

# Preparation of Epoxy Functionalized Hybrid Nickel Oxide Composite Polymer Particles

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**Abstract** Preparation of inorganic–organic hybrid materials is attracting much interest as such elaboration produces remarkable improvement in properties and versatility in application potential. In this investigation, nanosized nickel oxide (NiO) particles were first prepared by calcination of nickel hydroxide precursor obtained by a simple liquid-phase process. The produced NiO particles were functionalized with oleic acid followed by silane coupling agent. Finally attempt was made to prepare epoxy functional hybrid NiO/poly(methyl methacrylate-glycidyl methacrylate) abbreviated as NiO/P(MMA-GMA) composite polymer particles by seeded copolymerization in ethanol media. The composite particles were colloidally stable, and the obtained particles were characterized by Fourier transform infrared, scanning electron microscopy, X-ray diffraction, and thermogravimetric analyses. Electron microscopy showed that the composite particles are doped with nanosized NiO.

**Keywords** Inorganic-organic Composite, Epoxy Functionality, Seeded Copolymerization, NiO, Doping

## 1. Introduction

Hybrid polymer composites are a class of materials that combine the properties of inorganic materials with the easy processability, flexibility, elasticity, lighter weight and improved colloidal stability of organic polymer particles. The fabrication of hybrid inorganic/organic polymer composites offers advantages especially when those applications depend on mechanical and surface properties. Now a day researchers are showing much interest on the preparation of hybrid polymer particles because one can enhance or even incorporate novel properties (e.g. mechanical, electrical, catalytic, rheological, magnetic and optical) by independently altering the composition, dimension and structure. These inorganic-organic hybrid polymer particles find wide application potentials in catalysis, electrical, optical and electronic or photonic devices, biomaterials, flame-retardant and coatings[1-9].

There is an increasing interest in the synthesis of nanosized metal oxides because of their large surface area unusual adsorptive properties, surface defects, fast diffusivities and quantum size effects which are different from those of bulk materials[10-15]. Metal (oxide) nano particles smaller than about 20 nm have received widespread interest recently because of their envisioned applications in

electronics, optics, and magnetic storage devices. Among the various oxide materials, nickel oxide (NiO) is a very important semiconducting oxide material extensively used in catalysis, battery cathodes, gas sensors, electrochromic films, and magnetic materials[16–25]. NiO catalyst exhibits good low temperature catalytic performance for oxidative dehydrogenation of ethane to ethylene reaction[26]. In addition NiO has received considerable interest for its catalytic properties in decomposition of ammonium perchlorate,[27] hydrocracking reactions, reforming of hydrocarbons, and methane for production of synthesis gas, the removal of tar followed by the adjustment of the gas composition in biomass pyrolysis/gasification, in cellulose pyrolysis, and so forth[28]. These mechanical and physical properties as well as application potentials can be further improved by reducing the size and size distribution in the nano-range[29].

Despite the wide application potential of hybrid NiO composite polymer particles in different industries only few reports are available on the designing of such polymer particles. López *et al.* reported the preparation of poly(methyl methacrylate) (PMMA)/NiO nanocomposites by in situ bulk polymerization[30]. Prior to this, they treated the NiO particles with silane coupling agent. In another research, polyaniline/NiO/sodium dodecylbenzenesulfonate composite microspheres were synthesized in micelles by using ammonium peroxydisulfate initiator[31]. In an early work the author studied the feasibility of seeded emulsion polymerization to encapsulate NiO nano particles within PMMA shell layer using different NiO/MMA ratio

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(w/w)[32]. However, this results in some free NiO particles though the encapsulation efficiency was pretty good.

In the present investigation attempt was made to prepare hybrid epoxy functional NiO/poly(MMA-glycidyl methacrylate (GMA)) composite polymer particles via seeded polymerization in presence of silane coupled functionalized NiO particles. The introduction of easily transformable epoxy group would give reactive starting material for the design of whole range of compounds with various functional groups. These groups can be employed directly or via modification using cationic, anionic, chelate-forming, or fluorescent routes for immobilization of biopolymers, dyes and other sensitive compounds[33-36]. Additionally PMMA has some interesting properties like exceptional optical transparency, good mechanical properties, good weatherability, acceptable thermal stability, moldability and easy shaping[30,37]. The preparation scheme of NiO/P(MMA-GMA) composite polymer particles are depicted in **figure 1**.

## 2. Experimental

### 2.1. Materials

MMA and GMA of monomer grade purchased from Fluka, Chemika, Switzerland were distilled under reduced pressure to remove inhibitors. Oil soluble initiator 2,2'-azobis(isobutyronitrile) (AIBN) from LOBA Chem., India was recrystallized from methanol, vacuum dried at 5°C and preserved in the refrigerator before use. Poly(vinyl pyrrolidone) (PVP) of molecular weight  $3.6 \times 10^5$  gmol<sup>-1</sup> from Fluka, Chemika, Switzerland were used as a steric stabilizer. 3-(Methacryloyloxy)propyltriethoxysilane (MPS) from Alfa Aesar, UK, was used as a coupling agent. Ethanol was dehydrated and distilled before use. Other chemicals

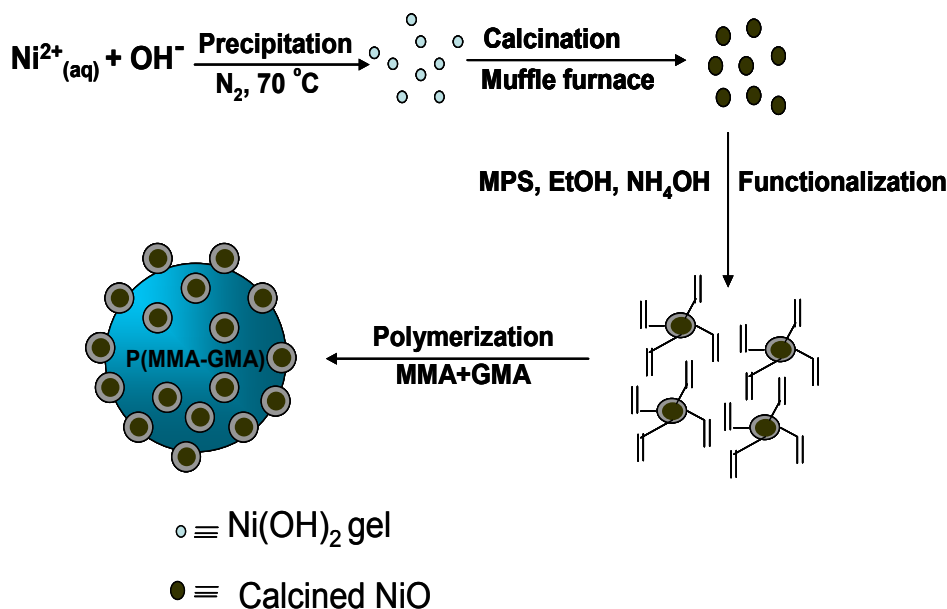
such as oleic acid, Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O, NH<sub>4</sub>OH were of analytical grade. Deionized water was distilled using a glass (Pyrex) distillation apparatus.

### 2.2. Instruments

Scanning electron microscope or SEM (LEO Electron Microscopy Ltd, Cambridge, UK) was used to see the morphology and particle size distribution. IR spectrophotometer (Perkin Elmer, FTIR-100, UK), proton NMR (JEOL spectrometer, 400 MHz, JNM-LA400, Japan) and table top high speed centrifuge machine (TG 16-WS, China) were used in this study. The X-ray diffraction (XRD) patterns of the powder samples were taken on Scanning X-ray Diffractometer (D8 Bruker AXS) using Cu K- $\alpha$  radiation (0.1542 nm) in parallel geometry (not focused). The intensities were measured at 2-theta values from 4.5° to 100° at a continuous scan rate of 10°/min with a position sensitive detector aperture at 3° (equivalent to 0.5°/min with a scintillator counter). Thermal analysis was carried out using a thermogravimetry analyser (TGA) from TGA Iris, TG 209 F1, Netzsch (Selb, Germany).

### 2.3. Preparation of Nanosized NiO Particles

Aqueous solution (0.6 M) of Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O was heated at 80°C, and the temperature was gradually increased to 100°C over 2 h under magnetic stirring. NH<sub>4</sub>OH was added slowly into the solution until pH 7 was reached. A green colloid solution was obtained, which was filtered and washed several times with deionized distilled water. The residual part was then dried at 100°C for 10 h to obtain the asprepared precursor, Ni(OH)<sub>2</sub> powder. The powder was calcined in platinum crucible at 400°C for 3 h to obtain the nanosized NiO particles.



**Figure 1.** Schematic representation for the preparation of NiO/P(MMA-GMA) composite polymer particles

## 2.4. Functionalization of NiO Particles

In order to improve the dispersibility, oleic acid stabilized NiO particles were prepared. First calculated amounts of NiO nano particles were dispersed in deionized distilled water by sonication for 1 h. Then 13 mL of oleic acid per unit mass of nano particles were added dropwise into the NiO nano particles dispersion at 80°C over the course of 2 h under vigorous magnetic stirring. The oleic acid stabilized NiO particles were washed repeatedly by serum replacement using water followed by ethanol to remove the excess oleic acid. 1.5 g of MPS per unit mass of oleic acid stabilized NiO particles was finally added and the dispersion was magnetically stirred at ambient temperature for 48 h under a nitrogen atmosphere. The MPS functionalized NiO particles were washed repeatedly by serum replacement using ethanol through repeated centrifugation.

## 2.5. Preparation of NiO/P(MMA-GMA) Composite Particles by Seeded Copolymerization

0.5 g of functionalized NiO particles dispersed in 50 g ethanol was taken in three necked round bottomed flask dipped in a thermostat water bath maintained at 70°C. 1.75 g of MMA and 0.75 g of GMA were added into the flask and 0.1 g of PVP was used as steric stabilizer. Polymerization was started by adding oil soluble AIBN (0.05 g) as initiator under a nitrogen atmosphere while the reaction was continued for 12 h.

The produced composite particles were washed three times by serum replacement to remove any unreacted monomer and initiator fragments.

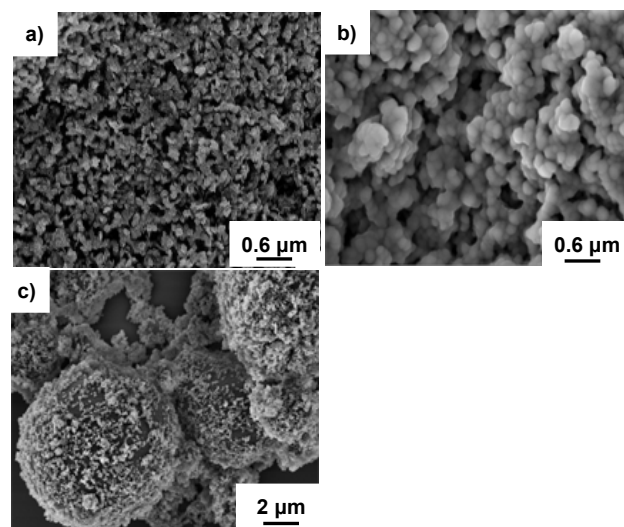
## 2.6. Thermogravimetric Analysis (TGA)

Thermal properties of the dried powder were measured by heating samples under flowing nitrogen atmosphere from 20°C to 800°C at a heating rate of 5°C/min and the weight loss was recorded.

## 3. Results and Discussion

SEM images of NiO, functionalized NiO and NiO/P(MMA-GMA) composite polymer particles are illustrated in **figure 2**. The average diameters of bare NiO particles are in the range of 30-70 nm and the average size of MPS modified NiO particles visibly increased to almost 200-300 nm. The increase in size is attributed to the functionalization of NiO particles by oleic acid and MPS. SEM image of NiO/P(MMA-GMA) composite polymer particles clearly shows deposition of smaller NiO particles on the surface of several micron-sized P(MMA-GMA) copolymer particles. From the image it is not clear whether some NiO particles are encapsulated within the polymer shell layer during seeded copolymerization though it was expected that presence of vinyl group in the silane coupling agent would favour copolymerization of MMA and GMA on the surface of functionalized NiO particles. The mechanism of formation

of NiO doped composite particles is not yet clear. However, during seeded copolymerization in ethanol medium the prior formation of P(MMA-GMA) primary particles from secondary nucleation can't be ruled out due to comparatively faster copolymerization reaction between MMA and GMA. As the concentration of MMA and GMA decreased towards the end of the copolymerization functionalized NiO particles participated in reaction through vinyl groups, resulted in NiO doped composite particles. It is also evident that NiO/P(MMA-GMA) composite particles are bit polydispersed ranging from four to several microns. Some small clusters of free NiO particles not participated in seeded copolymerization are also visible.

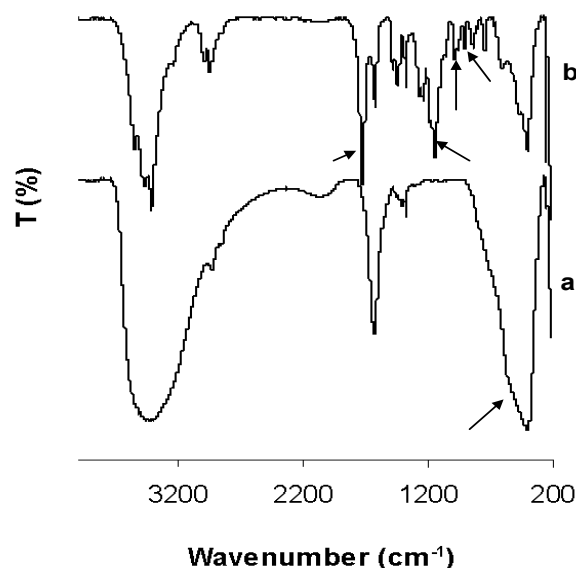


**Figure 2.** SEM photographs of NiO particles a), functionalized NiO particles b), and NiO/P(MMA-GMA) composite polymer particles c)

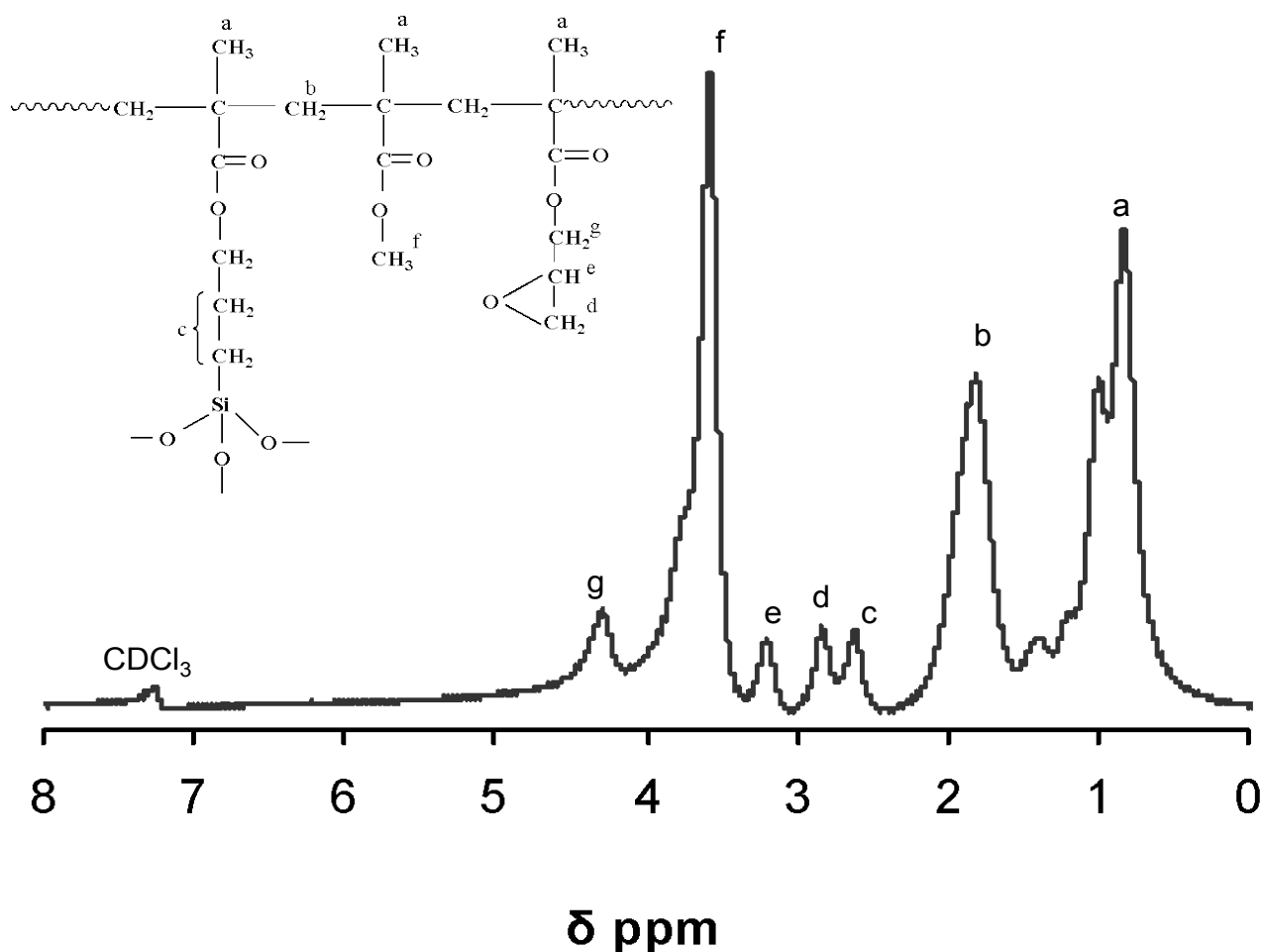
FTIR is an established tool to detect the surface properties of polymer particles[38,39]. **Figure 3** shows the FTIR spectra of functionalized NiO and NiO/P(MMA-GMA) composite polymer particles. These spectra were recorded for washed samples in KBr pellets. The FTIR spectrum of functional NiO shows absorption signal in the region 250-870  $\text{cm}^{-1}$  which indicates the formation of metal-oxygen bond (Ni-O). Additionally the broad and diffuse bands appeared at around 3383  $\text{cm}^{-1}$  and 1630  $\text{cm}^{-1}$  represent the existence of water in NiO. The identification of Ni-O bond in the FTIR spectrum confirmed the structure of NiO. The small absorption signal at 2912  $\text{cm}^{-1}$  band represents the aliphatic C-H stretching vibration derived from oleic acid and MPS. The spectrum of NiO doped composite polymer particles shows strong absorption signals at 1724  $\text{cm}^{-1}$  and 1142  $\text{cm}^{-1}$  due to characteristic C=O and C-O stretching vibration of ester derived from MMA and GMA comonomers. The weak absorption bands at around 960  $\text{cm}^{-1}$  and 833  $\text{cm}^{-1}$  represent the presence of epoxy group from GMA. The signal intensity of aliphatic C-H stretching vibration appeared at 2950.7  $\text{cm}^{-1}$  increased after the copolymerization reaction. Additionally the observance of comparatively weak and signal in the region 265-660  $\text{cm}^{-1}$  due to metal-oxygen bond in NiO indicates the formation of

NiO doped composite polymer particles. The change in chemical environment slightly shifted this absorption signal. The presence of free NiO particles not removed despite repeated washing may also contribute partially to this signal.

NMR is a better instrumental tool to check the composition of polymer.  $^1\text{H}$  NMR spectrum of NiO/P(MMA-GMA) composite polymer recorded in  $\text{CDCl}_3$  is shown in **figure 4** after removing the sedimented insoluble NiO. The signals at 0.81 and 1.78 ppm represent the C-CH<sub>3</sub> and C-CH<sub>2</sub>-C proton signals of MPS, MMA and GMA. The CH<sub>3</sub>-O-CO- proton signal of MMA and the -CH<sub>2</sub>-O-CO- proton signal of GMA appeared at 3.56 and 4.25 ppm respectively. The chemical shifts appeared at 3.16 and 2.80 ppm indicated the presence of epoxy protons from GMA. Both signals are shifted to upfield due to shielding of protons as the exact conformation of the soluble polymer is unknown. It is known that the chemical shifts are variable depend not only on the chemical environment but also on concentration, temperature and solvent.

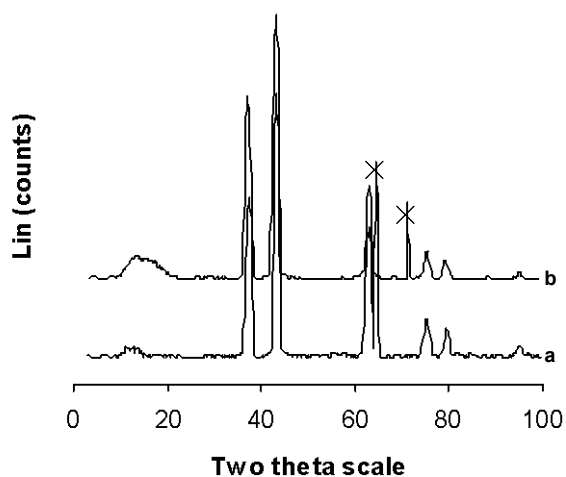


**Figure 3.** FTIR spectra of functionalized NiO particles a) and NiO/P(MMA-GMA) composite polymer particles b) recorded in KBr pellets



**Figure 4.**  $^1\text{H}$  NMR spectra of NiO/P(MMA-GMA) composite polymer particles taken in  $\text{CDCl}_3$

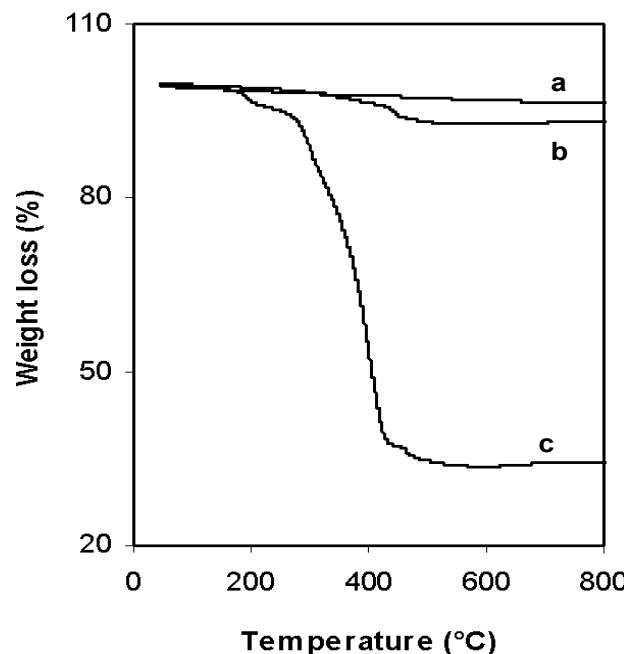
The NiO particles used to prepare functional hybrid composite polymer particles are appreciably crystalline in character as shown even for MPS modified NiO particles. The XRD spectrum of functionalized NiO particles presented in **figure 5** clearly shows the characteristic peaks of cubic crystals located at  $37.5^\circ$ ,  $43.5^\circ$ ,  $62.8^\circ$ ,  $75.5^\circ$  and  $79.8^\circ$  corresponding to the (111), (200), (220), (311) and (222) planes[30,40]. The structure of polymer matrix can be best explained by considering the spectra obtained at low angle region that gives idea about the amorphous property. In functionalized NiO a weak but broad signal that appeared at  $14.37^\circ$  indicated the presence of MPS and oleic acid layer and the intensity of this peak further increased with the incorporation of P(MMA-GMA) component. This behaviour suggests that amorphous character increases due to the formation of hybrid inorganic/organic polymer composites. Additionally the strong characteristic signal of crystalline NiO supports the doping of NiO on the surface of composite polymer particles as visibly observed from SEM image. It is to be added that signals appeared at  $65^\circ$  and  $71.38^\circ$  (represented by a cross sign) in functional NiO and NiO/P(MMA-GMA) composite particles respectively are artefacts due to problems with the detector.



**Figure 5.** X ray diffraction patterns (XRD) of functionalized NiO particles a) and NiO/P(MMA-GMA) composite polymer particles

TGA was used to confirm the incorporation of organic polymer in hybrid composite particles. It is expected that as the composite particles are heated from ambient temperature to  $800^\circ\text{C}$  the organic part would be burned off and the remaining part after calcination would represent the percentage of inorganic mass i.e. NiO content. TGA curves are illustrated in **figure 6**. Compared to bare NiO particles (reference material) a sharp but small transition is observed at around  $470^\circ\text{C}$  in functional NiO and the weight loss is increased by around 3.29%. This weight loss represents the inclusion of oleic acid and organic fraction of MPS in NiO. It is also evident that relative to functionalized NiO seeded copolymerization of MMA and GMA produced a big sharp transition in NiO/P(MMA-GMA) composite polymer particles at around  $287^\circ\text{C}$ . Considering the complete decomposition of organic part

in the measured temperature range, the percentage of residual inorganic NiO content in NiO/P(MMA-GMA) composite polymer particles is calculated to be roughly 34%. This numerical percentage is pretty high and some free NiO not removed during washing may also contribute to this higher value.



**Figure 6.** TGA thermograms of unmodified NiO particles (reference material) a), functionalized NiO particles b), and NiO/P(MMA-GMA) composite polymer particles

## 4. Conclusions

A simple three step process was attempted to prepare epoxy functional NiO hybrid composite polymer particles. The particles were named as NiO/P(MMA-GMA) composite particles. Nanosized NiO particles were first prepared by liquid-phase process. NiO particles were then functionalized with silane coupling agent and finally seeded copolymerization of MMA and GMA was carried out in ethanol media. NiO doped epoxy functional composite particles were obtained as confirmed by electron micrographs, FTIR, NMR, XRD and TGA analyses.

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