



Article Synthesis and Performance Evaluation of Graphene-Based Comb Polymer Viscosity Reducer

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Abstract: The high viscosity of heavy oil makes it difficult to realize its economic value. Therefore, improving the fluidity of heavy oil can effectively improve the economic benefit of the development of heavy oil resources. Oil-soluble viscosity reducers can utilize functional groups in monomers to break up asphaltene aggregates to improve the flow of crude oil. Graphene can be used to insert and split asphaltene aggregates through sliding phenomena and π - π interaction with colloidal asphaltene, thereby improving the fluidity of heavy oil. In this study, a graphene nanocomposite viscosity reducer. The net viscosity reduction rate reached 80.0% at 400 ppm. Compared with a polymer viscosity reducer, the viscosity reduction effect of a graphene nanocomposite viscosity reducer was improved by about 7%. Structural characterization of a graphene nanocomposite viscosity reducer was characterized with infrared spectroscopy and a thermogravimetric test. The mechanism of a graphene nanocomposite viscosity reducer splitting asphaltene aggregates was verified with scanning electron microscopy. This study provides a theoretical and practical basis for the research and development of a novel nanocomposite viscosity reducer.

Keywords: heavy oil; nanocomposite viscosity reducer; comb polymer; graphene

1. Introduction

With the rapid development of the global economy, the demand for petroleum products is increasing. At present, due to the shortage of conventional crude oil reserves, people are beginning to attach importance to the exploitation, processing, and utilization of unconventional crude oil. According to statistics, the reserves of unconventional crude oil in the world account for 70% of the total crude oil [1–3]. There are high contents of resin, asphaltene, and wax in heavy oil, which makes the heavy oil solid at room temperature and improves the difficulty of exploitation and transportation [4]. To solve this problem, it is of great practical significance to develop a high efficiency and energy-saving viscosity reduction method for the development of petroleum industry. At present, the viscosity reduction methods that have been developed include heating [5], blending diluting oil [6–8], emulsification [9,10], the microbial method [11], catalytic modification [12,13], and adding an oil-soluble viscosity reducer [14–16].

The method of adding an oil-soluble viscosity reducer has attracted wide attention due to its advantages of low cost, low energy consumption, good viscosity reduction effect, and no subsequent processing required, in which the active components are mainly comb polymers containing benzene rings and polar groups [17]. The strong polar groups



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Copyright: © 2023 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). introduced into the polymer will form hydrogen bonds with colloid and asphaltene, which can not only separate the aggregate structure of colloid and asphaltene layers in heavy oil but also play the role of polymer skeleton support [18]. The long-chain alkyl can attract resin and asphaltene to form a solvated layer [19] and can also interact with paraffin to destroy the three-dimensional structure of wax crystals [20,21]. Although the viscosity reduction rate of polymer for low-viscosity heavy oil is high, the effect on the fluidity of super heavy oil with a large viscosity base is not significant. To solve this problem, nanomaterials are applied in the field of viscosity reduction in heavy oil [22]. The sliding phenomenon [23–25], interface effect [26–28], and surface effect [29,30] of nanomaterials themselves can provide a nucleation site for waxes and can promote polymer groups to interpenetrate into resin and asphaltene sheets. The combination of nanomaterials and polymers improves the physical properties of polymers [31], and the properties of nanomaterials themselves also play a role in reducing viscosity. Sharma et al. [32] investigated the properties of poly (methyl methacrylate)-graphene oxide (PMMA-GO) as a point depressant for waxy Indian crude oil. It was indicated that the composite has better thermal stability and viscosity reduction properties than the polymer PMMA. The long-term stability of the nanocomposite-treated crude oils revealed that their rheological properties undergo lesser degradation with time, as evident by having the least increase in apparent viscosity. Liu et al. [33] grafted ethylenevinyl alcohol copolymer (EVAL) onto a GO surface to prepare EVAL-GO for Daging waxy crude oil. It was indicated that EVAL-GO could make the wax crystals form spherical crystals, and the increased void space promotes more liquid oil to be released, which significantly improves the low-temperature fluidity of Daqing waxy crude oil.

Graphene is a kind of material with a two-dimensional honeycomb lattice structure. Some studies have shown that the excellent physical properties of graphene are affected by geometric shape, size, interaction, and dispersion [34,35]. The hexagonal lamellar structure and smaller particle size of graphene determine the physical properties of the polymer nanocomposites [35,36]. Currently, GO is widely studied because its surface is rich in oxygen-containing groups and can be easily modified. However, the presence of oxygencontaining groups on the surface of GO can disrupt the conjugated structure of graphene itself and distort the surface with folds, which affects the properties of the material itself. Moreover, the preparation of GO utilizes harsh conditions leading to a costly and nonenvironmental preparation process. In this article study, the graphene nanosheets (GNs) were modified with lipophilicity using the wet ball milling method. During the ball milling process, GNs were further exfoliated, and the modifier octadecylamine (ODA) modified the GNs by electrostatic adsorption. The lipophilic modification prevents graphene from aggregating through its own van der Waals forces, allowing it to be better dispersed in heavy oil. Moreover, the complete conjugated structure of GNs ensures that they can better generate π - π interaction with resin and asphaltene lamellae to disaggregate the aggregates [37]. The modified GNs were compounded with ternary polymers by solution mixing, and their viscosity reduction effect was evaluated.

2. Materials and Methods

2.1. Materials

Toluene was purchased from Tianjin Fuyu Fine Chemical Co., Ltd. (Tianjin, China); octadecyl methacrylate, styrene, acrylamide, fumaric acid, and maleic anhydride were purchased from Shanghai Aladdin Biochemical Technology Co., Ltd. (Shanghai, China); and ODA was purchased from Shanghai McLean Biochemical Technology Co., Ltd. (Shanghai, China). The GNs used in the experiment were obtained 10 times by the supercritical CO₂ stripping of graphene raw materials. The oil samples used in this work were taken from Liaohe heavy oil, whose composition and physicochemical properties are shown in Table 1.

Analytical Wax	Saturated	Aromatic	Resin	Asphalt	Wax
Point (°C)	HC (%)	HC (%)	(%)	(%)	(%)
28.54	35.51	22.61	24.09	4.05	8.37

Table 1. Physicochemical properties of Liaohe heavy oil.

2.2. Preparation of Graphene Nanosheets

The experiment was carried out on a self-developed supercritical fluid device [36]. A total of 10 g of graphene raw material was added to the reactor, injected with a gaseous intercalation medium (CO₂) to the pressure of 16 MPa, and stirred at 60 °C for 1 h. After CO₂ seeped into the graphite layer, the pressure of the device was reduced from 16 MPa to 4 MPa. When the pressure of the system reached normal pressure, the collection tank was opened to remove the single stripped sample. The above operation was repeated 10 times to obtain the required powder from the GNs.

2.3. Preparation of Comb Polymer

Octadecyl methacrylate, styrene, and polar monomer (acrylamide, fumaric acid, or maleic anhydride) were added into a nitrogen-filled three-necked flask and mixed thoroughly with a mixed solvent of toluene and anhydrous ethanol (V:V = 1:1). Then, AIBN, which was 1 wt% of the total monomer, was injected into a three-neck flask and reacted at 80 °C for 5 h. The product took anhydrous ethanol as a precipitant, and the three products were named as polymers 1, 2, and 3. Taking polymer 1 as an example, the preparation process is shown in Figure 1.



Figure 1. Synthesis of graphene-based comb polymer (ODA-GNs/Polymer 1).

2.4. Preparation of Graphene-Based Comb Polymer

ODA-modified graphene nanosheets (ODA-GNS) were obtained by adding a certain mass ratio of graphene and ODA into a ball mill tank containing DMF and milling at a speed of 400 r·min⁻¹ for 12 h. A certain mass ratio of ODA-GNs and a comb polymer were added to a beaker containing toluene and stirred at 65 °C for 12 h. After stirring, the solvent was evaporated by heating, washed several times with anhydrous ethanol, and dried overnight at 30 °C under a vacuum. The preparation process of the graphene-based

comb polymer 1 (ODA-GNs/Polymer 1) nanocomposite viscosity reducer is shown in Figure 1.

2.5. Fourier-Transform Infrared Spectra

The test samples were placed in a mortar with potassium bromide in the ratio of 1:100 for thorough grinding, after which the mixtures were placed in a test mold for pressing, and the structures of polymers 1/2/3, GNs, ODA-GNs, and ODA-GNs/Polymer 1 were tested in the transmittance mode (TR) over the range of 4000 to 500 cm⁻¹.

2.6. Thermogravimetric Analysis Determination

The TGA temperature is set within the range of 50–800 °C, the heating rate is 10 °C/min, and the gas flow rate is 100 mL/min. ODA-GNs and ODA-GNs/Polymer 1 were tested, respectively. The thermal stability of the samples and the amount of grafting on graphene were judged by the change of mass with temperature.

2.7. Dispersion Stability Analysis

Using 10 mL of toluene as the solvent, GNs, ODA-GNs, and ODA-GNs/Polymer 1 samples were prepared with a concentration of 1000 ppm, respectively. After ultrasonic dispersion for 1 h, the samples were placed for 3 days to observe the stability of the samples in toluene.

2.8. Scanning Electron Microscope

Changes in the microscopic morphology of GNs, ODA-GNs, and ODA-GNs/Polymer 1 before and after compounding can be observed using scanning electron microscopy.

To further observe the change in the internal structure of Liaohe heavy oil before and after adding additives and analyzing its influence, scanning electron microscopy was taken. Respectively, a small amount of heavy oil added with comb polymer, heavy oil added with ODA-GNs/Polymer 1, and heavy oil without viscosity reducer were taken and placed in a small porcelain boat. They were calcined at a high temperature of 800 °C in a tube furnace under an argon atmosphere for 12 h. Observation and analysis were carried out using SEM with a high magnification of 6000.

2.9. Viscosity Reduction Effect Evaluation Test

The viscosity of the treated heavy oil was measured using a Physica MCR 301 rotational rheometer according to the China Petroleum Industry Standard (Standard No. SY/T0520-2008) [38,39]. The heavy oil was placed in the thermostat at 80 °C for 12 h to eliminate the thermal history of the heavy oil. Different masses of viscosity reducer were added to toluene to prepare different concentrations of viscosity reducer solution. Then, 20 g of Liaohe heavy oil was weighed into a 25 mL small beaker, and 2 g of viscosity reducer solution was added to the heavy oil. At the shear rate of 10 s^{-1} , the viscosity of the heavy oil was tested between 25 °C and 70 °C. The viscosity reduction performance of the prepared viscosity reducer was evaluated based on the viscosity reduction rate and the net viscosity reduction rate.

3. Results

3.1. Graphene Micromorphology Analysis

Figure 2 displays the microscopic morphology of graphene raw materials, GNs, GNs treated by ball milling, ODA-GNs, and ODA-GNs/Polymer 1. As shown in Figure 2a, the graphite material was thick and tightly stacked, showing a totally opaque state. From Figure 2b, after exfoliation, the stack structure of the GNs became loose and showed a translucent state, which was in sharp contrast with the graphite raw material. It showed that the graphite material was effectively separated. As shown in Figure 2c, the GNs were modified by ball milling with the addition of ODA. It was observed that the stacking structure was further loosened, and the size of the graphene was reduced. But the flake structure

was fully preserved, resulting in lipophilic graphene ODA-GNs with strong orientation. As shown in Figure 2d, the graphene with a flake structure in ODA-GNs/Polymer 1 prepared by solvent blending was mostly encapsulated on the surface of the polymer.



Figure 2. The microscopic morphology of (**a**) graphene raw materials, (**b**) GNs, (**c**) ODA-GNs treated by ball milling, and (**d**) ODA-GNs/Polymer 1.

3.2. Fourier-Transform Infrared Spectra

Infrared structure characterization of three polymers, ODA-GNs, and ODA-GNs/ Polymer 1 are shown in Figure 3. It could be seen from the infrared spectrum of the polymer in Figure 3a that the symmetric and asymmetric absorption peaks of methylene (–CH₂–) in the long alkyl carbon chain of octadecyl methacrylate and the absorption peak of tensile vibration of the benzene ring skeleton were displayed. The vibration absorption peak of the amide group in polymer 1 appeared at 3447 and 1723 cm⁻¹. The absorption peak of carboxyl (–COOH) in fumaric acid in polymer 2 appeared in the broad peak at about 3560 cm⁻¹. The absorption peak of maleic anhydride in polymer 3 (O=C–O–C=O) appeared at 1784 cm⁻¹. Figure 3a showed that the three polymers were successfully prepared.

It can be seen from the infrared spectrum of Figure 3b that the ODA-GNs/Polymer 1 had a stretching vibration peak related to the amide group at 3465 and 1730 cm⁻¹, indicating that the amide group exists in the product. The $-CH_2$ - symmetric and asymmetric absorption peaks of the long alkyl chain in the polymer were at 2922 and 2850 cm⁻¹. The absorption peaks of the benzene ring appeared at 1599 and 1450 cm⁻¹. The absorption peak of C–OH in GNs appeared at 1235 cm⁻¹, and 1050 cm⁻¹ was the absorption peak of the carbon nitrogen electrostatic adsorption force between $-NH_2$ - in the modifier ODA and GNs. In conclusion, the ODA-GNs/Polymer 1 nanocomposite viscosity reducer was successfully prepared.



Figure 3. FTIR spectral analysis of (**a**) three polymers and (**b**) samples of GNs, ODA-GNs, and ODA-GNs/Polymer 1.

3.3. Analysis of Viscosity Temperature and Rheological Properties of Heavy Oil

The viscosity of Liaohe heavy oil at different temperatures is shown in Figure 4a. Under the same shear rate, the viscosity of heavy oil decreased rapidly with the increase in temperature and then tended to be flat. The viscosity of heavy oil increased sharply when the temperature was lower than 45 °C, and 45 °C could be regarded as the turning point of sudden change of viscosity.



Figure 4. (a) Viscosity of Liaohe heavy oil at different temperatures (shear rate of 10 s^{-1}) and (b) viscosity of heavy oil under shear rate step conditions.

The shear rate step experiment in Figure 4b shows that the viscosity of heavy oil did not change with the change of shear rate when it was close to the waxing point or higher, and it showed Newtonian fluid behavior. Therefore, the subsequent viscosity temperature curves all used constant temperature drop to measure the instantaneous viscosity.

3.4. Thermogravimetric Analysis and Dispersion Stability Test

The ODA-GNs and ODA-GNs/Polymer 1 were determined using thermogravimetric analysis. The results are shown in Figure 5. From Figure 5, in the temperature range of 250–380 $^{\circ}$ C, the ODA molecules in the ODA-GNs underwent a thermal decomposition reaction, and the mass decreased rapidly. In the temperature range of 380–640 $^{\circ}$ C, the

mass decline trend tended to go from gentle to stable. This phenomenon indicated that the ODA molecule has gradually completed decomposition, and the weight loss rate in this process was about 12.3%, indicating that the ODA-GNs were successfully prepared. It can be seen from Figure 4 that the weight loss rate of ODA-GNs/Polymer 1 was 34.6% in the temperature range of 260–800 °C due to the continuous decomposition of organic molecular chains under high temperatures. This showed that the content of organic groups on the surface of GNs was improved after compounding and modification, thereby enhancing its lipophilicity. Through comparison, it was found that the weight loss rate of ODA-GNs/Polymer 1 was smaller than that of ODA-GNs in the temperature range of 250–380 °C, and the existence of modified graphene improved the stability of the polymer chain sealing end termination. This showed that the thermal stability of ODA-GNs/Polymer 1 was further improved.



Figure 5. (a) Thermogravimetric analysis and (b) dispersion stability test.

Using toluene as a solvent, the dispersion stability of the GNs, ODA-GNs, and ODA-GNs/Polymer 1 were determined with a sedimentation experiment, and the observed phenomenon is shown in Figure 5. The presence of ODA groups on the surface of GNs weakens the van der Waals force between GNs, ffe presence of the ODA group is not only conducive to the composite of GNs but also improves the lipophilicity of GNs. The results of the sedimentation experiment showed that the comb polymer was successfully attached to the surface of ODA-GNs, and the ODA-GNs/Polymer 1 was successfully prepared.

3.5. Viscosity Reduction Performance

As shown in Figure 6, although the polar monomers in the polymer were different, their influence on the viscosity reduction in the comb polymer had the same trend. With the increase in the proportion of polar monomers in the polymer, the viscosity reducing energy first increased and then decreased. The strong polar group in the polar monomer destroyed the hydrogen bond in the heavy oil accumulation structure and formed a stronger hydrogen bond with the polar group in the structure, releasing the light component wrapped in the aggregate and resulting in a reduction in the viscosity of the heavy oil. With the increase in polar monomers in the polymer, the solubility of a comb polymer in heavy oil decreased, which led to a decrease in viscosity reduction.

The influence of different polar monomers on the viscosity-reducing ability of a comb polymer is shown in Figure 6. In comparison, the viscosity reduction effect of polymer 1 was higher than that of polymers 2 and 3, as the stronger the polarity of the group, the stronger the ability to form hydrogen bonds with heavy oil. The order of polarity from high to low was carboxyl, amido, and anhydride. The fumaric acid contained two carboxyl groups, but its viscosity reduction effect was slightly lower than that of

acrylamide. The reason may be that the strong polarity of fumaric acid affected the oil solubility and permeability of the viscosity reducer, thereby affecting the viscosity reduction effect. Structurally, acrylamide has a smaller steric resistance than fumaric acid, which can support the polymer framework, so as to better disperse the heavy oil aggregates, reduce the viscosity of heavy oil, and improve the fluidity of heavy oil. Through the analysis and research on the viscosity-reducing properties of comb polymers 1, 2, and 3, considering their viscosity reduction rate and net viscosity reduction rate, the comb polymer 1 with the monomer ratio of 9.0:1.5:0.3 was selected to be combined with the ODA-GNs to prepare the ODA-GNs/Polymer 1 viscosity reducer.



Figure 6. (a) Effect of acrylamide, (b) fumaric acid, and (c) maleic anhydride on viscosity reduction.

The viscosity reduction effects of different graphene to ODA ratios are shown in Table A1, and the viscosity reduction effect increased and then decreased as the ratio of ODA increased. The reason for this phenomenon may be due to the fact that ODA-modified graphene is a single-layer physical adsorption modification. When the amount of ODA was too much, ODA formed an unstable and easily desorbed multilayer covering structure on the graphene surface. The ODA under the resultant desorption may form a competitive relationship with the ODA-GNs/Polymer 1, which reduces the viscosity reduction effect of the material. The best viscosity reduction effect of the material was achieved when GNs:ODA = 5:1, and all subsequent discussions were carried out under this condition. The effects of the ratios of ODA-GNs to polymer 1 on the viscosity reduction effect are shown in Table A2, where the viscosity reduction performance of the materials showed a tendency to first increase and then decrease as the ratio of polymer 1 increased. When the content of polymer 1 was too much, it led to polymer 1 being more easily desorbed on the surface

of ODA-GNs, and the percentage of molecular structure with viscosity reduction effect was reduced, resulting in a decrease in the viscosity-reducing effect. The best viscosity reduction effect of the materials was achieved when ODA-GNs:Polymer 1 = 1:1, and the subsequent discussions were carried out under this condition.

The viscosity reduction performance of ODA-GNs/Polymer 1 in heavy oil was investigated at different viscosity reducer concentrations, and the results are shown in Figure 7. In the range of 0–1000 ppm, the viscosity reduction effect increased first and then decreased. When the dosage was lower than the optimal dosage, the separation ability of asphalt aggregates was weak due to the low content of active components. When the dosage was higher than the optimal amount, the effective components were surplus, and some effective molecules interacted with resins and asphaltenes, or the molecules themselves intertwined and agglomerated, which reduced the fluidity of heavy oil. Figure 7 showed that when the dosage of ODA-GNs/Polymer 1 was 400 ppm, the viscosity reduction rate and the net viscosity reduction rate were 95.7% and 80%, respectively, which reduced the viscosity of heavy oil from 2426.7 mPa·s to 484.6 mPa·s. Compared with polymer 1, the net viscosity reduction rate of ODA-GNs/Polymer 1 was about 5% higher, and its dosage was reduced by 200 ppm. The main reason was that ODA modification improved the compatibility of GNs and comb polymers, and the two had a synergistic effect, which further improved the viscosity reduction effect of ODA-GNs/Polymer 1.



Figure 7. Different dosages of Polymer 1 and ODA-GNs/Polymer 1's (**a**) viscosity reduction and (**b**) net viscosity reduction.

In addition, Figure 8 depicts viscosity curves of heavy oil, 10% toluene solvent of heavy oil, 400 ppm of polymer 1, and ODA-GNs/Polymer 1 at different temperatures. Compared with polymer 1, the ODA-GNs/Polymer 1 further reduced the viscosity of the Liaohe heavy oil. At room temperature, the net viscosity reduction rate increased to 80%, which reduced the viscosity of heavy oil from 2426.7 mPa·s to 484.6 mPa·s. The graphene nanocomposite viscosity reducer performance was superior to the polymer viscosity reducer, which may be due to the following two reasons. Graphene is a lamellar structure, similar to the structure of colloidal asphaltene. When colloidal asphaltene aggregates loosen the lamellar structure at a high temperature, graphene could be inserted into the aggregate lamellar layer through the π - π interaction, preventing the aggregation of colloidal asphaltene when the temperature was lowered, thus lowering the viscosity of the heavy oil. The nanoparticles have the function of heterogeneous nucleation, and the wax crystals could be precipitated with the nanomaterials as the nuclei, which enhanced the dispersion of the wax crystals and thus enhanced heavy oil fluidity.



Figure 8. (a) Viscosity temperature curve of heavy oil before and after adding additives; (b) effect of different temperatures on net viscosity reduction.

In Figure 8b, when the temperature was higher than 45 °C, the net viscosity reduction rate of polymer 1 decreased more than that of the ODA-GNs/Polymer 1. Moreover, at 45 °C, the net viscosity reduction rate of ODA-GNs/Polymer 1 could still reach 75.4%, which was still very obvious in reducing the viscosity of Liaohe heavy oil. This was mainly because the molecular coils of the macromolecule comb polymer will expand with the increase in temperature, which will make the heavy oil have a negative effect on viscosity, and the fluidity becomes poor. Whereas ODA-GNs/Polymer 1 did not suffer from such problems.

3.6. Heavy Oil Microtopography Analysis

Figure 9 displays the microscopic morphology of heavy oil without viscosity reducer, with polymer 1 added, and with ODA-GNs/Polymer 1 added. Figure 9a shows that the morphology of the heavy oil sample without viscosity reducer was irregular and repeatedly stacked large layered particles, making the edge of the heavy oil morphology rough. Figure 9c compares with Figure 9b, where the morphology of the heavy oil sample after adding the ODA-GNs/Polymer 1 was more regular than that of the heavy oil sample after adding the polymer 1. The degree of the particle blocky and layered accumulation was smaller, and the particle blocky surface was smoother as Graphene lamellae underwent a lamellar slip phenomenon and π - π interaction in resin and asphaltene aggregates. Carrying the polymer into the aggregates while disassembling the aggregates ensured that the reactive groups in the polymer formed new hydrogen bonds with the resin and asphaltene or were embedded in the aggregate accumulation, and at the same time, the surface of the block particles was smoother, preventing resin and asphaltene from gathering again and reducing the viscosity of heavy oil.



Figure 9. Cont.



Figure 9. The microscopic morphology of heavy oil (**a**) without viscosity reducer, (**b**) with polymer 1 added, and (**c**) with ODA-GNs/Polymer 1 added.

4. Conclusions

In this paper, the effects of a graphene nanocomposite viscosity reducer on the rheological behavior and viscoelasticity of heavy oil at different dosages and temperatures were studied. The graphene nanocomposite viscosity reducer was successfully prepared by compounding ODA-GNs and polymer 1 using the solution blending method. When the amount of ODA-GNs/Polymer 1 was 400 ppm, the best viscosity reduction effect was achieved, its viscosity reduction rate was 95.7%, and the net viscosity reduction rate was 80%. Compared with polymer 1, the net viscosity reduction rate of the polymer nanocomposites was increased by about 7%. The action mechanism of polymer nanocomposites may be that the addition of ODA-GNs further breaks up the aggregate of resin and asphaltene and participates in the wax nucleation process. While the ODA-GNs break up the aggregate, the light components coated in it are released, thus further reducing the viscosity of heavy oil. Under the above optimal composite conditions, the ODA-GNs/Polymer 1 were characterized using FTIR, dispersion stability, and SEM tests, and it was found that the ODA-GNs/Polymer 1 had a strong lipophilicity, dispersion stability, and thermal stability. This study provided a theoretical and practical basis for the research and development of a novel nanocomposite viscosity reducer.

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Abbreviations

FTIR	Fourier-transform infrared spectroscopy
TGA	thermogravimetric analysis
SEM	scanning electron microscopy
GO	graphene oxide
PMMA-GO	poly (methyl methacrylate) -graphene oxide
EVAL	ethylene-vinyl alcohol copolymer
GNs	graphene nanosheets
ODA	octadecylamine
ODA-GNs	ODA modified graphene nanosheets
ODA-GNs/Polymer 1	graphene-based comb polymer 1
TR	transmission mode

Appendix A

Table A1. Influence of different amounts of ODA on viscosity reduction.

GNs/g	ODA/g	Viscosity/mPa·s	Viscosity Reduction/%	Net Viscosity Reduction/%
5.0	0.5	534.2	95.3	78.0
5.0	1.0	484.6	95.7	80.0
5.0	2.0	588.4	94.8	75.8
5.0	4.0	666.3	94.1	72.5

Table A2. Effect of different ratios of ODA-GNs and polymer 1 on viscosity reduction.

ODA-GNs/mg	Polymer 1/mg	Viscosity/mPa·s	Viscosity Reduction/%	Net Viscosity Reduction/%
500	250	512.8	95.5	78.9
500	500	484.6	95.7	80.0
500	750	566.6	95.0	76.7

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