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Mathematical simulation of surfactant flushing process to remediate diesel contaminated sand column

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Abstract. This paper presents a numerical model based on a UTCHEM simulator to simulate surfactant flushing process to remediate diesel contaminated sand column. For this purpose, we modeled remediation process under 10000 and 20000 ppm initial concentrations of diesel. Various percent-mass sodium dodecyl sulfate (SDS) considered in our model. The model results indicated that 0.3 percent-mass of SDS at 10000 ppm and 0.1 percent-mass of SDS at 20000 ppm initial diesel concentration had maximum removal perdition which is in agreement with the experiment results. For 10000 ppm diesel concentrations, the coefficient of determination (R^2) and index of agreement (IA) between the model result and the experimental data were 0.9952 and 0.9695, respectively, and for 20000 ppm diesel concentrations, R^2 and IA were 0.9977 and 0.9935, respectively. The sensitivity analysis of permeability illustrated that in all diesel concentrations and SDS percent-mass with increasing permeability the model resulted in more removal efficiency.

Keywords: numerical model; UTCHEM simulator; diesel; SDS; sensitivity analysis

1. Introduction

Hydrocarbons are one of the most important soil contaminations in the world, especially in oil-producing countries. Several methods exist for the remediation of organic and hydrocarbon contaminated soils. Soil washing is one of remediation methods which could be enhanced by using different kinds of surfactants. Because of capability in transforming pollutants, surfactants are used extensively in the soil washing process. Every surfactant molecule has a hydrophilic and a hydrophobic head and they are classified according to the charge in dissolved form in water with pH=7 (Asadollahfardi *et al*. 2013). Surfactants act as an interface between air and water. Surfactants enhance organic contaminant removal from soils by mobilization and solubilisation processes (Delshad *et al*. 1996, Vreysen and Maes 2005). Mobilization occurs at surfactant concentrations below the critical micelle concentration (CMC), while solubilization occurs at

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surfactant concentrations beyond the CMC of the surfactant (Vreysen and Maes 2005). Therefore, concentration is one of the most important characteristics of surfactants that affect many of the surfactant's properties including the formation of emulsions, foaming, and surface/interfacial tensions that are important for a proper choice of surfactant for soil washing (Asadollahfardi *et al*. 2013). Different types of surfactant exist, anionic, cationic, nonionic (neutral) and natural (bio-surfactant). Anionic surfactants are less adsorbed in soil compared with cationic surfactants and also ionic surfactants may also affect sediments (Asadollahfardi *et al*. 2013). Bio-surfactants as compared with artificial surfactants have superior surface activity and because they are produced from living organisms, they are less poisonous (Dyke *et al*. 1993, Francy *et al*. 1991).

Different studies based on experimental and numerical modeling have been conducted to understand the fundamentals and feasibility of the soil washing process. Deshpande *et al*. (1999) evaluated an experimental test of remediation of a petroleum contaminated soil with an anionic and non-anionic surfactant. They found that in low CMC the non-ionic surfactants were appropriate choices for remediation nevertheless in other cases anionic surfactant were the best choice due to the lower adsorption and higher solubilization potential they had. Vreysen and Maes (2005) performed the experimental process of leaching of diesel from self-contaminated sandy-loam soil. In their study, they used low concentration non-ionic surfactant solution of Tergitol NP-10 (10^{-6} to 10^{-3} mol/l). They stated without surfactant only 0.3% of diesel was leached from the soil and when they used surfactant, 20% of the diesel oil was removed. Zhu *et al*. (2005) focused on remediation of a diesel oil contaminated soil with aliphatic polyethenoxy ether (AEO_9) and sodium alcohol polyethoxylated ether sulfate (AES) as fluid flashing. They expressed that AEO_9 was more effective than AES in remediation of diesel oil from the contaminated soil. Urum *et al*. (2006) investigated the effect of different type of surfactant on crude oil removal from a contaminated soil in soil washing process. In their study, they used rhamnolipid, saponin and sodium dodecyl sulfate (SDS). They indicated that rhamnolipid and SDS had the same efficiency for crude oil removal and saponin was the less efficient than rhamnolipid and SDS. Khalladi *et al*. (2009) studied the washing process of a diesel contaminated soil with SDS. They focused on water flow rate and the contamination age in their study. They concluded that SDS was significant beyond a concentration of 8 mM and also oil washing process at a rate of 3.2 mL/min had removed 97% of the diesel fuel. Couto *et al*. (2009) conducted an experimental setup of diesel washing from contaminated sand which enhanced by using surfactant solutions, regular foams, and colloidal gas aphrons. They stated high percentages of diesel removal from contaminated sandy soils were achieved employing remediation with regular foams and aphrons and removal efficiency was significantly lower when only surfactant solution was employed as remediation fluid.

For the purpose of modeling, several studies have been presented. Delshad *et al*. (1996) described a three-dimensional, multicomponent, multiphase compositional model, UTCHEM for simulating the contaminant transport and surfactant enhanced remediation of non-aqueous phase liquid (NAPL) pollutant. Ouyang *et al*. (2002) investigated the formation and flow of micro-emulsions during surfactant flushing of a NAPL-contaminated soil with UTCHEM simulator based on finite difference method. They verified their simulation with their laboratory experimental datas. Christ *et al*. (2005) probed the effect of dimension reduction on simulated dense non-aqueous phase liquid (DNAPL) saturation distributions using a suite of three-dimensional, statistically homogeneous, non-uniform permeability fields that are representative of a natural formation. They stated reduction in dimensionality from 3-D to 2-D was shown to have a reasonably consistent impact on the predicted characteristics of the DNAPL source zone.

Bernardez *et al*. (2009) presented a numerical model based on UTCHEM simulator for diesel removal from the sand column with different micellar solutions containing the surfactant Hostapur SAS-60 (SAS), and two alcohols, n-butanol (nBuOH), and n-pentanol (n-PeOH). They stated the simulation results agree with experimental measurements and showed the entire residual diesel in the sand column was recovered after the downward injection of 5 pore volumes of the micellar solution. Lee (2010) focused on modeling NAPL transportation during surfactant-enhanced aquifer remediation in heterogeneous fields. For his simulation purpose, he used UTCHEM simulator and the results illustrated the long-term persistence of NAPL, the associated tailing off of effluent organic concentrations, and increases in the pressure drop at the injection well in highly heterogeneous fields. Asadollahfardi *et al*. (2013) modeled the treatment of a crude oil contaminated sand column with saponin as a biological surfactant. For purpose of modeling, they used UTCHEM simulator and found their results had good agreement with the experiment. Al-Shalabi *et al*. (2015) investigated the main mechanism and contributor to the low salinity water injection (LSWI) effect on oil recovery from carbonate rocks using the UTCHEM simulator. They indicated that LSWI has a negligible effect on water relative permeability.

This paper presented a numerical simulation of diesel removal from a contaminated sand column by using anionic surfactant SDS. For this purpose, we used UTCHEM simulator. We also carried out a sensitivity analysis on permeability to identify the effect of permeability on our model results.

2. Materials and methods

2.1 Experimental data

We used the data reported by Salehian (2007) for diesel removal of the sand column enhanced by (Sodium Dodesyl Sulfate) SDS as an anionic surfactant. The soil polluted by diesel with two different concentrations 10000 and 20000 ppm. 50 grams of diesel were added to 5 kilograms sandy soil to reach 5 kilograms sandy soil with 10000 ppm initial diesel concentration. Also, 100 grams of diesel were added to 5 kilograms sandy soil to reach 5 kilograms sandy soil with 20000 ppm initial diesel concentration. Water was added to the mixture of soil and diesel to gain uniform concentration of diesel in the sand. The soil was put out in a dry condition for 20 days to have a dry sandy soil by mentioning concentrations. Eventually, the high-performance liquid chromatography (HPLC) was carried out to calculate the diesel concentration in the soil to control the initial concentration of diesel. The sand column apparatus designed for the purpose of one-dimensional analysis with 50 cm height and 4 cm diameter; in addition, fluid injection was carried out in downward condition in experimental tests. Fig. 1 indicates the pilot experiment, which experimental soil washing process was carried out by this apparatus. Pore volume of soil obtained according to experimental measurement equal to 75 cm^3, also Table 1 indicates the sand properties and Fig. 2 presents grain-size distribution of the sand used in the present study. The experimental test was conducted until pore volume 10 and in pore volume 2, 4, 6, and 10 the concentrations of total petroleum hydrocarbon of effluent were measured. Soil washing experiment conducted under different concentration of SDS 0.1, 0.2, 0.3, 0.4 percent-mass for 10000 ppm and 20000 ppm diesel concentration. Table 2 indicates the anionic surfactant characteristics used in soil washing experiments. All the mentioned experimental work was carried out by Salehian (2007). We used the characteristics of the material and the results of his experimental work to simulate the removal of diesel from sandy soil polluted by diesel.

Fig. 1 Sand column apparatus of soil washing process (Salehian 2007)

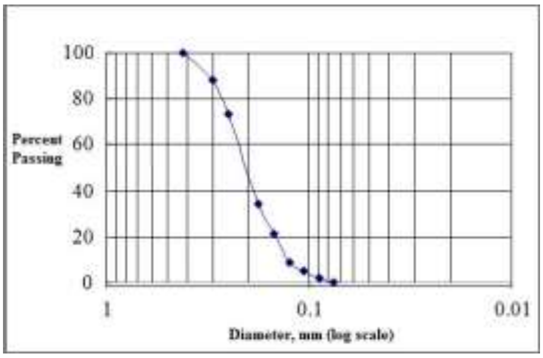
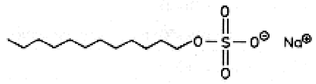


Fig. 2 Grain size distribution of sand which was used in the soil washing experiment

Table 1 Soil properties used in the current study reported by Salehian (2007)

Properties	Value
Soil Type	Sandy Soil
Porosity	0.4
Mobile Porosity	0.075
Void ratio	0.68
Permeability (cm/s)	3.2E-3
Dry density (g/cm^3)	1.55
pH	9
G_s	2.66
Conductivity (µs/s)	158
Organic Matter (%)	0

Table 2 Characteristics of the SDS

Surfactant Name	Dodecyl Sulfate Sodium Salt
Type of Surfactant	Anionic
Chemical Formula	$C_{12}H_{25}NaO_4S$-$C_{12}H_{25}OSO_2ONa$
CAS Number	151-21-3
CMC %	0.173-0.23
Molecular Weight	288.38 g/mol
Density	1.1 g/cm^3 (20℃)
pH	6-9 (10 g/l, H_2O, 20℃)
Structure	

2.2 Model description

To simulate the injection SDS in sand column polluted with diesel and potential dissolution and mobilization of the diesel, we used UTCHEM simulator version 9.82 (2000). UTCHEM model is a three-dimensional, multicomponent, multiphase compositional finite difference model for contaminant transport and surfactant-enhanced aquifer remediation (Ouyang *et al*. 2002). Originally UTCHEM developed as an enhanced oil recovery model, it has been adapted for using in environmental modeling (Qin *et al*. 2007, Schaerlaekens *et al*. 2006). The model solves pressure and energy balance equations; as well as, mass conservation equations for up to 19 components (including water, oil, air, surfactant, polymer, alcohol), which can be present in four pseudo-phases (aqueous, oleic, air, microemulsion) (Bernardez *et al*. 2009). The assumptions made while developing the flow equations are local thermodynamic equilibrium excluding for tracers and dissolution of the organic component, motionless solid phases, fairly compressible soil and fluids, stationary solid phases, Fickian dispersion, the ideal mixing, and Darcy's law (Asadollahfardi *et al*. 2013). The boundary conditions are no flow and no depressive flux across the impermeable boundaries (Delshad *et al*. 1996). Here instead of thorough formulation overview we presented the main mathematical formulation. Mass balance for component *k* is

$$\frac{\partial}{\partial t}\left(n\tilde{C}_k\rho_k\right) + \nabla\left(\sum_{l=1}^{n_p}\left[\rho_k\left(C_{kl}\mathrm{u}_l - \widetilde{D}_{kl}\right)\right]\right) = Q_k \tag{1}$$

Where n (-) is the porosity, $\tilde{C}_k$ (L^3/L^{3}PV) is the total volume of component *k* per unit pore volume including the adsorbed phases, C_{kl} (L^3/L^3) is the volume concentration of component *k* in phase *l*, ρ_k (M/L^3) is the density of pure component *k*, u_l (L/T) is the phase velocity, $\widetilde{D}_{kl}$ (L^2/T) is the dispersive flux of component *k* in phase *l*, n_p is the number of phases, and Q_k(L^3/T) is the source (+) or sink (-) term. The phase velocity u_l is given by Darcy's law (Brown *et al*. 1994)

$$\mathrm{u}_l = -\frac{k_{rl}K}{\mu_l}\left(\nabla P_l - \rho_l \mathrm{g}\nabla h\right) \tag{2}$$

Where k_{rl} (-) is the relative permeability, K (L^2) is the intrinsic permeability tensor, μ_l (ML^{-1}T^{-1}) is the viscosity, P_l (M/LT) is the phase pressure, ρ_l (M/L^3) is the density of phase *l*, g (L/T^2) is the acceleration of gravity, and h is the vertical coordinate. The dispersive flux is computed assuming that mechanical dispersion is Fickian

$$\overline{\mathrm{D}}_{kl} = -nS_l K_{kl} \cdot \nabla C_{kl} \quad (3)$$

Where S_l (-) is the saturation of phase l. Here K_{kl} is the dispersion tensor, which is given as (Bear 1979)

$$K_{klij} = \frac{D_{kl}}{\tau}\delta_{ij} + \frac{\alpha_T}{nS_l}|\mathrm{u}_l|\delta_{ij} + \frac{(\alpha_L - \alpha_T)}{nS_l} u_{li} u_{lj} / |\mathrm{u}_l| \quad (4)$$

Where D_{kl} (L^2/T) is the molecular diffusion coefficient of component *k* in phase *l*; τ (-) is the tortuosity; δ_{ij} is the Kronecker delta; α_L and α_T (L) are the longitudinal and transversal dispersivities respectively; u_{li} and u_{li} (L/T) are Darcy velocities of phase *l* in directions *i* and *j*, respectively. Pressure equation can be developed by substituting Darcy's law for a phase velocity term in the continuity equation. The resulting equation can be written explicitly in terms of aqueous phase (phase 1) pressure as

$$nC_t \frac{\partial P_1}{\partial t} + \nabla \cdot K \cdot \lambda_{rTc} \nabla P_1 = -\nabla \cdot \sum_{l=1}^{n_p} K \cdot \lambda_{rlc} \nabla h + \sum_{l=1}^{n_p} K \cdot \lambda_{rlc} \nabla P_{cl1} + \sum_{k=1}^{n_{cv}} Q_k \quad (5)$$

Where C_t $(ML^{-1}T^{-2})^{-1}$ is the total compressibility, K (L^2) is the permeability tensor, λ_{rlc} and λ_{rTc} $(ML^{-1}T^{-1})^{-1}$ are the relative and total mobilities, respectively, P ($ML^{-1}T^{-2}$) is the phase pressure and P_{cl1} ($ML^{-1}T^{-2}$) capillary pressure between the *l* and aqueous phases. UTCHEM first solves the pressure Eq. (5) implicitly and then the mass conservation Eq. (1) are solved explicitly for total concentration of each component (Bernardez *et al*. 2009, Roeder and Falta 2001). Table 3 indicates the parameters which we used to simulate the soil washing process by UTCHEM simulator.

2.2 Model efficiency

We calculated the coefficient of determination (R^2) and index of agreement (IA) between model predictions and experimental results (Krause *et al*. 2005, Saeidnia *et al*. 2016).

Table 3 Properties of fluids which was used in the UTCHEM simulator

Parameter	Value
Density (kg/m3)	
Water	1000
Diesel	831.6
Viscosity (mPa-s)	
Water	1
Diesel	2.21
Surfactant	4.08
Interfacial Tension (mN/N)	
Water-Diesel	27.7
Surfactant-Diesel	1
Contact Angle Between Water and Diesel ϴ	30

$$R^2 = \left(\frac{\sum_{i=1}^{n}(O_i-\bar{O})(F_i-\bar{F})}{\sqrt{\sum_{i=1}^{n}(O_i-\bar{O})^2}\sqrt{\sum_{i=1}^{n}(F_i-\bar{F})^2}} \right)^2 \quad (6)$$

$$IA = 1 - \frac{\sum_{i=1}^{n}(O_i-F_i)^2}{\sum_{i=1}^{n}(|F_i-\bar{O}|+|O_i-\bar{O}|)^2} \quad (7)$$

Where O is the observed value; $\bar{O}$ is the mean value of O; F is the predicted value and $\bar{F}$ is the mean value of F.

3. Results and discussion

Fig. 3 represents the UTCHEM model prediction for diesel removal from the sand column with an initial concentration of diesel of 10000 ppm at different percent-mass of surfactant. The trend of remediation process is ascending. The amount of diesel remediation of the different pore volumes is observable. As it is obvious pore volume 10 had the maximum remediation for flushing process in different percent-mass of surfactant. For 0.1, 0.2, 0.3 and 0.4 percent-mass of SDS (in pore volume 10) the remediation was 21.35, 24.12, 26.83 and 14.8 percent respectively. At 0.3 percent-mass of SDS, maximum diesel recovery was obtained. The highest remediation was for 0.3 percent-mass of SDS and when we increased the SDS concentration to 0.4 the remediation efficiency intensively decreased. The hydrocarbon remediation is a function of equilibrium between pollution concentrations, pH, and surfactant percent-mass. The specific percent-mass of surfactant could result in the maximum remediation, then more and less surfactant percent-mass has undesirable effects. Therefore, 0.4 percent mass of SDS is not the optimum amount of SDS for remediation process and redundant reactions between surfactant and diesel and soil particle occur and efficient remediation process is not accessible.

Fig. 4 demonstrates the diesel remediation in the 20000 ppm concentration from the sand column. Similar to 10000 ppm concentration of diesel (Fig. 3) maximum remediation was obtained in pore volume 10. In addition, the slopes were ascending up to pore volume 10 when

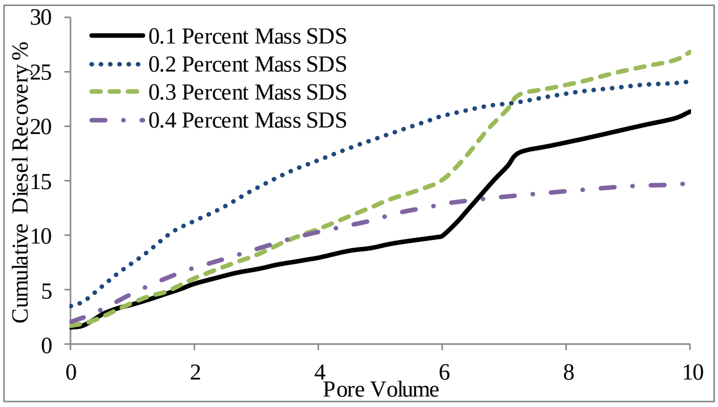


Fig. 3 Simulation results for diesel remediation with 10000 ppm concentration in soil column with different percent-mass of SDS

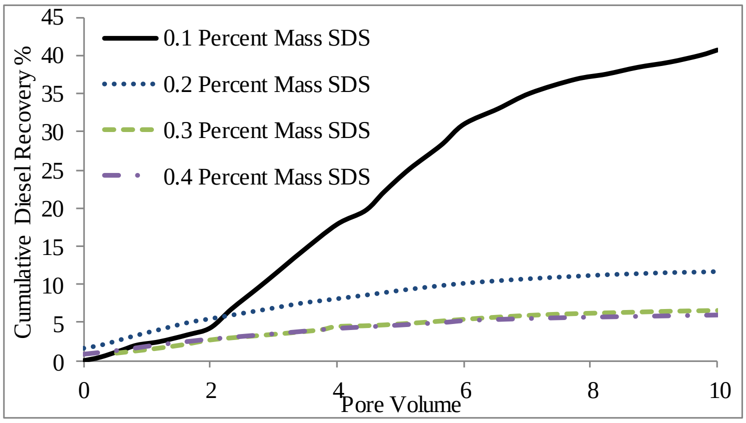


Fig. 4 Simulation results for diesel remediation with 20000 ppm concentration in soil column with different percent-mass of SDS

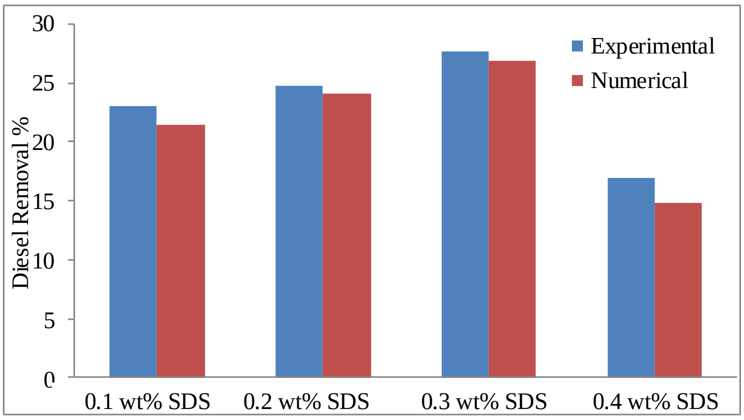


Fig. 5 Comparison between model prediction and experimental measurement for diesel removal (10000 ppm initial diesel concentration)

reached maximum diesel recovery. For 0.1, 0.2, 0.3 and 0.4 percent-mass of SDS the remediation is 40.7, 11.67, 6.64 and 5.97 percent respectively. The maximum efficiency for remediation occurred in 0.1 percent mass of SDS. Therefore, 0.1 percent-mass of SDS in 20000 ppm diesel initial concentration was the optimum amount and by increasing the SDS concentration the redundant reactions increased and the model tended low-efficiency result.

Figs. 5 and 6 illustrate the comparison between our numerical predictions, in pore volume 10, with experimental measurement for 10000 and 20000 ppm diesel concentration respectively. The R^2 and IA between the predicted and experiment data for 10000 ppm initial diesel concentration were 0.9952 and 0.9695 respectively. In addition, for 20000 ppm initial diesel concentration the R^2 and IA are 0.9977 and 0.9953, respectively. The mentioned parameters indicated a good agreement between our numerical simulation and experiment.

Solublization and mobilization mechanism in diesel recovery process depends on critical tie line which is the tangent to a miscibility curve at plait point. Above critical line mobilization

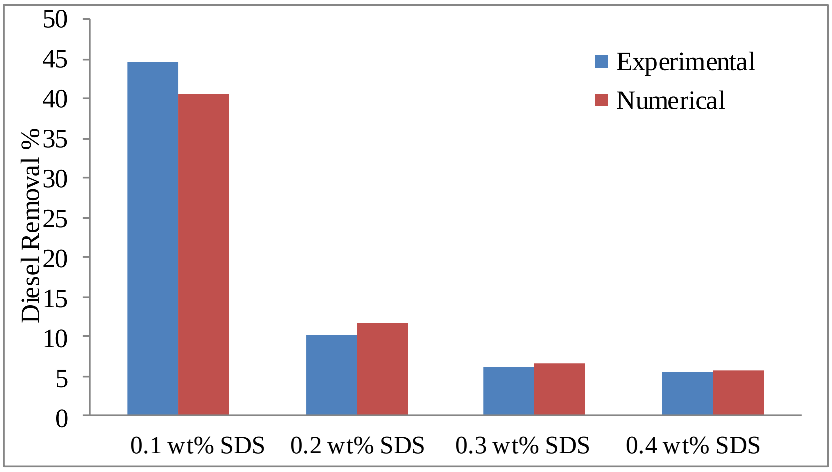


Fig. 6 Comparison between model prediction and experimental measurement for diesel removal (20000 ppm initial diesel concentration)

may be governed on the recovery process. Whereas, below the line multiple process coincide (St-Pierre *et al*. 2004).The forecasting of governing recovery process from the tie slopes and critical line in phase diagram is precise and can be replicated in a sand column (St-Pierre *et al*. 2004). We only simulated the process of diesel removal from contaminated sand column using UTCHEM simulator and did not carry out any experiment.

During the numerous mathematical simulation for hydrocarbon removal from contaminated soil based upon UTCHEM simulator, Bernardez *et al*. (2009) worked on modeling diesel removal from the sand column with UTCHEM. They conducted different experimental tests to examine the diesel removal from sand column under downward and upward injection using micellar sollutions. With the comparison between their numerical result and experiment, the R^2 and IA were more than 0.95 in all conditions.

4. Sensitivity analysis

We carried out the sensitivity analysis by changing the permeability to identify the role of permeability in model results. We increased or decreased the permeability value by 20 percent, while the other input parameters were kept unchanged, then the role of permeability in the prediction of the diesel removal of the sand column was investigated. For this purpose, we examined the effect of permeability on diesel removal in 10000 and 20000 ppm concentrations in different SDS' percent-mass. Figs. 7 and 8 illustrate the effect of permeability on 10000 ppm diesel with 0.3 percent-mass SDS and also on 20000 ppm diesel concentration with 0.1 percent-mass SDS in different pore volumes. In both figures, the trend is increasing and by increasing the permeability better removal efficiency was achieved. In Figs. 9 and 10 indicate the cumulative diesel removal in different percent-mass of SDS for 10000 and 20000 ppm diesel respectively. Figs. 9 and 10 demonstrate in all diesel and SDS concentrations with increasing permeability the removal efficiencies were increased.

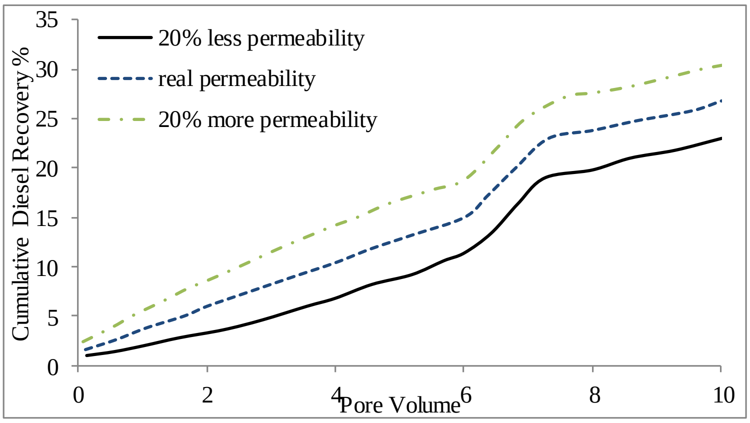


Fig. 7 Analysis of permeability effects on the diesel remediation from sand column (10000 ppm initial diesel concentration, 0.3 percent-mass SDS concentration)

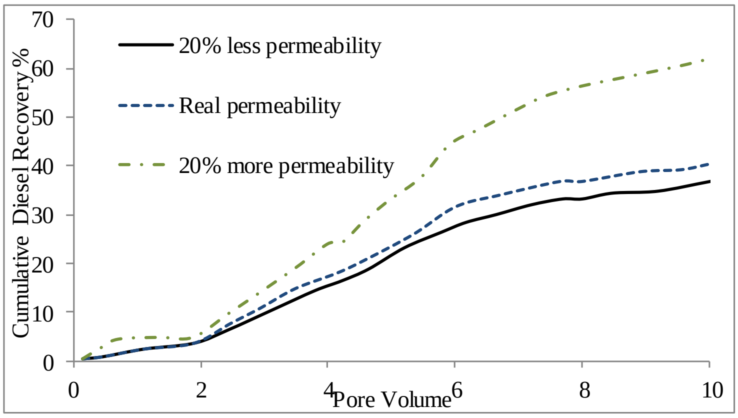


Fig. 8 Analysis of permeability effects on the diesel remediation from sand column (20000 ppm initial diesel concentration, 0.1 percent-mass SDS concentration)

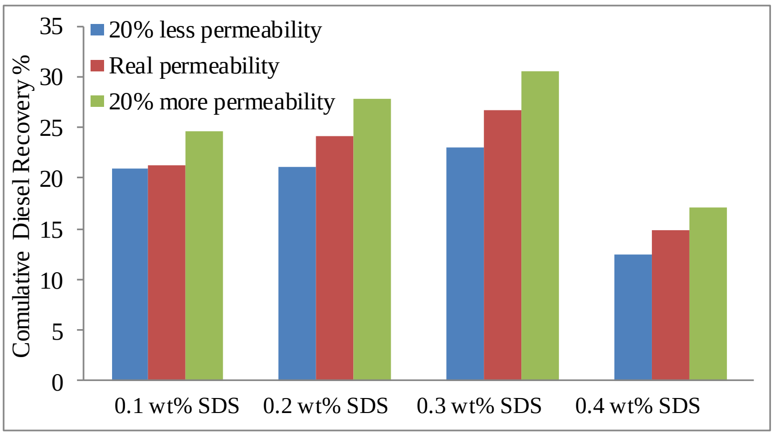


Fig. 9 Comparison between diesel removal efficiency of model by different permeability (with 10000 ppm initial diesel concentration)

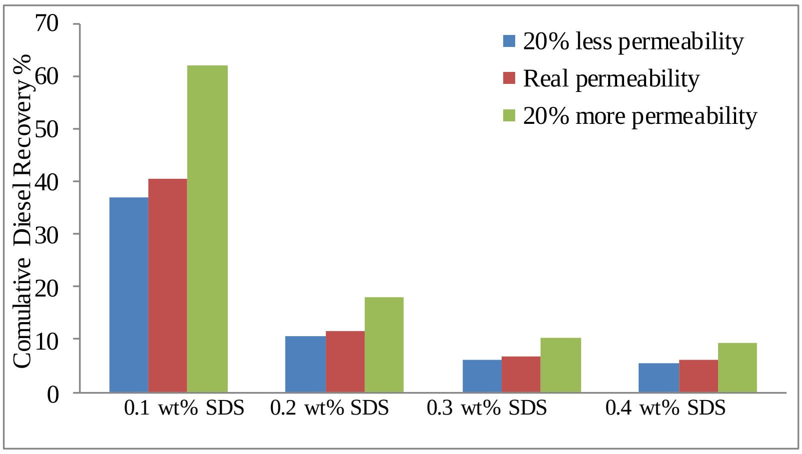


Fig. 10 Comparison between diesel removal efficiency of model by different permeability (with 20000 ppm initial diesel concentration)

5. Conclusions

Based on numerical analysis using UTCHEM simulators for modeling surfactant flushing to remediate diesel from sand column, we summarized the following conclusions:

1- Our model indicated 0.3 percent-mass of SDS results maximum diesel removal (26.83%) close to measurement (27.71%) in 10000 ppm initial diesel concentration. Furthermore, the calculation of R^2=0.9952 and IA=0. 9695 indicated model had a good agreement with the result.

2- For 20000 ppm diesel initial concentrations, our model indicated 0.1 percent-mass of SDS results maximum diesel removal (40.7%) close to experimental measurement (44.56%). In addition, the R^2 and IA calculation were 0.9977 and 0.9953 respectively, which indicate the accuracy of our model.

3- The role of soil permeability changing was evaluated to determine the effect of permeability on predicting the diesel removal from polluted soil. The results indicated that by increasing permeability, the predicting of diesel removal increased and by decreasing this parameter the model tended to low removal efficiency.

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