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Notes and experiments on the statics of capillary columns

G S Barozzi¹, M A Corticelli¹, D Angeli² and E Salerno¹

¹ DIEF - Dipartimento di Ingegneria “Enzo Ferrari”, Università di Modena e Reggio Emilia, Via P. Vivarelli 10 - 41125 Modena (Italy)
² DISMI - Dipartimento di Scienze e Metodi dell’Ingegneria, Università di Modena e Reggio Emilia, Via Amendola 2 - 42122 Reggio Emilia (Italy)

Email: giovanni.barozzi@unimore.it

Abstract. Experimental results are summarized, concerned with the statics of liquid columns in capillary tubes under non-standard conditions. Three configurations are considered: 1. inclined capillary tubes; 2. capillary effect in the horizontal branch of an L-shaped tube; 3. capillary columns in a vertical tube. The effect of inclination on capillary rise (1.) had already been explored in [1] using water in glass-tubes, and it was found that the vertical rise progressively reduces for increasing the tube inclination. This behaviour is now confirmed for n-Hexadecane (C₁₆H₃₄). For L-shaped capillaries (2.), the length of the horizontal branch of the tube occupied by the liquid is detected, as a function of the elevation of the branch itself over the feeding pool. The statics of suspended liquid columns (3.) is investigated for two configurations, namely: i. freely suspended columns, and, ii. edge-ending columns. In the latter case the evaporation transient is also tracked. Even if the experimental basis is limited, the results are sufficient to highlight some of the peculiar features of the statics of capillary columns under the above conditions. In particular, it is shown that the contact angle hysteresis plays a fundamental role in all the cases considered.

1. Introduction

Capillary effects play a crucial role in a number of technological applications, such as oil recovery, drug delivery, preparation of paints, pesticide production, food processing, micro-fluidics, wicking fabrics, dyeing of textile fabrics, and ink printing, just to mention a few. On the other hand, the rise of sap in plants, the locomotion of insects on the surface of water, or the wetting of the cornea are just examples of the ubiquitous importance of capillary forces in our daylife. This motivates the extremely huge research effort devoted to the analysis of the capillary forces and the interaction processes between liquids and solid surfaces throughout the world.

Capillary rise phenomena, in particular, have been widely investigated along more than three centuries. However, they encounter a widespread scientific interest, mainly dedicated in the recent years to the dynamic aspects of capillary rise, and, more specifically, to the time-rate evolution of the liquid column [2–15].

The theoretical bases of capillary statics in tubes are quite well grounded, and, for this reason, this topic is receiving much less attention in the present time. The recent extension of the capillary rise theory to arbitrarily shaped capillaries by Wang et al. [16] constitutes an exception in this context.

The rise (or fall) in a thin vertical tube immersed in a liquid pool at one end is in fact extremely well predicted by the Jurin’s law, formally established in 1728 [17]. For circular tubes it reads:
Here, $\gamma$ and $\rho$ are the surface tension of the liquid, and the liquid-to-air density difference, respectively; $H$ is the elevation of the meniscus apex over the free surface of the bath; $R$ is the capillary inner radius; $g$ is the gravitational acceleration, and $\theta_e$ is the equilibrium contact angle specific of the liquid-solid surface coupling. Equation (1) holds for both hydrophilic ($\theta_e < 90^\circ$) and hydrophobic ($\theta_e > 90^\circ$) couplings (in the latter case a negative rise of the liquid in the capillary occurs), subject to the condition $R \ll H$ (thin tubes). Equation (1) has been validated by hundreds of experiments, and can be theoretically derived following different and independent approaches. It is worth noting that the equation predicts an infinite $H$-value under zero gravity conditions. This result is consistent with the idea, underlying any of the derivation procedures of equation (1), that the hydrostatic equilibrium of the liquid column within a capillary tube derives by the balance of two actions, one of which is linked to the presence of the gravitational field, the other being provided by the capillary force.

This is, in fact, a statement of general validity to explain the static behaviour of a capillary column whatever it is the system geometry and orientation. In practical terms, however, a number of very basic questions remain unanswered as soon as we move from the basic case to which Jurin’s Law refers. For example, no criterion is available to predict the capillary rise in an inclined tube, or the horizontal spread of the liquid in an L-shaped capillary with its short vertical leg immersed in a liquid pool. In the former case, equation (1) predicts no difference from the vertical orientation, thus omitting the effect of the gravitational field on the meniscus shape. In the second case, under the same assumption, the liquid will move indefinitely up to the open extremity of the horizontal branch of the tube. That this is not the case in practice is self-evident, but no criterion is available to predict the static asset of the system. In view of the frequent occurrence of similar situations in real world, it is curious that no reference to them can be retrieved from the open literature.

In this work we summarize the results of a series of simple experiments, relevant to three non-standard static conditions for liquid columns in cylindrical capillary tubes. These are:

1. Inclined capillary tubes
2. L-shaped capillaries
3. Suspended liquid columns in vertical capillary tubes

It is anticipated that for none of cases considered the experimental data to be presented can be considered as definitive, both in view of the elementary experimental tools available, and the limited number of cases considered. In particular, all the experiments were carried out with borosilicate-glass tubes, with inner diameters ranging from 1 to 2 mm, and distilled water was used in most of the cases.

2. The effect of inclination on capillary rise

A first mention to the effect of inclination on capillary rise can be found in [18], where, however, the final equilibrium state of the system was assumed to be coherent with equation (1).

Experimental data on this case were provided first by Barozzi and Angeli [1]. The vertical elevation of the meniscus on the feeding pool free-surface, $H_o$, was measured for water in a series of tubes for values of the declination angle, $\alpha$, ranging from $0^\circ$ (vertical orientation) to $88^\circ$. Tubes of inner diameter $\Phi = 1.92$ mm were used. The equilibrium contact angle, as estimated by equation (1) for $\alpha = 0^\circ$ was $\theta_e = 47^\circ \pm 2^\circ$.

The experiment demonstrated that for this specific system $H_o$ does not deviate from Jurin’s law prediction, equation (1), up to $\alpha$-values around $50^\circ$. Thereafter, however, the capillary rise was found to decrease monotonically for increasing $\alpha$, the deviation from the reference level ($H_o = 10.3$ mm) becoming quite well evident and definitely out of the uncertainty range of the data. The axial extension of the capillary column
\[ L_\alpha = \frac{H_\alpha}{\cos \alpha} \]  

(2)

Reduced accordingly, and could be fitted by the equation

\[ L_\alpha = \frac{H_0}{(\cos \alpha + k \sin \alpha)} \]  

(3)

where \( k = 0.021 \). Results from equation (3) should be compared with predictions from the standard formula \( L_\alpha = H_0 / \cos \alpha \).

A successive series of experiments has been carried out with two tubes of the same series as above, using n-Hexadecane (\( \text{C}_{16}\text{H}_{34} \)). Relevant data for the operating fluid were directly measured by a Wilhelmy Plate Tensiometer (at 25 °C we had: \( \rho = 766.25 \text{ kg/m}^3 \), and \( \gamma = 22.657 \text{ mN/m} \)). The measuring technique was similar to the one reported in [1]. The capillary rise for the vertical orientation resulted to be \( H_0 = 6.20 \text{ mm} \) (\( \theta_e = 10^\circ \)), and \( H_0 = 6.26 \text{ mm} \) (\( \theta_e = 8^\circ \)) for capillary C1 and C2, respectively, with an estimated uncertainty of ± 0.06 mm. Results for the vertical capillary rise \( H_\alpha \) as a function of the declination angle \( \alpha \), are reported in figure 1. The mean values of the capillary rise ratio \( H_\alpha / H_0 \) for the four runs are plotted in figure 2, where results for water [1] and hexadecane are compared.

\textbf{Figure 1.} Capillary rise \( H_\alpha \) vs. \( \alpha \) for n-Hexadecane (\( \text{C}_{16}\text{H}_{34} \)).

\textbf{Figure 2.} Capillary rise ratio \( H_\alpha / H_0 \) vs. \( \alpha \) for n-Hexadecane (\( \text{C}_{16}\text{H}_{34} \)) and water [1].
It can be noted that the effect of inclination is quite well evident in both the cases, even if it becomes detectable only for relatively high values of the inclination ($\alpha > 75^\circ$), for which the capillary rise reduces of more than 10% towards the vertical reference.

This effect seems to be directly linked to the deformation of the meniscus produced by the component of the gravitational force normal to tube axis. This is made evident by the reproduction of the meniscus acquired with a Dataphysics contact angle goniometer (OCA) and given in figure 3, where the contact angle at the lower edge of the meniscus appears to be much lower than the one at the upper side. The loss of axial symmetry of the meniscus was already evident for $\alpha = 60^\circ$, where, however, no measurable effect on $H_\alpha$ could be observed.

![Image of meniscus](image)

**Figure 3.** OCA image of the meniscus for n-Hexadecane, $\alpha = 85^\circ$.

3. **Statics of L-shaped capillary tubes**

When immerging vertically the short-leg end of a L-shaped capillary tube in a liquid pool, the liquid rises along it, up to the $H$-value corresponding to Jurin’s Law prediction, $H_0$, provided that $H_0 < H_B$, being $H_B$ the elevation of the axis of the horizontal branch of the tube over the pool free surface.

When, on the contrary, the condition $H_B < H_0$ is fulfilled, the liquid will overcome the L-elbow and invade the horizontal tube-branch. It is then possible that the horizontal capillary elongation, $L$, be so high that the liquid reaches the open extremity of the tube. This would in fact be always the case if an ideal surface (rigid, perfectly smooth, non-adsorbing, and inert [19]) is considered, and the effects of gravity on the meniscus shape can be overlooked. The capillary force, in fact, could not be counteracted by any action.

It is intuitive that this does not happen in real world, provided that the horizontal branch of the tube is long enough.

An experiment was set up to have evidence of the above behaviour, using L-bended borosilicate-glass tubes directly provided by Hilgengeng GmbH (Germany). Data provided by the producer were: $L_1 = 30$ mm (short leg), $L_2 = 300$ mm (long leg), $\Phi = 1.05 \pm 0.05$ mm (inner diameter), and $S = 0.22$ mm (wall thickness). The level of tolerance on the inner diameter was confirmed by SEM-measurements on samples taken by different capillaries in the series. Distilled water was used in the experiments. The vertical rise was preliminarily determined as $H_0 = 28.7 \pm 0.2$ mm, to which corresponds a mean value of the equilibrium contact angle $\theta_e \approx 15^\circ$.

Experiments were carried out on an anti-vibration table. The experimental set-up is shown in figure 4. The free-surface level of water in the feeding pool was varied progressively, thus varying $H_B$. The metal scale shown was used to convert lengths into pixels of a digital photo-camera, and the horizontal lengths were thus retrieved by inverse conversion from digital images. Water was found to overcome the L-elbow for $H_B$-values just below the reference value $H_0$, and a short part the horizontal branch appeared to be invaded by the liquid already for $H_B = 28$ mm.
The results of the experiment reported in figure 5 are exemplificative of the general trend of the results. The capillary elongation is found to reduce progressively for increasing $H_B$. However, this holds true only for $H_B > 22$ mm, and $H_B < 14$ mm, while in the intermediate range ($14 < H_B < 22$ mm) a plateau region appears, where $L$ remains constant for decreasing $H_B$. In general terms, such a trend corroborates the idea that, when reducing the elbow height over the free surface of the bath, an increasing fraction of the capillary force potential is made available to push the liquid in the horizontal part of tube.

Analyzing the statics of the system, if, as a first choice, we exclude pinning effects at the triple line, the vertical equilibrium of the liquid column can only be ensured by the creation of a vertical resultant of the capillary force at the triple line, $F_{cv}$, equivalent to the weight of the vertical liquid column, according to the equation:

$$F_{cv} = \rho g H_B \pi R^2$$ \hspace{1cm} (4)

Such a vertical force must necessarily correspond to a deformation of the meniscus, which loses its axial symmetry and produces a re-distribution of the contact angle values along the triple line. Higher values of the local contact angle, $\theta_\psi$, are expected to occur in the upper part of the tube, so as to provide a vertical upward resultant.

The above mentioned meniscus deformation is not easy to track with the available visual tools. The sample OCA-images reported in figure 6 are in fact insufficiently resolved to allow for the quantification.
of the contact angles at the upper and lower edges of the section. However, they are sufficient to demonstrate that a meniscus deformation is actually present in figure 6.a ($H_B = 26$ mm, $L = 32$ mm), while it disappears in the plateau zone ($H_B = 22$ mm, $L = 150$ mm), in figure 6.b. It is evident that in the latter case, the statics of the systems is not led by the contact angle hysteresis, but rather by some pinning effect, since, in fact, the meniscus appears to remain axial-symmetric. This observation is of interest in itself, since it demonstrates that pinning can produce reactive forces in any direction.

![Figure 6. OCA images of the meniscus in the horizontal part of the capillary tube (water) in the proximity of the bend (a) and in the plateau zone (b).](image)

4. Statics of a liquid column in a vertical capillary tube

A liquid column can remain suspended within a vertical capillary tube in virtue of the contact angle hysteresis. As schematized in figure 7.a, at the upper triple line the contact angle reduces, while it increases at the lower meniscus. The vertical equilibrium of the liquid column under gravity is ruled by the equation [20]

$$\frac{2\gamma}{R} (\cos \theta_1 - \cos \theta_2) = \rho g H_C$$

(5)

Here, $R$ is the capillary radius, $\theta_1$ and $\theta_2$ are the non-equilibrium contact angles at the upper and lower triple line, respectively, and $H_C$ is the height of the suspended column. In other words, an upward directed vertical force, $2\pi R \gamma \cos \theta_1$, balances the capillary downward force $2\pi R \gamma \cos \theta_2$ at the lower triple line, and the column weight, $\rho g \pi R^2 L$. The limit for stability is dictated by the conditions $\theta_1 \geq \theta_R$ and $\theta_2 \leq \theta_A$, where $\theta_R$ and $\theta_A$ are the recession and advancing contact angles, respectively.

The limiting condition to be necessarily satisfied is

$$\frac{2\gamma}{R} (\cos \theta_R - \cos \theta_A) \geq \rho g H_C$$

(6)

Equation (6) states that equilibrium is impossible for ideal surfaces, for which contact angle hysteresis is excluded. Rigid, high energy, non-adsorbing, and very smooth surfaces usually approximate the ideal behaviour (quasi-ideal surfaces), and glasses normally match these conditions. The hysteresis range should be very restricted for surfaces of this kind. This, however, ceases to be true for dirty or rough surfaces, in which cases the difference $(\theta_A - \theta_R)$ can become as high as $50^\circ$ [20]. The above considerations are convincing, but seem not to have been corroborated by experimental evidence in the open literature. In addition, equation (6) does not include any consideration of the effect of the discontinuity created by the ending edge of the tube, whose role is evident in everyday experience, but has never been quantified. The case is represented in figure 7.b.
Results of experiments for water in a borosilicate-glass capillary of inner diameter $\Phi = 1.04$ mm are reported here. Two sets of experiments were carried out: i. freely suspended columns (figure 7.a), and, ii. edge-ending columns (figure 7.b). In the first case, a bubble of water was initially sucked by immersion in a water pool, and then reduced in volume by extracting a portion of mass with a syringe. The capillary tube containing the suspended liquid column was positioned vertically on the OCA optical bench, and the upper and lower contact angles were observed and directly measured. In the second series of experiments the liquid column was left to adhere to the lower capillary-end border. In this case, the lower meniscus results to be convex, and emerges from the capillary end. This specific arrangement was found to be prone to evaporation. The evolution of the upper and lower contact angles was followed over time periods ranging from 75 to 150 minutes, and the effect of the end-discontinuity on the equilibrium of the liquid column was observed in the course of the evaporation transients. Similar to the previous cases, the static contact angle was preliminarily measured for the capillary tube, by either direct observation at the OCA optical bench, or, indirectly, by inversion of equation (1). The estimated value of the static contact angle was $\theta_e = 62^\circ \pm 2^\circ$.

**4.1. Freely suspended columns**

Overall, seven series of data were collected for $H_c$ ranging between 2 and 20 mm. The lowest $H_c$-values resulted to be very uncertain, since the relative error on $H_c$ obviously increases for reducing the column height. Stable columns were found to be increasingly difficult to obtain for $H_c$ over 14 mm. This was thus assumed to be the stability limit of the suspended column. Only results for $H_c$ included within the range 4-14 mm were therefore considered to be acceptable, and these are reported in figure 8, in terms of the mean values of $\theta_1$ and $\theta_2$ vs. the measured $H_c$-value. It is worth to point out that the right and left values of the contact angles of the same meniscus, as estimated from the OCA images, could widely differ, probably due to the difficulties encountered in the manipulation and positioning of the tube. It was therefore decided to accept only the tests where the difference between the left and right $\theta$-readings did not exceed $5^\circ$.

Figure 8 indicates that the contact angle at the lower meniscus increases slightly for increasing $H_c$ from 4 to 14 mm, $\theta_2$ passing from around $63^\circ$ to around $68^\circ$. Over the same range, $\theta_1$ decreases more rapidly, from $55^\circ$ to $45^\circ$. The hysteresis range of $\theta$ is therefore not less than $23^\circ$. It is worth of note that the data-fittings of $\theta_1$ and $\theta_2$ converge to $60^\circ$, quite close to the measured equilibrium value, $\theta_e$. 

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**Figure 7.** Statics of suspended capillary columns: freely suspended column (a), and edge-ending column (initial condition) (b).
Figure 8. Statics of suspended capillary columns: upper ($\theta_1$) and lower ($\theta_2$) contact angles vs. $H_C$. The dashed lines indicate the linear fitting of the two datasets.

For the acceptable assays, the vertical force balance between the total capillary force, $F_c$, and the column weight, $F_w$, was finally checked using equation (7)

$$\Delta F = F_c - F_w$$

where

$$F_c = 2\pi R \gamma (\cos \theta_1 - \cos \theta_2)$$

$$F_w = \rho g H_C \pi R^2 + W_{1,2}$$

Here, $\Delta F$ is the closure error, and $W_{1,2}$ is a correction term accounting for the mass of liquid enclosed between the horizontal plane at the meniscus apex, and the meniscus surface. The results demonstrate that $\Delta F$ is relatively large, with a maximum deviation of order 40%, and is systematically negative. This suggests that significant pinning effects are present at the triple lines.

4.2. Edge-ending columns
A series of experiments was finally scheduled to observe the effect of the lower-end border of the capillary tube on the statics of the liquid column. As above, distilled water was used, leaving the suspended liquid column to emerge from the lower capillary end, as shown in figure 7.b.

As anticipated, during the experiments it was observed that a relatively fast evaporation process occurred, and it was decided to monitor it. Results are presented for a set of four experiments, carried out for values of the environmental temperature and the relative humidity within the range $22.4-22.6 ^\circ C$, and 55-60%, respectively. The column height, and the upper and lower contact angles were measured every ten minutes, starting from different initial conditions (i.e. different values of $H_{C0}$).

The evolution of the water-column height is reported in figure 9 for all the runs. It can be noted that the trends are quasi-linear; the evaporation rate (50-60 $\mu g$/min) is therefore almost insensitive to the $H_C$-value, as well as the menisci’s shape.

During the transient, the upper contact line moves downwards, while the lower contact line remains hooked to the tube edge. The time-evolution of the lower and upper contact angles is shown in figure 10.
The data plots in figure 10 are very interesting; in fact, they highlight the different evolution of the two menisci during the evaporation transient: starting from an initial value between 61° and 68°, $\theta_1$ slowly decreases, levelling down to a common value of 40°; $\theta_2$, on the contrary, starts from values of order 100-120°. It decays very rapidly, reducing to less than 90° in the first ten minutes, then assuming a quasi-linear trend, eventually reaching a common threshold value of 20°.

This is well illustrated by the three last snapshots in figure 11, where a typical sequence of the lower meniscus evolution is reported.
Figure 11. Edge-ending columns: time-evolution of the lower meniscus.

The last two images (for $t = 150$ min, and $t = 160$ min, respectively) also show that in the final stages of the observation, the meniscus rises along the capillary, $\theta_2$ remaining unchanged at its end-value, 20°. Such an unexpected behaviour was observed systematically in all the experiments.

For the scope of checking the vertical force balance, equation (7) was enforced as in the previous case. Figure 12 reports $\Delta F/F_w$ vs. time. The data show that the unbalance between the capillary and gravitational components is quite high and positive at the beginning of the transient, the capillary force thus prevailing largely in this phase. Later on, however, due to the different evolution of the two contact angles (as from figure 10), not only the capillary force decays more rapidly than the column weight, but, at one point, it reverses in sign, giving rise to a very strong downward resultant.

All of this demonstrates that a third force must necessarily be present, aside the capillary and gravitational terms, to preserve the statics of the liquid column.

Such a balancing force is likely to be provided by the adhesion of the lower contact line to the capillary tube edge, and, by evidence, has a reactive nature, in that it can either be directed upward or downward, according to the direction of the capillarity-gravitational resultant to be counteracted. The most curious effect in this context is however represented by the fact that the border-adhesion-force continues to act even when the lower meniscus seems to detach from the tube edge. It is argued here that this detachment is only apparent, and that a thin liquid layer continues to connect the lower meniscus to the tube edge.
The experiment indicates that the triple line pins to the capillary edge, and such a pinning effect definitely controls the stability of the column by introducing a very strong stabilizing force.

Along the evaporation process both the contact angles undergo a progressive reduction, eventually approaching an asymptotic value. In the second half of the transient, in particular, the capillary resultant reverses in sign, and sums-up to the column weight. The above stabilizing force may therefore become as high as 150% of $F_w$ or more. In the final stages of the observations the lower meniscus seems to detach from the edge, but the pinning force remains active anyway. All of this has little effect on the evaporation rate.

5. Final remarks

Only provisional conclusions can be drawn from the above experiments, due to the limits of the data collated, both in quantitative and qualitative terms. The experimental basis certainly needs to be extended for firm conclusions to be gained.

Keeping this premise in mind, all the results highlight anyway the effect of gravity on capillarity in tubes under non-conventional conditions. The capillary length scale $l_c = (\gamma/\rho g)^{0.5}$ is normally used to decide whether gravitational effects on the meniscus shape can be overlooked. For water and n-Hexadecane $l_c \approx 2.7$ mm, and $\approx 1.7$ mm respectively; these are therefore comparable to the tubes’ diameter size (1-2 mm). Extension to lower-sized capillaries would then be useful, since it is expected that meniscus deformations observed in the inclined and the L-shaped tubes reduce for reducing the tube diameter.

The experiments presented here, in fact, demonstrate that gravity modifies the meniscus shape and affects the contact angle value or its distribution along the contact line, so as to provide the mechanical stability of the system. The contact angle variation should be included within the advancing-receding range typical of the solid-liquid couple. In the case of the edge-ending columns the upper contact angle was found to change of 20° or more, and a similar variability range was found for the freely suspended column. This would indicate that that glass surface was pretty far from being ideal. Note, however, that contact angle hysteresis has been demonstrated to be possible even for perfectly ideal surfaces, under partial wetting conditions [21, 22].

The thermodynamic stability of the above states remains questionable, since, in fact, only the equilibrium value, $\theta_e$, is considered to correspond to a state of thermodynamic equilibrium. It is then possible that the observed states are meta-stable or non-unique.
The presence of pinning effects can obviously offset any other reasoning, and these have been observed (or can anyway be present) in the experiments. Pinning of the triple-line exercises reactive forces. When applied to the capillary edge, pinning has evident and even surprising effects.

As for the effect of the gravitational field on the capillary rise in inclined tubes, it can now be stated with reasonable certainty that the vertical capillary rise reduces progressively for increasing the tube inclination from the vertical. The experiments for the L-shaped capillary show, in turn, that the horizontal elongation of the liquid column is related to the difference in height between the horizontal tube and the free surface of the feeding-pool.

A comment is now in order, on the possibility of modelling numerically cases of this kind, where the contact angle hysteresis plays a fundamental role in the mechanical stability of the hydrostatic system. An attempt was in fact carried out [23] to predict the statics of the inclined capillary used in [1], using a commercial CFD code, including the VOF algorithm for the tracking of the meniscus shape. Even if a reduction in the vertical rise of the column was actually obtained, it was also noted that the deviations of the contact angles from the static value were very limited, and fell within the numerical errors. This was later confirmed by analysing with the same method the case of an inclined capillary-slit. A slight meniscus deformation was found at the end of transient, but no deviation of the lower and upper contact angles from the equilibrium value. As a consequence, no reduction in the capillary rise could be observed. It can then be concluded that numerical tools not including a specific model for contact-angle hysteresis can not give any valid prediction in this field.

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