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# 1 On the Transport of Emulsions in Porous

## 2 Media

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### 6 Abstract

7 Emulsions appear in many subsurface applications including bioremediation, surfactant-  
8 enhanced remediation, and enhanced oil-recovery. Modeling emulsion transport in  
9 porous media is particularly challenging because the rheological and physical prop-  
10 erties of emulsions are different from averages of the components. Current modeling  
11 approaches are based on filtration theories, which are not suited to adequately ad-  
12 dress the pore-scale permeability fluctuations and reduction of absolute permeabil-  
13 ity that are often encountered during emulsion transport. In this communication,  
14 we introduce a Continuous Time Random Walk based alternative approach that  
15 captures these unique features of emulsion transport. Calculations based on the  
16 proposed approach resulted in excellent match with experimental observations of  
17 emulsion breakthrough from the literature. Specifically, the new approach explains  
18 the slow late-time tailing behavior that could not be fitted using the standard ap-  
19 proach. The theory presented in this paper also provides an important stepping  
20 stone towards a generalized self-consistent modeling of multiphase flow.

21 *Key words:* emulsion, porous media, colloidal transport, memory effects

## 22 1 Motivation

23 Transport [1] of (micro)emulsions in porous media is relevant to several subsur-  
24 face applications. Nearly all enhanced oil recovery processes involve emulsion  
25 formation and flow in some form or other [1]. Recently, it has been shown  
26 that emulsification of edible oils prior to injection into groundwater (as a  
27 substrate for enhanced bioremediation) improves its spatial distribution in  
28 the contaminated zone [2,3]. Subsurface uses of microemulsions, which possess  
29 large interfacial area and have the ability to solubilize otherwise immiscible  
30 liquids [4], include cleanup of chlorinated solvents from groundwater [5] and  
31 enhanced oil-recovery [6–8].

32 Emulsion transport in porous media can be broadly classified according to  
33 (a) the stability of the emulsion and (b) the drop size of the dispersed phase  
34 relative to the pore size of the medium [9]. One class involves dilute, rela-  
35 tively unstable emulsion with very small average drop-size to pore-size ratio.  
36 A second class involves relatively stable emulsions with drop-size to average  
37 pore-size ratio on the order of unity. A third class, which is the subject of  
38 this paper, involves stable emulsions with relatively small drop-size to average  
39 pore-size ratio.

40 Current approaches of modeling the latter class of emulsions capitalize on  
41 the similarity of the transport and attachment-detachment processes of the  
42 dispersed-phase droplets with those of colloids [10,3]. In general, most col-  
43 loidal transport models are extensions of the advection-dispersion equation  
44 (ADE), which assumes colloid and grain size homogeneity at all scales. As

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45 a result, these models predict a fast exponential concentration decay and do  
 46 not capture slow late-time elution of colloids satisfactorily. Recently, we intro-  
 47 duced a Continuous Time Random Walk (CTRW) based filtration model for  
 48 biocolloid transport [11] that captures small-scale heterogeneity in an effective  
 49 stochastic framework. The proposed approach provided excellent match with  
 50 experimental breakthrough curves, including the late-time slow tailing. The  
 51 objective of this communication is to extend the CTRW based approach to  
 52 the transport of emulsions.

## 53 2 Theoretical Considerations

54 Soo and Radke [10] described the transport of emulsion in porous media as  
 55 analogous to transport of a colloidal phase. Transport of colloids is commonly  
 56 modeled by colloid filtration theory (CFT) [12] as an extension of the ADE

$$57 \quad \frac{\partial c}{\partial t} + \frac{\rho_b}{\theta} \frac{\partial s}{\partial t} = \mathbb{F}[c(x, t)], \quad (1)$$

58 where lengths are made non-dimensional with respect to the column length  $L$ ,

$$59 \quad \mathbb{F}[c(x, t)] \equiv -v \left[ \lambda c + \frac{\partial c}{\partial x} - \alpha \frac{\partial^2 c}{\partial x^2} \right] \quad (2)$$

60 is the classical filtration operator (with  $v$  the pore velocity and  $\alpha$  the disper-  
 61 sivity) acting on the colloid concentration  $c(x, t)$ , and

$$62 \quad \frac{\partial s}{\partial t} = \frac{\theta}{\rho_b} k_f c - k_r s \quad (3)$$

63 where  $\theta$  is the porosity,  $\rho_b$  is the colloid density,  $s$  is the colloid concentration  
 64 adsorbed reversibly on solid surfaces,  $k_f$ , and  $k_r$  are the forward and reverse  
 65 sorption rates, respectively. The parameter  $\lambda$  is the filtration coefficient, the

66 fraction of irreversibly sorbed dispersed phase, which can be calculated as

$$67 \quad \lambda = -\ln(f) + \alpha \ln(f^2) \quad (4)$$

68 where  $f$  is the eluted fraction [13]. An extensive review of the filtration coef-  
69 ficient  $\lambda$  and its parameters can be found in [14] and [15]. Eq. (3) adequately  
70 describe the initial advection-dispersion dominated breakthrough, but not the  
71 late-time elution of the dispersed phase. Laboratory and field experiments of-  
72 ten display power-law decaying of the breakthrough curve tails, which cannot  
73 be captured by the ADE-based filtration models that predict an exponen-  
74 tial decay of the dispersed-phase concentration. The misrepresentation of the  
75 tails is rooted in the definition of the forward and reverse sorption rates in  
76 (3) as mean values over some volume. The inherent heterogeneity of natural  
77 porous materials, however, induces a wide spectrum of rates, which can only  
78 be represented by some probabilistic distribution function (pdf).

79 Physico-chemical attachment and detachment of the dispersed phase can be  
80 attributed to small scale heterogeneities of the pore-surface dispersed-phase  
81 properties. In particular, surface charge heterogeneities on both, the dispersed  
82 and solid phases play a fundamental role in the transport of the dispersed  
83 phase. Furthermore, the dispersed-phase droplets can vary in shape, size,  
84 and surface heterogeneity. These multiscale heterogeneities, which are diffi-  
85 cult to characterize in a fully deterministic way, call for an ensemble averaged  
86 stochastic treatment of the unresolved level of heterogeneity. In order to ac-  
87 count for these heterogeneities, we propose a conceptual model that is similar  
88 to the filtration model described in (1) to the extent that it decouples the  
89 transport of the dispersed phase into advective and dispersive fluxes, and  
90 sorption and desorption mechanisms. Whereas the ADE approach ignores the

91 small heterogeneities, our proposed model treats these in terms of probabilistic  
 92 distributions. We have shown elsewhere [11] that a CTRW based filtration-  
 93 model provides excellent description of both the advection-dispersion domi-  
 94 nated early breakthrough and the late time elution of colloids. CTRW is an  
 95 effective stochastic transport theory based on local-scale ensemble averages.  
 96 At the core of the CTRW formulation is  $\psi(t)$ , the waiting time pdf for a  
 97 single droplet transition over time  $t$ . This function implicitly accounts for all  
 98 transport mechanisms (e.g., advection, dispersion, sorption, and desorption)  
 99 that displace a particle from one spatial location to another. For an extensive  
 100 review of the CTRW in hydrogeological applications see [16] and references  
 101 therein. The CTRW filtration equation can be written as

$$102 \quad u\tilde{c}(x, u) - c_0(x) = \tilde{M}(u)\mathbb{F}[\tilde{c}(x, u)]. \quad (5)$$

103 The tilde  $\tilde{\phantom{x}}$  in Eq. 5 indicates Laplace transformed quantities with respect to  
 104 time  $t$ , and  $u$  is the Laplace variable. The quantity  $\tilde{M}(u)$  is defined as

$$105 \quad \tilde{M}(u) = \bar{t}u \frac{\tilde{\psi}(u)}{1 - \tilde{\psi}(u)} \quad (6)$$

106 and plays the role of a memory function, which convolutes the classical filtra-  
 107 tion operator,  $\mathbb{F}[\tilde{c}(x, u)]$ .

108 Considering the similarity in  $\zeta$ -potentials between bio-colloids and oil droplets  
 109 in emulsions, we propose that (5) can be used to effectively describe emulsion  
 110 transport in porous media.

111 Following Margolin et al. [17], we define the waiting time pdf  $\psi(t)$  (which  
 112 describes how the droplets in the dispersed phase are temporarily trapped  
 113 and then released after some time  $t$ ) as a generalized convolution integral of  
 114 two contributions:  $\psi_0(t)$ , which is related to the complexity of the pore space

115 geometry, and  $\phi(t|\tau)$  (with  $\tau = (t - t')/T$ ), which defines the retention of  
 116 the dispersed droplets,  $\psi(\mathbf{s}, \tau) = \int_0^\tau \psi_0(\mathbf{s}, \tau - \tau')\phi(\tau'|\tau - \tau')d\tau'$ . In the limit  
 117  $\phi(t|\tau) \equiv \delta(t' - \Lambda\tau)$ , Eq. 5 is reduced to an ADE with simple retardation  
 118  $R = (1 + \Lambda)$ . More generally,  $\phi(t|\tau)$  accounts for multiple attachment and  
 119 detachment events of a droplet in the time interval  $\tau$ , and as such can be  
 120 defined by means of multiple convolutions of a large number of single “sticking  
 121 time” pdfs,  $\varphi(t)$ , (compound Poisson process). Denoting by  $\Lambda$  the average free  
 122 state sticking rate, it is possible to write  $\tilde{\phi}(u) \equiv \exp(\Lambda\tau(\tilde{\varphi}(u) - 1))$ . With  
 123 these definitions,  $\tilde{\psi}(u)$  reads as  $\tilde{\psi}(u) = \tilde{\psi}_0(u + \Lambda(1 - \tilde{\varphi}(u)))$ . When  $\tilde{M}(u) = 1$ ,  
 124 and  $\Lambda = 0$  the CTRW filtration PDE reduces to the classical filtration theory.  
 125 If  $\Lambda > 0$ , the memory  $\tilde{M}(u) \neq 1$ , i.e., the model exhibits memory effects as a  
 126 result of the time convolution. A detailed account of these developments can  
 127 be found in [17] and [11].

128 At this point we introduce some assumptions on the functional forms of  
 129  $\psi_0(t)$  and  $\varphi(t)$ . The  $\psi_0(t)$  models the impact of pore space heterogeneity  
 130 on the anomalous tailing of the dispersed phase due to pore shape hetero-  
 131 geneity and accounts for the anomalous tailing in the breakthrough of con-  
 132 servative tracers in homogeneously packed soil columns. To account for pore-  
 133 level geometric heterogeneity of a passive tracer experiments, it is sufficient  
 134 to adopt an asymptotic power-law model of the form  $\tilde{\psi}_0(u) = 1/(1 + u^\gamma)$   
 135 (with  $0 < \gamma < 1$ ), which yields a power law tailing [18]. For homogeneous  
 136 porous structures,  $\gamma$  approaches 1, and the waiting time pdf tends towards an  
 137 exponential,  $\psi_0(t) \sim \exp(-t/\bar{t})$ , i.e., passive tracer transport tends toward a  
 138 Fickian-behavior limit.

139 Small-scale heterogeneities in physico-chemical interactions can also be repre-  
 140 sented by a sticking time pdf of the form  $\tilde{\varphi}(u) \equiv 1/(1 + u^\beta)$ , where  $0 < \beta < 1$

141 [11]. From a physical point of view, this means that there is no single charac-  
142 teristic filtration time scale for this problem.

### 143 **3 Illustrative Example**

144 The experimental results used to illustrate the proposed theory were derived  
145 from Coulibaly et al. [3] (experiment FS-7%-#2). The experiment was con-  
146 ducted using a PVC column of length  $L = 0.8$  m and diameter  $D = 0.029$  m.  
147 The column was filled with medium to fine sand ( $D_{10} = 0.10$  mm,  $D_{50} =$   
148  $0.38$  mm,  $D_{60} = 0.45$  mm, and 6.9% by weight is finer than  $75\mu$  m) dry packed  
149 to achieve bulk density of  $1.44\text{kg/m}^3$ , equivalent to a porosity of  $\theta = 0.46$ . The  
150 emulsions used in this experiment were prepared by blending 33% by volume  
151 soybean oil, 62% tap water, and 5% premixed surfactant (38% polysorbate  
152 80, 56% glycerol monooleate from Lambent Technologies and 6% water) in a  
153 commercial blender at high speed for 5 min. The mean droplet size was  $1.2\mu\text{m}$   
154 (standard deviation  $1.3\mu\text{m}$ ). The  $\zeta$  potential of the oil droplets and sand in  
155 tap water (pH = 8.2 and specific conductance =  $190\mu\text{S/cm}$ ) were -18 mV  
156 and -24 mV, respectively, indicating unfavorable conditions for the negatively  
157 charged oil droplets to stick to the negatively charged sand [3].

158 The column was first evacuated and then saturated by flowing de-aerated wa-  
159 ter upwards at  $2.5$  mL/min for 2 hours. Subsequently, a nonreactive transport  
160 test was conducted by injecting 25 mL ( $\sim 0.1$  PV) of 175 mg/L NaBr solution  
161 through the column followed by 1000 mL of deaired water. Effluent samples  
162 were analyzed for Br by ion chromatography. The emulsion flow test was con-  
163 ducted by injecting 25 mL of 11% oil by volume emulsion ( $\sim 0.05$  PV of pure  
164 oil) followed by 1000 mL of deaired water. The effluent was collected every 30



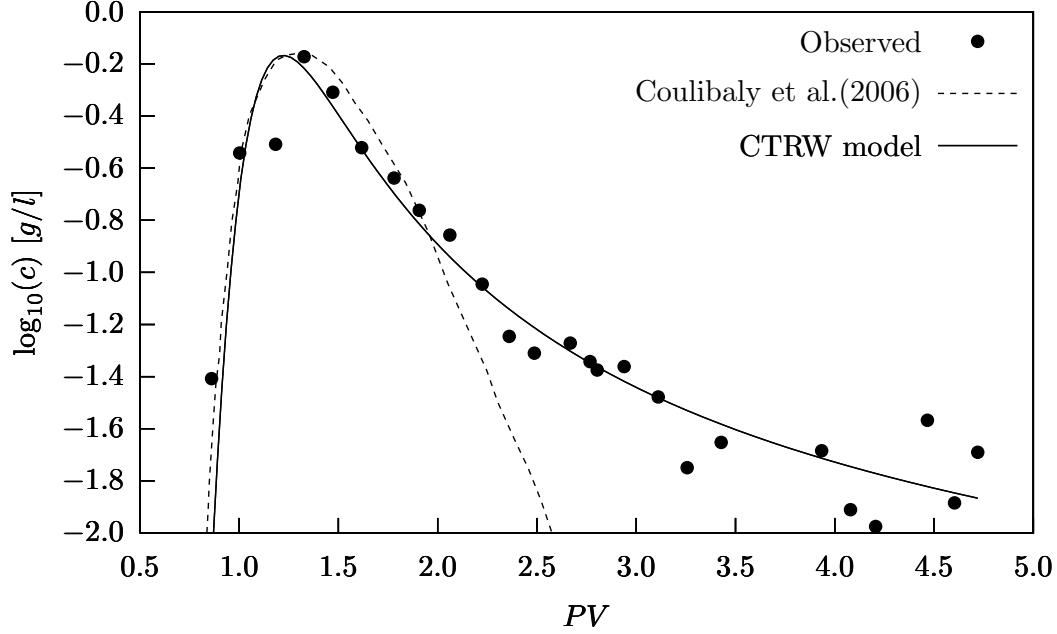


Fig. 1. Comparison of experimental results with model fits

165 mL and analyzed for oil content by volatile solids method.

166 The experimental oil BTC was fitted by means of (5). The time has been  
 167 rescaled in such a way that the BTC is plotted in pore-volumes. Lengths have  
 168 been rescaled by the column length  $L$ . The value of the filtration coefficient  
 169  $\lambda = 2.72$ , was obtained from (4) with the reported eluted fraction,  $f = 0.065$   
 170 and the value of the dispersivity  $\alpha = 0.1$  was obtained from the NaBr tracer  
 171 test [3]. This non-dimensional value of the dispersivity clearly indicates a scale  
 172 effect [19]. Considering that the porous medium used in the experiments was  
 173 made of sand grains of different sizes, it is legitimate to expect significant  
 174 small-scale heterogeneity of flow and transport properties. Long tailing in the  
 175 passive tracer BTC, which is typical in such porous media [2], might have been  
 176 misinterpreted as elevated dispersivity. Therefore, we introduce a model for the  
 177  $\psi_0(t)$  which assumes a long tailing, i.e.,  $\gamma < 1$  and a value of the dispersivity  
 178 of the same order of magnitude of the the median grain diameter-to-column  
 179 length ratio,  $D_{50}/L = 4.75 \times 10^{-4}$ . Given the distribution of diameter size

180 typical of the sand used in the experiment, we selected a value of  $\alpha = 1.5 \times$   
181  $10^{-3} \sim 3D_{50}/L$ . A sensitivity analysis indicates the appropriateness of this  
182 choice of  $\alpha$ . In contrast, using the value of  $\alpha = 0.1$  reported by Coulibaly  
183 et al. [3] and a value of  $\gamma = 1$  (Gaussian transport for the passive tracer),  
184 results in unsatisfactory CTRW model fit of the experimental BTC. The best  
185 fit values of the free parameters are  $\Lambda = 0.58$ ,  $\beta = 0.4$ , and  $\gamma = 0.9$ . The  
186 relatively high value for  $\gamma$  indicates that some geometrical disorder is present  
187 in the experimental column, as one would expect for natural soils. The small  
188 value of  $\beta$  is in agreement with that of transport of bio-colloids [18]. This value  
189 for  $\beta$  indicates a very high degree of physico-chemical disorder of the surface  
190 heterogeneity and a corresponding broad sticking time pdf  $\varphi(t)$ .

191 The best fit value for the transport velocity  $v$  indicates a retardation factor  
192  $R = (1 + \Lambda) = 1.58$ , in accordance to the sharply peaked  $\phi(t, t-t') \sim \delta(t' - \Lambda\tau)$   
193 analyzed earlier. The results are, thus, pointing towards a bi-modal type of  
194 transport for the dispersed phase: the first mode explains the attachment  
195 and detachment of the oil droplets to the pore walls and is characterized by  
196 a power law dominated sticking time pdf, whereas the second mode, clearly  
197 indicated by the overall retardation, can be attributed to a film-like flow along  
198 the surface of the pores.

#### 199 **4 Open Research Questions**

200 The CTRW based model for emulsion transport introduced in this paper pro-  
201 vided an excellent match to the experimental oil BTC of Coulibaly et al.  
202 [3]. In particular, the late-time long tailing was shown to be in remarkable  
203 agreement with a power-law dominated pdf for attachment and detachment

204 of the dispersed-phase (oil). This finding leads to interesting research ques-  
205 tions related to multiphase flow in porous media. We have shown here that  
206 the long time correlations underlying the power-law dominated pdfs need to  
207 be taken into account for a correct description of emulsion transport. The  
208 consequences of this microscopic behavior on other measurable macroscopic  
209 parameters needs further exploration.

210 We also recognize that the permeable pore space available for water flow is  
211 continuously changed as oil-droplets are lodged in pore-throats and pore-walls,  
212 resulting in transient effects in the small scale permeability to water. This ef-  
213 fect is different from straining of rigid colloids because the capillary forces  
214 that keep emulsion droplets in place can be readily overcome by hydrody-  
215 namic stresses. Moreover, the oil droplets in the dispersed phase inside the  
216 pore space are continuously moving giving the water phase a moving bound-  
217 ary for its flow. Recalling that the permeability of a porous medium at Darcy  
218 scale ( $k_0$ ) is defined as the volume average of the Stokes velocity flow field  
219 in the pore space available to the water phase, it becomes evident that when  
220 the fluctuation of the local  $k$  value (with respect to  $k_0$ ) are correlated in time,  
221 can induce macroscopically measurable changes in the effective permeability.  
222 Elsewhere, we have shown that long-range correlations in the spatial distribu-  
223 tions of diffusivity can induce memory effects and that these can be captured  
224 in the transient behavior of the effective permeability [20]. We argue that sim-  
225 ilar transient effects in the deviations from  $k_0$  are responsible for the changes  
226 in effective transient permeability observed in the emulsion transport exper-  
227 iments. Hence, the parameters of the CTRW filtration such as the free state  
228 sticking rate  $\Lambda$ , may be correlated to the observed drops in permeability. If we  
229 consider that a drop in permeability affects the velocity of the carrying fluid

230 (the water), and that the dispersed phase moves advectively with the water,  
231 it is plausible to draw a connection between the retardation factor observed  
232 in the oil BTC experiments and the drop in permeability. Assuming that this  
233 working hypothesis holds, the corresponding drop in the permeability should  
234 be given by  $k/k_0 \approx 1/R$ . For the experimental results reported in figure 1 this  
235 corresponds to  $k/k_0 = 0.63$ , which is of the order of magnitude of permeabil-  
236 ity drops observed in literature for this kind of porous media (e.g., [2]). This  
237 conjecture can be tested in multiple-column laboratory experiments.

238 The relation of the  $\beta$  exponent with the rheology of the emulsion remains  
239 poorly understood. Emulsion flow under simple experimental conditions ex-  
240 hibits non-Newtonian behavior such as aging and memory [21]. Further ex-  
241 ploration is needed to test whether these non-Newtonian characteristics  
242 are also responsible for the memory effects observed in the more complicated  
243 geometries of porous media.

244 Finally, the importance of microscopic transitions in emulsion transport high-  
245 lighted in this communication is likely to be important in other multiphase  
246 flow problems. Therefore, future research in multiphase flow theory need to  
247 address these effects in a self-consistent manner.

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