NOVEL POUR POINT DEPRESSANT (PPD) COPOLYMER SYNTHESIS, CHARACTERIZATION, AND PERFORMANCE EVALUATION FOR WAX DEPOSITION INHIBITION OF MALAYSIAN CRUDE OIL

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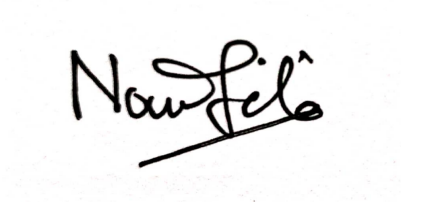
We hereby declare that We have checked this thesis and in our opinion, this thesis is adequate in terms of scope and quality for the award of the degree of Doctor of Philosophy.



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I hereby declare that the work in this thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at Universiti Malaysia Pahang or any other institutions.

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ABSTRAK

Pemendapan lilin di dalam paip pemprosesan dan tiub penyaluran dari pesisir pantai ke darat adalah sangat kritikal bagi industri minyak dan gas disebabkan oleh keadaan suhu yang rendah. Pendekatan yang paling popular untuk menyelesaikan isu ini ialah dengan memasukkan perencat lilin ke dalam saluran paip. Dalam kajian ini, novel poli stearyl acrylate-co-behenyl acrylate (SA-co-BA) bahan penurun takat curah (PPD) kopolimer telah berjaya disintesis menggunakan kaedah pempolimeran larutan radikal bebas. Proses pemilihan faktor untuk proses pengoptimuman diperoleh berdasarkan pra-penilaian faktor (OFAT). Kaedah gerak balas permukaan (RSM) digunakan bagi mengoptimumkan faktor ekperimen iaitu suhu tindak balas, kepekatan dan masa tindak balas pada nisbah jisim tetap (1:1). Keadaan optimum yang diperoleh adalah pada masa tindak balas 7 jam, kepekatan permula 1wt%, dan suhu tindak balas 90 °C dengan hasil sebanyak 97.31%. Analisis ANOVA memaparkan nilai R^2 sebanyak 98.44%. Penilaian prestasi SA-co-BA dengan dan tanpa penambahan partikel nano-silika pada PPD dan keatas sifat reologi minyak mentah Malaysia diselidik. Kopolimer SA-co-BA pada 1000 ppm memberi kesan kepada penurunan suhu takat curah dari 11 hingga 2 °C. Darjah pengurangan kelikatan (DVR) yang optimum untuk kedua-dua SA-co-BA PPD dan partikel nano SiO_2 adalah pada 1000 ppm sebanyak 89.76% dan 200 ppm pada 89.52%. Di samping itu, DVR tertinggi sebanyak 92% nisbah isipadu (3:1) pada campuran SA-co-BA /SiO_2. Penilaian prestasi novel SA-co-BA PPD dengan dan tanpa penambahan partikel nano-silika pada pemendapan lilin telah diuji. Dalam kajian ini, lima faktor telah dinilai iaitu kelajuan putaran dari 100 hingga 600 rpm, suhu jejari sejuk dari 5 hingga 25 °C, tempoh eksperimen dari 2 hingga 12 jam, kepekatan SA-co-BA dari 500 hingga 5000 ppm dan kepekatan nano-silika dari 100 hingga 800 ppm. Keputusan menunjukkan bahawa peratusan kecekapan perencatan tertinggi (PIE) bagi SA-co-BA PPD tanpa dan dengan penambahan partikel nano-silika masing-masing ialah 44.14% dan 52.25%. Keputusan RSM menunjukkan minimum mendapan lilin iaaitu 0.01 g diperoleh pada suhu 14 °C, 60 min dan 1200 ppm, dan ANOVA mengesan nilai R^2 ialah 0.9874. Bagi pengesahsahihan perencat lilin yang terpilih, simulasi dinamik molekul (MD) diperkenalkan untuk memahami interaksi antara kristal lilin dan perencat lilin pada peringkat molekul. Keputusan menunjukkan molekul lilin n-icosane terikat bersama bagi menghasilkan kristal lilin pepejal dengan interaksi van der Waals (vdW) antara H57 dan H60. Jejarian fungsi agihan (rdf) bagi interaksi H57•••H60 dalam sistem binari SA-co-BA dan NP secara individu teranjak daripada 2.75 Å kepada 3.25 Å. Interaksi H57•••H60 dalam sistem tertier lilin n- $C_{20}H_{42}$, – SA-co-BA /NP teranjak daripada 2.75 Å kepada 4.75 Å. Kesimpulannya, dapatan kajian ini boleh digunakan untuk membangunkan model yang boleh dipercayai bagi mengurangkan pemendapan lilin, kos dan masa pemprosesan.

ABSTRACT

Wax deposition in production pipelines and transportation tubing from offshore to onshore is critical in the oil and gas industry due to low-temperature conditions. The most significant popular approach to solving this issue is inserting a wax inhibitor into the channel. The objective of this study is to synthesis a novel poly stearyl acrylate-co-behenyl acrylate (SA-co-BA) pour point depressant PPD copolymer in order to improve the flowability of Malaysian crude oil. In this study, a novel (SA-co-BA) PPD copolymer was successfully synthesized by the free radical solution polymerization method. The selection of factor levels for the optimization process was obtained based on the pre-evaluation of factors using one factor at the time (OFAT). Response surface methodology (RSM) method was used to optimize the experimental factors which are reaction temperature, the initiator concentration and reaction time respectively, a constant mass ratio (1:1). The optimize conditions were obtained at reaction time of 7 h, initiator concentration of 1 wt %, and reaction temperature of 90 °C with the corresponding yield of 97.31%. The analysis of ANOVA detected R^2 value of 0.9844. Evaluation of the performance of SA-*co*-BA with and without the addition of nano-silica particles on PPD and rheological behaviour of Malaysian crude oil were investigated. The findings showed that SA-*co*-BA copolymer at 1000 ppm affects the pour point temperature from 11 to 2 °C. The optimum degree of viscosity reduction (DVR) for both SA-*co*-BA and SiO_2 nanoparticles were at 1000 ppm by 89.76% and 200 ppm 89.52%, respectively. In addition, the highest DVR was 92% at the volume ratio (3:1) of the SA-*co*-BA /SiO_2 blend. Evaluation of the performance of novel SA-*co*-BA with and without the addition of nano-silica particles on wax deposition was studied. In this study, five factors have been evaluated, which are speed of rotation from 100 to 600 rpm, the temperature of a cold finger from 5 to 25 °C, duration of the experiment from 2 to 12 h, SA-*co*-BA concentration from 500 to 5000 ppm and concentration of nano-silica from 100 to 800 ppm. The results indicated that the highest percentage inhibition efficiency (PIE) of SA-*co*-BA PPD without and with nano-silica particles were 44.14% and 52.25%, respectively. The result of RSM showed a minimum wax deposit of 0.01 g obtained at 14 °C, 60 min and 1200 ppm and ANOVA detected the value of R^2 was 0.9874. To further validate the wax inhibitor chosen, molecular dynamics (MD) simulation was introduced to understand the interaction between wax crystal and wax inhibitor at the molecular level. The results indicated the n-icosane wax molecules are bonded together to create a solid wax crystal with van der Waals (vdW) interaction between H57 and H60. The radial distribution function (rdf) of the H57•••H60 interaction in the binary system of SA-*co*-BA and NP individually are shifted from 2.75 Å to 3.25 Å. the H57•••H60 interaction in the tertiary system of n- $C_{20}H_{42}$ wax – SA-*co*-BA /NP is shifted from 2.75 Å to 4.75 Å. Therefore, the combinations in tertiary system had a greater possibility of inhibiting wax than their individual counterparts. As a conclusion, the findings of this study can be used to improve a reliable model for reducing wax deposits and the associated costs.

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