Research Journal of Applied Sciences, Engineering and Technology 6(17): 3209-3213, 2013
ISSN: 2040-7459; e-ISSN: 2040-7467

Submitted: January 15, 2013 Accepted: February 07, 2013 Published: September 20, 2013

The Study of PAM Adsorption and Migration Disciplines in Daqing Characteristics Soil

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Abstract: Through static and dynamic adsorption experiments, the Polyacrylamide (PAM) adsorption and migration behavior in two characteristics soils such as chernozem and saline soil in Daqing were studied. The migration of static adsorption of PAM in two different soils and different flow amount of PAM adsorption in the chernozem adsorption column were discussed. Experiments showed that the PAM adsorption equilibrium time in the chernozem soil was longer than in the saline soil and the adsorption amount in the chernozem soil was greater than in the saline soil. PAM was easy to reach saturation when the flow amount was large in the dynamic adsorption experiment. The introduction of inert tracer bromide ion simulated the migration of the PAM through the soil adsorption column in dynamic adsorption experiments, which provided datas for building the numerical model of PAM migration rule in soil.

Keywords: Dynamic adsorption, inert tracer bromide ion, PAM, static adsorption

INTRODUCTION

Environmental contamination by petroleum and its derivatives is a serious worldwide problem (Brahim *et al*., 2003). The oil an important industrial raw material and the people's life necessities occupies an important position in the development of world economy. But in the oil field production process, a lot of poisonous and harmful chemical substance caused great pollution on oil field soil that directly threat the ecological environment and human health (Lentz, 2003). Soil is the material base and environmental condition of human survival and development. From the view of environmental protection, soil is the ultimate receptor of pollutants. Soil pollution will cause and promote water, air, biological pollution further, so it is the important factors that the pollutants are intaked by human body thereby affect human body health. In the oil and gas field development and construction process, the waste water, gas and solid directly or indirectly emission into the soil environment which influence soil ecosystem. Once soil was polluted it will be difficult to control (Sirjacobs *et al*., 2000; Kazemi and Morris, 1998; Thomas *et al*., 2004; John and Sanders, 1999; Seifert and Peter, 2007). Avoiding the oil pollution and its harm to the Daqing city who takes the oil as the main industrial is extremely important. Therefore, studying the ways that the pollutants entering into the soil, the pollutants migration characteristics and the pollution range and also grasping the prevention and controlling method of soil pollution are helpful for the environmental protection and management of Daqing district.

At present, Polyacrylamide (PAM) as one of main substance in polymer flooding has large amount of applications (Comegna *et al*., 2001). Due to process, technology and some other reasons, PAM leaks in the process of production, transportation and application. A lot of PAM in environment causes a series of harm to the ecological environment (Kildsgaard and Engesgaard, 2002; Jyotsna *et al*., 2005; Zoi *et al*., 2008; Hojberg *et al*., 2005; Levy and Miller, 1999; Ho *et al*., 2002; Shokrollah and Abbasi, 2011; Sepaskhah and Mahdi-Hosseinabadi, 2008). However the impact, PAM in the important type of Daqing soil, is significant research to protect the soil ecological environment and sustainable development of Daqing Oilfield. Domestic and abroad related to PAM in soil environment behavior research is very few, especially the migration behavior of PAM in characteristics of oil pollution soil has no research at this aspect (Sepaskhah, 2006; Santos and Serralheiro, 2002; Sojka, 2000; Sojka *et al*., 2006). Therefore, the PAM migration and transformation in Daqing region important type soil has the important realistic significance to protect ecological environment. Investigating PAM static adsorption experiment in Daqing characteristics soil chernozem and saline soil and dynamic adsorption experiment adding bromide ion as a tracer establishes adsorption isotherm model.

EXPERIMENTAL

Materials and method: The chernozem came from the 0-15 cm surface soil of machinery science and engineering college of Northeast Petroleum University

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Table 1: The soil physical and chemical properties

Sample	Soil type	pH	Porosity	Moisture	Organic
1#	Chernozem	7.8	45%	2.2%	4.31%
2#	Saline Soil	8.6	43%	4.3%	6.37%

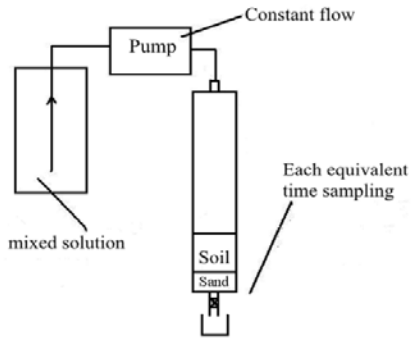

Fig. 1: Dynamic adsorption experiment device

(1#), the saline soil came from the 0-15 cm surface soil in the oil engineering institute of Northeast Petroleum University (2#). Removing the impurities, then according to the national standard determination of soil samples determined the particular physical and chemical properties displayed in Table 1.

Adsorption isotherm measurement: This experiment used maceration method that put solid adsorbent into a certain amount of known concentration of PAM solution to achieve adsorption equilibrium. At temperature 20ºC five conical flasks were respectively added the same volume solution, that the PAM mass fraction were 200, 400, 600, 800 and 1000 mg/L, respectively then put 1 mg soil sample in each conical flask. Constantly stirring, adsorption time 2, 4, 6, 8, 10, 12, 14, 16, 18 and 20 h, respectively. Reach conditions; take the supernatant liquid centrifugal 5 min (2200 r/min), extraction middle layer clear liquid using turbidity method determination of PAM mass concentration. Each group of experiments do three parallel sample, parallel sample determination results between the less than 3% when take its mean value. According to the above method to determine PAM in chernozem and saline soil adsorption equilibrium concentration and adsorption time.

Dynamic migration curve measurement: Configuration 300 mg/L of PAM and 120 mg/L of KBr mixed solution 20L. Dynamic adsorption experiment using soil was chernozem. Before the experiment, First step, determine the saturated water volume of soil in the adsorption column. The experiment measured the certain height of soil adsorption column, can contain the volume of water was half of the volume of soil. So soil saturated water volume was 50% soil volume. Second step, dynamic adsorption device installation was complete show that in Fig. 1, mixed solution flow into adsorption column. When the adsorption column below has first drops of solution outflow, continuously catch the same volume of liquid by a vial. Turbidity method and phenol red spectrophotometry respectively detection the PAM solution concentration and the bromide ion concentration. Third step, examine concentration and mixed solution concentration same then used clear water flowing into adsorption column. Continue continuously catch the same volume of liquid by a vial. Continue to examine concentration until PAM concentration was zero.

RESULTS AND DISCUSSION

PAM in different soil static adsorption situation: The data process from the soil static adsorption amount show that, when the equilibrium concentration reach ρ_e, PAM which was absorbed by 1mg soil was V (ρ_i - ρ_e) mg, this defined as Eq. (1):

$$\frac{V(\rho_i - \rho_e)}{m} = \frac{x}{m} \tag{1}$$

x : PAM adsorption capacity
ρ_i : PAM for initial concentration

For the same kind of soil speaking, experiment show that x/m was function of equilibrium concentration ρ_e and T, get Eq. (2):

$$\frac{x}{m} = f(\rho_e, T) \tag{2}$$

When the Temperature (T) was constant, the x/m was only function of ρ_e, get Eq. (3):

$$\frac{x}{m} = f'(\rho_e) \tag{3}$$

Using the equilibrium concentration as x axis and adsorption ratio as y axis, we can draw the adsorption isotherm. As shown in Fig. 2 that the PAM in different soil in the adsorption isotherm. In soil adsorption and migration of PAM was affect the environment behavior important mechanism. Through the static adsorption experiment, when arriving at adsorption equilibrium, we detect the adsorption capacity and adsorption time of PAM in chernozem and saline soil. In chernozem static adsorption experiments of PAM the initial concentration was 200, 400, 600, 800 and 1000 mg/L, respectively to achieve adsorption equilibrium that adsorption time was 16, 20, 24, 22 and 24 h, respectively equilibrium concentration was 49, 121, 264, 442 and 642 mg/L, respectively. In saline soil

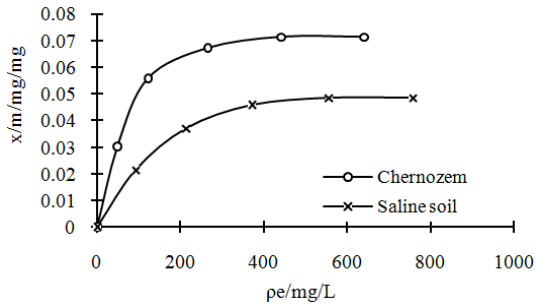

Fig. 2: PAM in two kinds of soil of the static adsorption isotherm

static adsorption experiments of PAM the initial concentration was 200, 400, 600, 800 and 1000 mg/L to achieve adsorption equilibrium that adsorption time was 8, 6, 4, 6 and 6 h, equilibrium concentration was 92, 214, 371, 557 and 757 mg/L, respectively.

On Fig. 2, through the analysis of data, PAM in chernozem and saline soil were all showed classical adsorption behavior, but also showed differences. Similarities were in two kinds of soil PAM adsorption quantity ratio increase in all along with the increasing balance of concentration. It was a significant increasing in the period of time. Increased equilibrium concentration, the PAM adsorption quantity ratio increase amplitude decreases. It shows that adsorption ratio was a certain value when different concentration of PAM in soil saturated adsorption. Difference that in chernozem the different concentration PAM saturated adsorption quantity ratio were greater than in saline soil PAM saturated adsorption quantity ratio, in chernozem the same concentration PAM saturated adsorption of equilibrium concentration were smaller than in saline soil PAM saturated adsorption of equilibrium concentration. According to a large number of studies suggest that PAM in soil adsorption force mainly the adsorbent surface between the Van Der Waals' force, hydrogen bonding force, surface metal ion and PAM molecules of rich electronic group was formed the coordination bond, in PAM of -COOH and adsorbent of Al^{3+} electrostatic force of attraction. The force blocking adsorption was the electrostatic repulsive force between the anion coming from adsorbent surface and negative charge on the PAM molecules, the balance of these two forces will affect the adsorption amount of PAM in soil. Experiment found in the study of mass concentration range PAM showed very classical adsorption behavior. With adsorption quantity ratio increasing the polymer quality concentration has increased dramatically, adsorption isotherm basic accord with Langmuir type. When in the polymer quantity concentration higher than 220 mg/L, emerge a platform area, this suggests that PAM in clay surface adsorption has reached saturation. The experiment shows that PAM in chernozem and saline soil of saturated adsorption ratio in turn was 0.072 and 0.049 mg/mg, respectively. According to the two adsorption curve change trend, the adsorption data using Langmuir isothermal adsorption equation regression fitting, found PAM in chernozem and saline soil the adsorption curve and the Langmuir isothermal adsorption regression curve has good anastomotic. PAM in chernozem adsorption the linear correlation coefficient $R^2 = 0.9963$, fitting Eq. (4):

$$y=\frac{0.00128\rho_e}{1+0.0016\rho_e} \tag{4}$$

PAM in saline soil adsorption the linear correlation coefficient $R^2 = 0.9916$, fitting Eq. (5):

$$y=\frac{0.000472\rho_e}{1+0.008\rho_e} \tag{5}$$

y : Adsorption Ratio, Unit-mg/mg

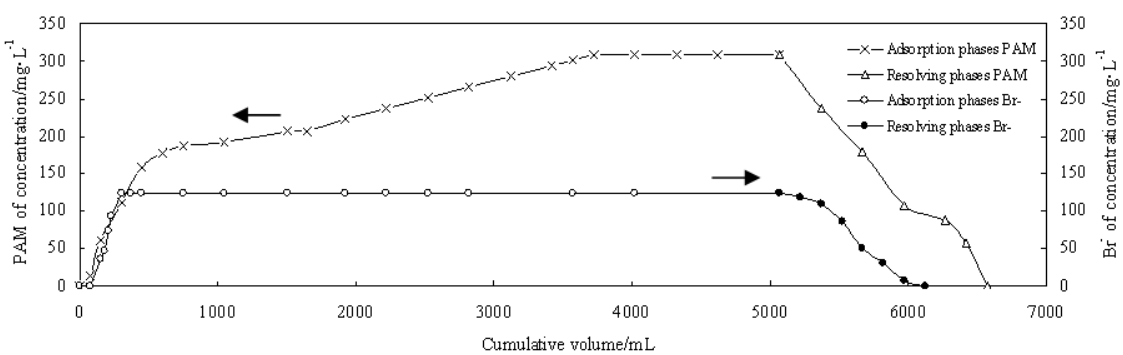

Fig. 3: PAM flow rate 10 mL/min in soil column adsorption migration discipline

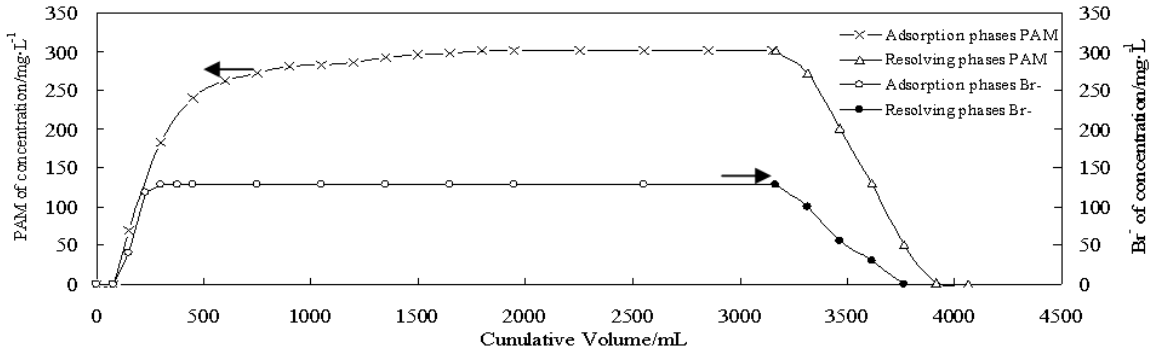

Fig. 4: PAM flow rate 50 mL/min in soil column adsorption migration discipline

PAM in same soil dynamic migration situation: PAM quality concentration was 300 mg/L and Br^- quality concentration was 120 mg/L, mixed solution flow rate were 10 and 20 mL/min, PAM in saline soil of dynamic migration disciplines. The experimental results are shown in Fig. 3 and 4.

The Fig. 3 shows that in same kind of soil, the initial quality concentration of PAM solution was 300 mg/L, the flow rate was 10 mL/min, need 3570 mL of accumulative total solution volume make soil adsorption column reach saturated adsorption, 357 min to achieve the same effect. Used water leaching has reached saturation soil adsorption column, need 1500 mL water make soil adsorption column of PAM concentration was zero. The Fig. 4 shows that in solution flow rate was 50 mL/min conditions, need 3150 mL of accumulative total solution volume make soil adsorption column reach saturated adsorption, 63 min to achieve the same effect. With water leaching has reached saturation soil adsorption column, need 900 mL water make soil adsorption column of PAM concentration was zero, as cleaning fully. When the flow rate was 10 mL/min, PAM quality concentration with total solution volume increased slowly growth, the cumulative volume reached 3570 mL and adsorption column of PAM quality concentration was same to mixed solution concentration. In the following sampling points, the concentration of PAM almost remains unchanged, at this time the PAM in soil adsorption column reached adsorption equilibrium, then kept constant velocity and used water to leach adsorption column, PAM concentration gradually reduced to almost zero, the adsorption column was fully cleaning. When the flow rate was 50 mL/min, PAM quality concentration with the increase of total solution volume rapid increase. Flow rate was 10 mL/min adsorption situation the main reason with the increase of the flow rate, the more solution of PAM in soil. The longer on the residence time, the more chances were soil adsorption and retention PAM in the soil. Lead to PAM in soil adsorption column of the concentration increase quickly. With the increase of the mixed solution, the concentration of PAM increased slower when it was closer to the adsorption equilibrium concentration. Because the soil adsorption column reached almost saturated adsorption, adsorption slows until the soil adsorption column of adsorption equilibrium. Reoccupy clear water to 50 mL/min flow rate cleaning adsorption column, PAM concentration reduces gradually until fully clean.

The Fig. 3 and 4, it was known that the flow rate was 10 and 50 mL/min, mixed solution of Br^- concentration with total solution volume increase. In accumulative total solution volume was 300 mL, bromide ion concentration and mixed solution of bromide ion concentration was consistent, after the testing concentration almost the same, that bromide ion reach saturated adsorption, Use water leaching adsorption column, bromide ion concentration gradually reduced until concentration was zero. In addition, in both soil migration experiments, bromide ion content in the saturated adsorption appeared more than saturated adsorption phenomenon. The phenomenon was bromide ion in the downward migration process, each layer of soil granule on it has certain entrapment effect, make bromide ion retention in the soil which has not been adsorption, cannot move down, again due to the limitation of the experiment device, this part of the retention in the soil of the bromide ion also counted in content in the soil, the bromine ion content more than saturated adsorption.

CONCLUSION

The following are the conclusions drawn from this study:

- In this study, through the static adsorption experiment of PAM, it could be conclude that the adsorption equilibrium time of PAM in saline soil was less than in chernozem and the equilibrium

concentration of PAM in saline soil was higher than in chernozem.

- The PAM adsorption quantity ratio in chernozem was more than in saline soil, thus the consequent in chernozem was better.
- The PAM dynamic adsorption experiment in chernozem showed that when PAM arrived at adsorption saturation the volume of mixed solution of 50 mL/min flow was less than 10 mL/min. It could be seen that the PAM dynamic adsorption experiment the migration tracing trajectory of tracer Br^- were basically the same in different soil.
- It provided data for the establishment of a numerical simulation model of PAM adsorption in different soil through the static adsorption and dynamic adsorption.

ACKNOWLEGMENT

This research was supported by China Postdoctoral Science Foundation funded project (Grant No. 2012M510303).

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