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Applications of Graphene in Oil and Gas Industry

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Abstract -*Graphene, in its pure or derivative form has been a topic of increasing importance in the scientific community for many years. However, it's application in oil and gas industry has been popularized in last few years, with bulk of research taking place within the last year or few. Due to graphene's unique and extra-ordinary chemical structure, electrical, and mechanical properties, it exhibits many applicability within oil & gas industry. Areas of application include Drilling, lubrication, Desalination, anti-corrosion coatings, cementing, oil-water separation, oil spill cleanup, and emulsion stabilization is few to be name. This paper reviews graphene and its derivatives as they apply to the oil and gas industry and describes how this magical substance will have impacts on the technology of the industry for years to come. The main factor preventing immediate implementation into the industry is not scientific, but rather economic and industrial. Scaling up laboratory results to a size that is applicable to oil and gas industry in a cost efficient manner will prove to be the largest obstacle moving forward.*

Key Words:**drilling, lubrication, de-salination, cementing, oil-water separation, oil spill cleann-up**

1.INTRODUCTION

The stable isolation of graphene was first conducted in a repeatable manner in 2004 by *Novoselov*(2004). *Novoselov* and *Giem* later earned a noble prize in physics for this work . Graphene , a single atom thick planar monolayer of graphite, is composed entirely of sp2 hybridized carbon atoms that are covalently bonded in a two-dimensional(2D) honeycomb lattice. The unique properties of graphene and its derivatives has made it the focal point for extensive research. Graphene exhibits high electrical conductivity with 7200 S/m as well as mechanical strength with thermal conductivity of 1800-5800 W/mK at room temperature (kuilla, 2010) (Balandin,2008). With a tensile strength of 130 GPa, young's modulus of 1.0 TPa, and breaking strength of 42 N/m, graphene can be considered one of the strongest materials ever measured (lee,2008). Graphene is 50 times stronger than steel in mechanical strength , yet has one-half the density of aluminium. Other properties that add to the utility of graphene include a resistivity of 10-60 ohm cm, electron mobility of 20,000cm2/Vs (at 300) and theoretical surface are of 2620 m^2/g . Graphene has been used in many fields including bio-degradation, flexible electronic devices, spintronics, photonics, optoelectronics, sensors, energy storage and conversion, and biomedical . Graphene has potential in nano-applications in many scientific and industrial fields including the study of nano-electronics, molecular separation, composite additives, catalysis, nano-sensors, and transport. Fig. 1 shows an image on graphene .

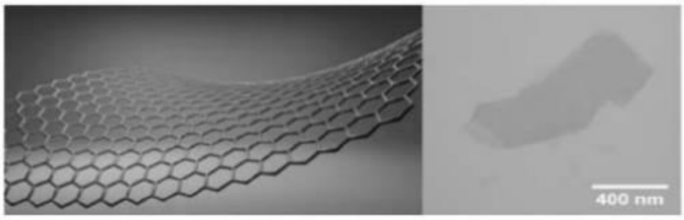

Fig. 1. Structure of graphene sheet and actual microscopic image (Advincula, 2014).

The oil and gas industry is a multi-dollar markets that has effected lives of any person who has ever driven a car, used electricity, walked on asphalt, or used polymeric products like plastics. It is estimated that liquid consumption projected to increase in the coming years and the amount of easily obtainable decreasing, applying new technology to this industry will be crucial to fulfilling the world's energy needs in the future (US Energy Information Administration, 2016). Nanotechnology including graphene and graphene derivatives have show potential in the oil and gas industry in the areas of drilling, shale inhibition, sensing, rheology modification, wellbore strengthening and stabilization, sensing, coatings, hydraulic fracturing, lubricity, emulsion stabilizers, desalination, and oil spills cleanup just to name a few (Genorio,2012) (Hoelscher,2013) (aftab,2016). Graphene derivatives, such as graphene aerogels, foams, sponges, modified membranes, and graphene oxide(GO) membranes have shown promising applications in oil spill cleanup technology, desalination process and emulsion stabilization. The properties like "*superhydrophobicity*" and "*superperoleophilicity*" of graphene's derived products allows for efficient oil-water separation. Desalination using graphene membranes could be a viable alternative to tradition desalination techniques because of the thickness, high mechanical strength. High water flux, and efficient ion rejection rate as well as a lower energy consumption and operating cost.The purpose of this paper is to shed light on technology of graphene and graphene derivatives and their applicability to the oil and gas industry. The paper will explain the major synthesis methods then it will introduce some if the main topics that are directly relatable to current oil field processes such as drilling, as well as larger industry wide issues, like desalination needs, oil-spill cleanup, and emulsion stabilization.

2.Graphene synthesis

There are multiple ways to synthesize the graphene. The majority can be classified into four categories; exfoliation and cleavage, chemical vapor deposition techniques, chemically derived techniques, and other synthesis method.

Exfoliation and cleavage methods involve the physical peeling of layers off pure, Graphite. The method was conducted by the *Viculis* group (Viculis.2003). This method later on perfected by Novoselov by using highly pyrolytic graphite sheets that were dry etched in oxygen plasma, stuck on a photoresist, and peeled off using ordinary scotch tape (Novoselov, 2004) also know as ' scotch tape method'. Since graphite is comprised of layers of uniformly stacked graphene bonded by van derWaal forces, it is conceptually feasible that those graphene layers can be separated and isolated. Fig.2 shows an example of the scotch tape method.

Fig. 2. Mechanical exfoliation of graphene from highly oriented pyrolytic graphite (HOPG) (Singh, 2011).

Chemical vapor deposition (CVD) methods include thermal CVD plasma enhanced CVDs, as well as thermal decomposition on substrates. The first CVD growth of planar few layer graphene on a metal surface was reported in 2006(Somani, 2006). Of the graphene methods currently being used, CVD methods are very common and seem to be the most promising for future large, scale production of monolayer graphene films. They involve first chemically etching a metal substrate, after chemical etching, the graphene layers are isolated, and removed.

By far the most researched and most popular chemically derived technique involves converting graphite to Graphene oxide (G0) and then using GO to obtain graphene or using the GO directly. The major GO synthesis method, known as " hummers methods" involves the oxidation of graphite using sulfuric acid, nitric acid, and potassium per-manganate (Hummers and Offeman, 1958). GO is then converted to a pure graphene through a variety of different routes.

3 Graphene applications in the oil and gas Industry

As mentioned before, graphene and its derivatives have been vigorously studied for many years. However, graphene as it pretains to the oil and gas industry is a more recent topic of interest. Despite its relativity short record, there has already been many applications of graphene and graphene derivatives to the energy industry with potential affects. Some of the main applications include drilling, cementing, enhanced oil recovery, desalination, oil spill cleanup, and emulsion stabilization, as well as other such as improvement to coatings and elastomers.

3.1. Drilling

Drilling is a important step in the recovery of hydrocarbons form the earth. A key factor in drilling is the use of drilling fluids. Drilling fluids carries the formation cuttings to the surface, lubricate the drill bit and controlling pressure in the well by maintains hydrostatic pressure. The uses of graphene and G0 can improve fluid loss control, rheology, lubrication, and shale stability. Nano-particles, such of graphene or GO are much smaller and far denser than bentonite. For this reason graphene has been studied as a alternatives drilling fluid in the use of fluid control loss. One studied concluded that even though graphene is small enough, its lateral size leads to flocculation and poor dispersion in well fluids (Xuan, 2013). However, modified graphene oxide maintains its morphology as well as resist permeation allowing it to act as a superior fluid loss additive which can be used in place of betonite with 10 time less additive.

With fluid loss in consideration, it is important to maintain rheological properties. Poor rheology will result in a lower penetration rate, hotter drill bit, and potentially more downtime. A study conducted using graphene Nano platelets compared to other additives in water based drilling mud by (Aftab and Ismail, 2016) showed that graphene has the best results on enhancing rheological performance in high temperature-pressure situations and found its enhanced properties is due to low friction between the Nano platelets.

When drilling shale formation, it is common for water in the drilling fluid to interact with clay materials in the shale causing swelling, thus reducing penetration rate. Due to this it is uncommon to add shale inhibitors to reduce the swelling. Graphene's derivatives has been tested as shale inhibitors and it is found that GO serves as an effective transport medium for the improved inhibitors as well as effective shale inhibitor for high temperature-pressure applications (Fiedheim, 2012).

3.2.Lubricant

A major contributing factor to the applicability of graphene derivatives to drilling fluids is the impermeable membrane properties as well as its lubricating properties. However, Graphene shows promise as a lubricant for many other things aside from drill bits, the very thin structure of graphene and its ability to be evenly distributed on a micro-scale and Nano-scale by simply using graphene flakes in solution allows graphene to be applied to much more than just drilling applications (Berman,2014). In multiple studies, computational methods were used to determine the changes in friction properties of graphene at a Nano-scale level. The studies showed that as the number of graphene layer increased the friction between the layers and the medium decrease and that as the number of graphene layers increased the friction between each graphene layer increase (Xu,2011) (Lui and Zhang,2011). A micro-study of graphene yielded similar results. The study conducted by Kim et al., found that adhesion and friction properties were reduced by using graphene, deposited on Ni and Cu substrates and fused silica and polydimethysiloxane lenses (Kim, 2011). When considering modified graphene, friction tend to increase while, adhesive properties are reduced or remain the same as the result of reduction in van der Waal's contact area (Balandin, 2008).

3.3 Desalination

Reservoir usually contain petroleum hydrocarbon in the form of liquid, gas, or both along with varying amount of water. It is generally believed that rock in the majority of oil bearing; formations were originally saturated with water prior to the introduction and trapping of petroleum through drainage or inhibitionmechanisms (Amyx and bass, 1960). Water, either produced or injected, is used in the many oilfield production applications including gas and liquid natural gas (LNG) operations, oil sands, steam assisted gravity drainage (SAGD), conventional, and unconventional (Adham,2015). The amount of water is utilized varies from application to application. For example, when dealing with oil sands, water is used as a lifting solution for the oil (Clariet,2013). This water intensive process requires around 4.5 barrels of water for each barrel of oil produced (International Desalination Association, 2013). SAGD operations require approximately 4 barrel of oil. When considering produced water as opposed to injected water, usually 3 to 4 barrels of water are produced for each barrel of oil for conventional operations (Adham, 2015). In unconventional operations, this ratio can be 7:1 or higher (Burnett, 2004). Generally the longer a well is being produced through primary recovery methods, the more water will be produced.

Produced water is the largest waste byproduct in the oil and gas industry (veil, 2004). Besides the cost of lifting water out of reservoir, the water must be processed, treated, re-injected, or disposed. Depending on the toxicity and composition of the produced water, it must be treated for oil contamination, organic materials, particles, and/or salts; with salts being the major contributor of toxicity (Ahmandun,2009). Discharging large volumes of contaminated water to the environment has become a growing concern and has resulted in more strict regulations and permitting by government (Tellez, 2005) (EPA,2017). The oil and gas industry represents 14% of the desalination market share, a multibillion-dollar market (Adham, 2015). Advancement in desalination technology will allow produced water to be transformed from an environmental and economic burden on energy companies to a valuable byproduct that can be used for re-injection. Irrigation, industrial uses, fire control, or in aquifers (Veil, 2004).

Graphene and its derivatives have had the focus of study in recent years for its application in desalination. Graphene based materials and membranes have proven to be extremely effective in salt re-injection (You, 2015). Graphene in desalination have potential to help solve some of the wastewater issues in its pure form is strongly impermeable to liquid and gases makes it a prime candidate for desalination technologies. The two main areas of graphene desalination research are the use of Nano-porous graphene membrane and the use of stacked graphene oxide sheets.

The general concept of using a Nano-porous, graphene membrane, for Desalination is as follows. Since graphene is impermeable to liquid and gases, even though it is extremely thin, it should be possible to create Nano-pores in the single layer graphene that allows the passage of water, but blocks the passage of larger ions. This concept arose from research on carbon Nano-tube membrane and their permeability to water using unique structure (Alexiadis, 2008). The pore selectivity would be two toned. First off, it is ions and the pore. This concept is summarized in Fig. 3.

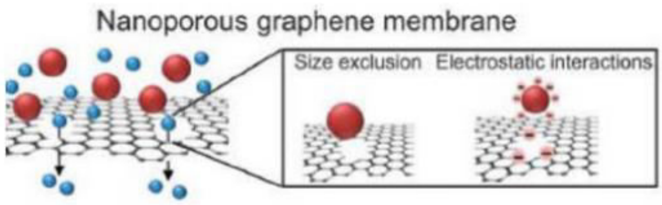

Fig. 3. Nanoporous graphene membrane selectivity (Perrearult, 2015).

Tests have shown that using Nano-porous membrane for water treatment can produced purification rates of 400-4000 Lm^2/hbar; a rate that substantially higher than reverse osmosis membranes (Cohen, 2012). The Nano-porous membrane can also reject salts at a comparable or higher efficiency than the osmosis membrane even with higher water volumes. More efficient membranes can result in overall higher water treatment volumes or smaller facilities can be utilized to achieve the same volume as current production. The largest issue to overcome with Nano-porous membranes is controlling the pore size in a consistent, reproducible manner. In addition, pure graphene is expensive especially in large quantities.

The stacked graphene oxide method was first used by the Nair group when they realized that stacking graphene oxide membrane allows water to pass through while blocking permeation of gases and non-aqueous solutions (Nair, 2012). Unlike graphene, graphene oxide is much cheaper to purchase or synthesize. By stacking graphene oxide membrane, hydrophobic "channels" are created from un-oxidized regions of each membrane. The stacked membranes have been showed to reject salts and other organic molecules in a similar fashion to a Nano-porous graphene membrane (Nair, 2012). Fig. 4 is a pictorial representation of this process.

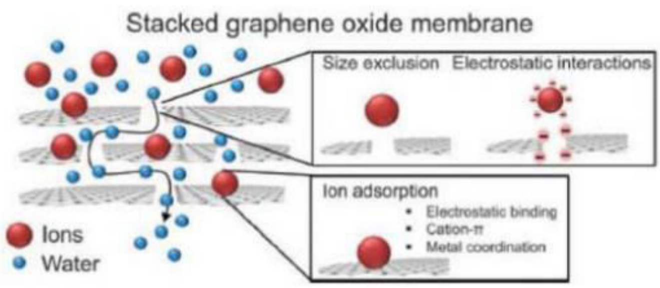

Fig. 4. Stacked GO membrane selectivity (Perrearult, 2015).

Tests have shown that using stacked GO membrane can achieve water treatment rates of 22-71 lm^2/hbar (Cohen, 2012). These rates are much lower than pure graphene but G0 membrane are also less expensive. Since the main mechanism of separation is pore size, there are issues with GO membranes that are currently being studied. The main issues are that channel widths are larger then have less selectivity of filtered water overtime using the GO membrane. A joint issue with Nano-porous and G0 membrane is being able to upscale the desalination process from a laboratory size to an industrial size.

3.4 Other Uses

Another use for graphene oxide is as an additive for cement. As a well is being drilled, casing must be insertedd in order to add structural stability and prevent an inward collapse. Each section of the casing must be cemented into place. A study on cement as an additives researched the effects of using graphene oxide as an additive to enhance the properties of cement and found to be positive, showing that adding quantities as low as 0.05% GO can result in an increase of 41%-59% in flexural strength, 13%-33% in compressive strength, and an increase in ductility (Mangadlao, 2014). The study also stated that GOmay helping prevent microbial induced corrosion, which is an issue in production wells over time.

Corrosion has been major issue in the oil and gas industry since its inception. It affects pipelines, production tubing, casing, and just about any type of down hole equipment that is not made of plastic. Graphene and its derivatives have been investigated for their applications as a corrosion resistant coating for down hole, metal equipment. The advantages of using graphene as a coating include its strong mechanical and electrical properties, its impermeability to gases and liquid, low chemical reactivity, as well as the fact that the coating could be substantially thinner than a traditional coating while in theory having superior performance, in one study, a 200nm thick graphene oxide hybrid coating was applied to stainless steel. The study concluded that the GO coating by itself did not provide much corrosion resistance, but as part of the hybrid blend, it provided excellent results (Mondal, 2015). A second study utilized an oil-based graphene oxide base a coating for old-rolled steel in a seawater environment (Singhbabu, 2015). The study concluded that in a high salt environment, the use of their graphene oxide coating was highly effective and reduced the corrosion rate by over 10,00 times when compared to the non-coated control group steel. In a final study conducted by the *Dumee* group, pure graphene Nano-flakes were grown directly on a stainless steel substrate (Dumee, 2015). The group successfully grew between 3 and 15 layers on the steel which resulting in enhanced corrosion resistant and electrical properties without affecting the property of the stainless steel substrate. The experimental growth can be seen in Fig. 5.

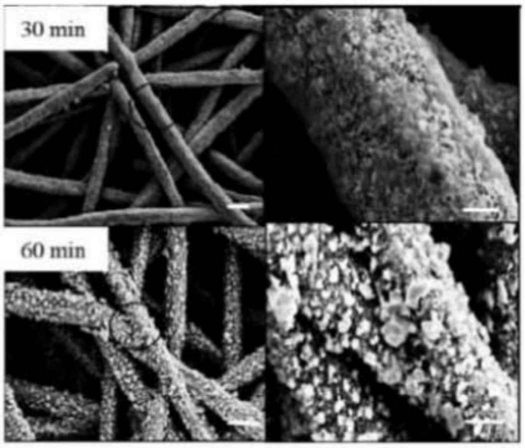

Fig. 5. Graphene growth on stainless steel as a function of time (Dumee, 2015).

Just as graphene, its derivatives have proven to be worthy additives. In cements and coating, they can also be used to enhance the properties of elastomers. Elastomers are used for a variety of different thongs in the oil industry. The most common use for elastomers are on down hole equipment such as packers, plugs, or components of artificial lift system. Regardless of their role, the elastomer must be able to withstand rigorous down hole conditions including high temperature and high pressure situation. With increase of SAGD operations, there is constantly a need for enhanced elastomer properties. In a study by Wei and Jacob, graphene oxide and reduced graphene oxide (RGO) was used to enhance a fluoroelastomer at standard and high temperatures (Wei and Jacob, 2013). The study concluded that by using GO and RGO as enhancements, stress resistance increased by 28% at room temperature and 49% at elevated temperature and the aspect and modulus ratios both greatly increased. Similar to coatings, graphene oxide and reduced graphene oxide function better as enhancement to elastomers additives as opposed to standalone additives.

3.5. Oil spill cleanup and separation

The largest critique of the oil and gas industry is its potential effects to the environment. Normal oilfields operations such as drilling, fracking, and pipeline expansion are constantly the target of environmental groups, personal activists, and government lobbyist. However, the largest environmental issue associated with oil and gas industry will always be oil spills, and more specifically oil spills in bodies of water. The BP oil pills, which was the largest spill in US history, resulted in over 200 million gallon of crude oil being dumped into the gulf of Mexico and damaging over 16,000 miles of coastline (Jarvis, 2010). After this major spill, oil cleanup technology and oil-water separation technology has become a hot topic both in the United States and abroad. Graphene and its derivatives have been identified as a viable option for both problems. Graphene membranes, foams, and aerogels are among the major areas of research for their applicability to this topic.

One application of oil-water separation in the oil and gas industry is the separation of oil-water emulsions. Emulsions separation, also known, as de-emulsification is the process of breaking crude oil into the oil and water phases. This is commonly used in refineries where the crude oil is processed into its constituent parts. Besides membrane filtration, other technologies exist to treat oil-water emulsions including; coagulation, ultrasonic separation, and flocculation (Stacka, 2005) (Bensadok, 2007). It has been studied that the use of graphene oxide to modify filtration membranes can result in better separation of the emulsion. In a study by the Hu group, an AL2O3 microfiltration membrane was modified using graphene oxide (Hu, 2014). The group prepared graphene oxide using the Hummers method and modified the membrane by transferring the graphene oxide via a vacuum method, which resulted in average pore size of 200nm (Chen, 2012). The experimental results showed that the modified membrane has a flux that was almost 28% higher than the unmodified membrane and the oil rate was 0.6% higher at 98.7% (Hu, 2014). The paper and experiment support the idea that graphene oxide can act as a membrane modifier for oil-water separation.

Instead of using graphene oxide as a membrane modifier, it is also possible to simply modify the Graphene oxide by the addition of functional groups in order to obtain a material suitable for oil-water separation. In a study from the carbon journal, a group altered graphene oxide into a 'superhydrophobic' material by using esterification with 'epoxy-functionalized' 'polyhedral oligomeric silsesquioxane' (ePOSS) (Lui, 2014). EPOSS is an organic-inorganic hybrid material with applications in biomedicine, catalyst, and stability enhancements that are hydrophobic (Kannan, 2005). Through esterification, a graphene oxide membrane can be altered to a GO/ePOSS membrane. Impurity free graphene oxide is characteristically hydrophilic with an air-water contact angle of less than 20 degree while ePOSS is characteristically hydrophobic with an air-water contact angle of around 115 degree. However, after the esterification of the two, the resulting GO/ePOSS membrane exhibited even more hydrophobic properties and an air-water contact angle of 145 degree (Lui, 2014). The study concluded that as long as the oil is less dense than the water than this method of separation is effective for at least 10 cycles of membrane use.

Graphene foam, which is a reduced derivative of graphene oxide with a variety of different agents, is a topic of considerable research due to its applicability for oil as well as organic liquid recovery. Graphene foam can be altered for absorption of gas, water, or oil because of changing the selectivity of the foam membrane. Graphene foam can be created in multiple ways but it is commonly synthesized by first making a metal foam (commonly Ni), then applying CVD growth to the metal foam and freeze drying it to foam a graphene foam (Sha, 2016) (Ping, 2017). Graphene foam is interesting because it has been shownto exhibit switchable wettability's in response to change pH (Zhu, 2014). The changes in properties, coupled with very high absorption capacities as well as high cycle lifespans, makes graphene foams effective for oil recovery and cleanup as well as organic solvent recovery. The key to the switching wettability properties is to first coat the graphene foam with an amphiphilic copolymer containing poly (2-vinylpyridine) and poly-hexadecyl acrylate which effectively results in a surface that can absorb oil when the surface is at a pH of 7 and can release the adsorbents at pH lower than 3 as a result of protonation and deprotonating(Zhu, 2014). When testing the absorbency of the graphene foam, it was shown gain between 40 and 196 times its own weight, which is higher than non-graphene foams and consistent or higher than other Nano-tube materials (Li, 2014) (Jai,2012).

The Kim group has applied reduced graphene walls to all sides of a cubical structure that will suck oil through the wall membrane while blocking the water. However, since only the outside of the vessel contains the graphene membrane it serves as a one way "check valve" (Kim, 2015). The prototype vessel created by the group floats in water, requires no energy source, collects oil with a purity of over 99.99%, and has a scaled up collection rate of over 20,000 l per square meter per hour. The prototype can be seen in Fig. 6.

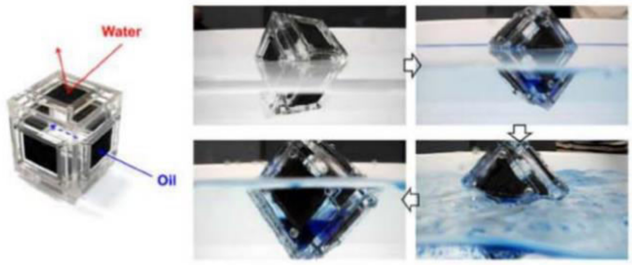

Fig : 6 Working Prototype of Graphene vessel

4. CONCLUSION

Graphene and its derivatIve have been a popular topic of research since its reproducible synthesis was discovered in 2004 (Novoselov, 2004). However, its application in the oil and has industry has only been in use in recent last few years. Due to graphene's unique chemical, structural, electrical, and mechanical properties, it shows applicability for many applications in the oil and gas industry. Graphene and graphene derivatives show promise in the areas of drilling, lubrication, desalination, anti corrosion coatings, cementing, oil-water separation and oil spill cleanup. This revolutionary substance will have huge impacts on the technology of the oil and gas industry in years to come. The main issue preventing implementation of graphene into the oil and gas industry is not scientific but rather economic and industrial. Figuring out how to scale up graphene laboratory research to be applicable to the petroleum industry in a cost efficient manner will be the largest obstacle to overcome moving forward.

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