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A Perspective on the Dynamics Properties in Polymer Nanocomposites

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Abstract Polymer nanocomposites (PNCs) usually have superior properties than pristine polymers. Understanding the dynamics properties in PNC system is crucial to reveal the mechanism of property change unpon the addition of nanoparticles (NPs), and therefore for a better design of the material properties. In this short perspective, we summarize recent advances mainly from theoretical and simulation studies of dynamics properties in polymer nanocomposite system. One is the "vehicle model" which reveals that diffusion dynamics of sticky NP is coupled to surrounding chain segments. Similarly, recent simulations demonstrate that such coupling also exists in all-polymer nanocomposite wich is composed of linear polymer chains and single-chain nanoparticles (SCNPs). These SCNPs have almost the same chemical composition as the matrix chain. Therefore, it is assumed that such all-polymer nanocomposite can act as a model system where there are no enthalpic interactions between NPs and polymer chains. Although the above dynamic coupling was found in the above two different systems containing inorganic NPs or relatively small organic SCNPs, it was found that the length scale of such dynamic coupling (the thickness of the matrix/NP interface) is comparable to the NP size, which is surprisingly consistent in the above two different systems. In addition, a chain-length dependence of the NP influence on the chain dynamics reported from a recent joint simulaiton and experimental study of all-polymer nanocomposite system, and a theoretical model developed for such phenomena are also reviewed. At the end, we give an outlook of this field, especially for possible chain-length dependence of complex dynamics in sticky-NP systems.

Keywords Polymer nanocomposite; Dynamics properties; Interfacial interaction

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INTRODUCTION

In the past few decades, the researches on polymer/nanoparticle composite (PNC) materials have been at the forefront of the polymer field. The incorporation of nanoparticles (NPs) into polymers not only brings better properties to polymer materials, it also greatly expands the application prospects of functional polymer materials in optics, gas separation, capacitors and insulator dielectric materials.[1–4] However, with the addition of NPs, the factors influencing material properties turn to be multi-scale, multi-components and multi-variables. For instance, not only the NP size and shape, surface chemistry, but also the relative size of the NP to the character size of the polymer chain, *i.e.*, Kuhn length and the chain radius of gyration can all be crucial factors determining the interactions between NP and polymer matrix and therefore the macroscopic material properties. Such complexity of the PNC system renders it difficult for the experimental investigation of various microscopic interaction mechanisms between NPs and polymer chains.

In this perspective, we present some recent progresses, from both literature and our group, in theoretical and simulation studies of dynamics properties in PNC systems. To have a full picture of the topic, understanding of the dynamics properties of both components, polymer chains and NPs, are important. Especially for the former, we have a well-known scaling relation of $\eta \sim N^3$ for entangled system according to the reptation theory by de Gennes or Doi-Edwards tube model, where η is the melt viscosity. Although these models have been proven very successful in describing entangled linear chain dynamics in homopolymer systems, they could not describe the effect of the addition of NPs on the dynamic properties of PNC systems. Empirically, in order to ensure the dispersity of NPs in a polymer melt, NPs are often sticky and have attractive surfaces to matrix. As a consequence, it has often an increase in the viscosity of the resulted PNC system.[5–7] Such viscosity increasement can be well described by the so-called Einstein-Batchelor (E-B) theory,[8,9] where viscosity follows a power law relation of $\eta = \eta_0(1 + 2.5\phi_c + 6.2\phi_c^2)$. However, such a theory is developed on the basis of small molecular solvent, it does not consider the chain length dependence for polymer chain dynamics. Therefore the specific scaling relation of N^ν for polymer system is inherently absent.

However, there are many experimental systems where the results are contrary to the prediction of the above E-B theory. For instance, Mackay and coworkers reported a so-called non-Einstein like viscosity reduction in an all-polystyrene PNC sys-

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tem, where NPs were built by an intramolecular collapsing of linear polystyrene chains induced by cross-linking reactions along the chain backbone, therefore they had the same composition as matrix linear chains. In such an idea model system, complex enthalpic interactions between polymer chains and NPs are considered to be absent. We also have to note that such anomalous viscosity reduction has also been widely reported in different PNC systems, where NPs can be inorganic nanoparticles,[10–13] fullerene,[14] grafted SiO_2[15,16] and dendrimer nanoparticle.[17] Many explanations have been proposed to explain such an anomalous phenomena, such as free volume effect, disentanglement effect, and NP induced shear thinning, *etc*. However, it has been proven that all of these possible models are not capable of fully explaining the above anomalous viscosity reduction effect.[17] Therefore, the underlying mechanism still remains a major challenge in the field. In addition, will the presence of NP influence the *N*-dependence of the polymer chain dynamics?

MECHANISMS OF INTERFACE DYNAMICS

Vehicle Model

In order to unravel the underlying mechanism, the interaction mechanism between polymer chain and NP at the interface region and the NP dynamics are considered to be crucial.[18–21] Sokolov *et al*.[22,23] combined small-angle X-ray scattering, atomic force microscopy, dielectric spectroscopy and other characterization methods to study the interfacial dynamics and interfacial thickness in PNCs. Theoretically, a so-called "vehicle model" has been proposed to describe the diffusion dynamics of embended NPs in polymer matrix.[24] When NP is much larger than the size of the matrix polymer, the diffusion dynamics of NP obeys the conventional "core-shell" model. There is a "shell" of polymer chains adsorbed on NP surface (Fig. 1a), they diffuse as a whole and the dynamics obeys the Stokes-Einstein relation. While for ultra-small NPs ($R_a \ll R_g$, Fig. 1b, where R_a represents the radius of NPs and R_g the radius of gyration of polymers, respectively) the adsorption layer is composed of many interpenetrating segments, and its diffusion mechanism is dominated by the "vehicle mode", as demonstrated in Fig. 1(c). At the initial moment, the diffusion of NP is related to the surrounding red chains (picturedly called "vehicle"). As the diffusion proceeds, NP will randomly "escape" from the original "vehicle" and jump randomly to other "vehicles" around (the green polymer chain in the picture). Sokolov *et al*.[25] added OAPS nanoparticles to poly(propylene glycol) (PPG) with

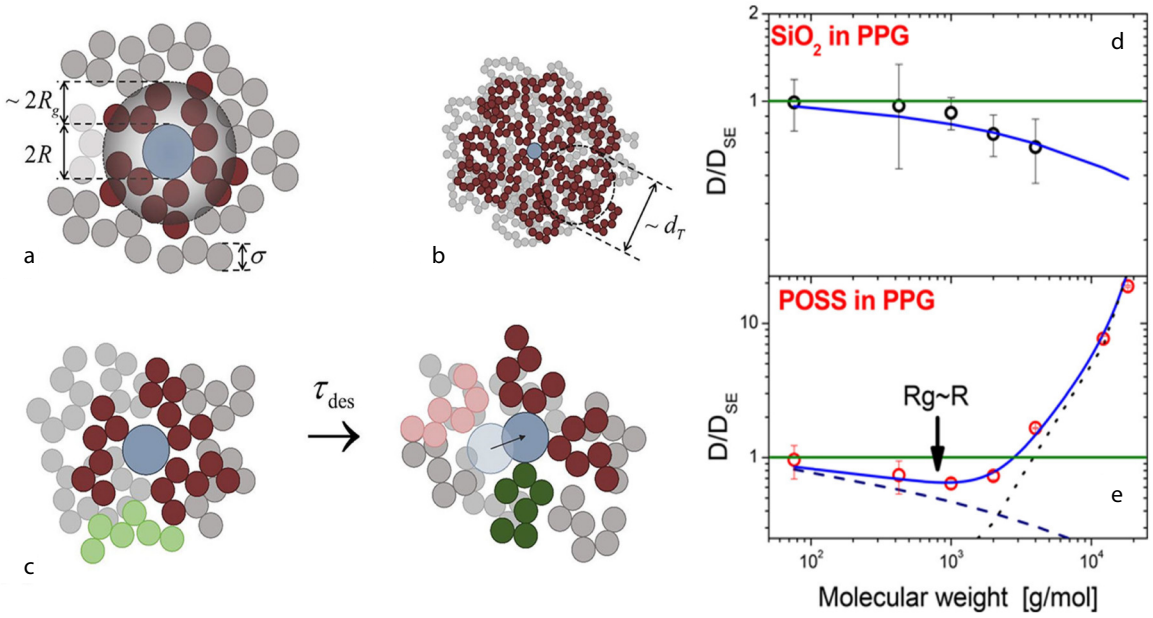

Fig. 1 Schematic of one sticky NP in a polymer melt where matrix chain segments (gray and red circles) are adsorbed onto the NP surface (blue circle) and (a) the NP radius (R_a) is larger than radius of gyration (R_g) of the chain, the adsorbed chains (red circles) form a dense layer around NP surface creating a core–shell object of effective radius $R_{eff} \sim R_a + R_g$ (shaded black region) and (b) $R_a \ll R_g$ where nonadsorbed segments significantly interpenetrate the adsorbed layer and the effective particle picture breaks down. For entangled melts an additional length scale, the tube diameter (d_t) is indicated. (c) Schematic representation of the "vehicle" mechanism. Initially, a NP follows the adsorbed chain motion (red circles, shown in left panel) using it as carrier or vehicle. The characteristic desorption time τ_{des} is defined as the elapsed time of elementary event of a NP desorbing from a segment and randomly finding a new vehicle chain (green circles, right panel). (d) Diffusion of SiO_2 NPs in PPG melts scaled by D_{SE} (symbols) versus molecular weight (MW) of the polymer chains. The blue solid curve is the prediction of the core-shell mechanism with $R_{eff} = R_a + R_g$. The green solid line denotes where the diffusion of NPs follows the Stoke-Einstein equation. (e) Diffusion of the OAPS particle in PPG melts scaled by D_{SE} (symbols) versus MW of the polymer chains. The black dashed curve is the prediction of the core-shell model with $R_{eff} = R_a + R_g$, while the black dotted curve corresponds to the diffusion constant (MW independent) predicted by the vehicle mechanism for relatively short τ_{des}. The solid blue curve is the sum of these two diffusion coefficients. The arrow marks the molecular weight when $R_g \sim R_{OAPS} \sim 0.9$ nm. The error bars are of the order of the symbol size for the points where they are not visible. (Reproduced with permissions from Refs. [24, 25]; Copyright (2018) American Chemical Society).

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different molecular weights, and found that the diffusion mechanism of nanoparticles changed from the core-shell model to the vehicle model as the chain length increased, as shown in Fig. 1(d). Importantly, this transformation happens at the point where the R_g of the melt chain is equivalent to the effective radius of the NP, which also shows the length scale of interface thickness between such adsorbed nanoparticles and the melt is equivalent to the radius of NP.

Dynamic Properties of All-polymer PNC System

Although the above models for the dynamics of sticky NPs are interesting, they cannot be used to interpret the above mentioned anomalous non-Einstein like viscosity reduction effect, especially possible models for chain dynamics are still missing. Recently, our group has been devoted to the study of dynamic properties of all-polymer PNC system.[26] In such a system, it was assumed that there is no complex enthalpic interactions between matrix linear polymer chain and NP since they both are chemically identical with each other. In such a model system, *in situ* characterizaiton of interfacial properties experimentally are difficult. We started at the investigation of the interfacial interaction mechanisms in the system[27] by using molecular dynamics simulations. In particular, a systematic coarse-grained polystyrene model[28] was used, which facilitates the use of large time and length scales in the simulations for the PNC system. Our simulation results demonstrated that the interaction range (*i.e.*, interface thickness) of the single-chain nanoparticle (SCNP) with linear matrix chains is comparable to its own size, implying that the interface is mainly composed of chain segments comparable in size of SCNPs, independent of the overall chain length of the matrix polymer. From the density distribution of PNCs in Fig. 2,[29] we found that there is a free volume release effect at the interface region. In this area, the overall density is smaller than that in the bulk phase. The release of free volume will inevitably lead to the dynamic acceleration of the melt chain in this region. In addition, we also investigated the diffusion dynamics of SCNPs and compared with that of the matrix linear chain. We found that for the chain segments which have comparable size to SCNP, they have almost identical mean square displacement (MSD) and displacement probability distribution as SCNP at a time scale smaller than the relaxation time of SCNP, which implying that the diffusion dynamics of SCNPs are dynamically coupled to the surrounding size-comparable matrix linear chain segments, independent of the overall viscosity of the system.[30] This dynamic coupling was also verified by studying the matrix chain length dependence for the SCNP diffusion coefficient.[31] In the short chain system, the diffusion coefficient D of the SCNP decreases with the increase of the melt chain length, showing the behavior predicted by the S-E relation. When the chain length continues

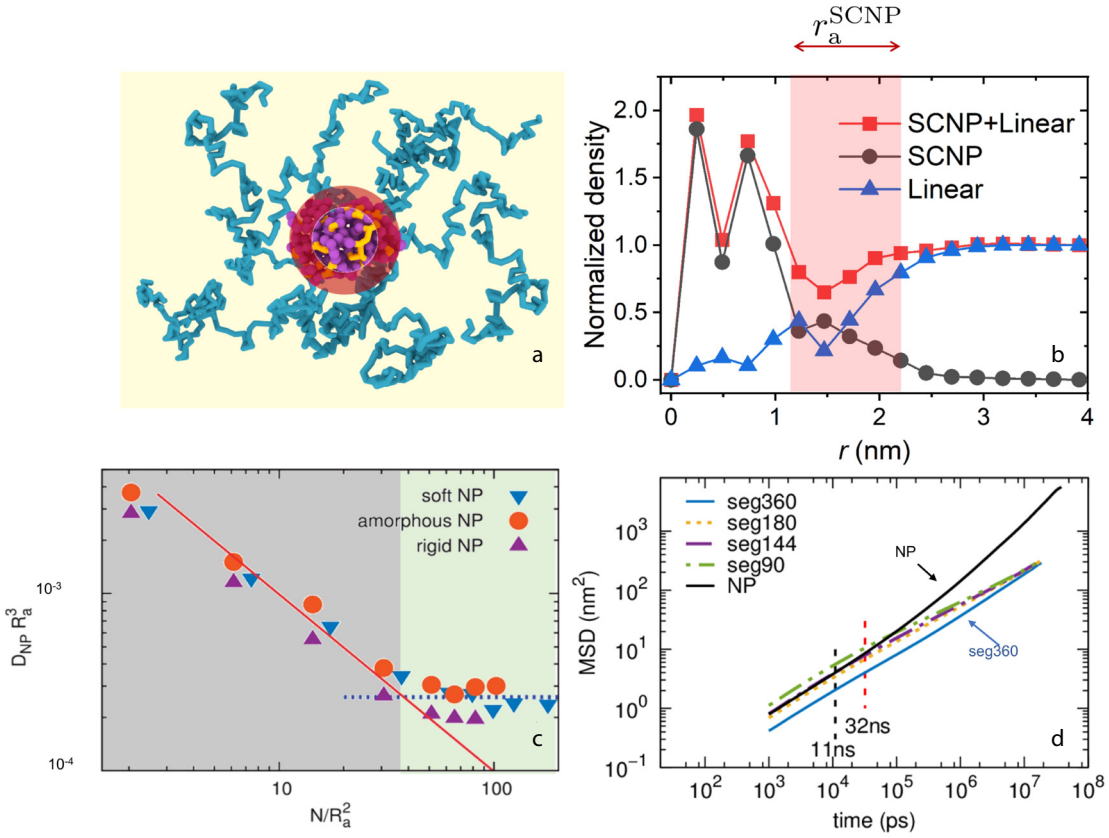

Fig. 2 (a) Schematic diagram of the nanoparticle-melt interface. (b) Normalized density distribution of monomers belongs either to SCNP or to melt chains at interface region. Shaded regions indicate the interface region. (c) Diffusion coefficients of the NPs in PS melts with different chain lengths. Red solid and blue dotted lines are guide lines with scaling factors of −1 and 0, respectively. (d) Mean-square-displacement (MSD) of NP250 and polymer segments with different lengths. (Reproduced with permissions from Refs. [30, 31], Copyright (2017, 2016), AIP Publishing, Elsevier).

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to increase, especially beyond the entanglement length, the decrease in the diffusion coefficient D of SCNP ceases, and the transition point occurs around chain length comparable to the size of SCNP.

Theoretical Model of Non-Einstein Viscosity Reduction

More importantly, after analysing the acceleration effect induced by the presence of SCNP on different length scales, we found an unexpected N-dependence.[32] We found that systems with longer matrix linear polymer chains will have a larger acceleration effect at a fixed loading of SCNPs. In order to testify such a prediction from the simulaiton, we have also conducted experimental rheological (small amplitude oscillatory shear) measurements on PNC system at a fixed SCNP loading of 2%, but with different matrix molecular weight. The experimental results demonstrated that systems with longer matrix chain will have a larger reduction in zero-shear viscosity, in good agreement with simulation. Such an N-dependence can be related to the dynamic coupling between SCNPs with their surrounding size-comparable chain segments that we discussed in the above section. Since SCNP is folded from a single chain, this results in a decrease in its internal degrees of freedom, which leads to a decrease in friction exerted on surrounding matrix chain segments. Such an effect can be transmitted along the polymer chain backbone through dynamic coupling with the size-comparable chain segments. In this studied system, the segment length corresponding to the nanoparticle size is ~144, which is close to the entanglement length of PS melt. Based on the above analysis, we proposed the following molecular mechanism to explain the viscosity reduction of the PNCs system:[32]

$$\eta = \frac{\pi^2 k_B T}{4 v_0 N_e} \tau_e \left(\frac{N}{N_e}\right)^3 f(\phi; N) = \eta^{pure} f(\phi; N) \quad (1)$$

where η_{pure} is the viscosity of the pure melt, ϕ is the SCNP fraction, N is the chain length of matrix polymer chains, and $f(\phi; N)$ counts the reduction of friction along the chain caused by adding NPs:

$$f(\phi; N) = \frac{N_e}{N} \sum_{i=1}^{N/N_e} \exp(-\delta \phi \boldsymbol{A}) : \mathbf{1}_{Z \times Z} \quad (2)$$

where δ describes an effective reduction in monomeric friction and $\boldsymbol{A}$ is a matrix whose element $A_{ij} = \exp(-|j - i|/a)$ describes the correlation between blobs or the contribution from neighboring j blobs along the chain backbone to blob i and a is the correlation length along the chain backbone. A least squares fitting of experimental data using the relation in Eq. (1) giving a correlation length ~13 entanglement blobs for this system, as shown in Fig. 3. Overall the N-dependence of the viscosity reduction is captured very well by our theory.

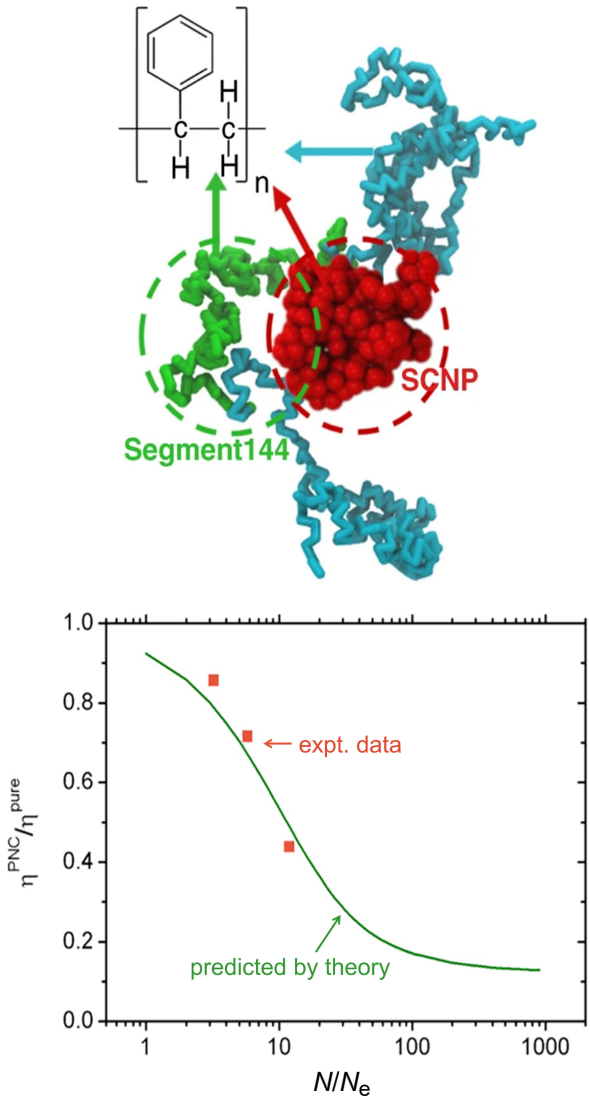

Fig. 3 Experimentally measured viscosity ratio between composites and pure PS melt, η_{PNC}/η_{pure}, plotted as a function of N/N_e. The solid line indicates a least squares fitting of experimental data points (symbol) using Eq. (1). On the right is a schematic depiction of the length scale of the interactions between SCNP (in red) with polymer chain. The entanglement blob which has a similar size with SCNP is plotted in green.

PERSPECTIVE

In fact, such viscosity reduction was widely reported in various PNC system. For instance, by adding polymer-grafted SiO_2 NPs into a polymer matrix, Mangal *et al.*[15] found an apparent speeding up of relaxation of the polymer matrix. Goldansaz *et al.*[17] added dendritic polyethylene NPs into polystyrene melt, and found an anomalous reduction in viscosity. The maximum reduction in viscosity was reached at 5% content of dendritic polyethylene NP. Continuing to increase the amount of NPs would lead to an increase in viscosity, but still less than the viscosity of pure polystyrene melt. In contrast to all-polystrene systme we discussed above, These systems usually have relatively strong heterogeneity. In all-polystyrene PNC system we studied, the SCNPs were constructed from intrachain crosslinking reactions between benzocyclobutene (BCB) units. Although these BCB units are chemically similar with the polystyrene chain backbone, they are chemically distinct. Therefore we can speculate that such chemically dissimilarity introduced inherently by cross-linking agent might be an important factor influencing the observed viscosity reduction effect. If this is the case, one may further manipulate the viscosity reduction effect to an optimum level, since tuning the NP surface chemistry has been now a mature technology. A

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systematic investigation on this point is ongoing in our group, hopefully detailed results on this topic will be reported shortly.

Interestingly, we also note that all-polystyrene PNC system we studied share the same interaction mechanism as the "vehicle" model for sticky NP, *i.e.*, the results of both systems demonstrate that the interaction range between NP and surrounding matrix polymer chains are on the order of NP size itself. Therefore, a direct question arises: is the N-dependence found in all-polystyrene PNC system general, especially also to the sticky NP system? Such an N-dependence generated from a friction decrease transmitted along polymer chain backbone in all-polystyrene PNC system, the friction increase effect can also be definitely transmitted along the chain backbone in sticky NP system. However, if the N-dependence also exists for deceleration effect in sticky NP systems, we would be curious how large can be the increament in the viscosity? Our recent molecular dynamics simulations using general Lennard-Jones potentials already show that such N-dependence do exists. More interestingly, along with such deceleration effect, there are some obvious subdiffusion behavior and new interesting relaxation modes due to the addition of attractive NPs in the system. More detailed analysis is still ongoing in the group.

In summary, although polymer/nanoparticle composite materials have achieved rapid development in recent years, due to the complexity of the system, theoretical models are still lacking. As an effective supplement to experiments, computer simulation is of great help to the study of PNC systems. Taking the interface mechanism as a good starting point, we studied the interface structure and dynamics of an all-polystyrene PNC system, and found that the interface thickness is equivalent to the size of NP. The NP and matrix chain segments of comparable size are dynamically coupled, leading to an N-dependence in the dynamically acceleration effect. Starting from the understanding of the variation in the monomeric friction coefficient, we proposed a theoretical model describing the effect of nanoparticle addition and the N-dependence of the NP influence on system dynamics, which explains very well the non-Einsteinian viscosity reduction effect reported in literature. Such N-dependence is also expected to be general to sticky NP systems, where the viscosity of the system will increase upon adding of NPs. However, some preliminary simulations and analysis (not shown here) demonstrate that in such a system, diffusion and relaxation dynamics in the system are very complex, and a detailed study of such an system will be interesting for a future study.

Conflict of Interests

The authors declare no interest conflict.

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