传递现象

含硅聚合物中小分子扩散行为的分子模拟

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摘要　　选择PCFF和COMPASS分子力场对橡胶态聚合物PDMS 和玻璃态聚合物PS1体系进行模拟。COMPASS力场模拟得到的体系密度，O2和N2在PDMS与PS1中扩散系数更接近实验值。在模型大小一定时，Group-based求和法中截断距离越长，耗用机时越长，但对计算结果改进不大；截断距离为1.3 nm时计算结果最好。Ewald方法耗时多而对计算结果却无明显改进。体系大小对扩散系数的计算值影响甚微。体积越小的分子，在聚合物中运动的范围越大，扩散系数越大。氧气和氮气分子在PDMS与PS1中运动轨迹不同，在PS1中氧气运动范围远大于氮气，而在PDMS中氧气运动范围稍大于氮气。小分子运动轨迹基本与聚合物自由体积分布对应，自由体积分数大，扩散系数也大。

关键词　　分子模拟　扩散系数　分子力场

分类号

Molecular simulation of transport behavior of penetrant through silicon-containing polymers

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Abstract

 PCFF and COMPASS force fields were used to describe PDMS and PS1 polymeric systems and to estimate the diffusion coefficient of N2 and O2 through the PDMS and PS1 matrices at 298 K by molecular dynamics simulation.It was found that the COMPASS force field was better in describing the transport behavior of the penetrants.The calculated densities after refinement were in good agreement with the experimental results.The group-based and Ewald summation skills were used to estimate the non-bonded interaction between atoms.Calculation using the Ewald summation method took much longer time without bringing in obvious improvement in density estimation.Various cut-offs in using the group-based summation method did not produce densities with much difference, and the cut-off of 1.3 nm was the best.Two types of diffusions of the small molecules in the polymers were discussed.The diffusion of O2 and N2 in PDMS could be transformed from anomalous to normal motion in 30 ps; while their diffusion in PS1 would take 300 ps transforming from anomalous to normal state.The trajectories of diffusion of N2 and O2 in PDMS or PS1 were different.The motion area of O2 in the PS1 was much larger than N2; however, the former in the PDMS matrix was only slightly larger than the latter.The diffusions of O2 and N2 in the PDMS and PS1 were consistent with the free volumes of polymers.

Key words FONT-FAMILY: 'Times New Roman' mso-bidi-font-size: 12.0pt mso-font-kerning: 1.0pt mso-ansi-language: EN-US mso-fareast-language: ZH-CN mso-bidi-language: AR-SA molecular simulation')">mso-fareast-font-family: 宋体">molecular simulation diffusion coefficient force field

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