1.1 Introduction

Graphene is an individual sheet of graphite. It has remarkable properties, including incredible strength, optical transparency, and ballistic electrical conduction. It also represents a platform material for the enhancement of various technologies. However, for the practical application, the mass production of graphene is required. For this, the major routes for graphene material synthesis are the chemical exfoliation of graphite. It requires overcoming graphite interlayer bonding energy, allowing for separation of individual sheets.

In contrast, graphene oxide (GO), a chemically modified material made from natural graphite, which can be reduced in various ways to give graphene-like platelets, commonly known as reduced graphene oxide (rGO), is solution processable in environmentally friendly solvents (i.e., water). However, the chemical modification required to form both GO and rGO platelet causes a reduction in some of the properties of graphene. However the properties of GO and rGO enable us to understand the structural difference between GO and rGO sheets, which is useful in understanding the formation and removal of oxygen functional groups such as carboxylic acid groups (-COOH), hydroxyl groups (-OH) and an epoxy group (-C-O-C) on both edges and hybridized surface of the basal plane through sp³ hybridization. Reduced graphene oxide (rGO) possesses exciting properties (H.Lu, et al. 2014; G.Ni,et al.2012; A.Kafy, et al. 2015). Generally, its properties depend on carbon to oxygen ratio of graphene oxide. The altering carbon to oxygen ratio of graphene oxide alters its properties from semi-metal to semiconductor (Q.Li, et al. 2010; J.Tian, et al. 2012).

Recently, carbon-based nanoceramic composites materials such as metal oxide in semiconducting nature have been used for organic pollutant elimination in wastewater (M.R.Para, et al.2015; E.Rokhsata et al.2016; J. Qin, et al.2017; J.A. Quek, et al.2018; Z.Ning, et al.2019). In recent literature, degradation of organic pollutant containing in wastewater has been degraded by GO-based photocatalysis material nanocomposites (S. K. Mandal, et al. 2019; Boruah, et al. 2015; F. Wang, et al. 2018). GO, and rGO is best in photocatalyst and adsorption application due to its large specific surface area and abundant functional groups. However, as a single rGO, having weak photocatalytic activity but its photocatalytic activity can be enhanced by combining it with other nanoceramic materials like metal oxide such as Cds, SnO₂, WO₃, SiO₂, Nb₂O₃, ZnO, TiO₂ (J.Qin, et al.2017; J.A.Quek, et al.2018). Among various photocatalyst, used TiO₂ and ZnO are the best with higher photocatalytic activity (A.Khataee et al. 2014; E.Rokhsata et al.2016; S.Benkara et al.2010; Q.Wang et al. 2018). Compared to TiO₂, and ZnO, ZnO as has attracted more attention because of its bandgap (3.37eV), long life span, and high chemical stability (A.Rezaee, et al. 2012; A.Khataee, et al. 2014; Y.Wang, et al. 2012; O.Yayapao, et al. 2013).

In the recent literature study, photocatalytic degradation of organic pollutants were influenced by several parameters, such as pH of the solution, initial concentration of organic pollutants, particle size and morphology of photocatalyst and its concentration, reaction temperature, light intensity, oxidizing agent, different photocatalysts (D. Fu, et al., 2012;L. Sun, et al., 2015;X. Du, et al., 2019;S.

K.Mandal, et al. 2019;D.R.Paul, et al.2019). In above mention fact, research is now focused on obtaining ZnO in a definite microstructure like nano-rods, hexagonal, nanowires, nanoflower nanobelts, nanotubes and nano-rings for the influence the catalytic potency (A.Bhattacharjee, et al.2016; L. Tingzhi, et al. 2015). In literature, Dongying Fu, et al. 2012 reported that low content of ZnO-rGO composites could not absorb the light completely, and an excess amount of ZnO-rGO reflects the light. Fengzhi Wang, et al. 2018 reported that nanorods like morphology of ZnO- rGO composites enhanced the photocatalytic performance. H.N.Tien, et al. 2013 reported that sphere ZnOSPs-rGO enhanced the degradation of methylene blue. S. K. Mandal, et al. 2019 reported that quantum dots ZnO- rGO completely degraded the organic pollutants. S.Xu, et al.2014 reported that flower-like ZnO-rGO enhanced the photogenerated carrier's recombination rate. X.Du, et al.2019 reported the effect of different pH and catalyst dosage on the photocatalytic activity.

From the above literature study, the morphology of ZnO with rGO is significant in consequence for the degradation of organic pollutant. In this regard, in the present thesis, lotus-likee, columnar-like of ZnO was prepare with GO by reduction process to obtain nanoceramic composite in the hybrid system. There are different method for synthesis of ZnO nanoparticles with particular morphology such as sol-gel method (A.E. Danks, et al. 2016), hydrothermal method (J. Ding, et al. 2015), assisted solution route (M.Darwishet, et al. 2016), two-step method (B. Bhushan et al. 2017), aqueous chemical route (M. Parra, et al.2014) and chemical precipitation method (Q. Yang, et al.2010). Among them, we have applied the sol-gel process for the synthesis of Lotus-like ZnO and two-step method for the columnar-like structure of ZnO. This has been discussed in the section of materials and

methods. A hexagonal plate-like morphology of ZnO was also prepared and their preparation method as discussed in the part of materials and methods.

1.2 Objective

As mentioned above, the excellent properties of graphene is attractive for composites. It is observed that the decoration of rGO sheets with different morphological structure of ZnO enhances the optical performance. In this consequence, it is imperative and challenging for developing the method that is fast, low-cost, environmentally friendly, and non-toxic for preparing particular morphology of ZnO with rGO. It is also complicated and challenging to control the size, morphological structure of ZnO on the surface of rGO. In recent literature, there is no report on the synthesis columnar and lotus-like of ZnO with nanocomposites of rGO.

To fulfil the above mentioned fact we achieve our research work objectives, as following

- To design and modify sol-gel techniques for preparing nanoceramic composites with reduced graphene oxide (rGO-ZnO) those are suitable for industrial application.
- 2- To identify suitable reducing agent and temperature to control morphology and size of the ceramic nanoparticles, this also can be used as industrial techniques to prepare these nanoceramic structures.
- **3-** Optimization of the sol-gel conditions such as pH value, reaction temperature, the reaction time of the solution, and



amount of ammonia solution and sodium hydroxide for preparation of ZnO nanoparticles.

- 4- Later on, utilizing the optimum sol-gel conditions for preparation of rGO-ZnO nanocomposites with different loading amount of graphene.
- **5-** The resultant composite can be used for photocatalytic degradation of phenolic compound and natural dye (ND).

