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# Synthesis of Graphene oxide nanocomposite and its spectral characterization

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**Abstract:** *In the present study, clay - graphene oxide nano composite catalysts were successfully used for the first time in the multi component one pot organic synthesis. The facile development of the hybrid clay - graphene oxide based materials has been achieved by a cost effective method without the use of any surfactants. The partial reduction of graphene oxide, upon incorporation of clay layers and subsequent heat treatment, is evident from the X-ray diffraction patterns and FTIR spectra of the samples. XPS and <sup>27</sup>Al and <sup>29</sup>Si NMR spectral analyses provide useful information regarding the interaction between clay layers and graphene oxide through Si-O-C and Al-O-C bonding. The deconvoluted spectrum of O (1s), Al (2p) and Si (2p) indicates the increased availability of acidic functionalities in the hybrid nanocomposite. FESEM and TEM photographs show the random distribution of the clay nanoflakes over the graphene oxide sheets and this could provide more of the active sites for catalysis. Synthesis of 3, 4- dihydropyrimidinones by the one pot Biginelli reaction was done over the present clay - graphene oxide heterogeneous catalysts with high product yield. Short time period of reaction and excellent reusability up to 8 repeated cycles under solvent free conditions are the key advantages of the present highly active hybrid nanocomposite clay - graphene oxide catalysts over most of the other reported catalysts used for Biginelli reaction.*

**Keywords:** *graphene oxide catalysts, nano composite catalysts*

## I. INTRODUCTION

Multicomponent reaction (MCR), one of the powerful tools for modern synthetic chemists (Weber, 2002), involves coupling of more than two starting materials in a one pot reaction condition to form a single complex product containing most of the atoms of the starting reagents (Hulme and Gore, 2003; Ugi et al., 1994)[1]. Biginelli reaction is a well-known MCR, which involves the synthesis of 3,4- dihydropyrimidinone (DHPM) by one pot condensation reaction of an aldehyde, ethyl acetoacetate and urea in ethanol (Biginelli, 1893; Kappe, 2000). The Biginelli analogues are famed for their biological activities such as antibacterial, antimalarial, anticancer, antiviral, anti-tuberculars, and anticonvulsants (Hentrich, 1932; Akhaja and Rava, 2011; Ramachandran et al., 2016; Prashantha Kumar et al., 2009). Also, DHPM is the core structure of natural marine alkaloid batzelladine which is used for the inhibition of the binding of HIV gp-120 to CD4 cell (Snider et al., 1996; Rama Rao et al., 1995)[2]. There are many reports on Biginelli reaction over various homogeneous and heterogeneous catalysts. The catalysts such as ion exchange resins (Joseph et al., 2006), YbCl<sub>3</sub> (Zhang et al., 2009), triphenylphosphine (Debache et al., 2008), propane phosphonic acid (Zumpe et al., 2007), phenylboronic acid (Debache et al., 2006), sulphamic acid (Chen et al., 2007), and sulphated carbon (Moghaddas et al., 2012) were used for the synthesis of DHPMs via Biginelli reaction. All these works are valuable; however, many of them encompass drawbacks such as the use of hazardous and/or expensive reagents, prolonged reaction time, low yield, tedious workup, and requirement of stoichiometric amounts of catalysts; in addition some of the reported procedures also cause environmental pollution. Nowadays, modern green practices give more attention towards the development of environmentally friendly, efficient and synthetic chemical procedures. In this perspective, in order to conquer the drawbacks of the above mentioned procedures and to accomplish most of the aspects of green and sustainable chemistry, the use of highly active low cost nonhazardous heterogeneous catalysts is essential (Herrmann and Cornils, 1997; Wilson and Clark, 2000)[3].

The thinnest material graphene has got much attention from various researchers because of its extraordinary thermal, electrical and mechanical properties (Geim and Novoselov, 2007; Geim and MacDonald, 2007; Novoselov et al., 2004)[4]. Owing to its high surface area, graphene is well established as a solid support for various metals and metal oxides and the prepared composite materials were effectively used as catalysts (Lu et al., 2009; Bruno and Machadoab, 2012; Liang et al., 2011). Among the various methods of preparation of graphene, better yield is obtained by the reduction of graphene oxide (GO) Park and Ruoff, 2009; Tapas et al., 2012. Even though the use of GO is exploited in various applications such as supercapacitors, photocatalysis, heterogeneous catalysis, electrocatalysis, fuel cells and solar cells (Kamat, 2011; Zhou et al., 2010; Seger and Kamat, 2009; Li et al., 2010), the use



of GO based materials as catalysts for multicomponent organic transformations is limited in number (Kundu and Basu, 2015; Xiong et al., 2014; Sengupta et al., 2014)[5]. Clay minerals are widely used heterogeneous catalysts due to the simplicity in workup, good performance, under mild experimental conditions, high product yield with excellent selectivity, low cost, eco-friendly nature and natural availability (Nagendrappa, 2002; Varma, 2002). Many modification methods are offered to improve the catalytic performance of clays[6]. Hybrid materials of clays with other inorganic/organic materials were also widely applied in catalysis (Yadav et al., 2004a; Binitha and Sugunan, 2008; Chen et al., 1995).

Even before the rediscovery of graphene in 2004 (Geim and Novoselov, 2007), there are reports on graphite like two dimensional carbon sheets stacked between the clay layers (Kyotani et al., 1988; Sonobe et al., 1991). Later on, Nethravathi et al. in 2008 had reported the preparation of bentonite clay – graphene nanocomposites (Nethravathi et al., 2008a)[7]. Surfactants such as octylamine and cetyltrimethylammonium bromide were used in the preparation for the co-stacking of these layered materials. Nethravathi et al. had also prepared graphite oxide intercalated anionic clay (nickel zinc hydroxyl salt) using the aqueous colloidal dispersions of negatively charged graphite oxide sheets and aminobenzoate intercalated anionic clay layers as precursors (Nethravathi et al., 2008b). Senthilnathan et al. had recently reported the preparation of nitrogen functionalized graphene/nanoclay hybrid via submerged liquid plasma approach (Senthilnathan et al., 2014)[8]. Clay – graphene nanocomposites prepared using sucrose as the graphene precursors were effectively used for hydrogen storage by Ruiz-Garcia et al. (2013). Zhang et al. had investigated the use of reduced graphene oxide – montmorillonite nanocomposite in hexavalent chromium removal from aqueous solutions (Zhang et al., 2015). Ascorbic acid was used as the graphene oxide reductant in that study. In addition, there are some reports on clay/graphene/polymer hybrid nanocomposites (Zaman et al., 2014; Longun et al., 2013). Recently, montmorillonite pillared GO was used as an adsorbent in the removal of Pb<sup>2+</sup> and methylene blue by Liu et al. (2014). Achari et al. reported the synthesis of highly water dispersible aminoclay–reduced graphene oxide hybrids by the in situ condensation of aminoclay over graphene oxide (GO) followed by reduction using hydrazine hydrate and had studied the biomedical applications of the composite (Achari et al., 2013). Behrouz et al. have reported the catalytic synthesis of 3,5- disubstituted isoxazoles in 80–92% yields using Cu/graphene/clay nanohybrid materials (Somayeh and Rad, 2015). Cu/graphene/clay nanohybrid was also used for the synthesis of carboacyclic nucleosides by 1,3-dipolar cycloaddition (Rad et al., 2015)[9].

To the best of our knowledge montmorillonite clay – GO nanocomposite catalyzed CR are not yet reported. In the present work, montmorillonite KSF clay (Mont.KSF) – GO nanocomposite is synthesized via a facile procedure and Biginelli reactions have been carried out in one pot over these composite materials. We have performed an economical method for the production of different weight percentage of clay loaded GO nanocomposite catalysts without the use of any surfactants. The catalytic MCR was executed under sustainable solvent free condition.

## II. EXPERIMENTAL SECTION

### A. Materials

Graphite flakes and Mont.KSF were purchased from Sigma Aldrich chemicals. Ethyl acetoacetate and methyl acetoacetate were procured from MERCK specialties Pvt. Ltd. and Loba Chemie respectively[10]. 4-nitro benzaldehyde and 2-nitrobenzaldehyde were purchased from Spectrochem and all other chemicals were obtained from NICE chemicals Pvt. Ltd, India. All reagents were of analytical grade and used as such without further purification. The reaction products were characterized by comparing their spectral (IR and NMR) and physical data with authentic samples.

### B. Preparation of Graphite Oxide(GO)

Graphite oxide was prepared by modified Hummers method (Hummers and Offeman, 1958). A mixture of graphite powder (2 g) and NaNO<sub>3</sub> (1 g) was added into concentrated H<sub>2</sub>SO<sub>4</sub> (96 ml) kept in an ice bath. KMnO<sub>4</sub> (6 g) was added gradually to the above mixture with vigorous stirring by maintaining the temperature below 20 °C. Then the mixture was stirred at 35 °C in a water bath for 18 h. The mixture turned into a brownish paste as the reaction commenced. 150 ml of distilled water was added to the paste in a drop wise manner while keeping the temperature below 50 °C. Thereafter the mixture was diluted with 240 ml of water and 5 ml of 30% H<sub>2</sub>O<sub>2</sub> was then added[11]. The colour of the diluted solution was changed to brilliant yellow along with bubbling in the solution. After continuous stirring for 2 h, the mixture was filtered and washed with 250 ml of 10% HCl, deionized water, and ethanol to remove ionic impurities. The resultant solid was dried at room temperature.



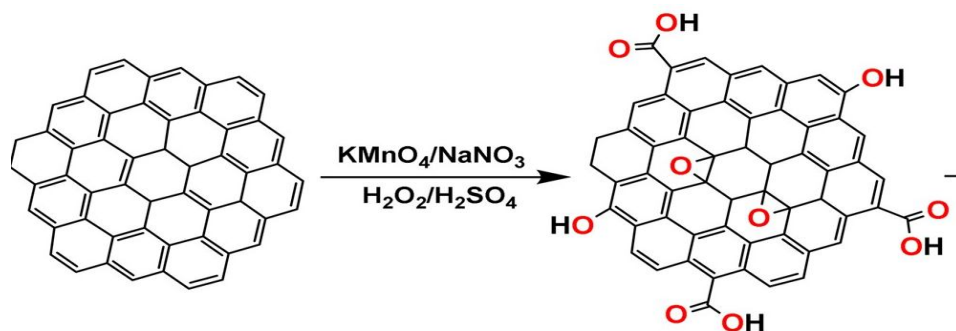


Fig.1. Preparation of graphite oxide(GO)

- 1) Preparation of Mont. KSF clay – GO Nanocomposite: In the preparation of Mont.KSF – GO nanocomposite (with clay:GO weight ratio of 10:1), initially graphene oxide solution was prepared by the sonication of 0.25 g graphite oxide in 500 ml water for 30 min. To the swelled Mont.KSF clay solution (2.5 g in 250 ml), GO solution was added in a dropwise manner with stirring at 80 °C (addition was done with 4 h duration and the mixture was stirred again for one more hour)[12]. The settled clay – GO composite was filtered, washed with dis- tilled water and treated at 200 °C for 3 h. The same procedure was repeated for the preparation of different Mont.KSF – GO nanocomposites having clay:GO weight ratios of 0.5:1, 1:1, 2:1, and 5:1. The catalysts were designated as CG(n:1) where n indicates weight percentage of Mont.KSF with GO.

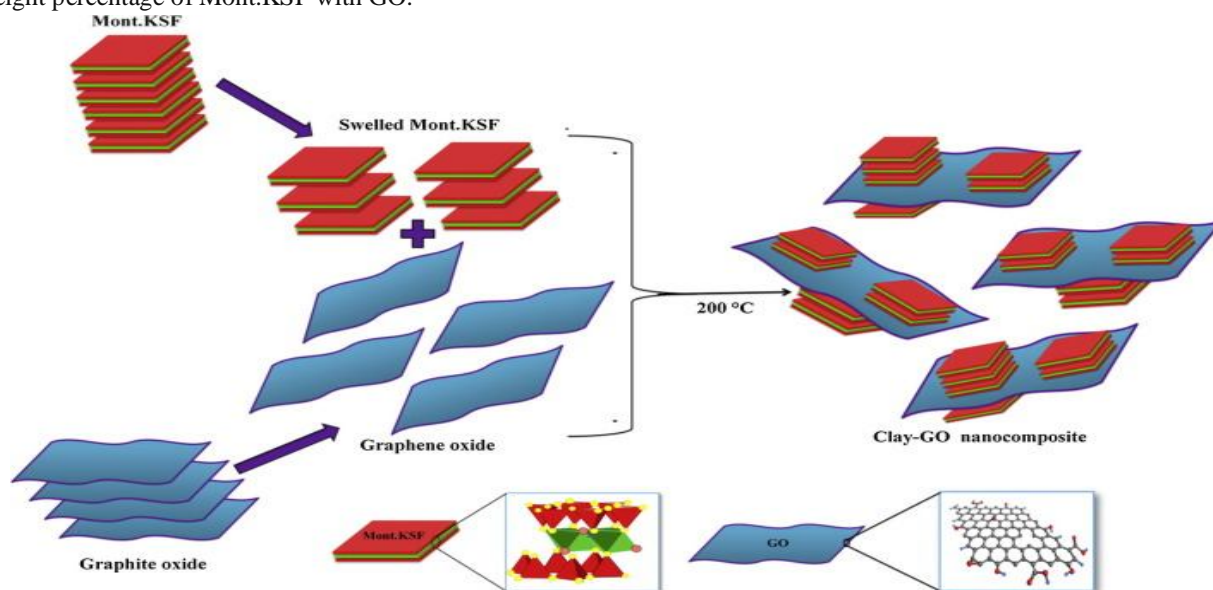


Fig.2 Formation of Clay-GO nanocomposite

### C. Preparation of Dihydropyrimidinones

The typical reaction condition selected for the preparation of DHPM is as follows unless otherwise mentioned. A mixture of aromatic aldehyde (1 mmol), ethyl acetoacetate (1 mmol), urea (1.3 mmol) and catalyst (0.1 g) was heated at 130 °C under stirring until the mixture got solidified, where the stirring became ineffective. Hot ethanol was added to this mixture and the catalyst was removed by filtration. Pure product was afforded by crystallization using ethanol. So as to investigate the reusability of CG(10:1) nanocomposite for the above reaction, the catalyst after each run was washed several times with hot ethanol and dried at 200 °C for 2 h[13].

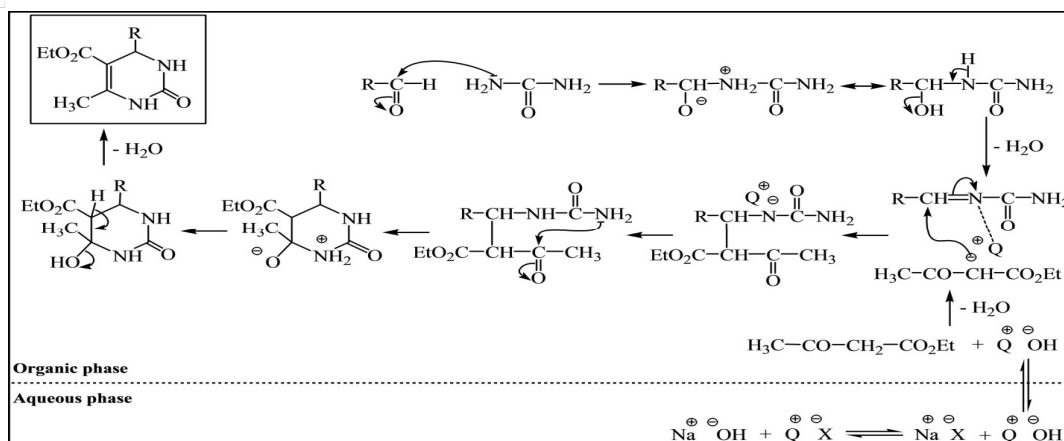


Fig.3. Preparation of dihydropyrimidinones

#### D. Characterization Techniques

X-ray diffraction (XRD) patterns of the catalyst samples were taken using Rigaku Miniflex 600 X-ray powder diffractometer equipped with Cu Ka radiation. FTIR spectra of different systems were recorded using KBr pellets method with Jasco FT/IR (4100) spectrophotometer. XPS analysis was done over AXIS ULTRA X-ray Photoelectron Spectrometer (KRATOS ANALYTICAL) with C1s at 284.6 eV as internal standard. Deconvolution of the XPS spectra of each element was done using Gaussian curve fitting. The solid state <sup>29</sup>Si and <sup>27</sup>Al NMR spectra were recorded on ECX400-Jeol 400 MHz High Resolution Multinuclear FT-NMR spectrometer. The <sup>29</sup>Si and <sup>27</sup>Al NMR were obtained at 79.42, 104.17 and 100.52 MHz respectively. The surface morphologies of synthesized nanocomposites were investigated by FESEM (Zeiss SUPRA55 scanning electron microscope at an operating voltage of 3 kV). TEM analysis of representative samples was taken on a Philips CM-12 instrument operated at 100 kV. FESEM/EDAX with elemental mapping of CG(10:1) was done on Carl Zeiss Microscopy (Model-UK & SIGMA). Raman spectral analysis was performed over WITEC AIFA300RA CONFOCAL RAMAN MICROSCOPE WITH AFM with 532 nm DPPS Laser excitation. The purity of Biginelli compounds was checked by TLC using silica gel and spots were visualized by exposing the dry plates in iodine vapours. <sup>1</sup>H NMR spectra of the Biginelli products in DMSO were recorded on a BRUKER 500 MHz spectrometer using TMS as the internal standard[14]. The melting points of the products were recorded on a melting point apparatus in open capillary tubes and are uncorrected.

### III. RESULTS AND DISCUSSION

Here, clay – GO nanocomposites were successfully prepared by a scalable and cost-effective method without the use of any organic modifiers. The schematic representation for the formation of clay – GO nanocomposites is shown in Scheme 1. Mixing of graphene oxide with swelled clay and further heat treatment leads to the formation of a uniform composite[15]. The prepared composite catalyst systems were characterized using different techniques to investigate the structural characteristics of these hybrid materials.

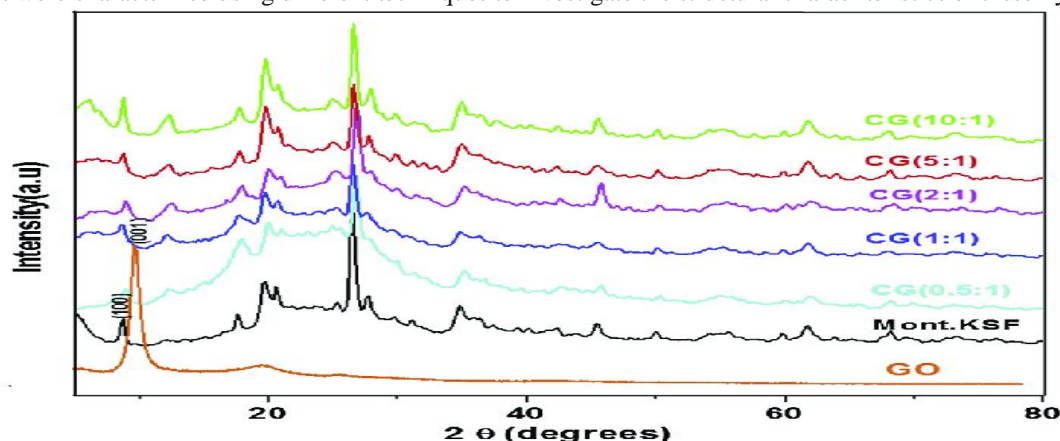


Fig. 4 shows the XRD patterns of different systems.

For Mont. KSF, the peak at  $8.9^\circ$  corresponds to diffraction from (100) plane with an interplanar distance of  $9.8 \text{ \AA}$  (Binitha and Sugunan, 2006; Farias et al., 2011). The prepared nanocomposites contain all the peaks of Mont.KSF clay, but with reduced intensity. A new peak around  $12.3^\circ$  in the nanocomposites may correspond to the characteristic diffraction from (001) plane of GO, indicating the presence of graphene in its oxidized form. Increase in the  $2\theta$  value when compared with that of bare GO (at  $9.9^\circ$ ) indicates decrease in the interlayer distance between graphene oxide sheets, which may be due to the elimination of some of the oxidized groups (Tapas et al., 2012; Guo et al., 2009a; Shabnam, 2013) during composite formation. The absence of graphitic peak around  $26^\circ$  additionally indicates the increase in the interlayer distance of all the graphite layers as a result of the incorporation of oxygen moieties in the sample (Shahriary and Athawale, 2014). The FTIR spectra of different systems are shown in Fig. 2. FTIR spectrum of Mont.KSF shows the characteristic vibration bands around  $3620 \text{ cm}^{-1}$ ,  $3450 \text{ cm}^{-1}$ ,  $1645 \text{ cm}^{-1}$  and  $1045 \text{ cm}^{-1}$  corresponding to the AOH stretching of lattice hydroxyl groups, AOH stretching from adsorbed water, AOH bending and Si-O stretching vibrations respectively (Binitha and Sugunan, 2006). In the FTIR spectrum of GO, band centred around  $3450 \text{ cm}^{-1}$  corresponds to AOH stretching vibration. The bands around  $1740 \text{ cm}^{-1}$ ,  $1630 \text{ cm}^{-1}$ ,  $1400 \text{ cm}^{-1}$ , and  $1040 \text{ cm}^{-1}$  indicate the presence of C=O, aromatic C=C, hydroxyl C-OH and epoxy C-O-C vibrations respectively (Guo et al., 2009a; Park et al., 2009). The vibration bands corresponding to clay as well as GO were retained in the hybrid materials with reduced intensities. The intensity of the vibration band around  $3620 \text{ cm}^{-1}$  increased with the clay content in the composites as expected. The carbonyl peak intensity is reduced in the nanocomposites indicating the reduction or conversion of the  $>\text{C}=\text{O}$  to other groups. The intensity of alcoholic C-OH band at  $1400 \text{ cm}^{-1}$  increases with increase in the clay content suggesting the role of clay in the partial reduction of carbonyl groups in GO.

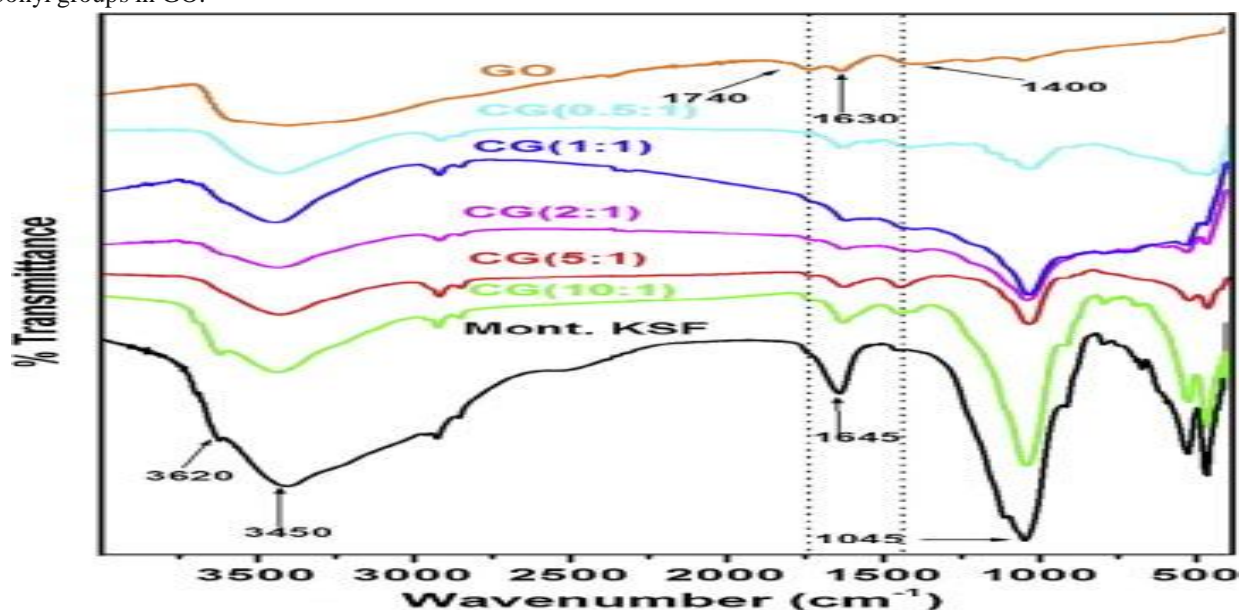


Figure 5 FTIR patterns of Mont.KSF, GO and CG(n:1) nanocomposites.

#### A. NMR Spectral Analysis

The  $^{29}\text{Si}$  NMR and  $^{27}\text{Al}$  NMR of montmorillonite KSF clay and CG(10:1) are shown in Fig. 4(a) and (b) respectively. Solid state NMR spectra of clays provide useful information about the type of coordination of Si and Al in the clay layers. From both the spectra, it is clear that the basic resonating peaks are observable in Mont.KSF and CG(10:1) indicating the retention of clay framework even after the composite formation. The  $^{29}\text{Si}$  NMR spectra of Mont.KSF show three resonating peaks at  $\delta$  values of 86.2, 91.2 and 107.56, corresponding to Q3 (2Al), Q3 (1Al) and Q3 (0Al) respectively. The Q3 (2Al), Q3 (1Al) and Q3 (0Al) can be attributed to the tetrahedrally coordinated silicon bonded to two Al atoms, 1 Al atom and 0 Al atom respectively with oxygen as rest of the coordinated species. The values at 107.56 and 110.0 ppm correspond to alpha quartz and crystabollite silica structure respectively. The  $^{29}\text{Si}$  NMR spectra of the CG(10:1) nanocomposite contain an intense peak with a deshielded  $\delta$  value of 89.9 (Q3(1Al)) and one broad peak with values 105.98 and 107.77 ppm. In CG(10:1), the peak corresponding to Q3 (2Al) is not distinct and is merged with the Q3 (1Al) peak.

In the  $^{27}\text{Al}$  NMR spectra, the chemical shift at 5 ppm for Mont.KSF and 6.8 ppm for CG(10:1) was attributable to the octahedrally coordinated aluminium (Al(VI)). Similarly for tetrahedrally coordinated aluminium (Al(IV)), the d value is obtained at 74.14 ppm with low intensity for both Mont. KSF and CG(10:1), which indicates the presence of low amount of tetrahedrally coordinated aluminium in the sam- ples. The small resonance bands on either side of the principal band indicate chemical shift anisotropy (Thompson, 1984; Ulicna et al., 2013; Fyfe et al., 1983).

It is discernible that in both  $^{29}\text{Si}$  and  $^{27}\text{Al}$  NMR spectra the d values are shifted to low field, which discloses the possibility of covalent bonding between the silicate and aluminate layer with GO. This shift of delta values to low field may be attrib- uted to the anisotropic deshielding effect of aromatic rings in the GO layers with the clay lamellae (Thompson, 1984).

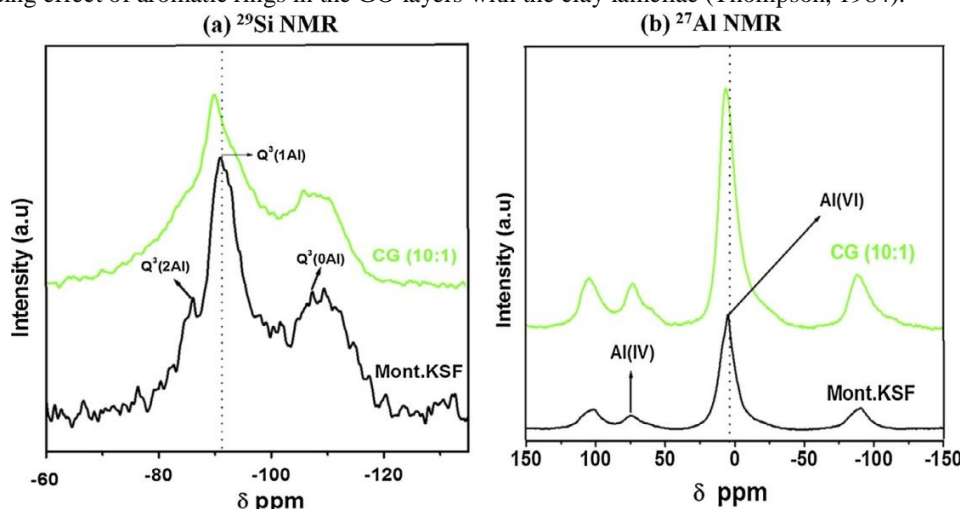


Figure 6 : (a)  $^{29}\text{Si}$  NMR spectra of Mont.KSF and CG(10:1), (b)  $^{27}\text{Al}$  NMR spectra of Mont.KSF and CG(10:1).

## REFERENCES

- [1] Achari, A., Datta, K.K.R., De, M., Dravid, V.P., Eswaramoorthy, M., 2013. Amphiphilic aminoclay-RGO hybrids: a simple strategy to disperse a high concentration of RGO in water. *Nanoscale* 5 (53), 16–5320. <http://dx.doi.org/10.1039/C3NR01108A>.
- [2] Akhaja, T.N., Rava, J.P., 2011. 1,3-dihydro-2H-indol-2-ones derivatives: design, synthesis, in vitro antibacterial, antifungal and antitubercular study. *Eur. J. Med. Chem.* 46, 5573–7559. [http:// dx.doi.org/10.1016/j.ejmech.2011.09.023](http://dx.doi.org/10.1016/j.ejmech.2011.09.023).
- [3] Bigdeli, M.A., Gholami, G., Sheikhhosseini, E., 2011. P-Dodecylbenzenesulfonic acid (DBSA), a Brønsted acid surfactant catalyst for Biginelli reaction in water and undersolvent free conditions. *Chin. Chem. Lett.* 22, 903–906. <http://dx.doi.org/10.1016/j.ccl.2010.12.030>.
- [4] Biginelli, P., 1893. Synthesis of 3, 4-dihydropyrimidin-2 (1H)-ones. *Gazz. Chim. Ital.* 23, 360–416.
- [5] Binitha, N.N., Sugunan, S., 2006. Preparation, characterization and catalytic activity of titania pillared montmorillonite clays. *Micro. Meso. Mater.* 93, 82–89. <http://dx.doi.org/10.1016/j.micromeso.2006.02.005>.
- [6] Binitha, N.N., Sugunan, S., 2008. Polyaniline/pillared montmorillonite clay composite nanofibers. *J. Appl. Pol. Sci.* 107, 3367–3372. [http:// dx.doi.org/10.1002/app.27353](http://dx.doi.org/10.1002/app.27353).
- [7] Bruno, F., Machadoab, P.S., 2012. Graphene-based materials for catalysis. *Catal. Sci. Tech.* 2, 54–75. <http://dx.doi.org/10.1039/C1CY00361E>.
- [8] Hausladen, M.C., Yang, R., 1995. Delaminated Fe<sub>2</sub>O<sub>3</sub>- pillared clay: its preparation, characterization, and activities for selective catalytic reduction of NO by NH<sub>3</sub>. *J. Catal.* 151, 135–146. <http://dx.doi.org/10.1006/jcat.1995.1016>.
- [9] Chen, W.Y., Qin, S.D., Jin, J.R., 2007. Efficient Biginelli reaction catalyzed by sulfamic acid or silica sulfuric acid under solvent-free conditions. *Synth Commun.* 37 (1), 47–52. [0.1080/00397910600977632](http://dx.doi.org/10.1080/00397910600977632).
- [10] Debache, A., Boumoud, B.M., Amimour, A., Belfaitah, S., Rhoutia, B., 2006. Carboni Phenylboronic acid as a mild and efficient catalyst for Biginelli reaction. *Tetrahedron Lett.* 47, 5697–5699. [http://dx. doi.org/10.1016/j.tetlet.2006.06.015](http://dx.doi.org/10.1016/j.tetlet.2006.06.015).
- [11] Debache, A., Amimour, M., Belfaitah, A., Rhouti, S., Carboni, B., 2008. A one-pot Biginelli synthesis of 3, 4-dihydropyrimidin-2- (1H)-ones/thiones catalyzed by triphenylphosphine as Lewis base. *Tetrahedron Lett.* 49, 6119–6121. <http://dx.doi.org/10.1016/j.tetlet.2008.08.016>.
- [12] Fana, X., Zhang, X., Zhanga, Y., 2002. Samarium chloride catalysed Biginelli reaction: one-pot synthesis of 3,4-dihydropyrimidin-2 (1H)-ones. *J. Chem. Res. (S)*, 436–438. <http://dx.doi.org/10.3184/030823402103172563>.
- [13] Farias, M., Martinelli, M., Rolim, G.K., 2011. Immobilized molybdenum acetylacetonate complex on montmorillonite K-10 as catalyst for epoxidation of vegetable oils. *App. Catal. A: Gen.* 403, 119–127. <http://dx.doi.org/10.1016/j.apcata.2011.06.021>.
- [14] Flament, O., Russat, J., Druet, E., 1990. X-ray photoelectron spectroscopy study of the interface between aluminum and spin-coated polyamic acid. *Adh. Sci. Tech.* 4 (1), 09–117. <http://dx.doi.org/10.1163/156856190X00135>.
- [15] Geim, A.K., Novoselov, K.S., 2007. The rise of graphene. *Nat. Mater.* 6, 183–191. <http://dx.doi.org/10.1038/nmat1849>.
- [16] Gopalakrishnan, A., Jeong, Y.T., 2010. A convenient one-pot Biginelli reaction catalyzed by Y(OAc)<sub>3</sub>: an improved protocol for the synthesis of 3,4-dihydropyrimidin-2(1H)-ones and their sulfur analogues. *Bull. Korean Chem. Soc.* 31 (4), 863–868. [http://dx. doi.org/10.5012/bkcs.2010.31.04.863](http://dx.doi.org/10.5012/bkcs.2010.31.04.863).
- [17] Guo, H.L., Wang, X.F., Qian, Q.Y., Wang, F.B., Xia, X.H., 2009a.





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