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ACKNOWLEDGEMENTS. We thank DST, New Delhi and the IISc Centre for Excellence in Nanoelectronics for funding. INI Centre at IISc is acknowledged for TEM facilities.

Received 17 July 2009; accepted 21 August 2009

Adsorption of non-petroleum base surfactant on reservoir rock

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Surfactant loss due to adsorption on the reservoir rock is one of the major concerns in enhanced oil recovery (EOR) processes. It weakens the effectiveness of the injected slug in reducing oil–water interfacial tension (IFT) and makes the process uneconomical. In this study, an attempt is made to investigate the adsorption of Na-lignosulphonate onto the porous media of Oil India Limited (OIL) petroleum reservoir rocks. The data were interpreted from the well-known models and it was found that the Langmuir model is a good fit for the pH and brine data over the entire range of variables. Adsorption increases with NaCl concentration and decreases with increase in pH.

Keywords: Adsorption, brine, Na-lignosulphonate, pH, reservoir rock.

SURFACTANT molecules adsorb well to solid interfaces such as the porous media found in petroleum reservoirs. The adsorbed surfactant layer represents both an additional resistance to flow as well as loss of surfactant properties and is therefore, of fundamental importance in the enhanced oil recovery (EOR) process that involves the flow of surfactant solution through porous media.

According to Austad and Milner¹, chemical flooding of oil reservoirs is one of the most successful methods to enhance oil recovery from depleted reservoirs at low pressure. However, on a volume basis, the greatest potential usage would be surfactant flooding for EOR². The 1970s and 1980s were active periods for research on surfactants for EOR and a large number of patents were issued.

Surfactant adsorption in the flow of surfactant solutions through porous media is usually accompanied by a variety of additional complex phenomena. Viscoelasticity has contributed to the increase in flow resistance, particularly at high-flow velocities^{3–6}. Consequently, the effect of adsorption on permeability reduction was often not clear. In addition, phenomena such as mechanical entrapment and hydrodynamically induced retention obscure the role adsorption plays in surfactant retention and mobility reduction^{7–12}. Much effort has been devoted to qualify the contribution of surfactant adsorption to mobility reduction and surfactant retention in both consolidated and unconsolidated porous media. The existing studies, however, do not quantify the resistance of the adsorbed surfactant layer to the flow of surfactant solutions.

Success or failure of a surfactant flood may depend on the degree of retention of surfactants during EOR operation and one of the possible mechanisms of the surfactant retention was the solid–liquid interface. Several papers dealing with the adsorption of commercially available surfactants have been published^{13–18}, but meaningful comparison of reported data was quite difficult because surfactants of various degrees of purity have been used.

Zaitoun *et al.*¹⁹ conducted a series of experiments on surfactant screening and evaluation for surfactant flooding in the Chihuido de la Sierra Negra field in Argentina. They developed a new anionic surfactant that provides good solubility in high salinities and low interfacial tension at low concentrations. Gogoi²⁰ described that sodium lignosulphonate formed as a waste in paper industries had the potential to be used in EOR.

Adsorption minimizes the loss of the high equivalent weight fraction that was most efficient in lowering the interfacial tension (IFT)²¹. Adsorption of surfactants considered for EOR applications have been studied extensively over the last few years^{22–27} and it has convincingly shown that it is possible to develop surfactant systems which displace oil from porous media almost completely when used in large quantities. Effective oil recovery by surfactant was not a question of technical feasibility but rather a question of economics. Clays in the rock have high surface area that can affect the surfactant flood in many ways. Bernard²⁸ has suggested that by ion-exchange, divalent ions were transferred to the surfactant solution from the clays resulting in precipitation of surfactant and loss of surfactant in the displacing fluid. It was found that for a particular sulphonate, the minimum tension between a salt solution containing the sul-

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phonate and an intermediate-paraffinic crude oil would occur at high NaCl concentrations and lower surfactant concentrations than for typical naphthenic crude²⁹.

Accordingly, a study on adsorption of the surfactant on to the porous rocks was carried out to assess its effect on EOR. This study was confined to adsorption equilibrium and the results were interpreted from the well-known theoretical models.

The adsorbents used for the study were porous media from the producing wells of NH₂ (depth 3856–3859 m) and BH₂ (3837–3847 m) of Oil India Limited (OIL), Duliajan. The adsorbate for the study was 0.34% Black Liquor (BL) in distilled water (DW) at different pH solutions. The main constituent of BL is Na-lignosulphonate [C₁₀H₁₄N₂Na₂O₈2H₂O] with a molecular weight of 372.

The absorbance of Na-lignosulphonate was examined in UV-VIS spectroscopy. A UV-VIS spectrum was collected from 190 to 350 nm, using a UV-VIS Spektralphotometer, SPECORD 205 P, Analytic Jena AG calibrated at 190–300 nm wavelength. Due to the high concentration of BL it was diluted by mixing 17 µl of BL with 4983 µl distilled water to make a solution of volume 5000 µl. The dilution factor was 5000/17, i.e. 294.12. This starting solution was further diluted to provide a series of standard solutions. UV-VIS spectrum of the BL sample was taken and λ_{max} the wavelength of the peak in the UV-VIS spectrum was recorded. Thanh³⁰ recorded λ_{max} of 198 nm, and in this study it was found to be 194 nm; this difference can be attributed to instrumental error. The absorbance at the reference wavelength for each standard solution was measured and recorded. In an excel worksheet, the concentration of each standard solution vs the absorbance was plotted to reveal the linear region.

Different amounts of the adsorbents (porous media) were taken in 15 ml graduated centrifuge tubes, to it was added 0.34% BL in DW. For the determination of adsorption equilibrium, different amounts of the porous media were weighed in amounts of 0.5–5 g and taken in 15 ml graduated centrifuge tubes. 10 ml of 0.34% BL in DW, saline concentrations of 1000, 2000, 3000 and 4000 ppm and different pH solutions of 4.8, 5.7, 7.1 and 8.3 were added; one solution at a time to the tubes containing different amounts of the porous media.

To obtain the equilibrium isotherms, the tubes were shaken in a Microshaker (Dynatech Product: Catalogue number 0029540800, Dynatech Laboratories Insurance, Chantilly, Virginia, USA) and in Coulter Mixer (made in England) for 24 h. After achieving equilibrium, the mixture was allowed to settle and the supernatant liquor was decanted and collected in test tubes. The test samples were centrifuged in Sorvall, RT6000B and Refrigerated Centrifuge (DUPONT) at 4000 rpm for 5 min and then filtered using filter paper (Whatman 42). By means of a micropipette of Eppendorf Repeater, USA, 4950 µl of DW or different pH solutions were taken in test tubes wherein 50 µl of the centrifuged liquor was added by

transferpipette to make a solution of 5000 µl. The solutions were transferred one by one to a curvette of 10 mm path length and mixed in a cyclomixer supplied by Remi, India. The solutions were analysed by the UV-VIS Spectrophotometer calibrated at 194 nm against the standard solution (DW or different pH solutions) in the UV range of 180–400 nm. Hence, knowing the measured absorbance and using calibration curve, the amounts of BL adsorbed per gram of porous media q_e (m mol g⁻¹) were calculated and the adsorption equilibriums estimated from a material balance. The data on adsorption were interpreted from Langmuir, Freundlich and Redlich–Peterson isotherms. The absorbance values of Na-lignosulphonate is given in Table 1.

The spectrophotometer absorbance of Na-lignosulphonate in DW, and in varying pH, and salinity solutions (nm) on porous media are given in Tables 2–4. The values of the experiments are plotted in Figures 1–3. Adsorption isotherm of Na-lignosulphonate on different porous media is shown in Figure 1. The amount of Na-lignosulphonate in millimol adsorbed in grams on the solid phase (i.e. the adsorbent or porous media) was more on NH₂ and BH₂ sand-pack. Adsorption isotherms obtained for DW and at different ppm of NaCl are shown in Figure 2. Addition of NaCl to the system is found to increase the solid phase concentration, which means that adsorption increases with increase in NaCl concentration. Figure 3 shows that the adsorption of Na-lignosulphonate on porous media decreases with increase in solution pH.

Table 1. Absorbance of Na-lignosulphonate

Serial no.	Absorbance (nm)
1	0.148
2	0.309
3	0.353
4	0.457
5	0.492
6	0.621
7	0.751
8	0.889
9	0.946
10	0.991

Table 2. Absorbance of Na-lignosulphonate on porous media

Porous medium	BH ₂	NH ₂
Absorbance (A) (nm)	2.3452	1.7982
	0.7786	0.4301
	0.4398	0.2348
	0.2834	0.1518
	0.2567	0.1072
	0.1909	0.0933
	0.2757	0.0863
	0.2129	0.08097
	0.1841	0.07941
	0.1723	0.07823

The adsorption equilibria are interpreted from Langmuir, Freundlich and Redlich–Peterson isotherms. The parameters are estimated by a nonlinear regression analysis using Levenberg–Marquardt’s method^{31–34} and computation is carried out in Pentium PC (Digital) using Numerical Recipes in C+. It appeared that Langmuir model provides the most satisfactory representation of the data almost at all pH and salinity values studied. The estimated parameters and correlation coefficients are shown in Table 5. It was found that *R*-square is close to unity and saturation value is high in the case of Langmuir model. So, the best fit model is the Langmuir model.

Table 3. Absorbance of Na-lignosulphonate with change in liquid phase salinity on NH₂

Salinity (ppm)	DW	1000	2000	3000	4000
Absorbance (A)	1.7982	1.5992	1.3448	1.2008	1.5992
(nm)	0.4301	0.4108	0.3745	0.2898	0.4108
	0.2348	0.2076	0.1809	0.1565	0.2076
	0.1518	0.1418	0.1511	0.1222	0.1418
	0.1072	0.0982	0.1482	0.1117	0.0982
	0.0933	0.09245	0.1345	0.1099	0.09245
	0.0863	0.08255	0.1284	0.1097	0.08255
	0.08097	0.07805	0.1192	0.0919	0.07805
	0.07941	0.07211	0.1191	0.0996	0.07211
	0.07823	0.07564	0.1104	0.0994	0.07564

Table 4. Absorbance of Na-lignosulphonate with change in liquid phase pH on BH₂

pH	5.1	5.3	7.1	8.3
Absorbance (A)	2.5083	2.9062	3.9862	4.5603
(nm)	0.6465	0.8374	1.2094	1.2012
	0.3098	0.4312	0.5722	0.5961
	0.2234	0.3667	0.3885	0.4385
	0.1907	0.2762	0.3576	0.3896
	0.1909	0.2355	0.2972	0.3272
	0.1808	0.2187	0.2341	0.3165
	0.1332	0.2098	0.2218	0.3092
	0.1241	0.2002	0.2587	0.2977
	0.1223	0.1904	0.2184	0.2849

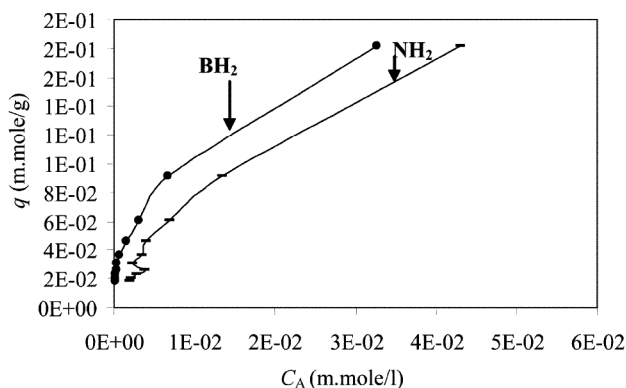


Figure 1. Adsorption isotherm of Na-lignosulphonate on different porous media (NH₂, BH₂).

This is perhaps the first demonstration of a favourable isotherm for the adsorption of Na-lignosulphonate on porous media obtained from OIL petroleum reservoir. The results of adsorption can be interpreted well by the well-known Langmuir model. It is observed that with the increase in pH, the adsorption of anionic sulphonate (Na-lignosulphonate) on the porous media decreases and as salinity of the aqueous phase increases the adsorption increases. The applicability of the Langmuir isotherm suggests the monolayer coverage of the adsorbate on the surface of the porous media.

Adsorption of surfactants on reservoir rock is an important consideration in surfactant flooding formulations. It is well known that high equivalent molecular weight surfactants are absorbed preferentially on the rock surface while the lower equivalent molecular weight surfactants show very little adsorption. Although the high equivalent weight surfactants are responsible for most IFT reductions, their losses decrease the surfactant slug’s ability to displace the residual oil in the reservoir. Solutions which reduce adsorption are added as sacrificial agents into the

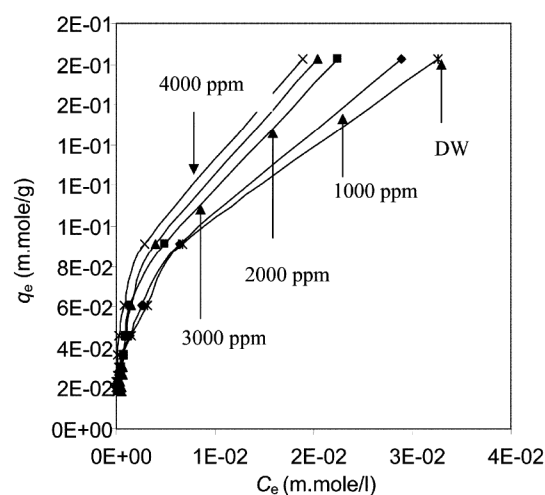


Figure 2. Adsorption isotherm of Na-lignosulphonate on NH₂ in different brine concentrations.

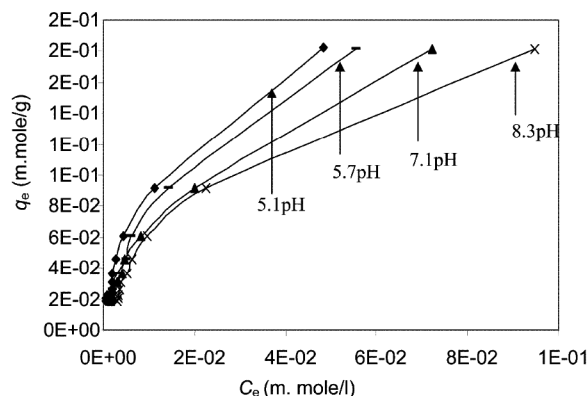


Figure 3. Adsorption isotherm of Na-lignosulphonate on BH₂ as a function of pH.

Table 5. Parameters of adsorption isotherms of Na-lignosulphonate

System	Langmuir model			Freundlich model			Redlich–Peterson model			
	C_1	K_d	R -square	K_f	n	R -square	K_R	P_1	P_2	R -square
Na-lignosulphonate on BH ₂	0.301	35.010	0.994	1.479	0.662	0.852	-1.591	2.482	0.727	0.933
Na-lignosulphonate on NH ₂	0.208	160.693	0.955	0.771	0.425	0.969	1.205	0.497	0.365	0.920
Na-lignosulphonate in 1000 ppm brine on NH ₂	0.209	176.594	0.985	0.777	0.414	0.954	2.470	0.315	0.292	0.878
Na-lignosulphonate in 2000 ppm brine on NH ₂	0.205	250.290	0.981	1.082	0.467	0.946	-3.128	2.004	0.546	0.907
Na-lignosulphonate in 3000 ppm brine on NH ₂	0.218	222.603	0.993	1.301	0.500	0.937	-11.202	5.818	0.682	0.903
Na-lignosulphonate in 4000 ppm brine on NH ₂	0.180	695.219	0.939	0.751	0.357	0.944	0.558	0.673	0.344	0.884
Na-lignosulphonate in 5.1 pH solution on BH ₂	0.222	81.881	0.988	0.852	0.507	0.926	-2.432	1.997	0.627	0.867
Na-lignosulphonate in 5.7 pH solution on BH ₂	0.241	51.707	0.995	0.924	0.558	0.977	-2.239	2.165	0.678	0.879
Na-lignosulphonate in 7.1 pH solution on BH ₂	0.235	41.632	0.989	0.770	0.549	0.967	-0.624	1.097	0.604	0.888
Na-lignosulphonate in 8.3 pH solution on BH ₂	0.237	33.017	0.996	0.677	0.553	0.942	12.281	2.187	0.727	0.895

K_d , Langmuir constant (1/m mol); K_f , Parameter of Freundlich model; C_1 , Parameter of Langmuir model, adsorption saturation capacity based on solid volume (m mol/g); C_e , Equilibrium liquid phase concentration; q_e , Amount of Na-lignosulphonate adsorbed at equilibrium; n , Parameter of Freundlich model; K_R, P_1, P_2 , Parameters of Redlich–Peterson model.

formulation or as pre-flood into the reservoir. It is also possible to adjust the molecular spectrum (equivalent molecular weight distribution) of the formulation to minimize the adsorption.

In addition to the technical feasibility of EOR surfactant flooding, economic feasibility must also be determined; however, the economic feasibility depends on the complex factors such as oil prices, international economies, and the cost of surfactants. But simply waiting for oil prices to increase will not necessarily make EOR economically feasible³⁵. Generally, the surfactant is the most expensive item in the EOR surfactant flooding process. The expenditure includes both the initial investment in purchasing the surfactant as well as the cost of replacing the surfactant which has been depleted by adsorption. Since Na-lignosulphonate taken in this study is synthesized from paper industry waste, it is cheap and easily available; so the question of cost of surfactant does not arise. This adsorption study provides a new tool for understanding the effectiveness of Na-lignosulphonate in EOR. Current work is directed towards the extension of this experimental method to the analysis of surfactant adsorption and retention in the porous media of OIL petroleum reservoir.

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ACKNOWLEDGEMENT. I thank my guide Prof. K. D. Gogoi for helpful discussions and suggestions.

Received 26 March 2009; revised accepted 1 September 2009

Carbon sequestration with special reference to agroforestry in cold deserts of Ladakh

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Global warming risks from emissions of greenhouse gases (GHGs) by anthropogenic activities have increased the need for the identification of ecosystems with high carbon sink capacity as an alternative mitigation strategy of terrestrial carbon sequestration.

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The agroforestry sector has received recent attention for its enormous potential carbon pools that reduce carbon emissions to the atmosphere. The Nubra Valley (Trans-Himalayan region) is covered with more than 575,000 agroforestry plantations (willow and poplar). These species have been found to sequester more than 75,000 tonnes of carbon. Every year these plantations are contributing 400 tonnes of leaf litter to the ground, which is one of the best sources of soil organic carbon. This communication attempts to suggest some appropriate management practices to improve soil organic carbon pools by analysing available information of agroforestry plants in the Nubra Valley by quantification of biomass contribution by willow and poplar species within the valley and CO₂ sequestration rates by the living biomass.

Keywords: Agroforestry, carbon sequestration, carbon sinks, cold desert lands, global warming, Trans-Himalayas.

THERE is much concern that the increasing concentration of greenhouse gases (GHGs) in general, and carbon dioxide in particular, in the atmosphere contributes to global warming by trapping long-wave radiation reflected from the earth's surface¹. Carbon sequestration, i.e. capturing and securing carbon that would otherwise be emitted and remain in the atmosphere might be a suitable alternative to control atmospheric emission of carbon. Plants capture CO₂ during photosynthesis and transform it to sugar and subsequently to dead organic matter. As the trees grow, they sequester carbon in their tissues, and as the amount of tree biomass increases, the increase in atmospheric CO₂ is mitigated². The ability of these plantations to sequester carbon has received renewed interest, because carbon sequestration projects in developing nations could receive investments from companies and governments wishing to offset their emissions of GHGs through the clean development mechanism (CDM) according to the Kyoto Protocol³. Following the Kyoto Protocol, many studies on carbon sequestration have been carried out in African countries. However, there is a dearth of studies in Asian countries, particularly in high altitude with special reference to agroforestry. Keeping this in view, the present study was carried out extensively for a number of agroforestry plantations (willow and poplar) and their sequestered carbon stock in the Nubra Valley, Trans-Himalayan cold desert.

The Nubra Valley in Ladakh comprises the Valley of Shyok River from its acute-angled bend down to its confluence with Nubra and further towards Indus (Figure 1) with Khardung-La pass (5605 m asl) as gate way of the valley. The area lies between two great mountain ranges, i.e. Ladakh (on the south) and Karakoram (on the north) with approximately 34°15'45"–35°30'N lat. and 76°55'–78°05'E long. There is a great variation in altitude and it ranges approximately between 2700 and 6000 m asl. The