Combined Effect of Pour Point Depressants and Magnetic Field on the Viscosity and Pour Point of Crude Oil

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Abstract- Pour point depressants (PPD) are widely used for mitigating wax deposition problems. But they are costly and environmentally hazardous. Another method which has attracted the attention is the magnetic field (MF) conditioning method. Though this method is environmentally friendly and economical, the results obtained have found to be controversial. The viscosity and pour point may increase or decrease. This has been attributed to number of reasons, the main reason being the specificity of crude oil and its interaction with the magnetic field. Hence an attempt has been made to investigate the combined effect of PPDs and MF for better viscosity and pour point reduction. It has been found that there is no substantial enhancement in the viscosity and pour point reduction with the combination. Hence pure PPD method dominates wax inhibition methods as of now.

Keywords - Crude Oil, Pour Point Depressants, Magnetic Field, Wax Deposition, Viscosity

I. INTRODUCTION

Transportation of crude oil by pipe lines is a difficult job owing to the viscous nature of the crude due to wax formation. If proper measures are not taken it may lead to tremendous economic loss as a result of decrease in production. The most commonly used methods are mechanical, chemical and heating methods. Chemical methods are more preferred due to the ease of implementation and the results obtained. But their use is environmentally hazardous. Also the cost of these proprietary chemicals is huge owing to the research efforts put in them. This method is the most widely studied method and new PPDs keep on pouring into the flow assurance market frequently.

II. LITERATURE

A. Pour Point Depressants

A pour point depressant can be linear polymer or co polymer with pendant hydrocarbon chain group. The different wax inhibitors that have been used traditionally include Ethylene Vinyl Acetate (EVA) and its copolymers, Maleic anhydride and methyl methacrylates [1][2][3][4]. Various wax crystal inhibitors have been synthesized by number of workers. Wei [5] has elaborately discussed recent advances in wax inhibition chemicals. Following are few of the synthesized polymers acting as pour point depressants. Copolymers of Maleic anhydride having polar functional group in their structure by Deshmukh and Bharambe [6], decyl acrylate and iso octyl acrylate by Ghosh and Das [7], reactive PPDs with anhydride group by Liu et al [8], Cocomide diethanolamine by Ridzuan et al [9], hexatriethanolamine mololeate by Sharma et al [10], Octyl, decyl and dodecyl 3-methyl-imidazolium Dedecylpyridinium Chloride by Subramanium et al [11], Triethanolamine (TEA) by Popoola et al and Taiwo et al [12], [13], maleic anhydride-α-octadecene copolymer and its derivatives with octadecyl (MAC), phenyl (AMAC) or naphthalene (NMAC) pendants by Xu et al [14], phthalimide and succinimide copolymers of vinyl acetate, styrene and methyl by Al-Sabagh et al [15], modified octadecene-co-maleic anhydride copolymers by El-Ghazawy et al [16] and poly (octadecyl acrylate) (POA)/clay nanocomposite PPD by Yao et al [17].

The mechanism of wax inhibition has been explained by different theories. These are the Incorporation- Perturbation theory, Nucleation Sequestration theory and the adsorption on pipe wall theory[18]. The nucleation theory suggests that the wax inhibitors act as nucleating agents. They aid in the formation of smaller wax crystals. They also adsorb on the surface of the wax nuclei leading to the formation of a defective wax surface. This weakens the interaction with the surrounding crystals [19]. The Incorporation- Perturbation theory states that when the wax crystallization process begins at a temperature lower than wax appearance temperature (WAT), the wax inhibitors incorporate on the growing surface of the wax crystals. In the pipe wall theory, oleophobic surfaces are created on the pipe walls by the inhibitors which prevent the adsorption on wax crystals. [20]

This has been supported by Behbahani [21] who says that flow improvers co-crystallize with wax thereby introducing a fault in the growing wax crystal. Chen et al [22] suggest that the structure of wax partly transformed from hexagonal to orthorhombic. Another theory by Binks et al claims that addition of PPD causes the critical volume fraction ϕ* to increase thus decreasing the pour point which is correlated to the axial ratio h/d. Thus effective PPDS work by changing the axial ratio of wax crystals that are precipitated [23]. Suryanarayana et al [24] found that flow improvers modify the aliphatic portion of resins, wax and asphaltenes and do not interact with the polar group in these fractions.

It has been observed that asphaltenes too influence the process of wax crystallization and affect to the effectiveness of the wax inhibitor. Kriz and Andersen [25] have shown that the crystallization of wax depends on the degree of dispersion or flocculation of asphlatenes. At low concentrations the asphaltenes are well dispersed. They increase the WAT as the concentration goes on increasing and reaches a critical value. After this point, the asphaltene molecules tend to flocculate among themselves. They co precipitate but cannot be incorporated in the wax network. This produces unorganized or random asphaltene-paraffin structure. This leads to decreased yield stress and WAT, but the dynamic viscosity increases. Perez et al [26] found the inhibition efficiency increased for oils having more asphaltene content but low wax content.

To overcome this, many techniques have been suggested. Lashkarbolooki et al [27] have suggested the use of diluents along with wax inhibitors like EVA to reduce asphaltene deposits. Liu et al [8] claim that anhydride groups of reactive PPDs react with the amino group of asphaltenes or resins and generate acylamino group. Chanda et al [28] have used Polybenenyl acylate (PBA) alongwith asphaltene soluble solvents like benzene, xylene and pyridine. Xu et al [14] have studied the influence of pendants in comb-type copolymers on the cold flow ability of crude oil. It was observed that small aromatic pendants improve the flow ability of waxy oils by adsorbing on the surface of asphaltenes, while large aromatic pendants impair the assembly of copolymers with asphaltenes by a higher steric hindrance.

B. Magnetic Field Conditioning

It is observed that the apparent viscosity of crude oil decreases when it is subjected to magnetic field. Early references suggest that this method which was used for the treatment of the inorganic deposits could also be used for the organic ones [29]. Thereafter number of workers have reported this phenomena.[30]–[41]. The basic mechanism proposes that magnetic field aligns the already disoriented paraffin molecules its direction which reduces the apparent viscosity. However there are reports which state that the viscosity of crude oil may increase [42] or may have no substantial change [31].

A number of mechanisms have been proposed for this reduction in viscosity. Charged species in petroleum give rise to Lorentz forces which destroy aggregation of paraffin particles. [29], [30], [37]. Presence of vanadyl complexes have been reported by Loskutova et al [40]. Iron oxides of colloidal nature and paramagnetic properties have also been discovered in oil samples from producing wells by Lesin et al [43] The reasoning for this contradictory behaviour has been attempted by some. Some say that it is due to aggregation of the paraffin molecules in a suspension [44] while others say that it is due to disaggregation of paraffin crystals [45], [46]. The suspension theory claims that crude oil is a suspension of paraffins in a solvent. If magnetic field is applied for a long time, the paraffin aggregate into long chains while aligning in the direction of the field and increase the viscosity. But if the oil is exposed to the magnetic field for a lesser span of time, the paraffin particles do not form long chains and reduce the effective viscosity. The disintegration theory states that magnetic treatment results in the disaggregation of the colloidal particles in the crude oil. Original crude oil has paraffin particles that are interlocked. When magnetic field is applied disaggregation occurs and the average size of the suspended particles decreases which results in decrease in viscosity. Owing to the complex nature of crude, the exact mechanism of action of magnetic field on crude oil is still under investigation. Very few attempts have been made to investigate the combined effect of chemical and magnetic field on crude oil behaviour [47].

The objective of this paper is to investigate the combined influence of magnetic field and pour point depressants on viscosity and pour point of crude oil. To the best of our knowledge, there has been no work done so far on such type of combination method.

III. **MATERIALS**

Crude oils of three different types named as C1, C2 and C3 were obtained from western parts of India. Three different commercially available PPDs viz. Ethylene vinyl acetate having vinyl acetate content 18% and 28% (EVA-18 and EVA-28) from Sigma Aldrich and Poly(methyl methacrylate) (PMMA) from LG Chemicals were chosen for study. These represented the classes of linear polymers and comb shaped polymers respectively. The magnet used for the study was an electromagnet. The magnetic field could be varied by changing the gap between the pole pieces and by changing the current to the magnet. A gauss meter was used to measure the magnetic field.

IV. **METHODS**

These inhibitors were mixed with the crude oil in concentrations of 200 ppm, 1000 ppm and 1500 ppm. Volume of the sample taken was 50 ml. Viscosities and pour points of these samples were measured. The samples were then subjected to electromagnetic fields of strengths 1000 gauss and 3000 gauss for a period of one minute. The time of exposure was chosen with reference to the observations of Tao and Xu [44]. This was held stationary between the pole pieces of the electromagnet. Viscosities and Pour points of these magnetically treated samples were measured using Brookfield DVII+ Pro Viscometer and ASTM D-97 method respectively.

The experiments were performed at a temperature close to the wax appearing temperature (WAT) or close to WAT. This is chosen as per the observations reported by Tung et al [37] and Chow et al [42]. Tung et al [37] had observed that the magnetic field had more pronounced effect when the oil was treated at a temperature 10^0C higher than the pour point, which is almost close to WAT. Also Chen et al [22] had found that the PPD effect vanishes at lower temperatures.

V. RESULTS AND DISCUSSION

TABLE 1
CRUDE OIL PROPERTIES

	C1	C2	C3
Density (g/cc)	0.8754	0.877	0.887
API	30.1	29.8	28
WAT (deg C)	30	43	50
Viscosity at WAT (cP)	69.3	55.4	82.1
Asphaltene content (%)	12.2	5	7.2
Wax content (%)	25.1	35.5	45.7
Pour Point (deg C)	18	27	33

TABLE 2
VARIATION IN VISCOSITY USING PPDS OF DIFFERENT CONCENTRATIONS AT DIFFERENT MAGNETIC FIELDS FOR CRUDE OIL 1.
TEMPERATURE = 30^{0}C, TIME OF EXPOSURE OF MAGNETIC FIELD = 1 MINUTE

	PPD Concentration (ppm)	Viscosity (cP)		
		0 gauss	1000 gauss	3000 gauss
C1	0	69.3	62	55.5
C1+ EVA-18	200	12.4	11.8	10.9
	1000	11.9	11.3	11.5
	1500	11.7	13.9	12.4
C1 + EVA-28	200	14.3	13.8	15
	1000	14.7	14.1	14.8
	1500	19.2	34.2	24.3
C1 + PMMA	200	10.5	11.8	9.7
	1000	9.5	10	9.2
	1500	11.6	13	10.5

TABLE 3
VARIATION IN VISCOSITY USING PPDS OF DIFFERENT CONCENTRATIONS AT DIFFERENT MAGNETIC FIELDS FOR CRUDE OIL 2.
TEMPERATURE = 45^{0}C, TIME OF EXPOSURE OF MAGNETIC FIELD = 1 MINUTE

	PPD Concentration (ppm)	Viscosity (cP)		
		0 gauss	1000 gauss	3000 gauss
C2	0	55.4	29.1	31.9
C2 + EVA-18	200	18.2	20.1	14.2
	1000	20.6	24.3	19.8
	1500	22.4	25.1	35.5
C2 + EVA-28	200	24.2	20.9	22.7
	1000	35.6	37.8	33.1
	1500	38.1	42.1	39.7
C2 + PMMA	200	10.5	11.2	10.9
	1000	10.3	10.9	9.5
	1500	10.2	10.3	9.6

TABLE 4
VARIATION IN VISCOSITY USING PPDS OF DIFFERENT CONCENTRATIONS AT DIFFERENT MAGNETIC FIELDS FOR CRUDE OIL 3. TEMPERATURE = 50^{0}C, TIME OF EXPOSURE OF MAGNETIC FIELD = 1 MINUTE

	PPD Concentration (ppm)	Viscosity (cP)		
		0 gauss	1000 gauss	3000 gauss
C3	0	82.1	47	50.3
C3 + EVA-18	200	19.1	17.1	18.5
	1000	19.4	18.2	18.8
	1500	19.8	16.9	17.4
C3 + EVA-28	200	22.5	21.5	23
	1000	23.6	22.9	22.3
	1500	25.1	24.3	24.8
C3 + PMMA	200	12.6	13.7	13.1
	1000	13.2	13.8	12.6
	1500	14.1	14.5	13.8

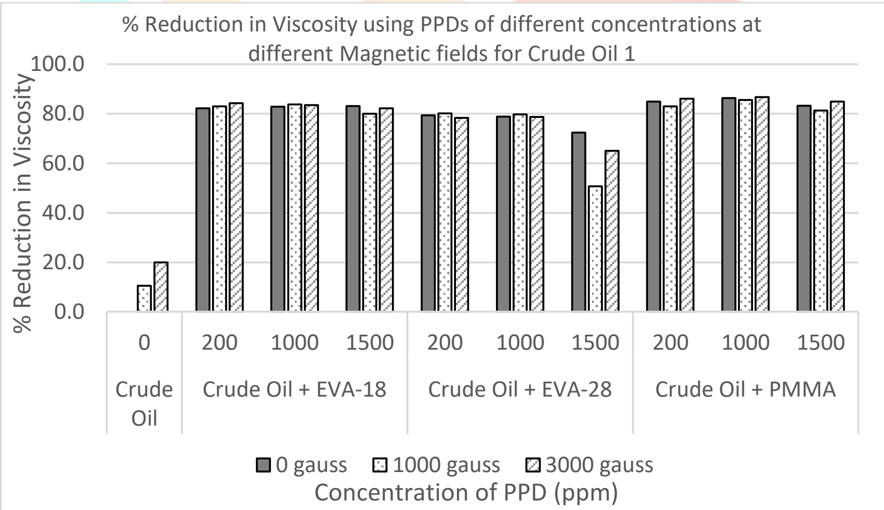

Fig 1: % Reduction in Viscosity using PPDs of different concentrations at different Magnetic fields for Crude Oil 1

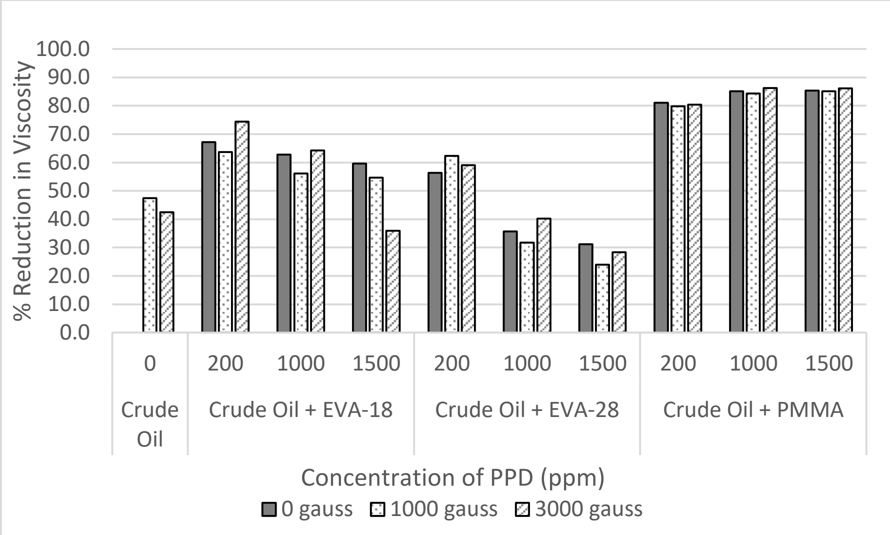

Fig 2: % Reduction in Viscosity using PPDs of different concentrations at different Magnetic fields for Crude Oil 2

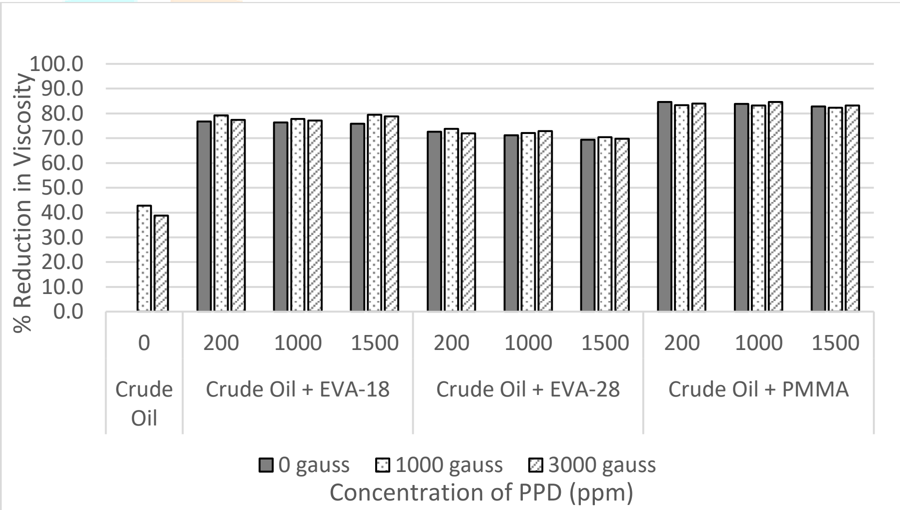

Fig 3: % Reduction in Viscosity using PPDs of different concentrations at different Magnetic fields for Crude Oil 3

TABLE 5
VARIATION IN POUR POINT USING PPDS OF DIFFERENT CONCENTRATIONS AT DIFFERENT MAGNETIC FIELDS FOR CRUDE OIL 1. TIME OF EXPOSURE OF MAGNETIC FIELD = 1 MINUTE

	PPD Concentration (ppm)	Pour Point (Deg C)		
		0 gauss	1000 gauss	3000 gauss
C1	0	18	15	14
C1+ EVA-18	200	15	14	13
	1000	15	13	14
	1500	16	15	14
C1 + EVA-28	200	10	12	13
	1000	17	18	17
	1500	18	18	16
C1 + PMMA	200	13	14	12
	1000	15	14	14

	1500	17	13	14

TABLE 6
VARIATION IN POUR POINT USING PPDS OF DIFFERENT CONCENTRATIONS AT DIFFERENT MAGNETIC FIELDS FOR CRUDE OIL 2. TIME OF EXPOSURE OF MAGNETIC FIELD = 1 MINUTE

	PPD Concentration (ppm)	Pour Point (Deg C)		
		0 gauss	1000 gauss	3000 gauss
C2	0	27	19	18
C2 + EVA-18	200	21	20	19
	1000	26	27	26
	1500	27	25	28
C2 + EVA-28	200	21	21	23
	1000	25	27	26
	1500	25	24	26
C2 + PMMA	200	27	27	26
	1000	26	25	25
	1500	27	25	24

TABLE 7
VARIATION IN POUR POINT USING PPDS OF DIFFERENT CONCENTRATIONS AT DIFFERENT MAGNETIC FIELDS FOR CRUDE OIL 3. TIME OF EXPOSURE OF MAGNETIC FIELD = 1 MINUTE

	PPD Concentration (ppm)	Pour Point (Deg C)		
		0 gauss	1000 gauss	3000 gauss
C3	0	33	24	26
C3 + EVA-18	200	28	26	27
	1000	27	26	27
	1500	26	26	25
C3 + EVA-28	200	25	28	27
	1000	26	26	25
	1500	28	26	27
C3 + PMMA	200	24	25	26
	1000	25	26	24
	1500	26	26	25

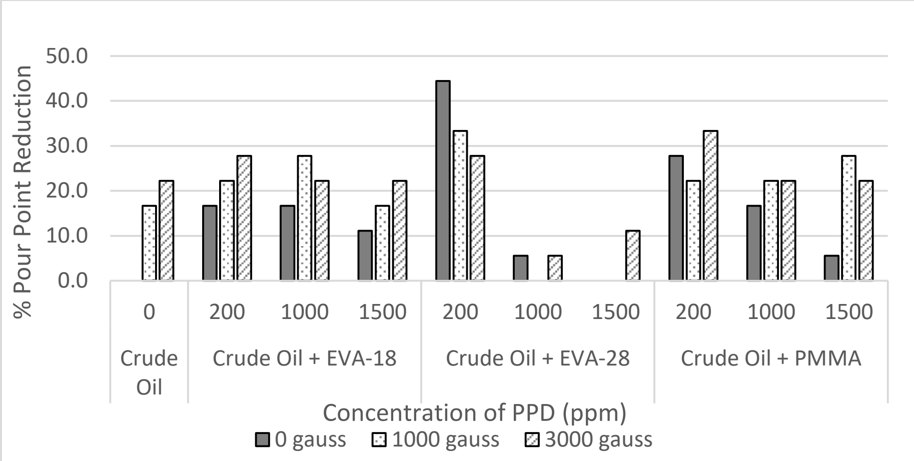

Fig 4: % Reduction in Pour Point using PPDs of different concentrations at different Magnetic fields for Crude Oil 1. Time of Exposure of Magnetic Field = 1 minute

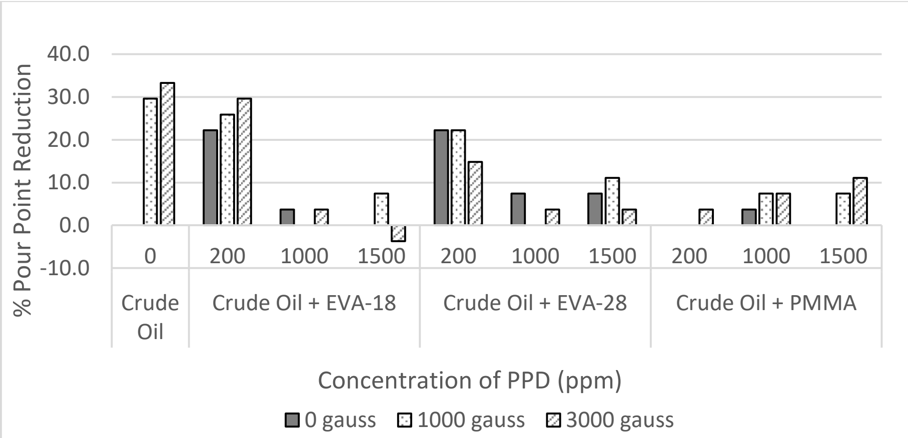

Fig 5: % Reduction in Pour Point using PPDs of different concentrations at different Magnetic fields for Crude Oil 2. Time of Exposure of Magnetic Field = 1 minute

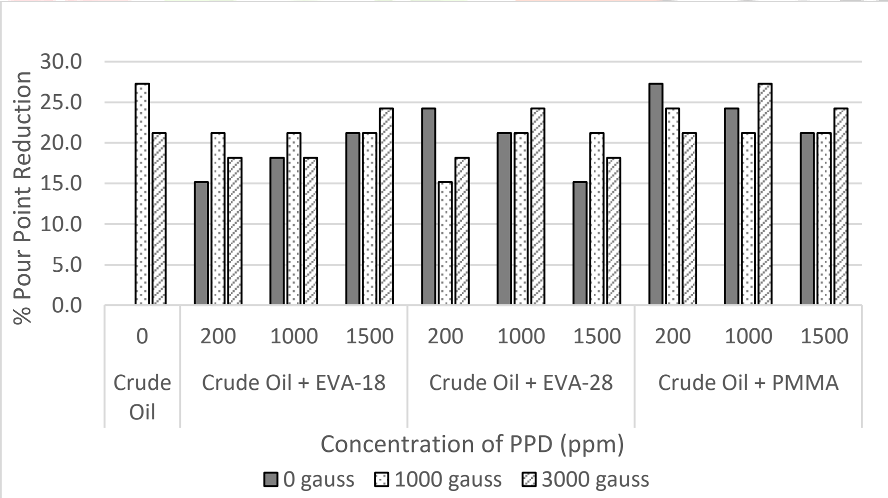

Fig 6: % Reduction in Pour Point using PPDs of different concentrations at different magnetic fields for Crude Oil 3. Time of Exposure of Magnetic Field = 1 minute

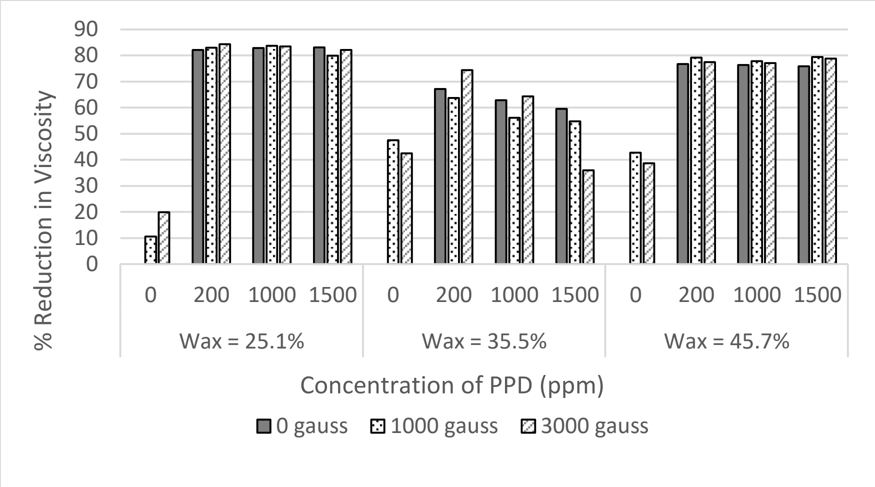

Fig 7: % Reduction in Viscosity vs Wax % in crude oil at different concentrations of EVA-18

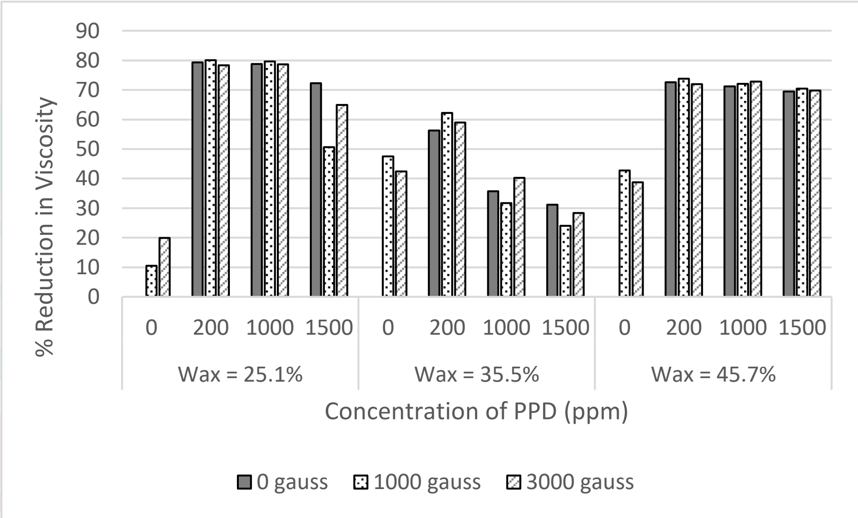

Fig 8: % Reduction in Viscosity vs Wax % in crude oil at different concentrations of EVA-28

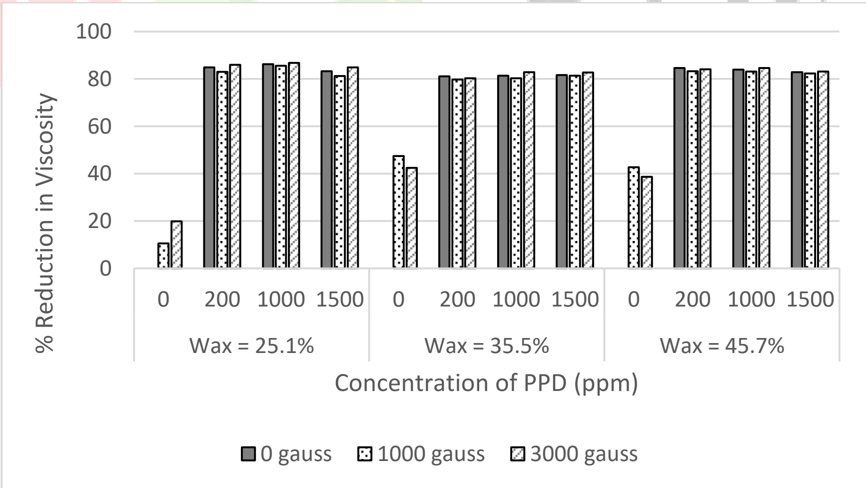

Fig 9: % Reduction in Viscosity vs Wax % in crude oil at different concentrations of PMMA

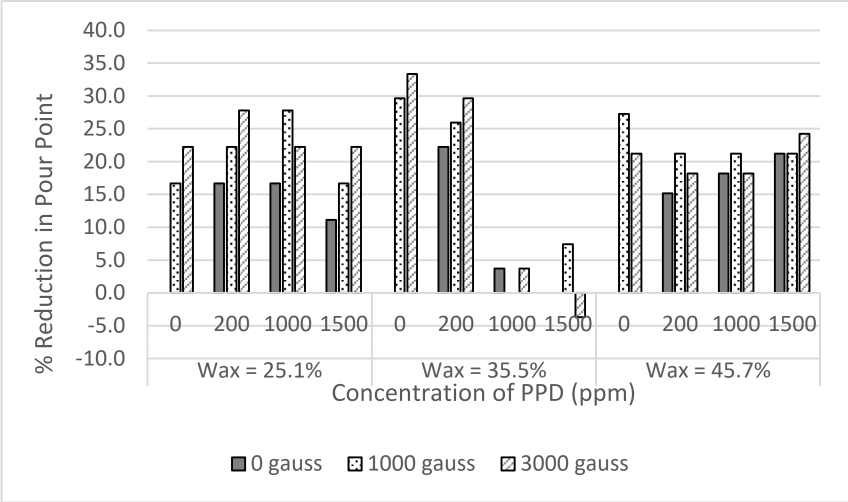

Fig 10: % Reduction in Pour Point vs Wax % in crude oil at different concentrations of EVA-18

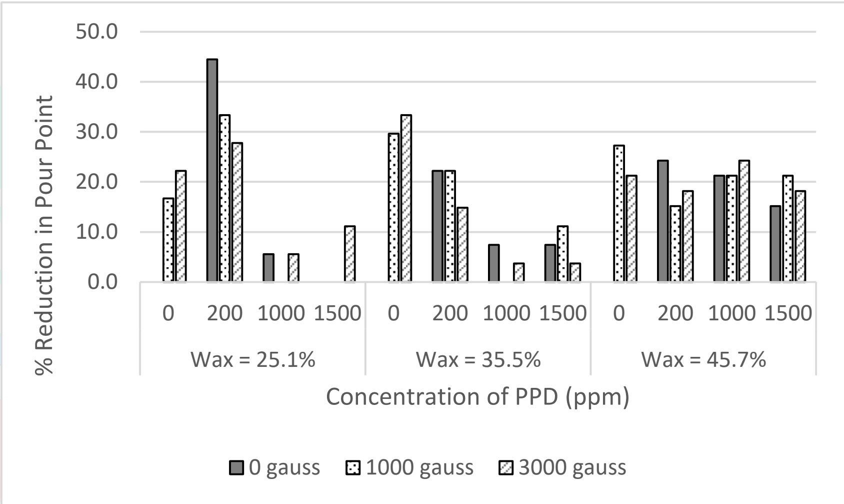

Fig 11: % Reduction in Pour Point vs Wax % in crude oil at different concentrations of EVA-28

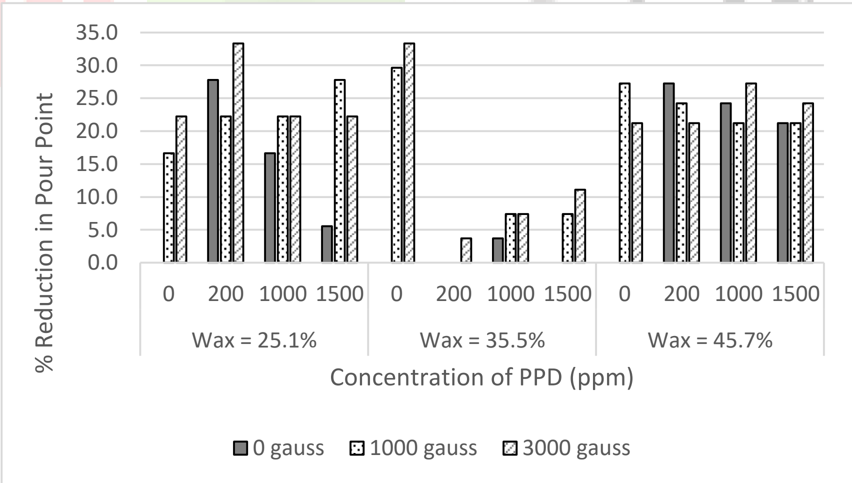

Fig 12: % Reduction in Pour Point vs Wax % in crude oil at different concentrations of PMMA

A. Effect of Magnetic field and PPDs on Viscosity

As seen from Fig 1, 2 and 3, maximum viscosity reduction was observed for PMMA followed by EVA-18 and EVA-28. If the concentration of PPD is increased, the reduction in viscosity decreases or remains same for linear polymers. The effect is more pronounced for EVA-28. Hence lower the concentration, better is the viscosity reduction. For comb polymers, it remains same or increases. It has been proved that PPDs associate themselves with wax crystals. If the concentration of PPDs is increased, they form a larger network with wax thereby reducing the flow ability[48].

The structure of copolymers, especially the proportion of polar and nonpolar segments, plays an essential role in affecting the rheological properties of crude oil[14]. Comb shaped polymers are found to be more effective as compared to linear polymers [49]. When magnetic field is applied it is observed that the linear polymer with lesser vinyl acetate (VA) content is affected more as compared to higher VA content.

B. Effect of Magnetic field and PPDs on Pour Point

Figs 4, 5 and 6 show that the reduction in pour point is seen better for EVA than for PMMA. Hence it is observed that linear polymers give better reduction in pour point as compared to comb type. For comb type polymers, the reduction in pour point decreases with increase in the PPD concentration. This is in agreement with Kuzmic et al [50] who had found that the methacrylate polymers are more effective at concentrations less than 1000 ppm. Also in case of EVA, the reduction in pour point decreases as the vinyl acetate content increases.

Literature suggests that the optimum concentration of vinyl acetate in EVA should be around 28% for effective pour point depression [2]. Increasing the VA content does not increase the effectiveness. Also Rosdi and Ariffin [51] had tested emulsions of EVA with different VA content viz. 12, 18, 28 and 40% and had found that lower VA content gives better flowability. Oliviera et al [52] had taken EVA with concentrations of VA as 19% and 28% and found that the effectiveness was dependent on the average molar mass of the chains. The polymer was more effective if the chain sizes were less than C_{28}. This is supported by Jin et al [53]. Since the characterization of wax in this experiment was not done, the concentrations of VA in the present experimentation were chosen as 18% and 28%.

One interesting observation is obtained in Fig 10 which shows that for EVA 18 with concentration of 1500 ppm the pour point increases. This is in accordance with Zhang Jinli et al [54]. They have found that some pour points are greater than that of crude oil even after the addition of the additive. They have found that minimum dosage of EVA depends on the total area covered by the EVA molecules and molecular weight. If it is not sufficient, the pour point increases.

When only the magnetic field is applied, maximum reduction in pour point is observed for crude oil 3 which has the highest wax content. At lower wax content, higher magnetic field is effective and vice versa. The pour point reduction is maximum for least asphaltene to wax ratio. As asphaltene to wax ratio increases, reduction in pour point decreases. Maximum reduction is pour point is around 33% and the least is 16%. The average reduction in pour points is 16%, 7.4% and 29.9% for C1, C2 and C3 respectively. There is no direct relation between the viscosity and pour point reduction [53], [1]. Co-crystallization was the main reason for pour point depression. Pour point reduction is seen in case of EVA but not in case of PMMA. On the other hand viscosity reduction is more for PMMA but not for EVA.

C. Effect of Wax % on Viscosity and Pour Point reduction

Crude oil 1 has the lowest wax content. The maximum reduction in viscosity with magnetic field alone is found to be between 10% and 20% for 1000 and 3000 gauss. For crude oils 2 and 3 which have higher wax content, the % reduction in viscosity ranges from 38% to 47%. For lower wax content a greater MF is found to be effective and for higher wax content, lower field is effective. There has to be a threshold wax where this transition takes place.

The results obtained for pure PPDs shows that the maximum reduction in viscosity of around 87% is obtained for PMMA. EVA-18 and EVA-28 have similar results which range between 70-80%. EVA is more effective for less wax %. As the concentration of wax increases, the % reduction in viscosity decreases for EVA-18 and EVA-28 but not for PMMA. For comb type polymers wax % does not seem to affect the viscosity reduction. This suggests that linear polymers work best with lower wax% as compared to comb polymers which work for higher wax content with hard phase. [18]

For higher magnetic field and higher PPD concentration, the pour point decreases appreciably. Also it is seen that this trend increased for higher wax %. Hence the combined effect is more pronounced for higher wax % when compared with PPDs.

D. Effect of Asphaltene content on Viscosity and Pour Point reduction

The asphaltene/wax ratio for C1, C2 and C3 are 0.48, 0.14 and 0.16 respectively. It is observed in case of PPDs, that less is the asphaltene to wax ratio less is the reduction in viscosity. The observations are supported by those of Perez et al [26]. They showed that inhibition efficiency increased for oils having more asphaltene content but low wax content. But in case of magnetic field, less is the asphaltene/wax ratio more is the reduction in viscosity.

E. Overall Change in Viscosity and Pour Point

Average reduction in viscosity is better obtained by PPDs as compared to magnetic field. In case of pour point reduction, magnetic treatment seems to be better as compared to PPDs but marginally. Crude 2 is an exception. This may be attributed to the lower asphaltene to wax ratio. For the combined treatment of PPDs + MF does not show much difference if compared to PPD treatment for both viscosity as well as pour point.

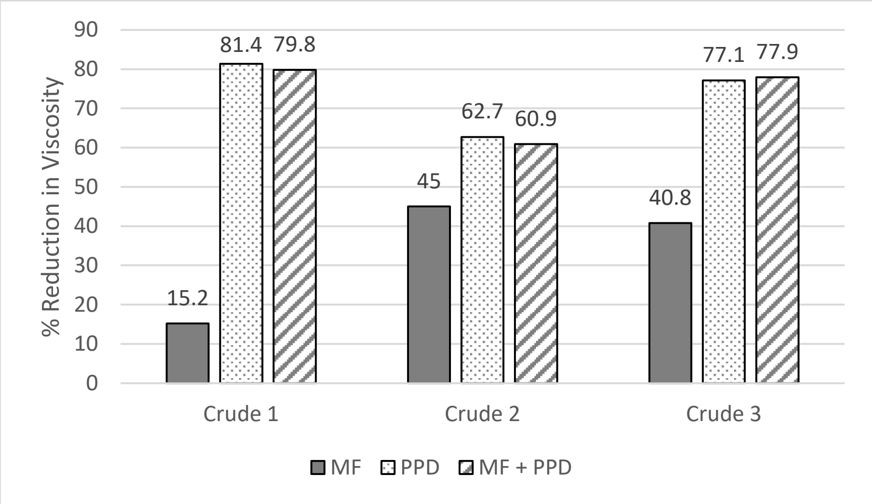

Fig 13: Average % Reduction in Viscosity for Different Crude Oils

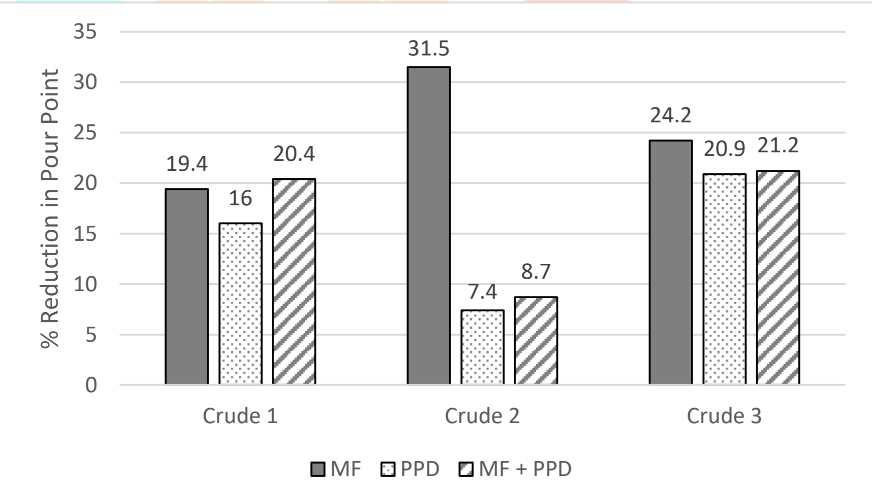

Fig 14: Average % Reduction in Pour Points for Different Crude Oils

VI. **CONCLUSIONS**

A comparison between three different methods viz. pour point depressant method, Magnetic Conditioning Method and combination of the two methods with respect to reduction in viscosity and pour point of 3 different crude oils has been presented. Viscosity reduction is effectively done by comb type polymers as compared to linear polymers. Also viscosity reduction is better with PPDs than with magnetic field. Pour point reduction is better with linear PPDs. Magnetic field gives slightly better reduction in pour point. Increase in wax% decreases the viscosity reduction of linear polymers. Comb polymers show no response. PPDs work best at lower wax content and magnetic field at higher wax%. Combined effect of the PPDs and MF more or less gives similar results as obtained by PPDs. It is more visible at higher magnetic field and higher PPD concentration. The asphaltene to wax ratio also plays an important role. For lesser ratios PPDs are more effective and for higher ones magnetic field is more effective. Thus the combined method does not seem to be promising for lower magnetic fields. It would be interesting to investigate the effect at higher magnetic fields.

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