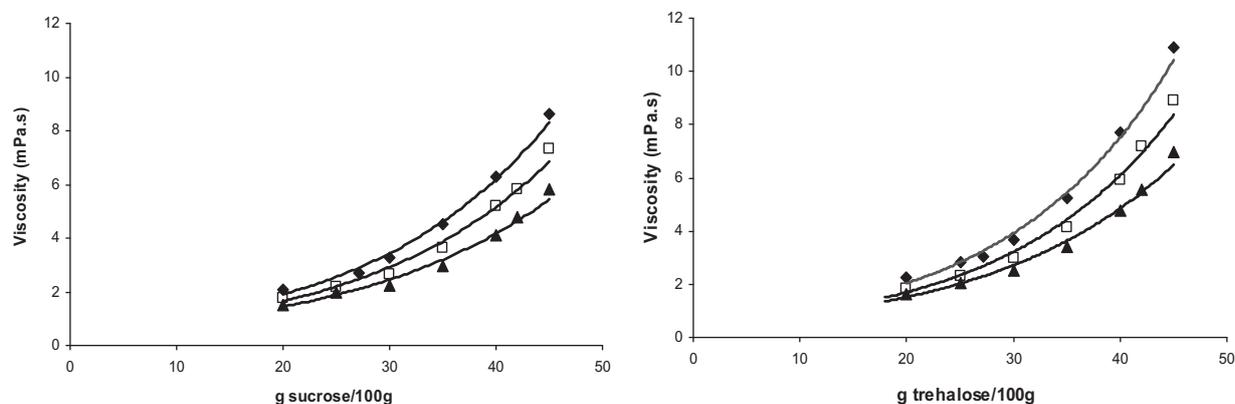


Fig. 2. Effect of temperature (20 to 34 °C) on the viscosity of trehalose and sucrose solutions. ◆ 20 °C; □ 27 °C; ▲ 34 °C.

Table 1
Correlation coefficients (R^2) and values for constants (A_0 and A_1) relating viscosity to concentration according to equation: $\text{Viscosity} = A_0 e^{(A_1 \cdot c)}$, where A_0 and A_1 are empirical parameters.

Temperature (°C)	Trehalose			Sucrose		
	A_0	A_1	R^2	A_0	A_1	R^2
20	0.554	0.065	0.989	0.586	0.059	0.989
27	0.474	0.064	0.989	0.528	0.057	0.987
34	0.474	0.058	0.988	0.500	0.053	0.984

Fig. 2 shows the viscosity of sucrose and trehalose at all three temperatures (20, 27 and 34 °C). Trehalose solutions showed a viscosity higher than that of sucrose ones at all temperatures; differences, however, became smaller as temperature increases.

The viscosity dependence on concentration could be explained by the following exponential empirical equation;

$$\text{Viscosity} = A_0 e^{(A_1 \cdot c)} \quad (1)$$

where A_0 and A_1 are empirical parameters and c is concentration expressed as g disaccharide/100 g. Values for A_0 and A_1 for sucrose and trehalose solutions were calculated by non linear regression analysis of experimental data shown in Fig. 2, and are shown in Table 1 for every measured temperature.

Values of A_1 were found to be higher ($p < 0.001$) for trehalose than for sucrose at every temperature; this would indicate a greater dependency of viscosity on concentration for trehalose. Rampp, Buttersack, and Lüdemann (2000) observed measurable differences in dynamic properties (viscosity and diffusion coefficient) between trehalose and sucrose solutions at concentrations higher than 30 g/100 g. At higher concentrations the direct contact between sugar molecules through hydrogen bonds becomes probable due to an increase in the number of molecules in the solution. The rigid carbon skeleton of the disaccharides then imposes sterical restraints which may influence these dynamic properties (Rampp et al., 2000). This reveals the importance of spatial configuration of each disaccharide in relation to their viscosity.

Disaccharide/water interactions could also explain the differences observed for viscosity. Branca et al. (2001) found that the trehalose/water system is characterized by the highest values of the interaction strength parameter of the hydration number obtained by ultrasonic and viscosity measurements. They supported the hypothesis of a highly strong thermodynamic character (high water-sugar interaction) of the trehalose/water system at low water content in comparison to sucrose. These observations were supported by Magazú, Migliardo, Mondelli, and Romeo (2003) who, by means of neutron scattering experiments, obtained a more rigid structure for trehalose/water respect to sucrose/water mixtures on a nanoscopic scale.

In this way, a higher interaction between trehalose molecules and water and the formation of a more rigid structure might explain the higher increase in viscosity for this sugar when compared to sucrose.

3.2. Effect of guar-gum addition

Addition of 0.1 g/100 g of guar gum to the sugar solutions resulted in a pseudoplastic behavior as observed for guar gum solutions. Fig. 3 shows relative viscosity in terms of shear rate for four mixed systems (sugar plus guar gum), being these the maximum and minimum concentrations studied (20 and 45 g disaccharide/100 g). While disaccharide solutions presented a Newtonian behavior, in mixed systems a pseudoplastic behavior

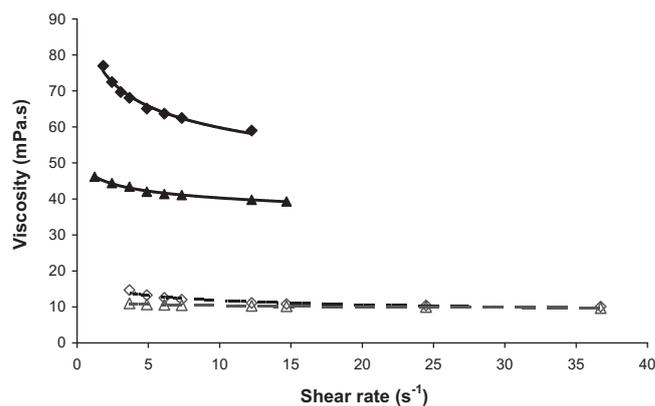


Fig. 3. Effect of adding 0.1 g guar gum/100 g on the viscosity behaviour of trehalose and sucrose solutions of different concentrations. \blacklozenge (45 g trehalose + 0.1 g guar gum)/100 g; \blacktriangle (45 g sucrose + 0.1 g guar gum)/100 g; \diamond (20 g trehalose + 0.1 g guar gum)/100 g; \triangle (20 g sucrose + 0.1 g guar gum)/100 g.

was observed, showing a decrease in viscosity as shear stress increased. Similar results have been previously found for other mixed (disaccharide + polysaccharide) systems (Cancela, Alvarez, & Maceiras, 2005).

At high concentrations there is a synergistic relationship between trehalose and sucrose with guar gum. It was observed that, while the viscosity of the 0.1 g/100 g guar gum solution was of 5.4 mPa.s at 12.2 s^{-1} (data not shown), the mixed model consisting of (45 gT + 0.1 g GG)/100 g had a value of 59 mPa.s at 12.2 s^{-1} and the (45S + 0.1GG)/100 g solution presented an apparent viscosity of 38.4 mPa.s at the same rotational speed.

Fig. 4 shows the difference between measured viscosity for trehalose + GG and measured viscosity for sucrose + GG at each sugar concentration (20, 30, 40 and 45 g/100 g). As sugar concentration increases the difference between sucrose and trehalose with added guar gum, also increases emphasizing a more synergistic relationship between trehalose and guar gum.

In order to evaluate the degree of the synergistic relationship between sucrose or trehalose and guar gum, the measured viscosity (apparent viscosity determined at the possible maximum shear stress) of each solution was compared to a calculated viscosity which was obtained by adding the viscosity of each component of the mixture measured separately. For this purpose, the individual

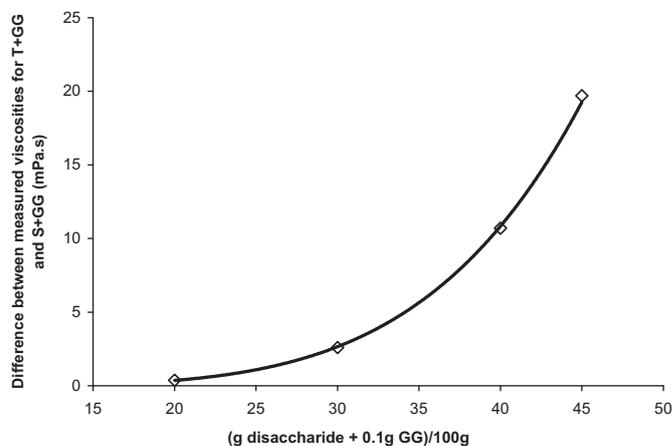


Fig. 4. Difference between measured viscosity of trehalose (T) + guar gum (GG) and sucrose (S) + guar gum (GG) at 20, 30, 40 and 45 g of sugar/100 g. Being the empirical equation that fits to data $y = 2E-07x^{4.8903}$; $r^2 = 0.9999$.

Table 2
Interaction effects on viscosity in the mixed systems disaccharide + guar gum.

System	Viscosity _{calc} (mPa s)	Viscosity _{meas} (mPa s)	Δ Viscosity (%)
(20 g S + 0.1 g GG)/100 g	9.28	9.15	-1.4
(20 g T + 0.1 g GG)/100 g	9.46	9.47	0.1
(30 g S + 0.1 g GG)/100 g	12.32	16.00	29.9
(30 g T + 0.1 g GG)/100 g	12.71	18.60	46.3
(40 g S + 0.1 g GG)/100 g	18.39	26.20	42.5
(40 g T + 0.1 g GG)/100 g	19.79	36.30	83.4
(45 g S + 0.1 g GG)/100 g	23.24	39.30	69.1
(45 g T + 0.1 g GG)/100 g	25.49	59.0	131.5

S: sucrose, T: trehalose, GG: guar gum; Viscosity_{calc}: sum of the viscosity of components in the system; Viscosity_{meas}: apparent viscosity at maximal shear rate (s⁻¹) measured; Δ Viscosity (%): [(Viscosity_{meas}/Viscosity_{calc}) - 1] x 100.

viscosity of gum suspensions was considered at its effective concentration in the mixture, taking into account only the quantity of water available in the mixture. The effective concentration of 0.1 g GG/100 g resulted 0.125, 0.143, 0.167 and 0.182 g/100 g for blends having 20, 30, 40 and 45 g/100 g sugar concentration. This correction was not made for the individual viscosity of sugar solutions since 0.1 g/100 g of GG has no major effect on the amount of water available for the sugar dissolution. Viscosity values for guar gum solutions in the range 0.1–0.3 g/100 g were calculated according to empirical (equation 1) with values of 1.537 for A₀ and 12.356 for A₁.

According to data presented in Table 2 systems formed by 20 g sugar/100 g and 0.1 g GG/100 g could be approximately explained by the sum of the individual viscosities. However, as the sugar concentration increased the calculated viscosity was lower than measured viscosity (*p* < 0.05) showing a synergistic interaction between disaccharides and guar gum was observed, since the difference between calculated and measured viscosity increased up to 69% and 131% for S + GG and T + GG, respectively.

Other authors have studied viscosity of mixed (disaccharide + polysaccharide) systems finding a different effect according to the disaccharide present in the solution. As an example, Sato, Kawabuchi, Irimoto, and Miyawaki (2004) studied the viscosity of solutions consisting of high-methoxyl pectins and one or the other; trehalose, maltose or sucrose observing that the systems containing trehalose showed the highest viscosity at 25 °C (550 mPa.s for a 27 g/100 g trehalose solution as opposed to 380 mPa.s for an equally concentrated sucrose solution). These authors proposed that the increase in viscosity observed in the mixed systems could be due to the effect of sugars on the molecular interaction among pectin molecules.

Richardson, Wilmer, and Foster (1998) also studied the viscosity of mixed solutions of sucrose and guar or locust bean gums, describing the different possible interactions between components depending on the concentration of sugar and polysaccharide in the solution (5–40 g/100 g), especially at high concentrations where they compete for water.

Other authors have suggested specific interactions via hydrogen bonds between the polymer and the sugar-OH groups. Bayarri, Durán, and Costell (2004) reported an increase in the storage (or elastic) modulus of κ-carrageenan gels in the presence of sucrose, suggesting that sugar increases and stabilizes the number of junction zones between the polymer chains. In the present study the results show a differential effect of trehalose and sucrose on the viscosity of mixed sugar/polysaccharide solutions.

As discussed for solutions of sugars alone, the different spatial structure and interaction forces between disaccharide-water could affect its interactions with the polysaccharide in the mixed systems, resulting in a greater synergism in the viscosity in the case of trehalose.

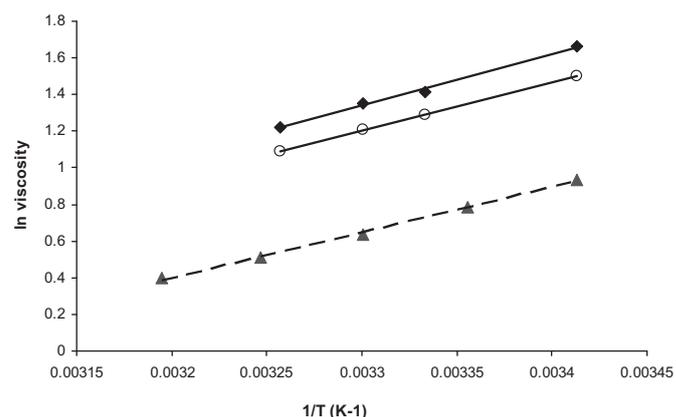


Fig. 5. Arrhenius plot for the effect of temperature on the viscosity. ◆ 35% (w/w) trehalose solution (*r*² = 0.996); ○ 35% (w/w) sucrose solution (*r*² = 1); both sets of data obtained in present work; ▲ 24.6% (w/w) trehalose solutions (*r*² = 0.998), data from literature (Sampedro et al., 2002) for the purpose of comparison.

A synergistic effect on the apparent viscosity was also observed in mixtures of polysaccharides with proteins, e.g. between guar gum and soy protein at concentrations of 0.15–0.3 g/100 g and 2 g/100 g respectively (Sanchez, Bartholomai, & Pilosof, 1995). In this study, the authors propose the formation of weak electrostatic interactions and hydrogen bonds, forming a network that allows for greater water retention and results in a bigger increase in viscosity than that developed by the individual components in the solution.

3.3. Temperature effect

To analyze the effect of temperature on viscosity, the activation energy (*E*_a) relating viscosity dependence on temperature was calculated for every disaccharide solution and their mixtures with 0.1 g/100 g guar gum. Fig. 5, shows the Arrhenius plot for sucrose and trehalose solutions at 35 g/100 g obtained from measurements at four different temperatures. A good fit to Arrhenius behaviour was obtained. Table 3 shows calculated values of *E*_a for both sugars and literature data showing similar results even with the use of a wider range of temperatures and different measuring methods. No significant difference was found among the values of *E*_a obtained for trehalose and sucrose. These results suggest that, even if both disaccharides may have different spacial structure and viscosities, the change with temperature occurs in a similar fashion.

Table 3
Activation energies (calculated from Arrhenius plots) for the different systems.

System	Concentration g/100 g	Temperature range, °C	Activation energy (kJ/mol)
Sucrose (this work)	35	20–34	22.0
Sucrose ^a	35	10–80	21.9
Trehalose (this work)	35	20–34	23.2
Trehalose ^b	30	0–30	23.3
Trehalose ^c	24.6	20–40	20.5
Trehalose + guar gum (this work)	45 T + 0.1 GG	20–35	30.6
Sucrose + guar gum (this work)	45 S + 0.1 GG	20–35	29.5
Guar gum (this work)	0.1	20–32	17.5

T = trehalose; S = sucrose; GG = guar gum.

^a From experimental data reported by Bubnik et al. (1995).

^b From experimental data (rotational viscometer) reported by Rampp et al. (2000).

^c From experimental data (falling ball viscometer) reported by Sampedro et al. (2002).

For mixed systems, the activation energy was also determined obtaining similar results for both mixtures; 29.5 kJ/mol for (45 g sucrose+ 0.1 g GG)/100 g and 30.6 kJ/mol for (45 g trehalose+ 0.1 g GG)/100 g (see Table 3). This increase in E_a could be due to an increase in the local concentration of sugar in presence of the polysaccharide; this effect has been observed with other disaccharides when working at concentrations near saturation (Rojas-Cazares, Delgado-Reyes, & Martínez Padilla, 2005; Rampp et al., 2000). The activation energy obtained for guar gum at 0.1 g/100 g was of only of 17 kJ/mol which would indicate a smaller dependence of viscosity on temperature for this polysaccharide.

4. Conclusions

Viscosity of sucrose and trehalose solutions (in the range 20–45 g/100 g) increased in an exponential way as sugar concentration increased. Trehalose solutions show a higher viscosity than those of sucrose at all temperatures examined. Differences between trehalose and sucrose viscosity increased with concentration and were less noticeable when raising temperature.

Addition of 0.1 g/100 g of guar gum revealed a synergistic relationship between both sugars and guar gum, being this effect more important for trehalose. For both disaccharides solutions and mixed systems viscosity dependence on temperature showed a good fit to Arrhenius behaviour. The E_a values were very similar for both sugars while for both mixed systems higher E_a values were obtained.

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