



# Functionalized graphene nanoplatelet-nanofluids for solar thermal collectors



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## ABSTRACT

This work reports on the characterization of new heat transfer fluids consisting in dispersions of functionalized graphene nanoplatelets (fGnP) in Havoline XLC Premixed 50/50 base fluid. Optical, stability and rheological properties have been considered, with the aim to assess the nano-suspension potential for volumetric direct solar absorbers. Nanoparticle addition leads to a considerably higher sunlight absorption with respect to the pure base fluid even at the investigated extremely low concentrations (0.005% and 0.05% wt). Rheology tests show a Newtonian behaviour in the shear rate range between 10 and 1000 s<sup>-1</sup>. Viscosity and thermal conductivities do not significantly increase with respect to the pure base fluid, showing that these new fluids will not require additional pumping powers to flow.

## 1. Introduction

Conventional solar collectors operating at low-mid temperatures consist of a sunlight absorbing coating deposited on a solid surface exchanging heat with a working fluid. To reduce energy losses due to thermal re-radiation by the heated absorber, a vacuum insulation of the absorbing surface is typically required. This scheme can be significantly simplified by the use of a dark fluid working both as volumetric light absorber and as heat exchanger. The first idea of a direct-absorption solar collector (DASC) dates back to 1975 and employed India ink [1]. However, organic dyes rapidly degrade when exposed to solar radiation. Thus in this field, the development of nanotechnology, allowing both the creation of new nanoparticles and their stable suspension in fluids (nanofluids), has a high potential. In fact, nanofluids have given a new pulse to DASC concept, as shown by the impressive growth of studies on this topic [2,3]. A large variety of nanoparticles and base fluids has been investigated in the literature. Among the investigated materials, the family of carbon-based nanostructures has emerged as particularly promising. From carbon black [4] to graphite [5], as well as carbon nanostructures e.g. single, multi-wall and functionalized nanotubes [5–8], carbon nanohorns [4,9–11], many nanofluids containing nanoparticles of carbon allotropes have been studied for solar energy applications. Graphene is one of most intriguing carbon allotropes [12]. Graphene nanoplatelets or nanosheets consist of small flakes of several-layer stacked graphene that partially inherit the good properties of

graphene with much lower production costs. The literature reports on the dispersion of relatively large graphene nanoplatelets in water for solar applications [13]. Nevertheless, to achieve a better dispersion process in most aqueous or organic solvents, besides to reduce as possible the particle dimensions, it is necessary to chemically modify the (hydrophobic) graphene surface, e.g. by oxidation [14] or functionalization by means a polycarboxylate chemical modification, like for nanoparticles investigated in the present study.

In this work, we present new heat transfer fluids with thermo-physical properties nano-enhanced by the dispersion of commercial functionalized graphene nanoplatelets in a novel base fluid, widely used up to now as coolant in the engine industry, and particularly in wind turbine cooling. Initially, we focused the attention on the stability of nanofluids determining zeta potential and pH values to propose nanofluids with enhanced capabilities. Optical properties were determined in the perspective to evaluate strong and weak points of this nanofluid for solar energy. Furthermore, it was performed a rheological analysis of the dispersions, key issue to understand the flow behaviour of these new fluids in this application.

## 2. Materials and methods

The heat transfer fluid, Havoline XLC Premixed 50/50, is a commercial mixture at 50:50% vol of Havoline XLC (93% wt ethylene glycol) and water, Hav/W 50/50. Sodium dodecyl benzene sulphonate,

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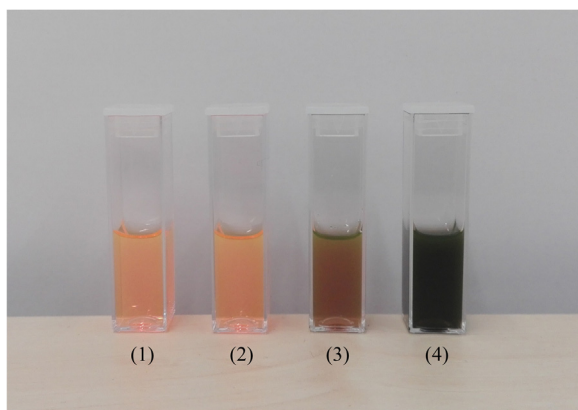


Fig. 1. Pictures of the studied fluids: (1) Hav/W 50/50, (2) (Hav/W 50/50 + 0.125% wt SDBS), (3) 0.005% wt nanofluid and (4) 0.05% wt nanofluid.

SDBS, supplied by Sigma-Aldrich (Louis, Missouri, USA), was added to Hav/W 50/50 at the optimized mass fraction of 0.125% to improve the long-term stability of the produced nanofluids [15]. Polycarboxylate chemically modified graphene nanoplatelets, fGnP, were supplied by NanoInnova Technologies S.L. (Madrid, Spain). SEM images of a drop of powder dispersed in analytical grade methanol and placed over a silica support were obtained by a field emission scanning electron microscope JSM-6700F (JEOL, Tokyo, Japan) with Yttrium Aluminum Garnet detector at 10.0 kV operating accelerator voltage.

Dispersions of fGnP in Hav/W 50/50 using SDBS as surfactant have been prepared following a two-step method. The amounts of each component necessary to obtain the desired nanoadditive mass concentrations 0.005% and 0.05% wt were weighted in a CPA225 electronic balance (Sartorius AG, Goettingen, Germany). The dispersions were firstly mechanically stirred and then sonicated by using an ultrasonic bath (Ultrasounds, JP Selecta S.A., Barcelona, Spain) for 240 min with a maximum sonication power of 200 W and a frequency of 20 kHz. Fig. 1 shows the nanofluids at the investigated concentrations.

Regarding material employed in the validation between literature data and values obtained in this work, distilled water and 99.5% purity ethylene glycol (Honeywell, Seelze, Germany) were used.

The zeta potential of the studied nanofluids was analysed in the temperature range from 293.15 to 323.15 K with a 10 K step through dynamic light scattering technique by using a Zetasizer Nano ZS (Malvern Instruments Ltd, Malvern, United Kingdom). pH values were measured by a Sension+ PH3 pH-meter (Hach, Colorado, United States) with a pH electrode Sension+ 50 11 T.

Optical transmittance spectra at room temperature in the range 300–2700 nm have been measured using a double-beam UV–VIS spectrophotometer Lambda 900 (Perkin Elmer Inc., Massachusetts, USA). Effective thermal conductivities were experimentally determined at 293.15 K by means of a KD2 Pro thermal analyzer (Decagon Devices Inc., Pullman, USA) equipped with a KS-1 probe (60 mm long and 1.3 mm diameter). The expanded uncertainty (0.95 level of confidence) of these measurements is 3% [16]. Densities were measured in the temperature range from (288.15 to 313.15) K with a 5 K step through a densimeter DMA 500 (Anton Paar, Graz, Austria). The expanded uncertainty (0.95 level of confidence) of these measurements is 0.1% [17]. Finally, the rheological behaviour was analysed in the temperature range from (293.15 to 323.15) K with a 10 K step by means of a rotational Physica MCR 101 rheometer (Anton Paar, Graz, Austria) equipped with a cone-plate geometry CP50-1 (49.974 mm diameter,  $-0.993^\circ$  cone angle and 102  $\mu$ m cone truncation). The expanded uncertainty (0.95 level of confidence) of these measurements is 3% [18].

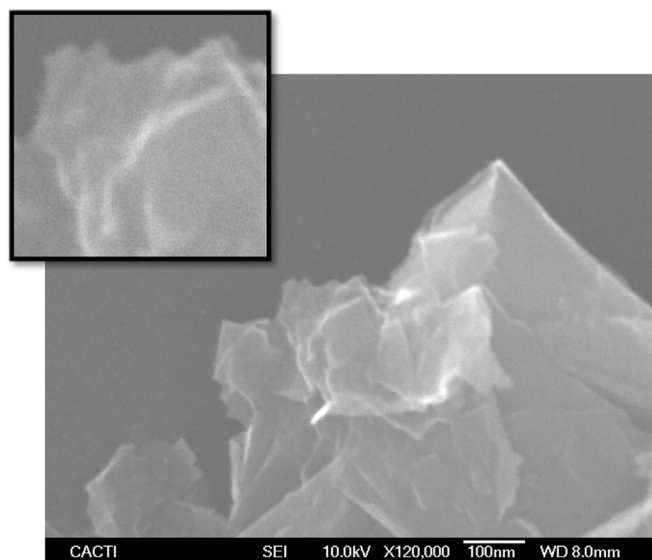


Fig. 2. SEM image of polycarboxylate chemically modified graphene nanoplatelets at 120000  $\times$  magnification.

### 3. Results and discussion

Fig. 2 shows a SEM picture of the dry nanopowder, in which it is possible to observe various agglomerated nanoparticles. The inset in Fig. 2 is a higher magnification area showing that the structure of nanoparticles consists of stacked graphene layers. Complete SEM analysis allows confirming that employed polycarboxylate chemically modified graphene nanoplatelets consist of 5–10 graphene stacks with 2–3 nm height per layer and main dimensions larger than 200 nm.

According to the literature, absolute zeta potential values higher than 30 mV denote good dispersion stabilities in aqueous solutions due to the stronger electrostatic repulsions among particles [19]. Since the zeta potential of the samples prepared by directly dispersing the graphene in water did not reach this value, sodium dodecyl benzene sulfonate, SDBS, was added to the base fluid. The negatively charged part of the SDBS is expected to be absorbed onto the graphene surface forming an inner Helmholtz layer. This surfactant layer increases electrostatic repulsion between graphene nanoplatelets and improves dispersion stability [20]. In this study, different mass fractions of SDBS in the range between 0.025% and 0.25% in relation to the amount of Havoline XLC Premixed 50/50 were considered. A mass concentration of 0.125% was selected for the preparation of the two fGnP concentrations because it ensures the stability in all cases with the minor modification of the commercial coolant properties. Fig. 3 shows the zeta potential of the finally studied fGnP nanofluids dispersed in the base fluid containing the cited mass fraction of SDBS, in the temperature range from 293.15 to 323.15 K. As it can be observed, zeta potential values are equal or lower than  $-30$  mV, for 0.005% wt concentration at 293.15 K this property exhibiting a value close to that limit. The zeta potential values do not show a clear trend with the temperature, being all of them in the range  $-30$  to  $-40$  mV with differences practically covered by the experimental uncertainty. It must also be remarked that the obtained pH-values for Hav/W 50/50, Hav/W 50/50 with SDBS, and the cited fGnP dispersions are practically equal ( $\sim 8.5$ ).

In Fig. 4 we show the extinction coefficient of fGnP suspensions and that of the base fluid in the spectral range 300–2700 nm, obtained from using the method described in [21]. In the same picture, the sunlight spectrum is also superimposed for reference (green curve). In the spectral region of high transparency of the base fluid, an addition of fGnP as low as 0.005% wt increases the extinction coefficient by more than four times in the interval 590–950 nm and by more than eight

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