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Kinetics of Green Reduction of Graphene Oxide via *Hibiscus Sabdarriffa L* Aqueous Solution

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Abstract

In this study ,a kinetics of green approach for the synthesis of reduced graphene oxide was investigated. The reduction of graphene oxide to reduced grapheme oxide was performed using aqueous solution of *Hibiscus Sabdarriffa L* (Roselle) leaves as reducing agent under mixing and sonication at 40kHz treatments. Various experimental conditions were investigated which involve effect of treatment time, pH , reaction temperature, extract % and grapheme oxide concentration. The kinetics of RGO formation were evaluated spectroscopy by measuring the absorbance at 660 nm based on that molar extinction coefficient of reduced graphene oxide is higher than graphene oxide by several times at this wavelength. It has been found that pseudo RGO rates formation were enhanced by high pH value, high reaction temperature, high extract amount, and high GO concentration. Also, the results indicated that conductive and spectral response properties of synthesized RGO were best in comparison with GO. Also, activation energy of RGO formation was determined. The optimum concentration of reduced graphene oxide was calculated. A good dispersion GO and RGO aqueous solution were achieved.

Key Words: green approach, reduced graphene oxide, *Hibiscus Sabdarriffa L*,kinetics,spectroscopic

1.Introduction

Reduced graphene oxide(RGO) has attracted a huge attention in recent years due to its two-dimensional conjugated chemical structure and owing to its unique and remarkable mechanical, electrical ,optical and thermal properties. A great potential was spent on applications of grapheme in various technological fields. Combined with synthetic materials,graphene open up new fields for materials, especially based composites that have attracted much attention in recent studies and have shown their usefulness in electronic,photocatalysis ,photovoltaic devices[1-5].Due to its high charge mobility and biocompability,graphene has a various application in bio sensing[6] , drug and gene delivery[7,8],clinical imaging[9],antibacterial[10] .Reduced graphene oxide can be synthesized by several methods such as mechanical exfoliation of graphite[11] ,thermal chemical vapor deposition[12],plasma enhanced chemical vapor deposition technique[13], solution-based chemical reduction of graphene oxide[14]**.** The chemistry approach is the most commonly adopted method to synthesis of GO, and the most common rout to exfoliate 3D network graphite is the use of strong oxidants such as sulphuric acid(H_2SO_4),phosphoric acid(H_3PO_4) ,potassium permanganate($KMNO_4$), potassium chlorate($KClO_3$) ,sodium nitrite($NaNO_3$) . The chemical reduction of graphene oxide is a more adopted reduction methods due to producing graphene in large quantity with high dispersion .The chemical reduction of garaphene have undesired features such as high toxicity of chemical reductantes and irreversible aggregation of product which making GO unusefull for biological applications, also some reductantes specially hydrazines may be explosive. Synthesis of high quality ,low cost, large quantity, and eco-friendly graphene is a subject under continuous research. Scientists developed various eco friendly methods which has more attention in recent years . There are several works

involved using of green nanotechnology to synthesis of RGO,such as using aqueous extract of *Amaranthus dubius* [15] ,green tea extract solution[16],rose water[17],glucose[18,19],bacteria[20,21] ,melatonin[22], L-ascorbic acid[23-26] , alcohols[27-28], wild carrot root[29], *Salvadora persica* L. root (Miswak) extract[30] , protein[31] ,amino acid[32,33] , fenugreek seeds[34],grape seeds[35] .In the present study, we reported a green approach for the synthesis method of reduce graphene oxide by using aqueous extract of *Hibiscus Sabdarriffa L* Roselle as natural antioxidant.The investigation of reduce graphene oxide was spectroscopy established through measurement of RGO concentration at 660 nm.

2.Experimental

2.1 Chemical used

Graphite fine powder was obtained from B.D.H.,Sulphuric acid , sodium hydroxide, and hydrochloric acid   were purchased from Scharlu.Potassium permanganate was supplied by Fluka.

2.2 Instruments used

Double beam- spectrophotometer(UV-1650, Shimadzu) was used to recoding the UV-visible absorption of grapheme oxide and reduced graphene oxide solutions using 1 cm path length quartz cuvette. A good dispersed GO solution was achieved by using sonication bath(LUC-410, Labtech). Ultracentrifuge(Triup international Corp) was used to separation of GO and RGO.Temperture was adjusted using K&K thermostating water bath model K-CWBBL.K&K ,K-VO27 vacuum oven was used to drying of GO and RGO,Suspension of graphite and GO were stirred by using Lab terch,LSG-120D mechanical stirrer. FTIR spectrum was recorded over the range 400-4000 cm^{-1} on a Shimadzu FTIR-8000 spectrometer using the KBr disc. AFM image

was acquired with AA2000 atomic force microscopy (Angstrom advanced Inc.). SEM image was obtained using Model JSM 6390 from JOEL Company, Japan .

2.3 Synthesis of graphene oxide

Graphene oxide was prepared from graphite powder by modified Hummer,s method[36] . 2.0 g of graphite powder was added to 100 mL of concentrated sulphuric acid   at room temperature .The mixture was cooled to 5^0C using an ice bath with magnetic stirring for 30 min and maintain the temperature of mixture below 5^0C.Then, $KMnO_4$ (8 g) was added slowly under stirring and cooling and the mixture temperature was not allowed to reach 10^0C.a . 100 mL of distilled water was added to the mixture under stirring for 1 hr and further diluted to approximately 300 mL with distilled water .The color of the mixture changed from dark purplish green to dark brown.Then,20 mL of 30% H_2O_2 was added to the mixture to stop the oxidation process by reduce residual $KMnO_4$ till the brown solution turned pale yellow colour, indicating a high oxidation level of graphite . The obtained graphite oxide was filtered , washed three times with 1 M of HCl aqueous solution to remove metal ions and repeatedly washed with deionized water until a pH of 7 was achieved. The washing process was carried out using simple decantation of supernatant via a centrifugation technique at 6000 rpm.The resulting graphite oxide was dried at 45^0C for 24 h using vacuum oven.

2.4 Preparation of *Hibiscus Sabdarriffa L*   extract

   10 g of plant leaves powder was added to 50 mL of dionized water with stirring on magnetic stirrer for 30 min till the temperature of boiling was achieved.Then,the mixture was filtered, and the filtrate was cooled to room temperature. Further diluted to 100 ml with dionized water to achieve 10% extract solution.

2.5 Reduction of graphene oxide to reduced graphene oxide

The reduction of GO to RGO was achieved by using Hibiscus Sabdarriffa L aqueous solution as reducing agent in 40kHz sonicator bath .Various experimental condition involved the effect of treatment time, pH value, reaction temperature, concentration of aqueous Hibiscus Sabdarriffa L extract and GO concentration were investigated on the rate of RGO formation. In a typical method, a known disperse of graphene oxide was obtained by sonication in aqueous solution for 30 min by using a 50 mL round bottomed flask. The dispersion was transferred to 100 mL round bottomed flask and a desired volume of Hibiscus Sabdarriffa L extract solution was added and stirring using mechanical stirrer for 150 min under sonication at adjusted temperature and pH . Within treatment time, the brown color of GO solution changes to black and ,a samples of suspension were analyzed spectroscopy at 660 nm to study of RGO formation rate. In all cases, absorbance was recorded after centrifugation of suspension at 500 rpm to remove aliquots and analysis of homogeneous dispersion.pH value was adjusted using 1M HCl and 1M NaOH.Also,a uniform GO disperse was reduced adopting mixing treatment excluding sonication under the same set of effects mentioned above.

2.6 Spectroscopic analysis of RGO formation

For the estimation of RGO formation under deterrent effects,UV-Vis spectroscopy was obtained.Baesd on relationship between absorbance(A) and concentration(C) as described by Lambert beer's law[37,38]:

$$A = \varepsilon x C_{RGO} x l \tag{1}$$

Where is the molar extinction coefficient, and l is the optical path length(1 cm).Above equation was used to determination of RGO homogeneous solution . it may be assumed that ε_{RGO} is α times larger than ε_{GO} at 660 nm wavelength as following[26] :

$$\varepsilon_{RGO}= \alpha x \varepsilon_{GO} \qquad\qquad (2)$$

and

$$R_{RGO}= (A_t- A_0)/(t) \qquad\qquad (3)$$

Where are A_0 and A_t are the absorbance of dispersion at 660 nm before and after reaction t time, R is the pseudo rate RGO formation,min^{-1}.

3.Results And Discussion

3.1charecterization

The concentration of graphene dispersion in solvents depends on a series of factors such as energy added(sonication,heat,UV …etc) ,pH,flake size ,thickness, and amount of reductant. In the present paper, a dispersed GO in aqueous solution was performed by sonication of GO using sonicator bath,and in a later step, GO reduction was performed using ultrasound treatment and *Hibiscus Sabdarriffa L* aqueous solution extract .For comparison, a mixing treatment was performed excluded ultrasound treatment. The exfoliated graphene prepared in this paper was previously characterized(thesis).Figure 1. shows UV-Vis absorption spectra of GO , RGO and *Hibiscus Sabdarriffa L* extract in aqueous homogeneous solutions. It is clear that RGO is more absorptive in comparison with GO. Based on figure 1. and previous literature[39-41] ,the absorbance above 600 nm is used for kinetic analysis of graphene formation. The UV-Visible spectrum of graphene oxide exhibits an absorption band at 212 nm which due to π-π*.Also ,the weak shoulder at 300 nm is corresponding to n-π* transition of carbonyl group. Adding of and *Hibiscus*

Sabdarriffa L assists a reduction of GO and causes red shift of 258 nm in UV-Visible spectrum of RGO.
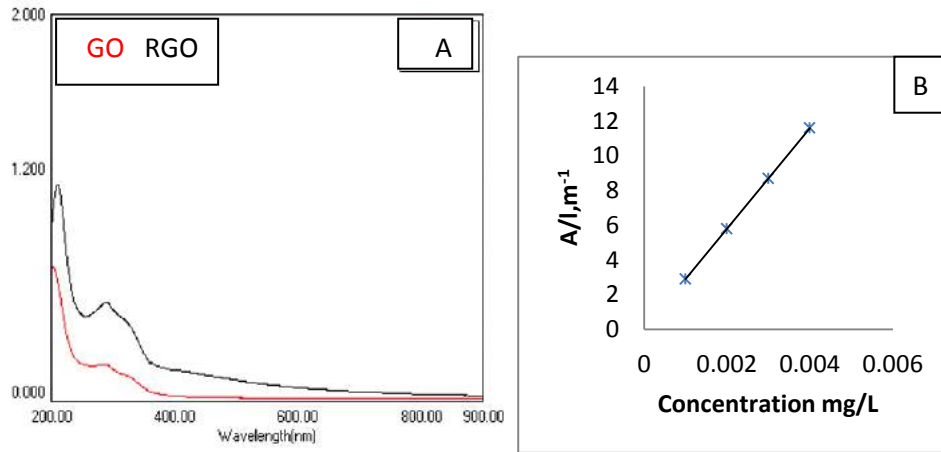

Figure 1.A-UV-Vis spectra of GO and RGO dissolved in water.B-.Linear relationship between absorbance and RGO concentration

Figure 2 shows FTIR spectrum of GO and RGO .The peaks at 1720 cm^{-1} and 1610 cm^{-1} is assigned to C=O and C=C groups. The decreasing of peak intensity at 1720 cm^{-1} is an indicator of reduction of graphene oxide and showing that graphene restores conjugated aromatic system.
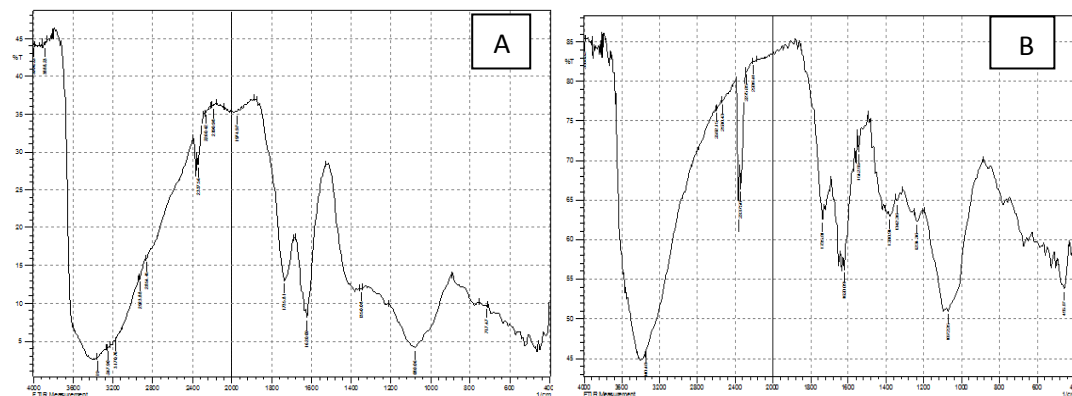

Figure 2.FTIR spectrum of :A- GO, B-RGO

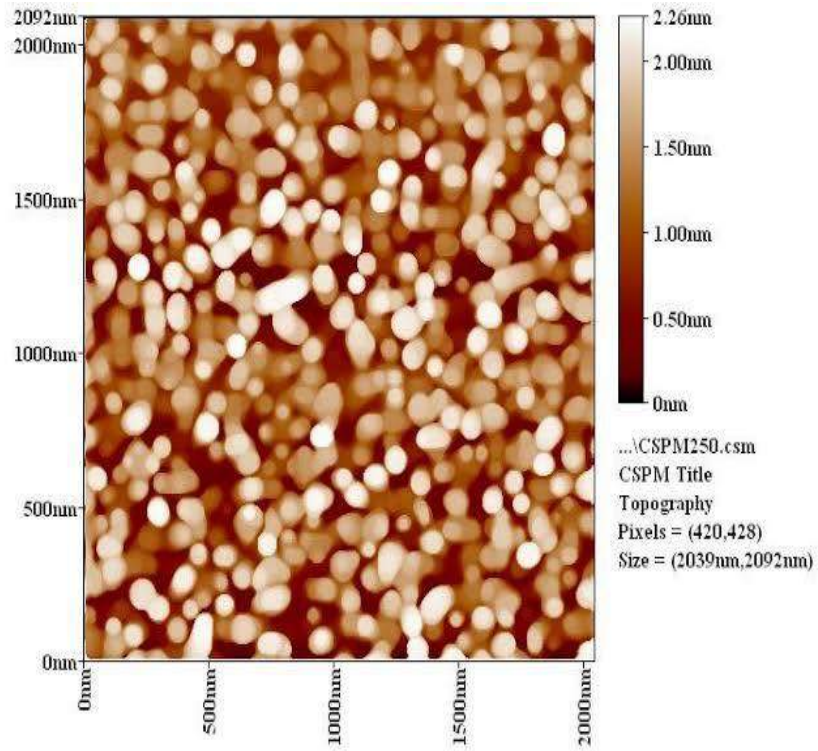The AFM image of RGO is shown in figure 3.It is clear the thickness of RGO layers is about 2.24 nm.

Figure 3.2D AFM image of RGO

Figure 4. shows the SEM image of RGO.The figure exhibits a transparent nanosheets of RGO layers. Also, RGO shows a crumpled and wrinkled morphology.
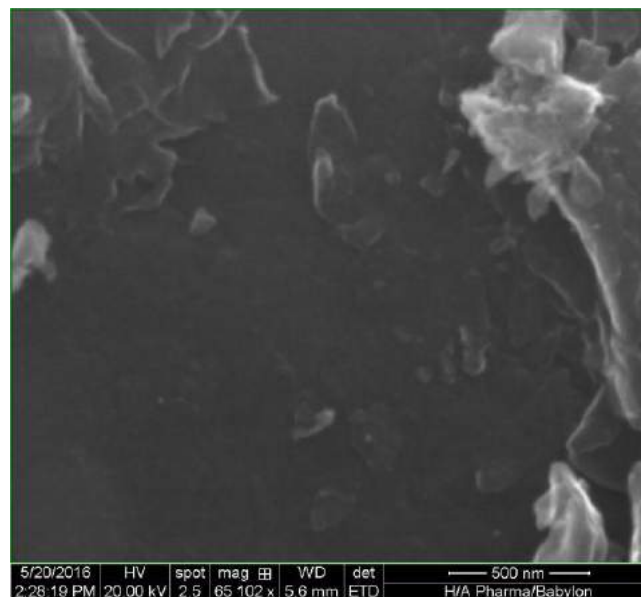

Figure 4.SEM image of RGO

3.2 Effect of pH

Figures 5,6, and 7. and table 1. show the effect of medium pH on the rate of RGO formation. it is clear that the increasing of ph enhances the reduction of GO. The obtained result is in acceptable agreement with the results obtained from other works[26].This may due to the fact that antioxidant is difficult to oxidized at lower pH values, so the higher pH values cause an acceleration of antioxidant transform to dehydrogenation state that held a high reducing potential[26]. RGO concentration and rate formation were calculated depending on the absorption coefficient that reported in our previous work(thesis) which displayed a mean value of 2900 L g^{-1} m^{-1} in distilled water.
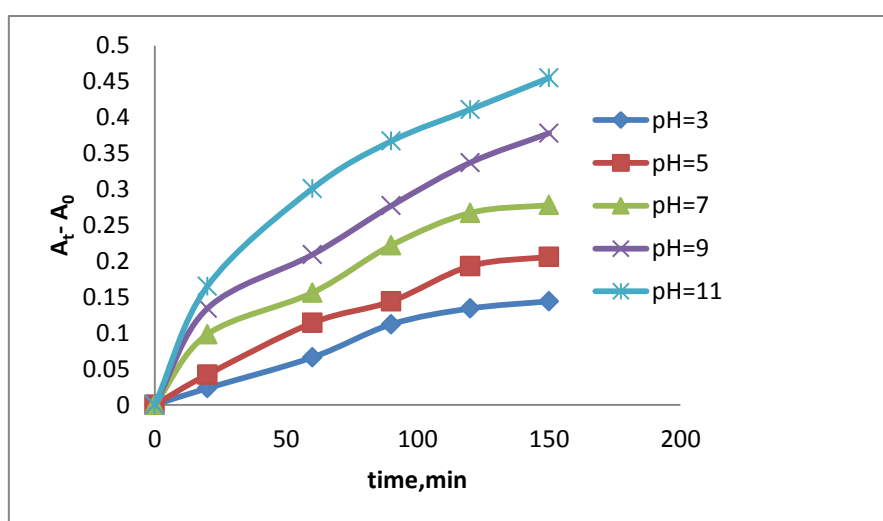

Figure 5.Change in A_t-A_0 as a function of reaction time under different pH media and sonication treatment. GO concentration 0.3 g/100 mL, extract conc.0.3%,temperature 298 K,ultrasound frequency 40KHz.
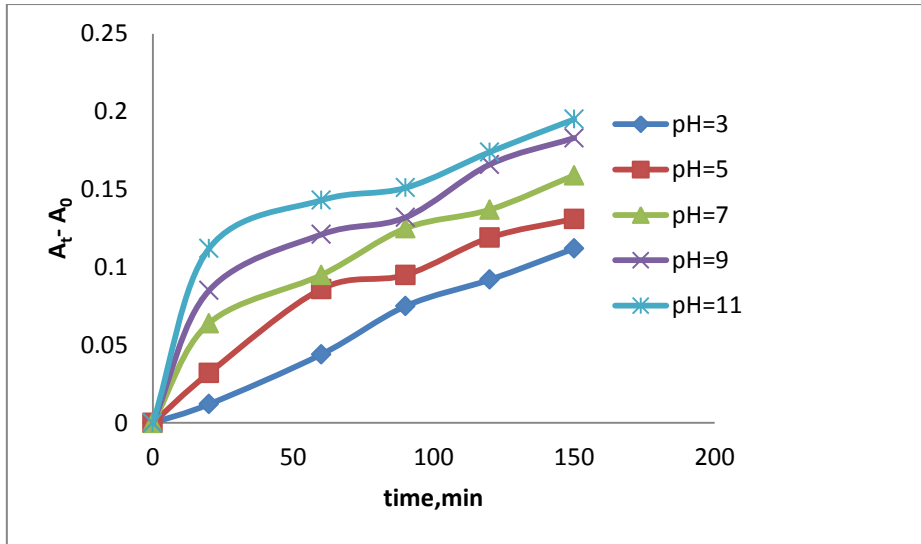
Figure 6.Change in A_t-A_0 as a function of reaction time under different pH media and mixing tratment. GO concentration 0.3 g/100 mL, extract conc.0.3%,temperature 298 K
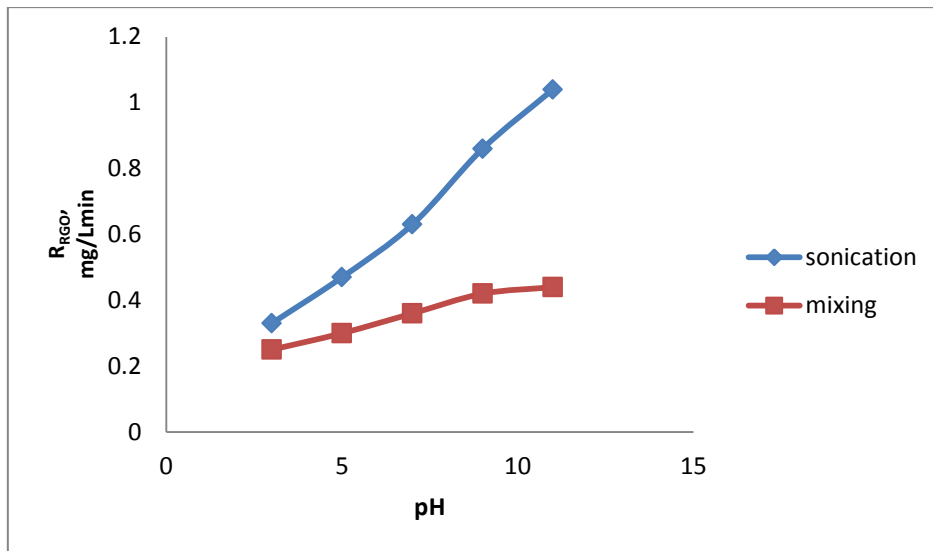

Figure 7.Effect of pH upon rate formation of RGO . Treatment time 150 min,GO concentration 0.3 g/100 mL, extract conc.0.3%,temperature 298 K,ultrasound frequency 40KHz(in case of sonication treatment)

Table 1.Effect of pH upon RGO concentration(mg/L). Treatment time 150 min,GO concentration 0.3 g/100 mL, extract conc.0.3%,temperature 298 K,ultrasound frequency 40KHz(in case of sonication treatment).

pH treatment	3	5	7	9	11
mixing	38.6	45.1	54.8	63.1	67.2
sonication	49.6	71.0	95.8	130.3	156.8

3.3 Effect of extract concentration

As known that reductants had a reducing potential more than oxidants, higher reductant concentration leads to higher formation rate of reduced material. Figures 8 and 9. and table 2. show that RGO rate increases as extract concentration increases. The reason may be described as an increasing of antioxidant molecules with fixation of oxidant concentration, produces more electrons that drive reduction process.
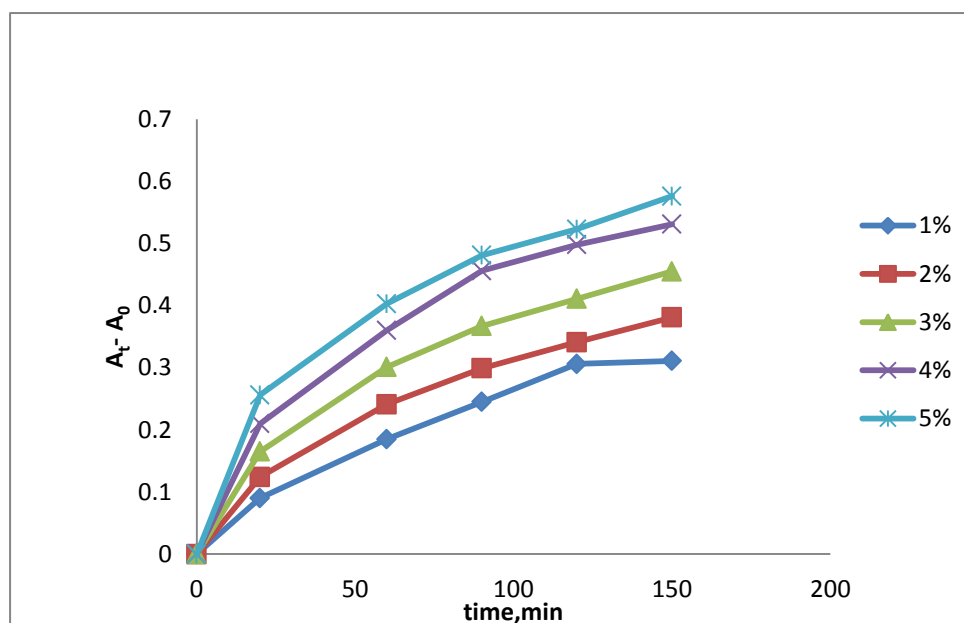

Figure 8.Change in A_t-A_0 as a function of reaction time under different extract concentrations and sonication treatment. GO concentration 0.3 g/100 mL, 0.3%,pH 11,temperature 298 K,ultrasound frequency 40KHz
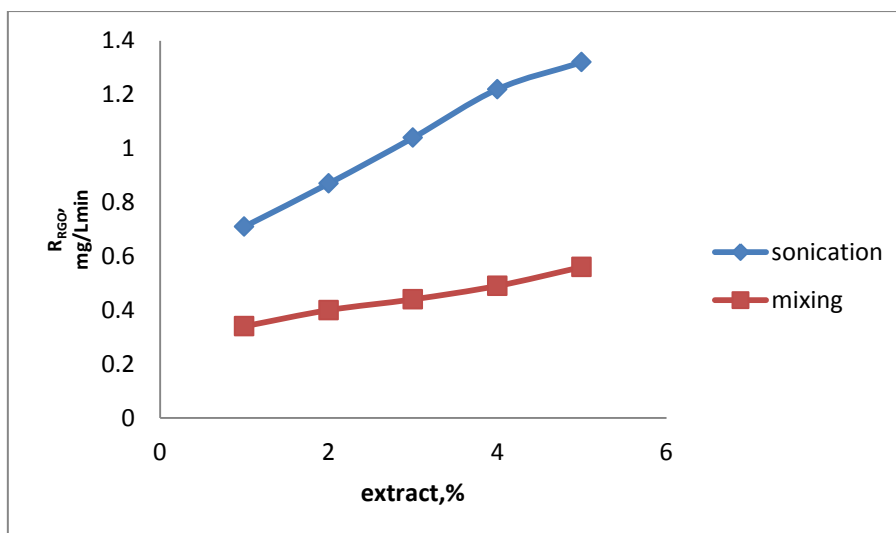
Figure 9.Effect of extract concentration upon rate formation of RGO . Treatment time 150 min,GO concentration 0.3 g/100 mL,pH 11,temperature 298 K,ultrasound frequency 40KHz(in case of sonication treatment)

Table 2.Effect of extract concentration upon RGO concentration(mg/L) . Treatment conditions : treatment time 150 min,GO concentration 0.3 g/100 mL,pH 11,temperature 298 K, ultrasound frequency 40KHz(in case of sonication treatment).

Extract,% treatment	1	2	3	4	5
mixing	52.4	60.3	67.2	73.7	84.4
sonication	107.2	131.3	156.8	183.1	198.6

3.4 Effect of temperature

Energy either heat or ultrasound power are an important factors that affected RGO formation[42-45] .Figures 10 and 11 and table 3. illustrate the relationship between A_t-A_o and time as a function of temperature changing. The results are clearly indicated that the increasing of solution temperature causing an increase in RGO rate formation. It considered that relationship between temperature and rate of RGO is positive and Arrhenius plot is obtained(figure).
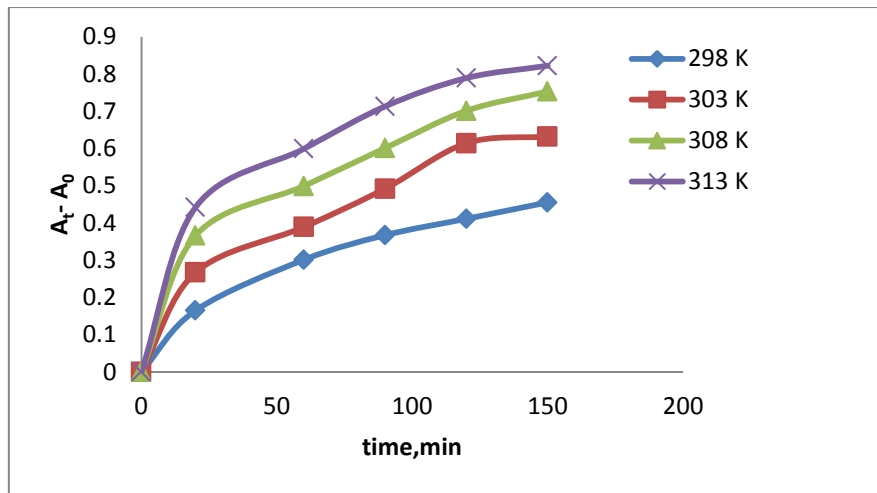
Figure 10.Change in A_t-A_0 as a function of reaction time under different temperatures and sonication treatment. GO concentration 0.3 g/100 mL, extract conc.0.3%,pH 11,temperature 298 K,sonication frequency 40KHz(incase of sonication treatment)
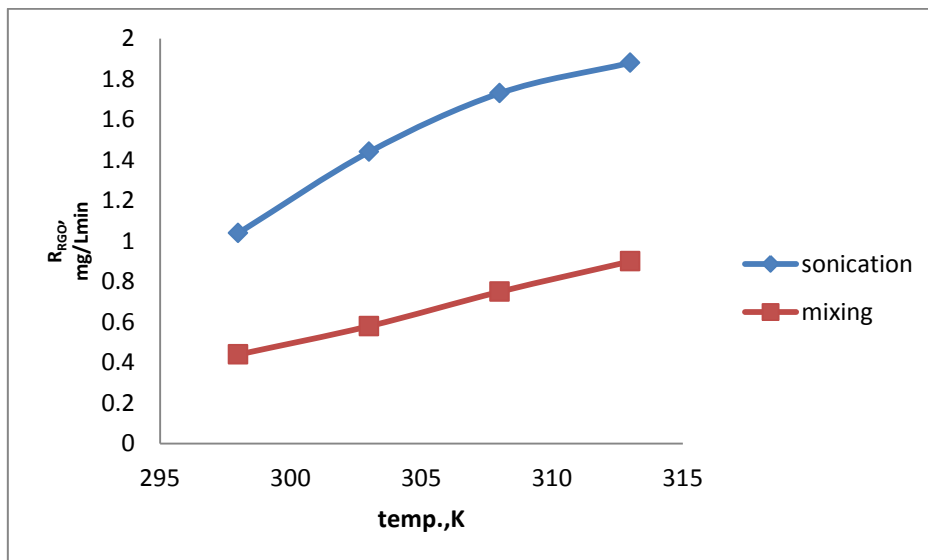

Figure 11.Effect of temperature upon rate formation of RGO . Treatment time 150 min,GO concentration 0.3 g/100 mL, extract concentration 3%,pH 11,ultrasound frequency 40KHz(in case of sonication treatment.

Table 3.Effect of temperature upon RGO concentration (mg/L). Treatment conditions : treatment time 150 min,GO concentration 0.3 g/100 mL,extract conc.0.3%,pH 11 ultrasound frequency 40KHz(in case of sonication treatment).

Temp.,K treatment	298	303	308	313
mixing	67.2	88.2	112.7	135.5

sonication	156.8	217.9	259.6	283.4

As reaction rate constant is directly proportional with reaction rate ,the ln k_{RGO} is replaced by ln R_{RGO} in logarithmic formula of Arrhenius equation as following[26] :

$$ln\ R_{RGO}=(-E_a/RT)+ln\acute{A} \qquad (3)$$

Where $\acute{A}$ is the constant related to the frequency factor in pristine Arrhenius equation, E_a is the pseudo activation energy(KJ mol^{-1}),R is the gas constant(8.314 J mol^{-1} K^{-1}),and T is the solution temperature(K).Pseudo activation energy calculated from plot shown in figure was() for ultrasound and ()for mixing respectively. It is noticed, that the temperature of ultrasound irradiated solution is higher than bath temperature that controlled by temperature regulator [26,47,48].The temperature change (ΔT) over time during sonication of GO suspension needs to correction. The correction of temperature was done as mentioned in table one ,and activation energy after correction was Kj mol^{-1}. The sonicator bath tends to heat up to 40^{0}C ,because a temperature over that, an evaporation is occur .Water level was continuously watched during irradiation.Also,a 20W output power of bath was adopted, which considered moderate power. The using higher output power, a degradation of solvent and reactants may be occur as a result of that water bath tends to heat significantly[38] .
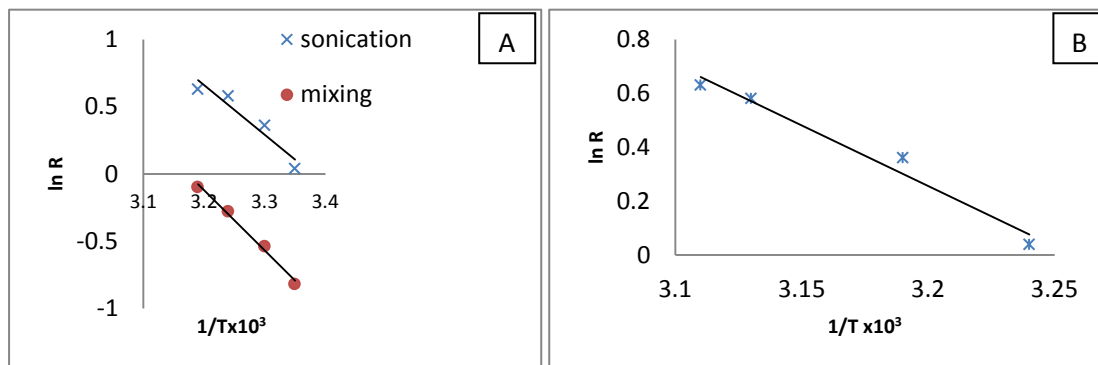

Figure12.Relationship between 1/T and ln R underg treatment time 150 min,GO concentration 0.3 g/100 mL,extract conc.0.3%,pH 11 ultrasound frequency 40KHz(in case of sonication treatment). A:ultrasound treatment before temperat ultrasound treatment before temperature correction

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