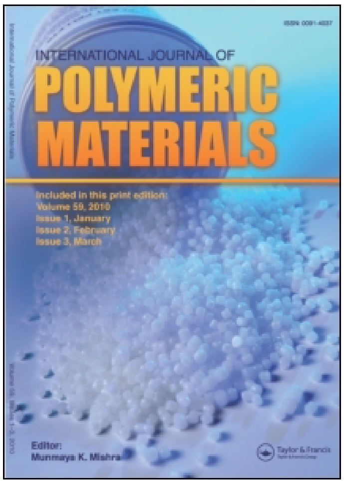

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Dielectric Properties of Closed Cell Microcellular Ethylene Propylene Diene Rubbers at Microwave Frequency

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Microwave dielectric properties of closed cell microcellular EPDM rubber in short-circuited wave-guide method have been studied at 9.69 GHz. Plots of dielectric constant (ε') with volume fraction of rubber shows a nonlinear relationship for both unfilled and silica filled microcellular rubbers obeying Weiner's inequalities. Several empirical relations have been tried to obtain a good fit. But for the solid vulcanizates other experimental values or points found to give a good fit. However the experimental fit found to deviate from the original empirical relation. A better fit was obtained for a proposed modified empirical relation suggested.

Keywords: Dielectric properties; Ethylene Propylene Dienes; Microwave frequencies; microcells

INTRODUCTION

The flexible cellular and microcellular materials have been widely used as radio-transparent construction materials in radar technique, as light electric insulator and packaging material in radio and electronic engineering, aviation, space applications [1, 2] and cable applications. [3, 4] EPDM has good aging and weathering properties as well as excellent thermal, electrical and dielectric properties. Dielectric properties of polymeric foams was studied by Stutov and coworkers. [5]

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The dielectric properties of polystyrene foams and its blends at X-band microwave region (9.375 GHz) were studied by Chaki *et al.* [6–9]. The electric and dielectric properties of polystyrene, polyethylene and polypropylene at microwave frequency have been reported in the literature [10–13]. Thus closed cell microcellular EPDM has immense scope for application as microwave dielectric materials. Very limited studies of microwave dielectric properties on closed cell cellular and microcellular flexible materials are found in the literature.

In the present study, the dielectric properties of closed cell microcellular EPDM have been reported at microwave frequency at room temperature. The effect of density and volume fraction of rubber on dielectric properties have also been reported.

EXPERIMENTAL

Materials

The EPDM rubber [Kelton 520, ethylene content 55 mole%, diene content 4.5 mole% (DCPD), specific gravity 0.86, manufactured by DSM chemicals, Holland] was used. The precipitated silica used as filler was manufactured by Degussa AG, Germany. Its characteristics were given as follows: Specific gravity, 1.9; BET surfaces area, 160–120 m^2/g; particle size, 10–20 nm. Dicumyl Peroxide (DCP) used was Percidol 540C (40% DCP on inert filler), manufactured by Chemoplast(I) Ltd. DNPT used as blowing agent was manufactured by High Polymer Labs, India.

Compounding and Sample Preparation

The rubber was compounded with other ingredients according to the formulations of the mixes (Table-I) and the blowing agent was added at the end. For obtaining cure characteristics of the compounds Monsanto Rheometer (R-100) was used. The compounds were molded at 160°C to 80% of their respective cure times. All sides of the mold were tapered to 30° to facilitate the expansion of the molded compounds to closed cell, nonintercommunicating, microcellular product with better mold release. Expanded microcellular sheets were post cured at 100°C for one hour in an electrically heated air oven.

TABLE I Formulations* of gum and silica filled vulcanizates

	Mix no								
	G_{10}	G_{11}	G_{12}	G_{14}	GS_{30}	GS_{31}	GS_{32}	GS_{34}	GS_{36}
EPDM	100	100	100	100	100	100	100	100	100
Silica	0	0	0	0	45	45	45	45	45
Paraffin Oil	2	2	2	2	12	12	12	12	12
Ethylene Glycol	0	0	0	0	3	3	3	3	3
DNPT	0	1	2	4	0	1	2	4	6

*Each mix contains ZnO–2phr, Stearic Acid–2phr, Dicumyl Peroxide (40%)–2phr.

Scanning Electron Microscope

SEM photomicrographs are obtained from Cam Scan Series 2 model Scanning Electron Microscope of razor cut surfaces of microcellular sheets. The surface of samples were gold coated for the testing.

Microwave Dielectric Measurements

Measurement of dielectric constant and loss of samples was done in a single frequency of 9.69 GHz in the microwave region by a short-circuited wave-guide method of Roberts and Von Hippel [14, 15] as modified by Dakin and Works [16] for application of low and medium loss samples, using hallow rectangular wave-guide at X-band as sample holder. In the shorted-line technique, a slotted section is used to measure the shift in minimum of a standing-wave and the change in standing wave ratio (which is the ratio of the maximum voltage to the minimum voltage or maximum current to minimum current). The minimum of the standing-wave pattern occur at intervals of one half wavelength from the short-circuit when the sample is absent. When the sample is inserted in front of the short-circuit, the minima shift towards the short-circuit as shown in Figure 1. The shift in minimum is a measure of the dielectric constant. The decrease in standing-wave ratio is a measure of the loss tangent. For calculation of dielectric constant and loss of samples the following formula was used:

$$\varepsilon' = (x\lambda/2\pi d)^2 + (\lambda/\lambda_c)^2$$

where λ is the wavelength of the frequency applied, d is the thickness of the sample, and λ/λ_c is the wave guide proportionality constant

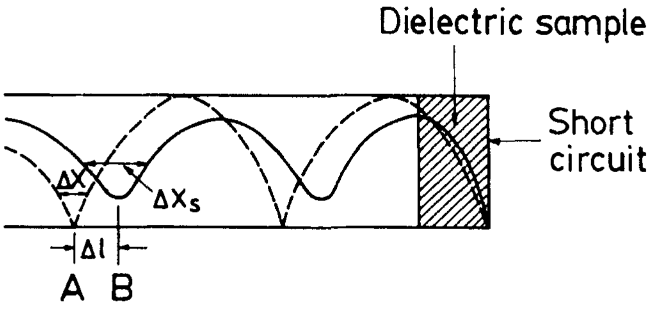

FIGURE 1 Standing wave in the wave guide with and without sample. (- -) without sample; (—) with sample.

which is a function of the wave guide dimensions and the mode of propagation. The value of x can be obtained using the relation,

$$\frac{\tan X}{X} = \frac{\lambda_g}{2\pi d} \cdot \tan \frac{2\pi(\Delta l + d)}{\lambda_g}$$

where λ_g is the guided wavelength in the absence of a sample and Δl is the shift of minima of the standing wave due to insertion of the sample.

RESULTS AND DISCUSSION

Cell size and cell size variation are studied from SEM photomicrographs (Fig. 2). These photomicrographs show that the cell size are spherical in nature and decrease with increase in blowing agent concentration. The number of cell increases with the decrease of density.

Closed cell microcellular rubbers are inhomogeneous composites of air and rubber. The extrapolated dielectric constant, ε', for microcellular rubber at zero density (zero volume fraction of rubber) which is predominantly decomposed gas (air), is to be unity. ε_0 is the dielectric constant corresponding to solid vulcanizates. The dielectric constants, ε', of the closed cell microcellular rubber samples as shown in Figures 3 and 4 exhibit a linear relationship with the density as well as volume fraction of rubber upto a limited range. As expected the extrapolated ε' for microcellular rubber at zero density is unity. Cellular

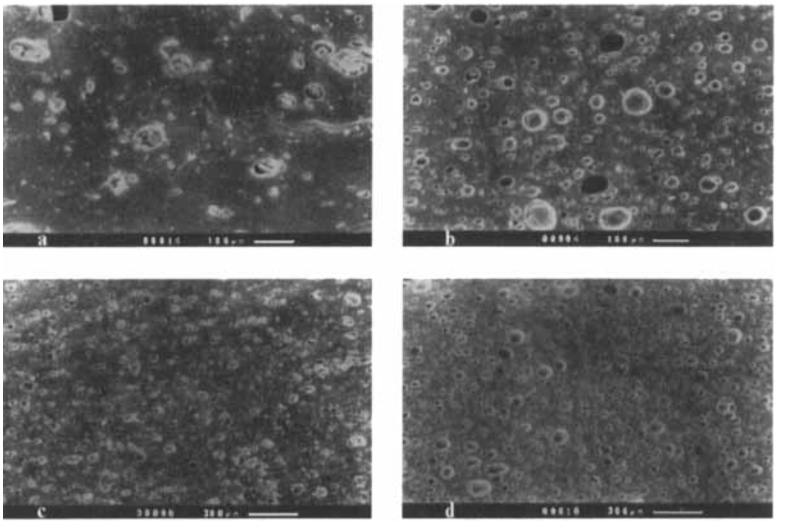

FIGURE 2 SEM photomicrographs of microcellular rubber vulcanizates a) G_{12} b) GS_{32} c) G_{14} d) GS_{34}.

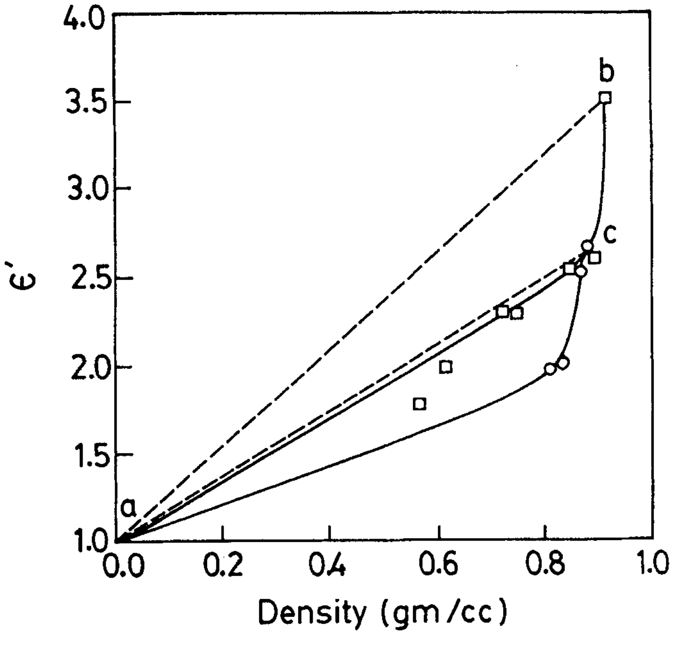

FIGURE 3 Plots of dielectric constant (ε′) with density. Unfilled (o); 45 phr silica filled (□).

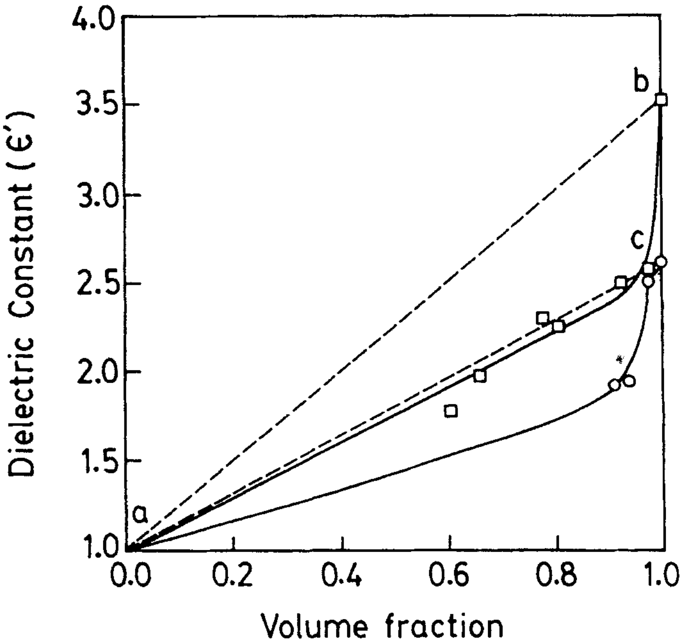

FIGURE 4 Plots of dielectric constant (ε') with volume fraction of rubber. Unfilled (o); 45 phr silica filled (□).

rubbers are chaotic or "statistic" mixture of several components. The true values of dielectric constant for these cellular or microcellular rubber obey the Wiener inequalities [17]:

$$\frac{1}{\sum_{i=1}^{m} \frac{x_i}{\varepsilon'_i}} \leqslant \varepsilon^* \leqslant \sum_{i=1}^{m} x_i \varepsilon'_i$$

where x_i is the volume fraction of component i in a statistical mixture of m number of components, ε'_i and ε^* are the permittivity of components and mixture respectively. The dielectric constant of the microcellular rubber under study lies between the limits depicted in the relation.

Plots of ε' vs density and ε' volume fraction of EPDM in the microcellular rubber have been depicted in Figures 3 and 4 respectively. The revealed that density and dielectric constant are linearly dependent on each other upto 0.82 gm/cc of density beyond which the relation is nonlinear. The slope of the experimental line is 2.753 where as the slope of the theoretical line obtained by joining *a* and *b* corresponding to 0 and 100% volume fraction of rubber respectively is 3.712. Similarly the volume fraction and dielectric constant are also linearly dependent upto volume fraction 0.90 beyond which nonlinear relation is evident. The slope of the line obtained from experimental data is 2.566 where as that of the theoretical line is 3.503. It is also seen that the relation between density as well as volume fraction of rubber with dielectric constant for gum vulcanizates is nonlinear.

Nonlinear dependence in the wider range of densities for volume fraction 0 to 100% illustrates Wiener's inequalities. Lichtenecker and Rother [17] developed an empirical relationship for the calculation of dielectric constant of statistic mixtures in the form of logarithmic law. A version of Lichtenecker and Rother's formula for the calculation of dielectric constant of foam and porous plastics is given by relation,

$$\varepsilon'_{\text{foam}} = \varepsilon_0^x$$

$$\text{or, } \log \varepsilon' = x \log \varepsilon_0$$

where, ε_0 is the dielectric constant of the unfoamed and ε' is the dielectric constant of the foam. In order to apply the same to microcellular rubber, we take ε_0 and ε' as the dielectric constant of solid and microcellular EPDM respectively. Figures 5 and 6 shows the plot of $\log \varepsilon'$ vs density and volume fraction respectively. The dotted line joining *a* and *b* corresponding to the density of air and solid vulcanizates is the theoretical line with a slope of 0.589 for 45 phr silica filled EPDM rubber. The experimental results shows linear relation excluding point *b* (dielectric constant to solid vulcanizates) with a slope of 0.47 (Fig. 5). Figure 6 shows that volume fraction of microcellular rubber bears a linear relationship with dielectric constant with a slope of 0.424 for 45 silica filled vulcanizates as against the theoretical slope of 0.544. Therefore, the experimental results show deviation from the above logarithmic relation. Low density microcellular gum vulcanizates could not be prepared because above 4 phr blowing agent loading blisters appear. These gum microcellular vulcanizates also do not obey the above logarithmic relation.

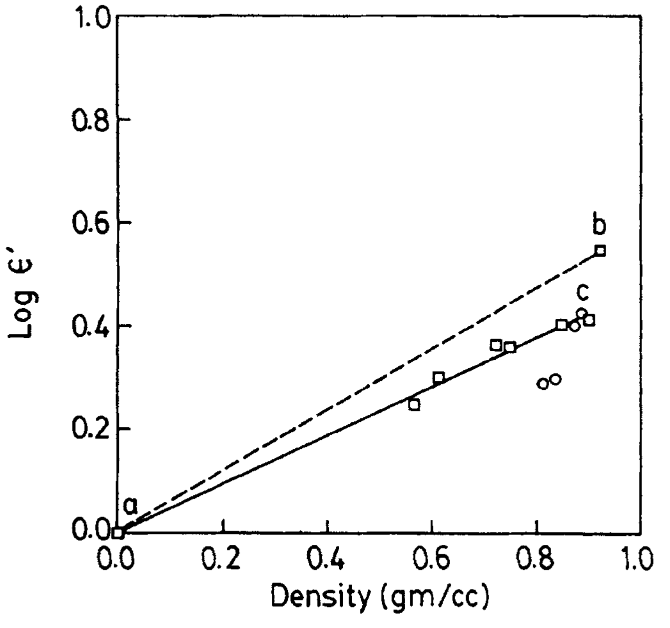

FIGURE 5 Plots of log ε′ with density of rubber. Unfilled (o); 45 phr silica filled (□).

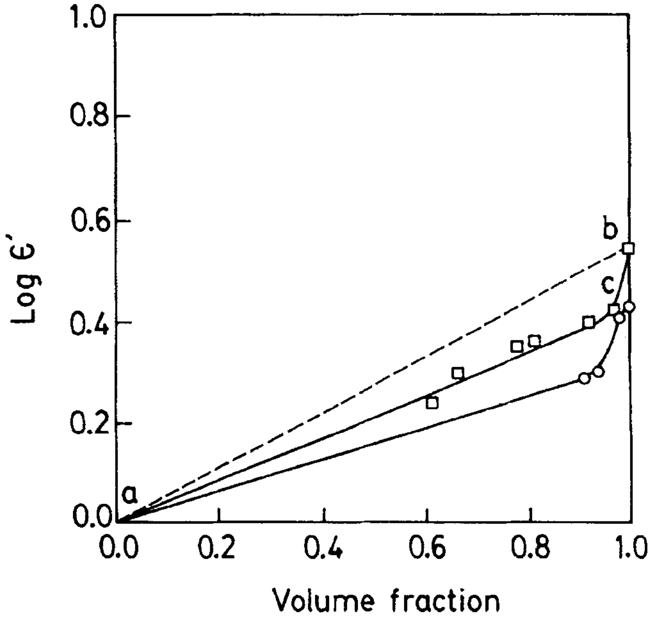

FIGURE 6 Plots of log ε′ with volume fraction of rubber. Unfilled (o); 45 phr silica filled (□).

Numerous other formulae pertaining to low density material namely simplified Landau and Lifshitz's [17] relation,

$$(\varepsilon')^{1/3} = 1 + x(\varepsilon_0{}^{1/3} - 1)$$

and simplified Beer's relation,

$$(\varepsilon')^{1/2} = 1 + x(\varepsilon_0{}^{1/2} - 1)$$

have been tried. However they do not open up any new vistas except showing some empirical agreement.

Figures 7 and 8 shows the plots of $(\varepsilon')^{1/3}$ and $(\varepsilon')^{1/2}$ vs volume fraction of rubber respectively for both gum and 45 phr silica filled microcellular vulcanizates. The plots of $(\varepsilon')^{1/3}$ and $(\varepsilon')^{1/2}$ vs volume fraction of rubber respectively pass through unity with respective slope of 0.391 and 0.629 as against 0.518 and 0.872 respectively. Thus both Landau and Lifshitz's and Beer's relation shows deviation from the theoretical line.

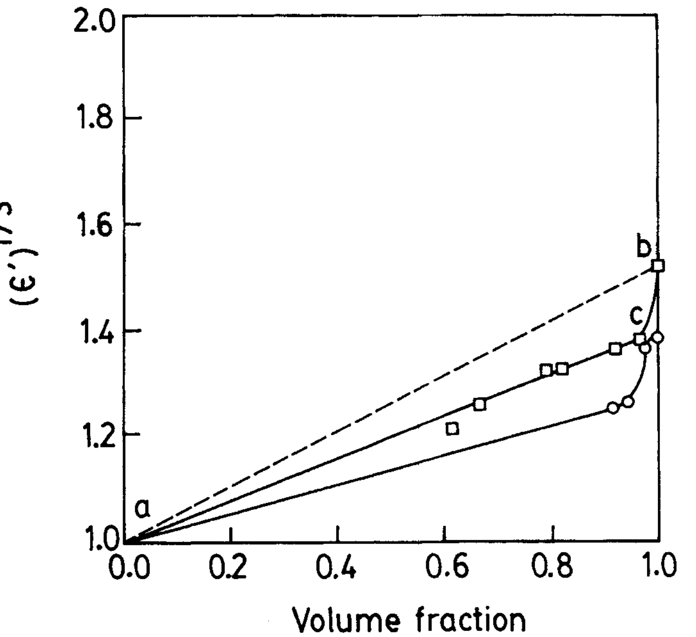

FIGURE 7 Plots of $(\varepsilon')^{1/3}$ with volume fraction of rubber. Unfilled (o); 45 phr silica filled (□).

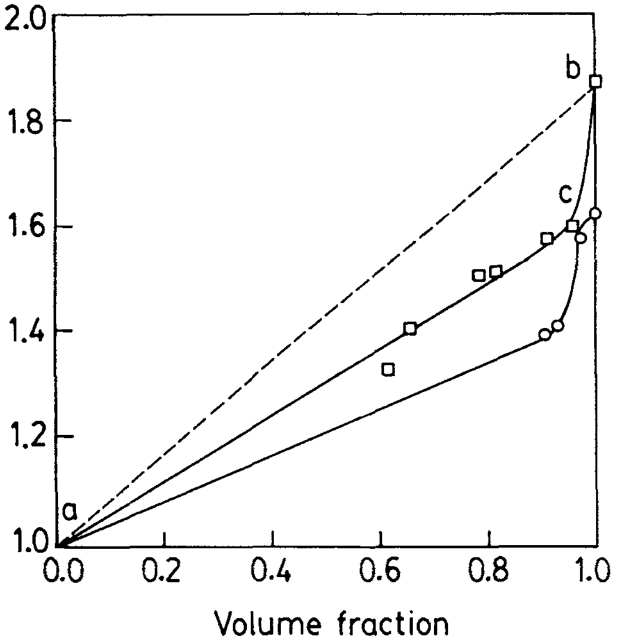

FIGURE 8 Plots of $(\varepsilon')^{1/2}$ volume fraction of rubber. Unfilled (o); 45 phr silica filled (□).

The above formulae used for calculation of dielectric constant of static mixture (composites) are derived on the basis of the various theoretical presumption and experimental data. The system consists of two different homogeneous dielectric dielectrics connected in parallel or series. For the model to follow parallel connection additivity rule should follow. Whereas if it follows series connection it should shows negative deviation. Dielectric relationship of the microcellular rubber lie in between parallel and series model. Therefore a model was proposed in line with the suggested model for series, [17]

$$1/\varepsilon' = 1 + x(1/\varepsilon_0 - 1)$$

Figure 9 shows the relationship between $1/\varepsilon'$ and volume fraction of rubber. It is observed from the figure that 45 phr silica filled microcellular rubber shows a linear relationship with volume fraction of rubber.

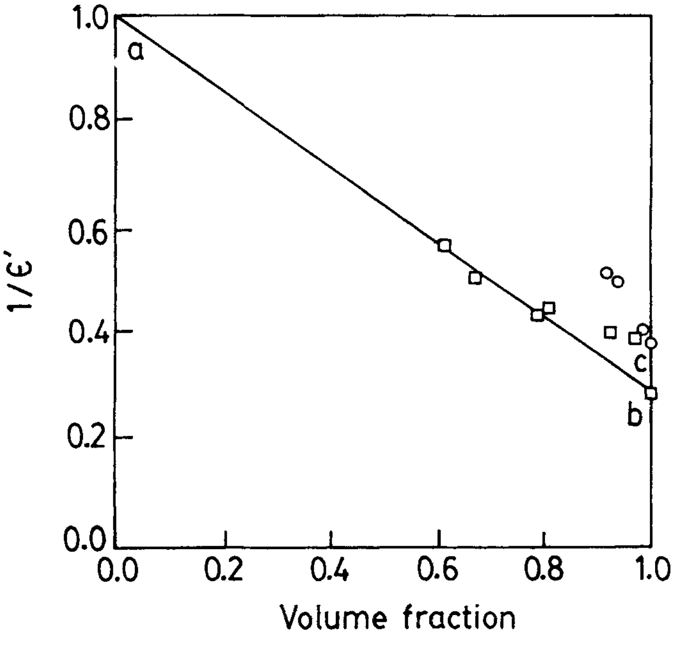

FIGURE 9 Plots of $1/\varepsilon'$ with volume fraction of rubber. Unfilled (o); 45 phr silica filled (□).

However this does not hold good for unfilled microcellular rubber vulcanizates.

CONCLUSIONS

It can be concluded that low loss dielectric microcellular rubber based on EPDM, suitable for microcellular application are possible to be made as described here. From the detailed study of the dielectric behaviour of the silica filled microcellular EPDM rubber with various composition it is seen that the dielectric relation of composite with volume fraction of rubber lie between the parallel and series model. The dielectric behaviour neither follow the logarithmic law of Lichtenecker and Rother nor the model proposed in the line with the series model.

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