



A new insight into the membrane-supported interfacial polymerization via Poisson Distribution



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ABSTRACT

In this theoretical study, the membrane-supported interfacial polymerization is distinctively considered as a Poisson process. We liken the film formation on the supporting membrane to the phenomenon that numerous water-waves created by raindrops hitting a pond expand on the pond surface gradually as they grow larger. Accordingly, a mathematical equation for the permeability of thin-film composite membranes, named as “Poisson Interfacial Polymerization” equation, is proposed. And the simulation results are in good agreement with the corresponding experimental results reported by many researchers. We believe that our approach offers new degrees of freedom in establishing mathematical models for interfacial polymerization.

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1. Introduction

Interfacial polymerization (IP) is a widely used preparation technique for thin-film composite (TFC) membranes [1]. Since 1972, a series of valuable TFC membranes, such as polyurea [2,3], polyamide [4–12], polyurea-amide [13–16], polyimide [17], polyester [18–22], and polysulfonamide [23,24], have been successfully fabricated by this facile technique, leading to an urgent need for a theoretical understanding of the membrane-supported IP process [25–27]. A systematic mathematical model for the IP process could provide a powerful guide for an effective control of the membrane performance [25,28,29]. Just as what Dhumal et al. [25] recently summarized from the existing mathematical investigations of IP, three basic approaches are often employed to theoretically treat the formation of the polymer film. First, the dissolved monomers diffuse to the phase-interface where the IP takes place. The entire formed polymer products contribute to the formation of a polymer film which is incompatible with the aqueous/organic phase [1,27]. In the second, IP is assumed to occur in a thin reaction zone which lies between the organic phase and the already-formed polymer film. The latter is excluded from the reaction zone as it grows [1,30–33]. In the third, the film growth is considered as a gradual thickening/gelification of the steady reaction zone which has a finite thickness [1,25,34]. Consequently, many extensive detailed,

parametric but highly effective theoretical models have hitherto been proposed successfully based on these three approaches [1,25–44]. These researches no doubt promote a better understanding of the membrane-supported IP process.

However, the existing theoretical researches usually encounter the difficulty in exactly tracking IP's non-steady state. The general exhaustion phenomenon of monomers probably leads to the transition from a reaction-controlled IP process to a diffusion-controlled one [28,29]. Besides, these researches usually need to introduce many parameters to establish the relationship between film performance and preparation parameters. It inevitably brings about highly numerous and complicated deducing processes from which the complex equations hence stem. Furthermore, from the standpoint of membrane engineering (or membrane industry) where the complex IP process is not the uppermost concern, a simple but effective model connecting the membrane performance directly with controllable and accessible preparation parameters is actually much more practical.

Motivated by the desire to build such an effective theoretical model based on a simple deducing process and equation, we have to shift our attention from the complex IP process to the property of the final resultant TFC membrane itself in the present study. To put it in another way, we attempt to divert our focus from “how” (how the preparation parameters influence the formation of TFC membranes) to “what” (what is the performance of the TFC membranes we could obtain under certain preparation parameters) based on the existing correct understanding [25,28,29,43,44] of the complicated membrane-supported IP process.

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Nomenclature

a_t	constant defined in Eq. (4)	X	a representative point on the surface of the supporting membrane
a_{mc}	constant defined in Eq. (5)	X'	a representative point on the pond surface (Poisson Raindrop Problem)
a_r	constant defined in Eq. (6)		
C_m	the original concentration of the investigated monomer in the aqueous/organic phase		
e	the base of natural logarithm		
f	permeability of the nonporous section on the supporting membrane		
f	permeability of the TFC membrane		
f_p	permeability of a partially covered membrane		
F	permeability of the supporting membrane, or the pore section on the supporting membrane		
I^*	the number of new water-waves appearing on the surface of the per-unit-area pond per unit time (Poisson Raindrop Problem); the number of new reaction points emerging on the per-unit-area supporting membrane per unit time		
n	the number of water-waves created by different raindrops which pass over a representative point X' on the pond surface up to time T (Poisson Raindrop Problem)		
$P(t)$	the probability that a representative point X on the supporting membrane belongs to the pore section on the membrane surface during the IP time t		
$P_n(t)$	the chance that the number of water-waves created by different raindrops which pass over a representative point X' on the pond up to time T is n (Poisson Raindrop Problem)		
t	interfacial polymerization time		
T	rain time (Poisson Rain Problem)		
		Greek letters	
		λ	the area of the water-waves on the per-unit-area pond during T (Poisson Raindrop Problem); the area of the polymer networks on the per-unit-area supporting membrane during t
		ν	the spreading rate of polymer networks on the supporting membrane
		π	the usual mathematical constant
		$\tau_{(i)}$	a certain moment throughout the IP process
		$\Delta\tau$	the time difference between τ and τ_{i+1}
		ϖ_{mc}	constant defined in the equation $\nu = \varpi_{mc} * C_m$
		ϖ_r	constant defined in the equation $I^* = \varpi_r t_i$
		Subscripts	
		BTEC	3,3',5,5'-biphenyl tetraacyl chloride
		HTC	cyclohexane-1,3,5-tricarbonyl chloride
		IP	interfacial polymerization
		PDMAEMA	poly[2-(N,N-dimethyl amino)ethyl methacrylate]
		PIP	Poisson Interfacial Polymerization
		PSf	polysulfone
		PVAm	polyvinylamine
		TEOA	trimesoyl chloride
		TFC	thin-film composite
		TMC	triethylamine

In the current study, a less parametric but practical mathematical model for the permeability of TFC membranes is tentatively established based on the Poisson Distribution. This result-oriented perspective mainly concentrates on the membrane performance rather than the extremely complicated IP process, which skillfully avoids the difficulty in tacking the non-steady state of IP process.

2. Theory

The mathematical model for the permeability of TFC membranes is developed via Poisson Distribution in this section.

The classical mean-field approach from the cross-sectional point of view is a long-held method to theoretically investigate the membrane-supported IP process. It inevitably leads to the conclusion that the growth of the ultra-thin polymer film much lies on its average thickness and the corresponding decrease in membrane permeability is mainly caused by this thickening process. However, the strong fluctuation of local monomer concentration would result in the phenomenon that the IP reactions at different points on the supporting membrane have different reaction rates. This is why the IP technique generally bestows a rough surface upon the as-obtained TFC membranes. Moreover, the different local morphologies and/or structures of the supporting membrane and hence the different local monomer-absorption conditions would also contribute to the formation of such a rough texture on the membrane surface. Therefore, the IP process could be reasonably considered as an assemblage of reactions with different reaction rates occurring simultaneously at various points on the supporting membrane.

Hereby, to facilitate the development of our mathematical model, we distinctively interpret these reactions as the ones with the same reaction rate taking place at different times throughout the IP process. Under this circumstance, a parameter I^* , referring to the number of new reaction points emerging on the per-unit-area

supporting membrane per unit time, is introduced into our following model in reason. Besides, from the surface point of view, the structure of the polymer film transits from a loose texture to a dense and even nonporous one as IP proceeds. In other words, it could be understood as the porous supporting membrane being covered gradually by crosslinked polymer networks which derive and then spread outwards from the corresponding initial reaction points. To put it more vividly, such a structure transition looks like numerous water-waves of different areas created by raindrops hitting a pond expand on the pond surface gradually as they grow larger (shown in Fig. 1). The “pond” stands for the porous “supporting

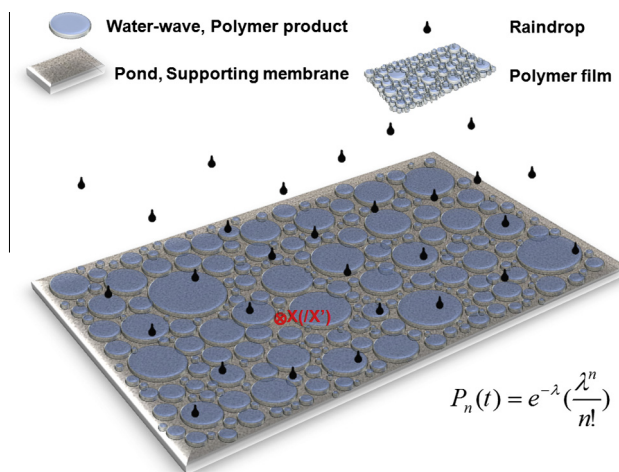


Fig. 1. Three-dimensional schematic representation of the Poisson Raindrop Problem, and the membrane-supported IP process viewed from the standpoint of structure transition on the membrane surface based on Poisson Distribution.

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