Electronic Supplementary Material (ESI) for Environmental Science: Nano.

Supporting Information for:

Efficient removal of viscous crude oil by a super-hydrophobic polystyrene/carbon black foam with photo-thermal conversion

Di Qiao [a, b], Yihao Guan [a, b], Dan Zhang [a, b], Bingqian Bi [a, b], Yiming Li [a, b, *]

[a] *Frontiers Science Center for Deep Ocean Multispheres and Earth System/Key Laboratory of Marine Chemistry Theory and Technology, Ministry of Education, Ocean University of China, 266100, Qingdao, P.R. China*

[b] *College of Chemistry & Chemical Engineering, Ocean University of China, Qingdao 266100, P.R. China*

[*] Corresponding author.
E-mail address: liym@ouc.edu.cn (Y.M. Li)

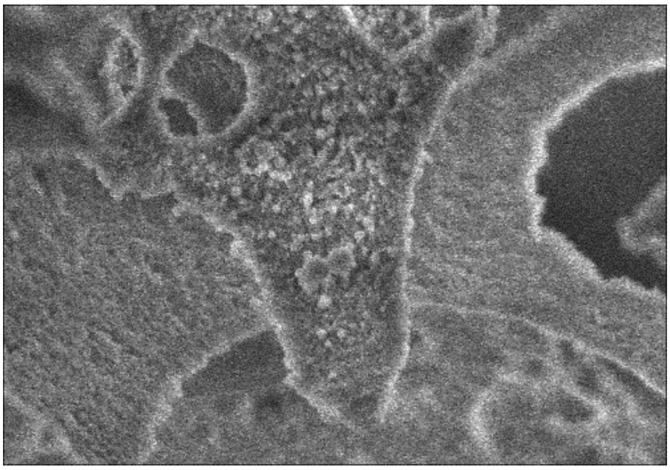

Fig.S1 Evolution of foam surface morphology after embedding carbon black.

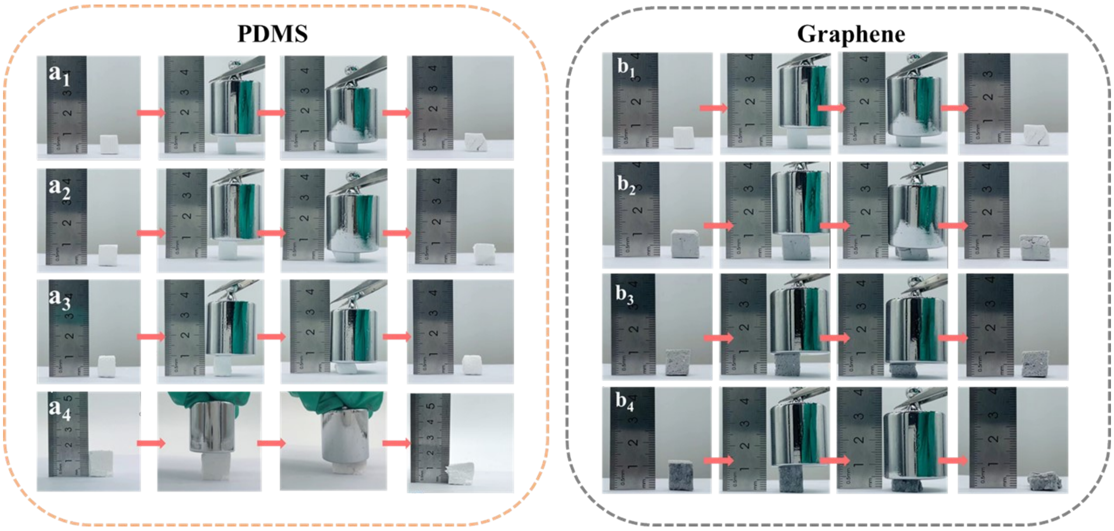

Fig.S2. Mechanical properties of foams. a_1-a_4 PDMS contents were 0 wt%, 0.15 wt%, 0.30 wt%, and 0.45 wt%, respectively; b_1-b_4 graphene contents were 0 wt%, 0.0058 wt%, 0.012 wt%, and 0.017 wt%, respectively.

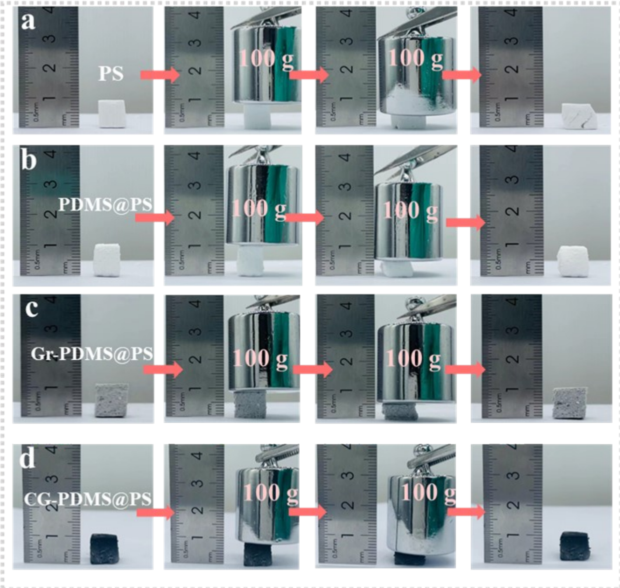

Fig.S3. Mechanical properties of foam. (a) Original PS, (b)PDMS/PS, (c) Gr-PDMS@PS, (d) CG-PDMS@PS.

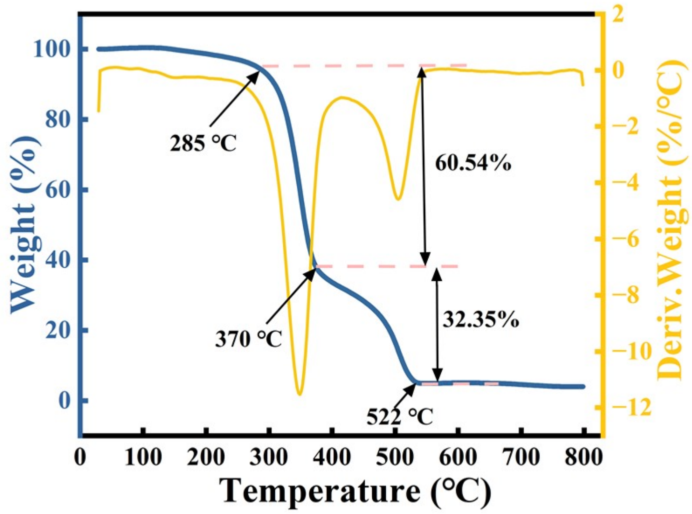

Fig.S4. TGA of CG-PDMS@PS foam.

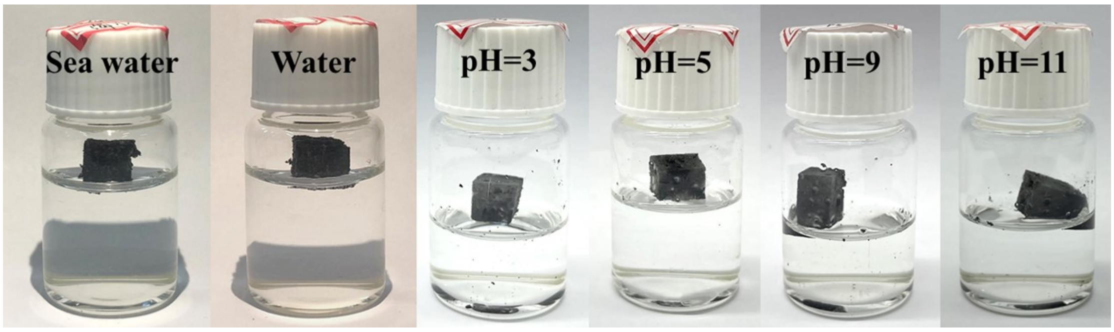

Fig.S5. Wettability of CG-PDMS@PS after soaking in different solutions for 7 days.

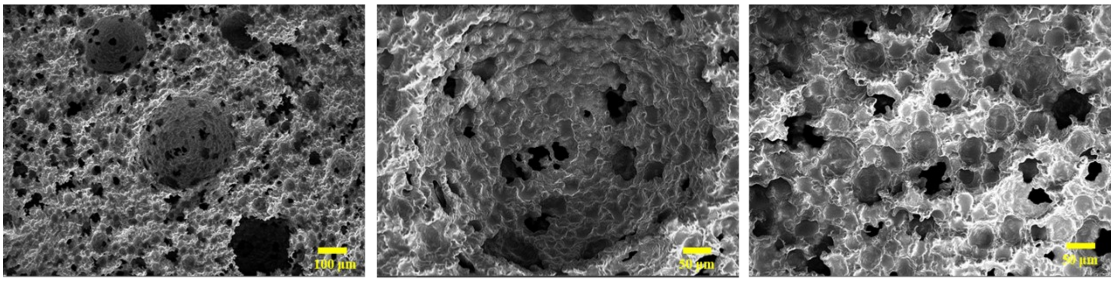

Fig.S6. SEM of dry foam filled with oil.

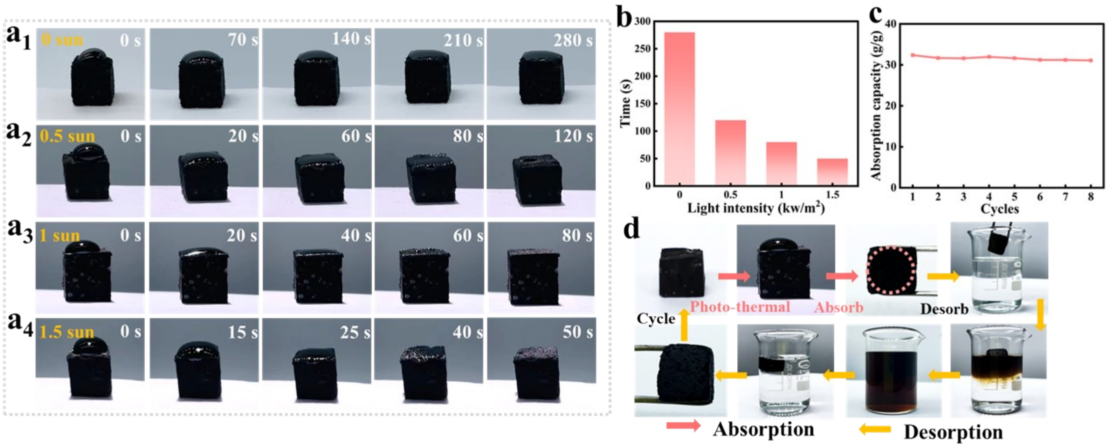

Fig.S7. (a) Optical images of the permeation process of CG-PDMS@PS on crude oil under different illumination conditions. (b) Permeation rate statistics. (c) Crude oil absorption cycle of CG-PDMS@PS. (d) Schematic diagram of the adsorption cycle of CG-PDMS@PS.

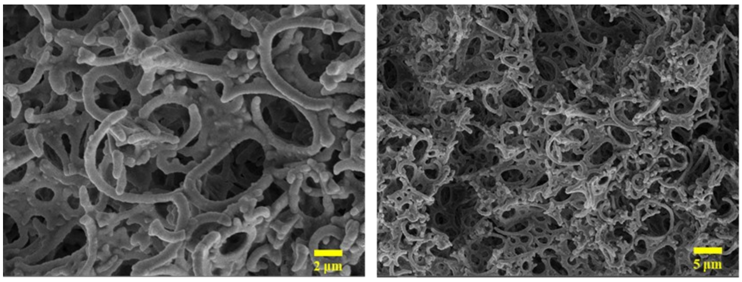

Fig.S8. SEM of the foam after the cycles.

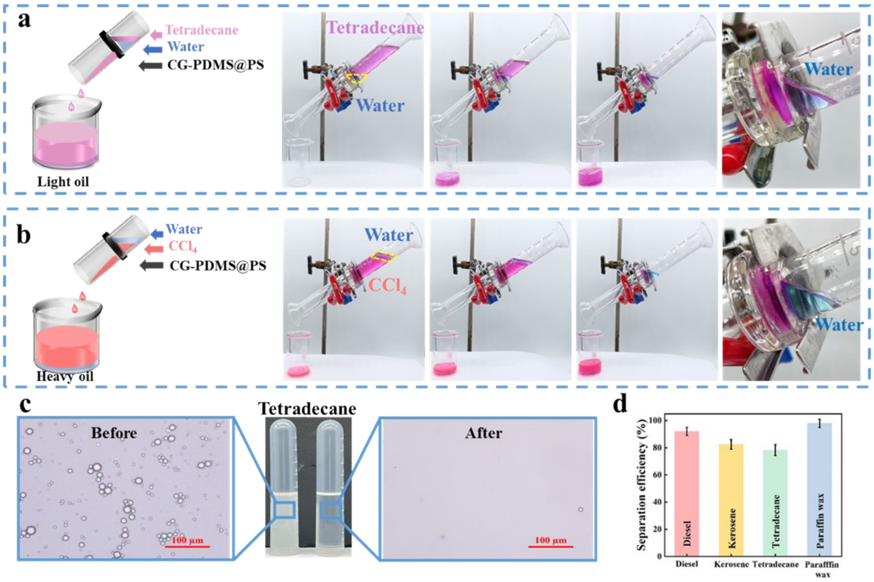

Fig.S9. Oil-water mixture separation capacity of CG-PDMS@PS foam. (a) Separation of light oil (tetradecane dyed with an oil soluble dye) mixture. (b) Separation of heavy oil (CCl4 dyed with an oil soluble dye) mixtures. (c) Comparison diagram of W/O emulsion before and after separation. (d) Separation efficiency of various W/O emulsion.

CG-PDMS@PS foam is uniquely suited for oil-water separation applications due to its exceptional super-hydrophobic properties, which enable it to effectively repel water and attract oil. Oil-soluble dye was added to CCl_4 (Fig.S9a) and tetradecane (Fig.S9b) to mimic heavy and light oil. To create an oil-water mixture, 2 mL water with methylene blue was combined with 8 mL of oil that contained an oil-soluble dye. The CG-PDMS@PS foam (5 mm) was placed in the filter interlayer and the mixture was allowed to separate naturally by gravity. Consequently, the foam's water-repellent features ensured that the water was effectively trapped in the filter cup, while the oil was collected in the beaker. Therefore, the CG-PDMS@PS foam's ability to separate oil and water was confirmed.

Except for high-viscous crude oil, the presence of oil-water emulsions in oily wastewater significantly exacerbates the challenges involved in treating oily wastewater. The super-oleophilic and porosity of CG-PDMS@PS foam hold significant potential for emulsion separation. The efficient and straightforward separation of emulsions continues to be a major challenge, especially when surfactants are present. As super-hydrophobic materials have proven to be effective in emulsion separation, we conducted experiments with CG-PDMS@PS foam for this purpose. A piece of foam (5 mm) was utilized as a filter without any pretreatment. Fig.9c showed the oil droplets in emulsion before and after separation. The initial emulsion consisting of tetradecane was opaque, but after filtration through the foam, the liquid turned transparent. In subsequent experiments, we tested the foam's effectiveness in separating different types of oils and observed that it was capable of separating all types of emulsions effectively (Fig.S10).

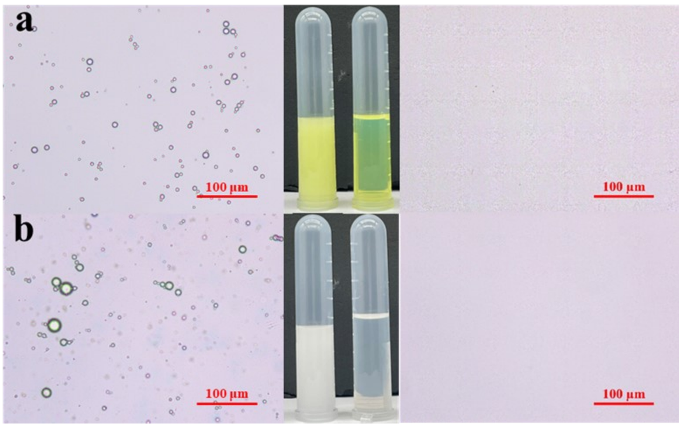

Fig.S10. Performance and separation mechanism of W/O emulsions. (a)diesel. (b) paraffin wax.

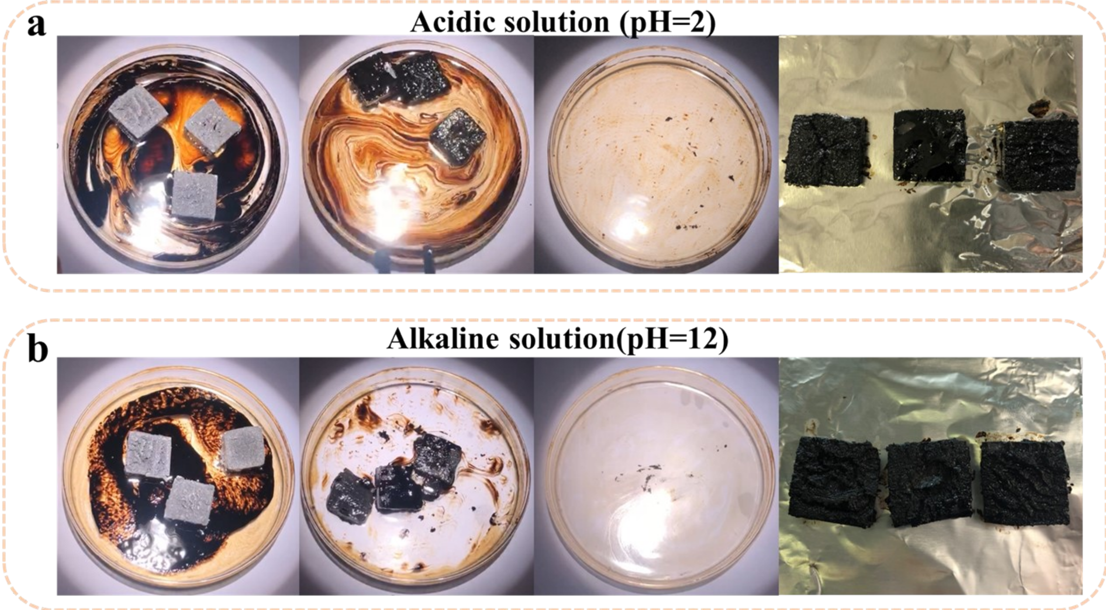

Fig. S11 CG-PDMS@PS foam recovering the crude oil on the different water under 1.0 kW/m^2.

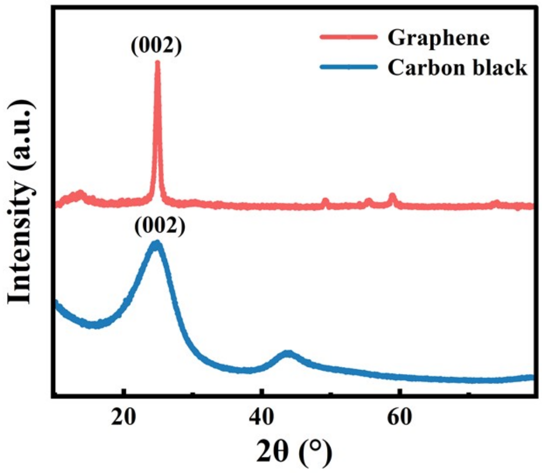

Fig.S12. XRD of carbon black and graphene.

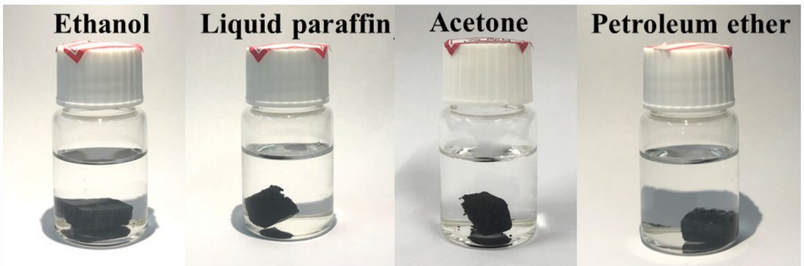

Fig.S13. The solvent resistance of CG-PDMS@PS.

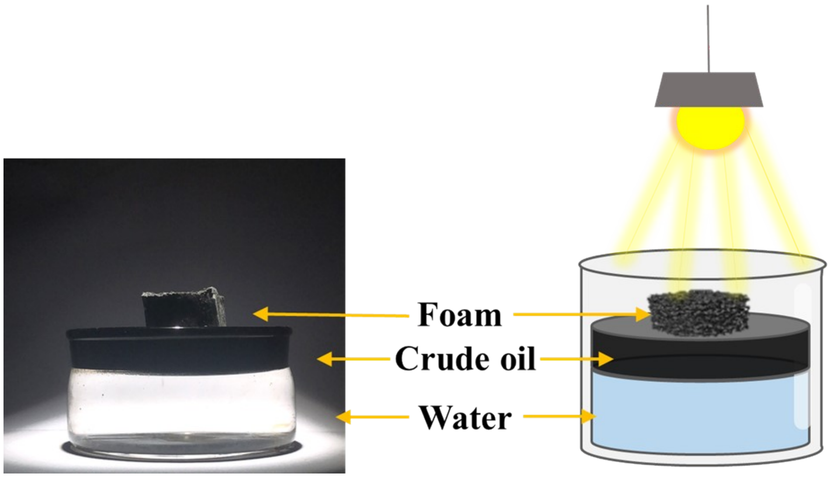

Fig.S14. Schematic diagram of foam adsorption process.

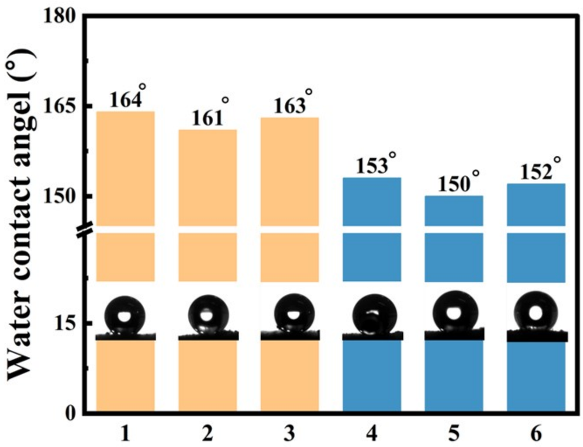

Fig. S15. The contact angels between foam and water, foam and seawater. From 1 to 3 are water, from 4 to 6 are seawater.

Table. S1. Pore information of PS and CG-PDMS@PS.

	Total intrusion volume (mL/g)	**Total pore area (m^2/g)**	**Median pore diameter (μm)**	**Average pore diameter (μm)**	**Porosity (%)**
PS	27.9	8.1	7.3	15.7	84.0
CG-PDMS@PS	29.0	14.9	2.4	17.8	92.2

Table.S2. Fitting curve of adsorption velocity of five different oils.

Oil type	**a**	**b**	**R^2**
N-Tetradecane	$0.023 \pm 7.05 \times 10^{-4}$	6.42×10^{-4}	0.999
Paraffin liquid	0.0203 ± 0.001	2.97 ± 0.0318	0.999
Soybean oil	$0.0736 \pm 2.91 \times 10^{-4}$	$3.03 \pm 1.75 \times 10^{-16}$	1
Olive oil	$0.128 \pm 6.87 \times 10^{-16}$	$3.41 \pm 2.36 \times 10^{-15}$	1
Engine oil	$0.427 \pm 1.9 \times 10^{-15}$	$3.28 \pm 1.97 \times 10^{-15}$	1

Table.S3. Detail information of oils.

Oil type	Viscosity (mPa s)	Density (g cm^{-3})	Source of oil
Diesel fuel	2	0.84	Local gas station
Paraffin	15	0.77	Sinopharm Chemical Reagent Co., Ltd. (China)
Tetradecane	2	0.75	Sinopharm Chemical Reagent Co., Ltd. (China)
Olive oil	61	0.84	Enwode Jingdong supermarket (China)
Engine oil	100	0.83	Enwode Jingdong supermarket (China)
Soybean oil	48	0.86	Enwode Jingdong supermarket (China)
Dimethicone	100	0.98	Sinopharm Chemical Reagent Co., Ltd. (China)

Table.S4. Details of crude oil[1].

No.	Crude oil	Density at 20 ^{o}C (g/cm^3)
1	Ashtart	0.8730
2	Ostra	0.9433
3	Bijupira	0.8744
4	Castilla	0.9774
5	Escalante	0.9043
6	Fionaven	0.8750
7	Heidrun	0.9036
8	Oriente	0.9115
9	Patos Marinza	0.9902
10	Upper Zakum	0.8510
11	Vasconia	0.8968

Reference

1. D. Stratiev, I. Shishkova, A. Nedelchev, K. Kirilov, E. Nikolaychuk, A. Ivanov, I. Sharafutdinov, A. Veli, M. Mitkova, T. Tsaneva, N. Petkova, R. Sharpe, D. Yordanov, Z. Belchev, S. Nenov, N. Rudnev, V. Atanassova, E. Sotirova, S. Sotirov and K. Atanassov, Investigation of Relationships between Petroleum Properties and Their Impact on Crude Oil Compatibility, *Energy & Fuels*, 2015, **29**, 7836-7854.