



# Clean-up of oily liquids, fuels and organic solvents from the contaminated water fields using poly(propylene glycol) based organogels

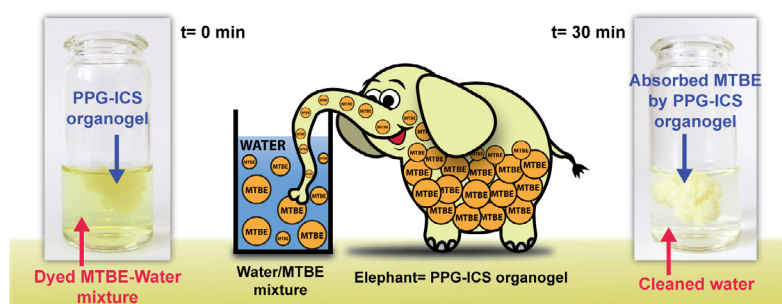
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## HIGHLIGHTS

- Spreading oily liquids can contaminate fresh water and soil.
- Highly absorptive PPG-ICS networks were successfully prepared for oily liquids.
- Outstanding MTBE cleanup from water bodies by PPG-ICS organogels.
- High rate of absorption and desorption operation.
- Effective reusable organogels at least for ten cycles.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Organogels with high and fast absorption properties, as well as reusability functions, were successfully prepared through the condensation reactions of different molecular weights of poly(propylene glycol)s (PPG)s and tris[3-(trimethoxysilyl) propyl] isocyanurate (ICS). The characterization of these organogels was determined by solid-state  $^{13}\text{C}$  and  $^{29}\text{Si}$  cross polarization magic angle spinning (CPMAS) nuclear magnetic resonance (NMR) and Fourier transform infrared spectroscopy (FTIR). Thermal gravimetric analysis (TGA) was used to describe the thermal behavior of the prepared organogels. Obtained organogels are reusable and can absorb a large amount of oils, such as dichloromethane (DCM), tetrahydrofuran (THF), methyl tert-butyl ether (MTBE), toluene, benzene, acetone, and gasoline. The capacity of the synthesized organogels did not change in either pure organic liquids or in organic liquid/water mixtures, these can be used at least ten times without loss of capacity.

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## 1. Introduction

Oil spillage is an inevitable incident during petroleum exploration, production, and transportation processes [1–4]. Various remediation methods have been proposed for oil and oil derivative organic solvents in an attempt to mitigate the catastrophic effects on marine life and aquatic systems such as bioremediation [5,6],

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chemical dispersants [7,8], absorbents [9,10], solidifiers [11], booms, skimmers, and in situ burning processes [12–14]. On the other hand, methyl tert-butyl ether (MTBE), a gasoline additive, has been accepted as a widespread and persistent groundwater and surface water pollutant, which is the second most frequently detected volatile organic compound in groundwater [15]. MTBE is considered to be toxic, skin and eye irritant; it can also affect the potable water quality by ruining its taste and odor [16]. Because of the high production grade of MTBE, it is often predicted that MTBE will be constantly found in the environment by virtue of

accidental fuel leakage during storage and transportation and, as mentioned above, this is also true of oil spillages. Also related to physicochemical features of MTBE is the fact that it has high water solubility, which can cause difficulty during MTBE removal from groundwater or surface water [17]. Remediation of MTBE from the water has been investigated by many scientists, who have used adsorption, the oxidation process, and biological treatment in their explorations; the removal of MTBE has raised significant concern [18,19]. Using the absorbents with hydrophobic features and porous structure for the removal of oil and volatile organic solvents such as MTBE, toluene, benzene, etc. is considered to be one of the most efficient, economical, and ecologically friendly, attracting a great deal of attention by scientist in the field of oil-water clean-up in the last two decades [1,12,20–23].

Organogels are cross-linked macromolecules that are an important class of soft materials; they can immobilize a large amount of organic liquids within the three-dimensional network structure by forming a network either of cross-linked or entangled chains for chemical and physical gels, respectively [24–26]. There exist a variety of different areas in which organogels can be used, such as controlled drug delivery systems [27], fragrances, surfactant industry, fire starters [28], hand sanitizers, and oil absorbents [29].

In existing literature, various synthetic absorbents have been investigated for the removal of oil and organic solvents from water surfaces. These include spongy graphene sorbents [30], porous oil sorbents based on PET fibers [31], carbon nanotube based organogels [32],  $\beta$ -cyclodextrin based oil sorbents [33], porous PDMS beads [34], amino acid based cross-linked polymer networks [35], eugenol based oil absorbent [36], cellulose based absorbents [37], porous carbon monolith [38], poly(orthocarbonate)s as an oil absorbent [10,39,40], poly(orthosilicate)s as an oil absorbent [41–43], poly(alkoxysilane) based sorbents [9,20,44], organic–inorganic hybrid gels [45], biphenyl-bridged crosslinked poly(alkoxysilane)s [46], coated kapok fiber [47].

Poly(propylene glycol) (PPG) or poly(propylene oxide) (PPO), which is usually produced by the ring opening polymerization of propylene oxide, is a polymer with alternate hydrophobic and hydrophilic groups in molecular chains. PPG displays some degree of hydrophobicity due to the additional methyl group in each repeating unit. It is insoluble in water, especially when the molecular weight is greater than 740 g/mol. PPG polymers can demonstrate highly hydrophobic properties at higher molecular weights. PPG is an important material in the polyurethane industry, owing to its terminal hydroxyl groups that undergo polycondensation reactions with diisocyanates. Additionally, it is used in nonionic surfactants, lubricants, anti-foaming agents, softeners, rheology modifiers, and organogels [48], and is frequently used in conjunction with poly(ethylene glycol) (PEG) [49–51]. Previously, PEG–tris[3-(trimethoxysilyl) propyl] isocyanurate (ICS) [2] and poly(tetrahydrofuran) (PTHF)-ICS [52] based sorbents were synthesized as an oil/solvent absorbent having high absorption capacity and reusability. The promising results especially belonging to PTHF-ICS based sorbents encouraged us to work further in this direction. Thus, PPG polymers were embraced to generate new high capacity sorbents by taking advantage of their hydrophobic methyl groups and elasticity in order to create better sorbents.

In this paper, PPG based organogels were synthesized through the condensation reactions of various molecular weights of commercially available PPG macromonomers with ICS to prepare oil/solvent absorbents in order to clean up the oil and volatile organic solvents from contaminated aqueous and terrestrial areas. By preparing this, new and the valuable usage area for PPG macromonomers has been proposed. The capacity, reusability, and water-oil separation efficiency of PPG based organogels were systematically investigated. Whole synthesized PPG-ICS organogels demonstrate a high capacity for oil absorption and are completely

stable in aqueous media. The removal of MTBE from the water is substantially improving due to synthesized PPG based organogels.

## 2. Experimental

### 2.1. Materials and equipment

Different molecular weight poly(propylene glycol)s (PPG 400, PPG 725, PPG 1000, PPG 2000, PPG 3000, PPG 4000) was purchased from Alfa Aesar and were used as received. Tris[3-(trimethoxysilyl) propyl] isocyanurate (ICS), potassium hydroxide (KOH) and the organic solvents; dichloromethane (DCM), tetrahydrofuran (THF), benzene, toluene, acetone, methyl *tert*-butyl ether (MTBE) were obtained from Sigma-Aldrich, and were used without further purification. 95 octane and euro diesel were supplied from British Petroleum (BP) and crude oil was obtained from TUPRAS (Turkish Petroleum Refineries Co.). Seawater was procured from Sea of Marmara in Turkey. Water absorption process was performed using distilled water.

### 2.2. Instrumentation

FTIR spectra were obtained with a Perkin Elmer Spectrum 100 FTIR Spectrometer. Solid-state  $^{13}\text{C}$  and  $^{29}\text{Si}$  cross-polarization magic angle spinning (CPMAS) nuclear magnetic resonance (NMR) were obtained with a Bruker Superconducting FT-NMR Spectrometer Avance TM 300 MHz WB. Thermogravimetric analyses (TGA) were operated under an argon atmosphere at 10 °C/min from 25 to 900 °C using a Mettler Toledo model TGA/SDTA 851. Polymer-solvent interaction was provided with an Innova 2000/platform shaker.

### 2.3. Production of organogels

A series of organogels was synthesized by the condensation of different molecular weight poly(propylene glycol)s as well as tris [3-(trimethoxysilyl) propyl] isocyanurate as a cross-linker.

Proper amount of each different molecular weight PPG was dissolved in DCM and then ICS was added into the mixture according to stoichiometric ratio with KOH under inert atmosphere. The reaction vessel was sunk in an oil bath at 50 °C which is the starting reaction temperature that was increased 10 °C every 10 min until gelation time. The detailed synthesis procedure for each organogel is given in [supporting information](#).

### 2.4. Removal of soluble fractions (SFs)

Soluble fractions, including monomers, cross-linkers, or some polymer chains which are not attached to three dimensional network structures can be kept inside the organogel. The removal of SFs is very important in order to obtain a clear understanding of the accuracy of absorption measurements. In order to eliminate the adverse effects of the SFs on absorption tests, the SFs were removed with the following method: A known weight amount of organogel was placed in DCM in order to solve the SFs without harming the network structure of the organogel to then extract the SFs, for 72 h, at room temperature. After the extraction of SFs is completed, the organogel was dried under a vacuum at room temperature for 24 h. The percentage of SF was calculated using Eq. (1).

$$SF (\%) = \frac{(W_o - W)}{W_o} \times 100 \quad (1)$$

where  $W_o$  and  $W$  are the weight of organogel before and after extraction, respectively.

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