

**Tata and Arora Reply:** In a recent Letter [1] we reported a novel vapor-liquid (VL) condensation in charged colloidal suspensions upon de-ionization. The results were interpreted as evidence for the presence of attraction in the particle pair potential. In their Comment [2] Palberg and Würth (PW) report the repetition of our experiments under three different sealing conditions and attribute the phenomena to be due to nonequilibrium concentration gradient of impurity ion concentration  $c_s$  and claim that the results can be explained on the basis of DLVO potential. The concern of PW about the gradient in  $c_s$  is legitimate and its consequences must be examined; however, we show with the help of additional experimental results that the phenomenon reported in [1] is not due to the gradient in  $c_s$  and is a genuine equilibrium VL condensation.

PW noted that only sample C, which is expected to have largest  $c_s$ , exhibited a stationary separation and attributed it to presumably large gradient in  $c_s$ . They argue that *this gradient leads to the particles moving towards the ion exchange resins (IEX) and results in the phase separation.* We have also been concerned about the possible gradients in  $c_s$  and its effects. In order to establish that the phenomenon reported [1] is not due to the gradient in  $c_s$ , we have independently carried out further experiments in cells where the IEX (contained in a gauze bag) were confined to the upper region of the cells (see [3] for sample preparation). Such an arrangement would reverse the direction of the gradient in  $c_s$ , if any. Note from Fig. 1 that VL phase transition occurs here also and under the action of gravity the concentrated phase is settled in the lower part of the cell. If the arguments of PW based on DLVO potential were valid, the particles should have moved towards the upper part of the cell. Further, if the phenomenon occurred due to the gradient in  $c_s$  the suspensions with  $n_p > 3 \times 10^{12} \text{ cm}^{-3}$ , which are also expected to have similar  $c_s$  gradient, should also have shown stationary separation. On the other hand, these suspensions are found to exhibit a reentrant homogeneous state [1]. We agree with PW that there may be some gradient in  $c_s$  within the cell; however, our new result unambiguously shows that the VL condensation is an equilibrium phenomenon arising due to a minimum in the particle pair potential. The microscopic investigations in a *gradient free* suspension [4] also suggest the presence of a potential minimum.

We now show that the observations of PW in cells C, A, and B, which have successively lower  $c_s$ , are understandable on the basis of the pair potential  $W(r)$  derived by Sogami [5] and are also consistent with those reported by us [1,6]. The position of the potential minimum  $R_m$  of  $W(r)$  depends strongly on the Debye screening length and hence on  $c_s$ . Sample C shows stationary separation similar to that reported in [1] as it has relatively large  $c_s$ . At low  $c_s$ , when  $R_m$  is greater than the average nearest neighbor (nn) distance  $R_a \approx n_p^{-1/3}$ , the system can be in a reentrant homogeneous state as observed in samples A and B. These observations are also consistent with our

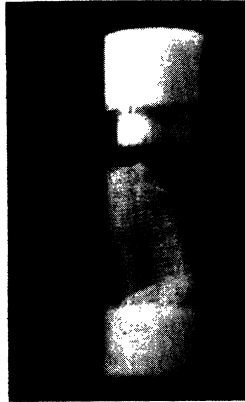


FIG. 1. Cell exhibiting VL condensation in aqueous polystyrene colloid (particle diameter 110 nm,  $n_p \approx 2 \times 10^{12} \text{ cm}^{-3}$ ) with IEX in the bag in the upper part.

Monte Carlo (MC) simulation results [7]. The simulations also predict that upon complete de-ionization ( $c_s \approx 0$ ) the suspension will be homogeneous (ordered), as reported by PW.

PW further argue that the homogeneous liquidlike or crystalline phases could be understood on the basis of DLVO potential. It must be emphasized that these phases are also predicted by the MC simulations using the potential  $W(r)$  at low  $c_s$  [7] and it is shown that both these potentials are equally good in explaining the structural ordering [3,8] in these suspensions.

To conclude, we have experimentally demonstrated that the VL condensation is not a nonequilibrium phenomenon arising due to gradient in  $c_s$ . The different behaviors in the cells A, B, and C of PW can be understood on the basis of the dependence of  $W(r)$  on the average values of  $c_s$  in the cells. Further, the excellent quantitative agreement [1] between the nn distance in the liquidlike phase and the  $R_m$  predicted by  $W(r)$  also confirms the validity of  $W(r)$ .

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B. V. R. Tata and A. K. Arora  
Materials Science Division  
Indira Gandhi Centre for Atomic Research  
Kalpakkam 603 102, India

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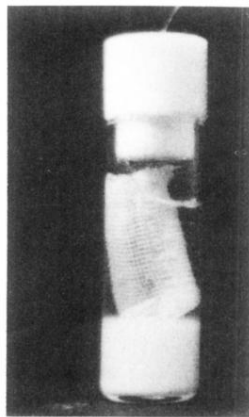


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