

# Synthesis, characterization, viscosimetric study and formulation of a new nanofunctional polymer's nanocomposite: Nanoglycidyl trihydrazine 4,4,4-tripropoxy of ethylene tribisphenol A

Rachid Hsissou, Atiqa Bekhta, Bouchra El Hilal, Jalila El Azaoui, Omar Dagdag, Kamal Abouelouafa, Ahmed El Harfi

**Abstract**— The objective this work is to synthesize a new nanofunctional epoxy nanoglycidyl trihydrazine 4,4,4-tripropoxy ethylene tribisphenol A (NGTHPTBAE) by chemical modification in two steps. We synthesized at first the triglycidyl ether ethylene of Bisphenol A (TGEEBA) and then we synthesized the nanofunctional resin by adding the hydrazine as nucleophile on the TGEEBA. After synthesizing the nanofunctional epoxy resin (NGTHPTBAE), we first characterized the new nanofunctional epoxy resin by the Fourier Transform Infrared Spectroscopy (FTIR). Its structure is confirmed by proton Nuclear Magnetic Resonance (<sup>1</sup>H NMR) and carbon (<sup>13</sup>C NMR). Secondly, we have studied its viscosimetric behavior, which plays a crucial role in the flow phenomena and the implementation of the macromolecular matrix by using a capillary viscometer of the Ubbelohd type. We have finally crosslinked and formulated our new thermosetting composite material in the presence of natural phosphate at various percentages (0%, 5%, 10% and 15%) as fillers and by adding methylene dianiline (MDA) as a hardener. The dispersion of the charges in the macromolecular matrix was monitored by using the Scanning Electron Microscope (SEM).

**Index Terms**—NGTHPTBAE, polyepoxides, TGEEBA, chemical modification, hydrazine, viscosimetric behavior, crosslinking, formulation and thermosetting nanocomposite

## 1 INTRODUCTION

Epoxy resins are macromolecular thermosettings with technological matrices which are widely used as hyperlinks in formulating composite materials [1]. They have an important place in the industry because of their thermal [2], mechanical [3], electrical [4], dielectric [5], physical, chemical [6-7], viscosimetric [8-9] and rheological properties [10-11-12]. Their uses affect several domains such as the textile industry (reinforcement of clothes and sails of boat ...), aeronautics [13] and aerospace industry [14] and sports equipment (cycling, navigation ...).

This has led to the development of the so-called thermostable epoxy resins in which the functionality is higher and also at high bonds density containing, for example, aromatic nucleus [15]. There are several methods to prepare epoxy resins, the most widely used is the epichlorohydrin condensation on structures containing at least two mobile hydrogens such as diacids, diamines, and polyphenols; by glycidation, via the condensation of glycidol on halogenated compounds [16-17-

18]; and/or by the oxidation of unsaturated compounds in the presence of peracids [19].

The polyepoxide resins are crosslinked by the following hardeners : diols, diacids, anhydrides and diamines. The latter are good hardeners for crosslinking polyepoxide prepolymers, such as methylene dianiline MDA, which have a very high glass transition temperature [19-20-21]. They may also have excellent thermal stability and good resistance to standard or formulated chemicals [22-23].

In this work, we mainly studied the viscosimetric properties of new nanofunctional polymer NGTHPTBAE. These properties play a primordial role in the implementation phenomena and that of the macromolecular matrix flow [24-25]. Subsequently, the charge dispersion in the macromolecular matrix was monitored by using the scanning electron microscope. The synthesis of ethylene tribisphenol A nanoglycidyl trihydrazine 4,4,4-tripropoxy (NGTHPTBAE) was carried out by chemically modifying the bisphenol A triglycidyl ether ethylene resin (TGEEBA) [24], and then by adding the hydrazine as a nucleophile.

## 2 MATERIALS AND METHODS

### 2.1 Hardware

#### 2.1.1 The used products

In this work, we have used several products among which we listed the methylene dianiline (MDA) which is used as cross-

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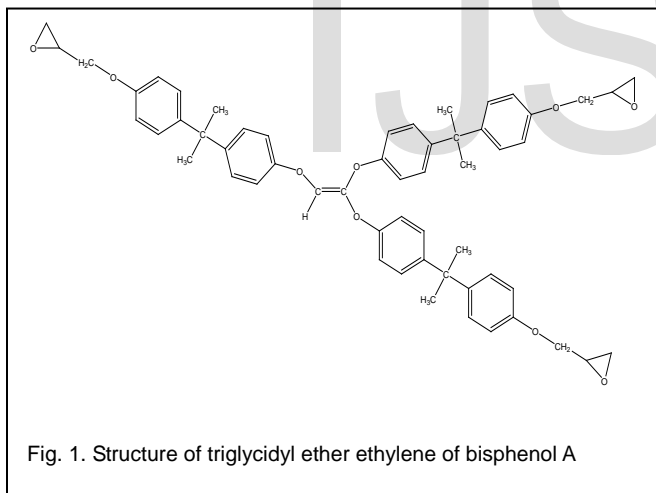
linker [19-22], and which is an aromatic primary diamine of formula  $C_{13}H_{14}N_2$ . Its functionality is equal to four. Thus, we have synthesized the following prepolymer in the laboratory namely the triglycidyl ether ethylene of bisphenol A, epichlorohydrin with a purity of 99%, methanol, toluene, triethylamine, hydrazine hydrate and natural phosphate used as filler. All of these commodities were supplied by Acros Chemical Co and Aldrich Chemical Co.

### 2.1.2 Synthesis of ethylene tribisphenol A nanoglycidyl trihydrazine 4,4,4-tripropoxy (NGTHTPTBAE)

In this study we followed two methods for the synthesis of ethane tribisphenol A nanofunctional epoxide nanoglycidyl trihydrazine 4,4,4-tripropoxy epoxy macromolecular binder.

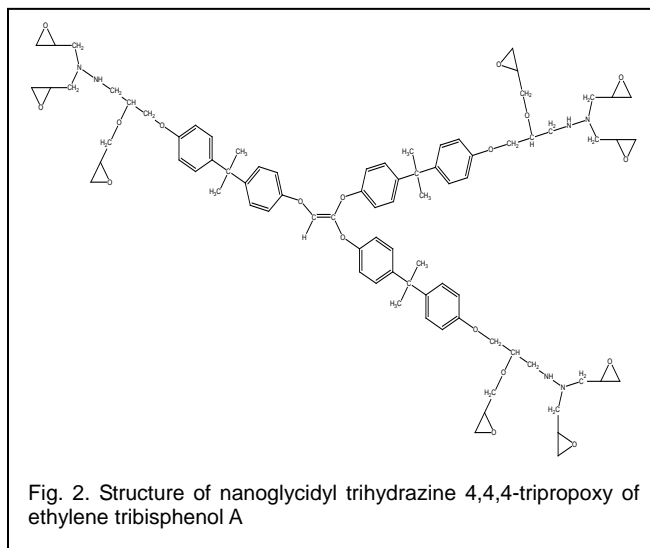
#### 2.1.2.1 The first method: the synthesis of triglycidyl ether ethylene of bisphenol A (TGEEBA)

In a flask equipped with a refrigerant, we placed 1 cm<sup>3</sup> of trichlorethylene and added 7.6 g of bisphenol A solution in methanol. The obtained mixture is then vigorously stirred with a magnetic stirrer at 100 °C for 4 hours. Then, 3 cm<sup>3</sup> of epichlorohydrin (in excess) was added and the mixture was brought to 70 °C for 4 hours with stirring. The mixture was then cooled to 40 °C and then 4.5 cm<sup>3</sup> of triethylamine was added and the mixture was stirred for 3 hours. The solvent was then removed by using the rotary evaporator. Finally, a viscous resin of brown color was obtained, figure 1.



#### 2.1.2.2 The second method: the synthesis of ethylene tribisphenol A nanoglycidyl trihydrazine 4,4,4-tripropoxy (NGTHTPTBAE)

In a flask equipped with a condenser, we condensed  $3.2 \cdot 10^{-3}$  mol of TGEEBA and  $7.4 \cdot 10^{-3}$  mol of hydrazine in a solution of 13 ml of methanol. The mixture was then heated to a temperature of 100 °C for 4 hours. Then, the mixture was cooled at 80 °C and 5.24 moles of epichlorohydrin were added to the reaction mixture (in excess) with magnetic stirring. Then, 6 cm<sup>3</sup> of triethylamine  $N(Et)_3$  was added and the mixture was brought to a temperature of 40 °C with magnetic stirring for 3 hours. The solvent was then removed by using the rotary evaporator. We finally obtained a viscous resin of black color as in figure 2.



## 2.2 Characterization methods used in the preparation of nanoglycidyl trihydrazine 4,4,4-tripropoxy of ethylene tribisphenol A

### 2.2.1 Fourier Transform Infrared Spectroscopy (FTIR)

The IR spectrometer used is BRUKER Fourier Transformed Spectrometer (FTIR). The light beam passes through the sample to a thickness of 2  $\mu$ m. The analysis is carried out between 4000  $cm^{-1}$  and 600  $cm^{-1}$ .

### 2.2.2 Nuclear Magnetic Resonance (NMR)

The nuclear magnetic resonance analyzes of the proton (<sup>1</sup>H NMR) and carbon (<sup>13</sup>C NMR) were obtained by using the BRUKER ADVANCED 300 MHz apparatus. The used solvent is DMSO and the chemical displacements are expressed in ppm.

### 2.2.3 Viscosimetric behavior

The viscosimetric analysis of the standard epoxy prepolymer was monitored by using the Ubbelohd VB-1423 capillary viscosimeter on liquid samples.

The measurement conditions used by the Ubbelohd VB-1423 capillary viscosimeter are as follows:

Viscosimeter of 1B size for a dilution series, capillary tube 0.46 mm in diameter and constant  $k = 0.051493$ ;

Measuring temperature in °C: 30 to 60;

Number of measurements: 3 each time;

Solvent: Toluene;

The Hagenbach correction was calculated according to the formula given in DIN 51562-1 January 1999 (Measurement of kinematic viscosity by means of the viscosimeter Ubbelohd); Solubilization of the epoxide prepolymers in toluene was carried out at 25 °C with magnetic stirring.

#### 2.2.3.1 The preparation of the samples analyzed by the Ubbelohd VB-1423 capillary viscometer

In order to determine the viscosimetric behavior of the synthesized nanofunctional epoxy prepolymer, namely nanoglycidyl trihydrazine 4,4,4-tripropoxy of ethylene tribisphenol A

(NGTHTPTBAE) according to concentration and temperature, we dissolved this prepolymer in toluene at different concentrations (5%, 10%, 15% and 20%). Then, we studied their viscosities by using the Ubbelohd VB-1423 capillary viscosimeter at temperatures ranging from 30 °C to 60 °C.

## 2.2.4 Scanning Electron Microscopy (SEM)

The scanning electron microscope was used to make photographic images and the observations were carried out on a JEOL-JSM-5500 microscope.

This technique is based on the use of electrons beam accelerated by a fixed potential which excites the sample's surface. The interactions of these primary electrons with the material lead to the emission of secondary electrons, backscattering electrons, X-rays and Auger electrons.

### 2.2.4.1 Preparations of samples analyzed by the scanning electron microscope

The crosslinking reaction is carried out by methylene dianiline (MDA) [22], known for its excellent mechanical properties and good thermal stability which it is provided to the final product, in view of other hardeners. Its chemical structure is given by the following figure.

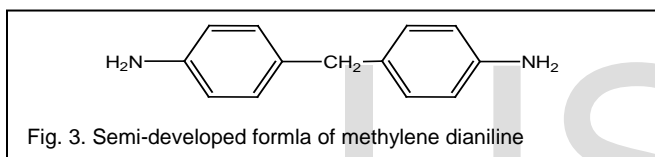


Fig. 3. Semi-developed formula of methylene dianiline

The protocol consists of preheating the prepolymer stoichiometric amounts and the hardener [19]. The methylene dianiline (MDA) is placed in an oven at 120 °C while the prepolymer is heated to 60 °C. Once it is melted, the MDA is mixed with the prepolymer to give a single fluid phase. The prepared samples were sealed in molds in the desired geometric form for 24 h at 70 °C. This hardener has a functionality of four, which means that during the crosslinking reaction with a nanofunctional resin (eg, NGTHTPTBAE), both amine functions theoretically react with eighteen epoxide functions when they are in stoichiometric proportions [24-25]. The final product is a chemically bridged three-dimensional network. After the chemical reaction, the crosslinked materials are hard, infusible and insoluble.

## 2.2.5. Ration's calculation

### 2.2.5.1. Stoichiometric coefficients' Calculation

In order to obtain the optimum properties, we cured the multifunctional polyepoxide macromolecular matrix in the presence of hardener (especially amines). It is desirable in this particular phase to make the prepolymer and the curing agent react in approximately stoichiometric amounts.

The calculation of the Epoxy Equivalent Weight (EEW) of the synthesized macromolecular matrix: nanoglycidyl trihydrazine 4,4,4-tripropoxy of ethylene tribisphenol A is as follow:

$$EEW = M_w(\text{NGTHTPTBAE})/f$$

With  $f$  as the functionality of the synthesized epoxy resin. From which:

$$EEW = 1486/9$$

$$EEW = 165 \text{ g/eq}$$

The calculation of the amine equivalent (Amine Hydrogen Equivalent Weight: AHEW).

AHEW then constitutes the mass of the hardener containing an amine equivalent.

$$AHEW = M_w(\text{MDA})/f$$

Methylene dianiline:

$$M_w = 198; f = 4$$

$$AHEW = 198/4$$

$$AHEW = 49,5 \text{ g/eq}$$

### 2.2.5.2. Calculation of the ration by weight

The ration by weight of the hardener relative to the resin is calculated in the majority of cases, per 100 parts of the resins or PHR (Parts per Hundered of Resin):

$$\text{Amin PHR} = (\text{Amin H eq/epoxy equivalent}) \times 100$$

In this case the NGTHTPTBAE:

$$\text{Amin PHR} = (49,5/165) \times 100$$

$$\text{Amin PHR} = 30 \text{ g/eq}$$

Therefore, 30 g of methylene dianiline per 100 g of nanoglycidyl trihydrazine 4,4,4-tripropoxy of ethylene tribisphenol A will have to be reacted to achieve optimum properties when the prepolymer has been cured with methylene dianiline.

### 2.2.5.3. Calculation of the ration to the quantity of the load:

We calculated the quantity of the desired charge according to the following equation:

$$y\% = x / (\text{resin} + \text{MDA} + x)$$

## 3. RESULTS AND DISCUSSIONS

### 3.1. Spectral characterization of the synthesized epoxy resin

We have carried out the structural analysis of the epoxy resin synthesized by Fourier Transform Infrared Spectroscopy (FTIR), whose structure is confirmed by the proton Nuclear Magnetic Resonance ( $^1\text{H}$  NMR) and carbon ( $^{13}\text{C}$ -NMR). The letters s, d, t, q, and m are respectively singlet, doublet, triplet, quadruplet and multiplet. The results of the structural analyzes, given below, confirm the structure of the synthesized macromolecular matrix.

### 3.1.1. Fourier Transform Infrared Spectroscopy (FTIR)

The synthesized polyepoxide matrix was characterized by means of a quadrant transform infrared analysis. The latter was exposed in its viscous state to infrared rays under ATR mode. The obtained results are illustrated in figure 4. The allocation of the different bands which are obtained by means of the analysis are given below:

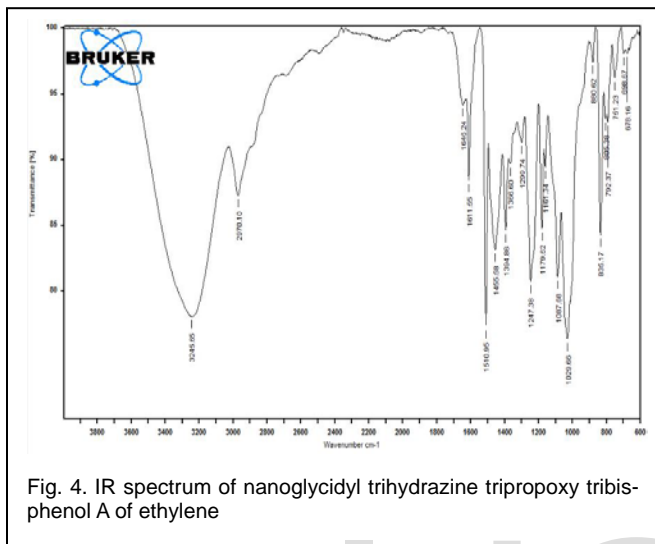


Fig. 4. IR spectrum of nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene

The band at 3245  $\text{cm}^{-1}$  corresponds to the elongation of the bond linked to the polymer; at 2970  $\text{cm}^{-1}$  it represents the vibration of the C-H of  $\text{CH}_3$  of the carbon  $\text{SP}^3$ ; at 1611  $\text{cm}^{-1}$  it corresponds to the vibration of the bond C=C of carbon  $\text{SP}^2$ ; at 1510  $\text{cm}^{-1}$  we found the vibration of the aromatic C=C bond of the para-substituted benzene cycle; at 1455  $\text{cm}^{-1}$  it corresponds to the deformation of the  $\text{CH}_2$  group in the plane; at 1394  $\text{cm}^{-1}$  it corresponds to the deformation of the CH bond in the plane. The band between 1179 and 1247  $\text{cm}^{-1}$  confirms the existence of elongation of the CN bond of the aliphatic amines while between 1029 and 1087  $\text{cm}^{-1}$  it corresponds to the vibration of the CO bond; at 805  $\text{cm}^{-1}$  it represents an epoxy group; finally between 792 and 835  $\text{cm}^{-1}$  it corresponds to the deformation outside the plane of the  $\text{RRC}=\text{CHR}$  bond.

### 3.1.2. Nuclear Magnetic Resonance NMR

#### 3.1.2.1. Proton Nuclear Magnetic Resonance

The spectrum of the proton's nuclear magnetic resonance of the synthesized nanofunctional macromolecular matrix is presented in figure 5, which confirms the structure of nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene. The distribution of the chemical displacements of the nanoglycidyl trihydrazine tripropoxy tribisphenol A resin of the observed ethylene is given below.

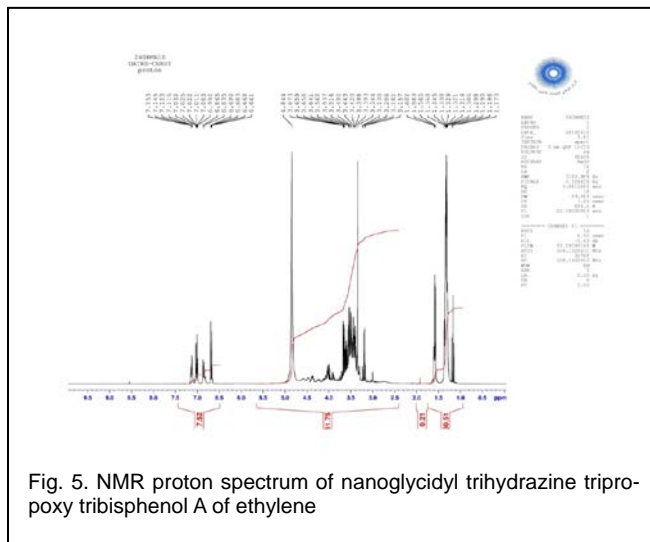


Fig. 5. NMR proton spectrum of nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene

1.67 ppm (s, 18H,  $\text{CH}_3$ ); 2 ppm (s, 3H, N-H); 2.38-2.63 ppm (d, 18H,  $\text{CH}_2$  oxirane); 2.77-2.86 ppm (m, 9H, CH-oxirane); 2.56-2.81 ppm (d, 12, oxirane-linked  $\text{CH}_2$  and nitrogen); 2.65-2.90 ppm (d; 6H, -CH-alpha and -O-C beta); 3.38-3.63 ppm (d; 6H;  $\text{CH}_2$  bonded to oxirane and oxygen); 3.58 ppm (s; 3H; CH alpha -O-C, beta -O-benzene and beta-N); 3.95-4.20 ppm (d; 6;  $\text{CH}_2$  alpha -O-benzene and beta-O-C); 5.28 ppm (s, 1H, CH ethylene) and 6.65-6.96-6.69-7.02 ppm (d, 24H; aromatic CH).

#### 3.1.2.2. Nuclear Magnetic Resonance of Carbon

The carbon's nuclear magnetic resonance ( $^{13}\text{C}$  NMR) spectrum of the epoxy resin nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene is shown in figure 6. These results confirm that of the proton's nuclear magnetic resonance spectrum. The chemical shifts of the different carbons of nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene are given below.

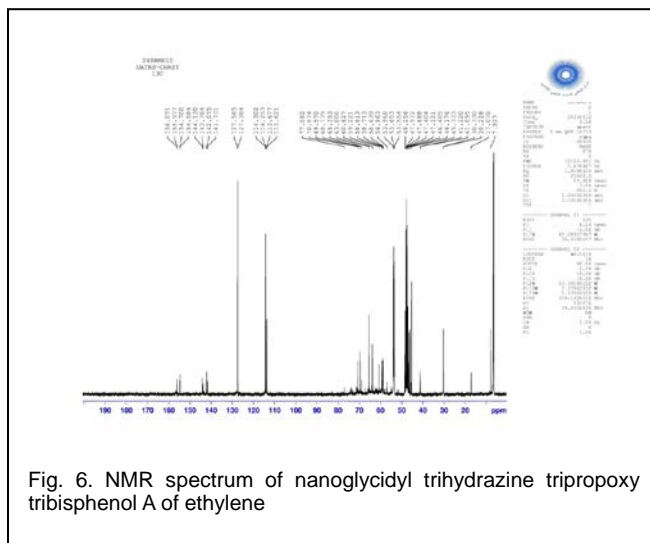


Fig. 6. NMR spectrum of nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene

31 ppm (S;  $\text{CH}_3$ ); 42 ppm (S; Benzene Alpha Quaternary Carbon and  $\text{CH}_3$  alpha); 44-45 ppm (S;  $\text{CH}_2$  of oxirane); 46-50

ppm (S; CH of oxirane); 50 ppm aliphatic (alpha -N, beta-O-C and gamma -O-benzene); 59 ppm (S; CH<sub>2</sub> bonded to oxirane and tertiary amine); 68 ppm (aliphatic S; CH<sub>2</sub> (alpha -O-benzene, beta -O-C and gamma -N)); 71 ppm (S; CH<sub>2</sub> bonded to oxirane and oxygen); 78 ppm (aliphatic S; CH (alpha-O-C, beta-O-benzene and beta-NH)); 92 ppm (S; CH ethylene); 114-117-128-129 ppm (S, aromatic CH); 137-140 ppm (aromatic S, C-C); 146 ppm (S, C ethylene) and 152-155 ppm (S; aromatic tertiary carbon bonded to oxygen).

### 3.2. Viscosimetric behavior

The different curves of the viscosity in view of the concentration and the temperature of the epoxide prepolymer synthesized nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene dissolved in toluene are respectively shown in figures 7 and 8.

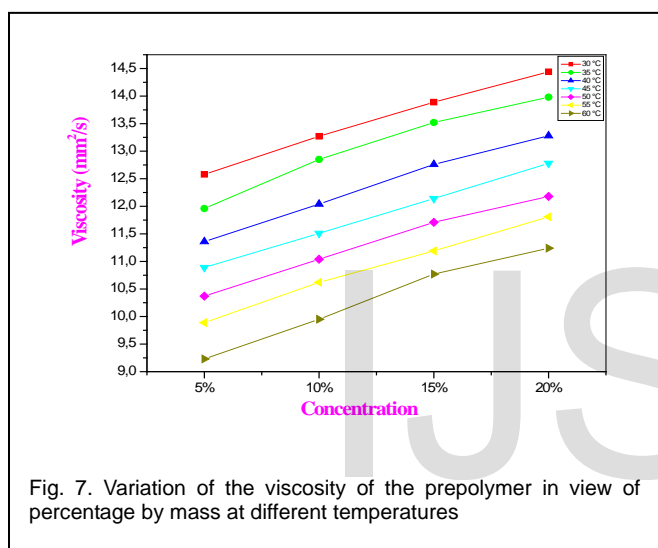


Fig. 7. Variation of the viscosity of the prepolymer in view of percentage by mass at different temperatures

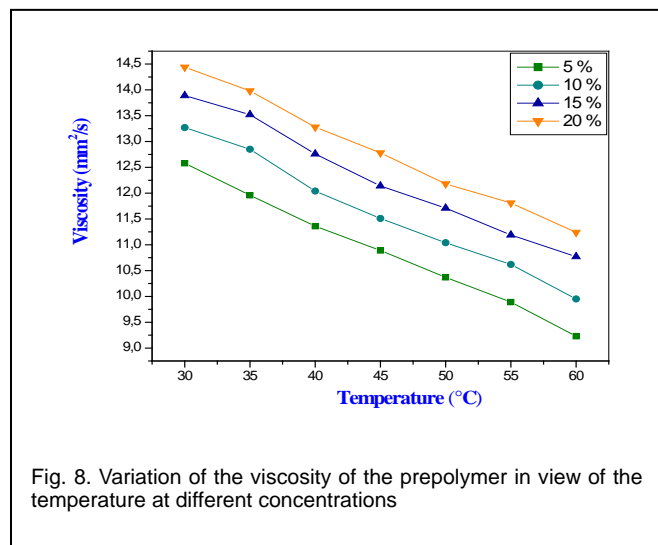


Fig. 8. Variation of the viscosity of the prepolymer in view of the temperature at different concentrations

From figure 7, it has been found that the values of the viscosity in view of the percentage by weight increase as the concentration of the prepolymer mass increases. This shows the progress of the homopolymerization reaction since the viscosity increases with the increase in the molecular mass of the so-

lute. This is related to the chemical transformations undergone by the prepolymer [9-26].

Indeed, the variation of the viscosity in view of temperature decreases with the increase of the temperature. This implies that the nanoglycidyl trihydrazine tripropoxy tribisphenol A prepolymer of ethylene passes from a viscous to a liquid state because the provided heat accelerates the process of depolymerization of the resin, which explains the drop in the observed viscosity (figure 8) [27].

### 3.3. Experimental determination of the molecular weight of NGTHTPTBAE by viscometer

The capillary viscosimeter mainly consists of three tubular parts (1, 2 and 3) of the capillary tube (6) with the measuring sphere (5). Above and below the measuring sphere (5), the annular marks M<sub>1</sub> and M<sub>2</sub> are printed on the tube (3) and they define the volume of the pure solvent and the solution (solvent + resin) for the experiments in figure 9.

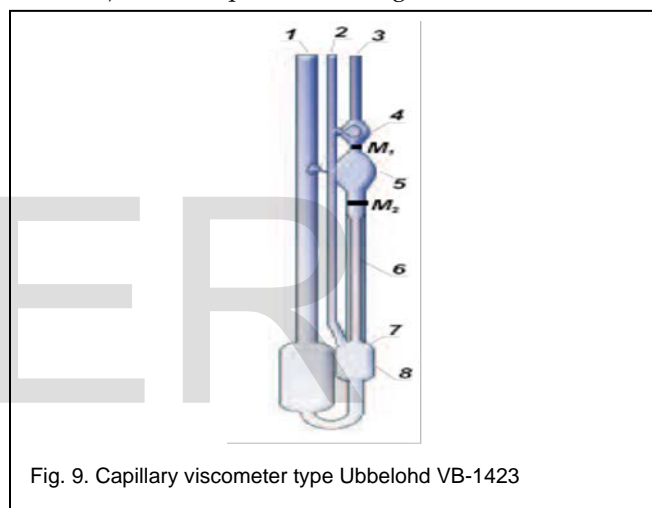


Fig. 9. Capillary viscosimeter type Ubbelohd VB-1423

We have defined the following viscosities:

$$\text{Relative viscosity } \mu_{rel} = \mu / \mu_0$$

$$\text{Specific viscosity } \mu_{sp} = (\mu - \mu_0) / \mu_0$$

$$\text{Reduced viscosity } \mu_{red} = (\mu - \mu_0) / \mu_0 C = \mu_{sp} / C$$

$$\text{Intrinsic viscosity } [\mu] = \lim (\mu - \mu_0) / \mu_0 C = \lim \mu_{sp} / C \text{ (when } c \text{ to } 0)$$

With  $\mu_0$  and  $\mu$  are respectively the viscosity of the pure solvent and the viscosity of the solution (solvent + resin).

#### 3.3.1. Relationship between viscosity and flow time

The flow of a fluid in a capillary tube of radius  $r$  and of length  $l_c$  is governed by the law of Poiseuille (mechanical fluid CF)  $dv/dt = \pi r^4 / 8 \mu l_c$

$dv/dt$ : Volume unit flowing at time  $dt$ ;

$P$ : Hydrostatic pressure;

$l_c$ : Capillary length;

$\mu$ : Viscosity of the solution;

$R$ : Radius of the capillary tube.

We have considered that a volume  $V$  of liquid flows at an instant  $t$ , while the equation of poiseuille becomes:

$$\frac{v}{t} = \frac{\pi \cdot \bar{p} \cdot r^4}{8 \mu l_c}$$

$\bar{p}$  : Average hydrostatic pressure  $\bar{p} = p \cdot g \cdot \bar{h}$

P: Density of the solution that flows into the capillary

g: Acceleration of gravity

$\bar{h}$  : Average value of the height of the liquid in the capillary

Thereby:

$$\frac{v}{t} = \frac{\pi \cdot p \cdot g \cdot \bar{h} \cdot r^4}{8 \mu l_c} \text{ is therefore } \mu = \frac{t \cdot \pi \cdot p \cdot g \cdot \bar{h} \cdot r^4}{8 v l_c}$$

Finally, the viscosity takes the form:  $\mu = K \cdot p \cdot t$

K: constant dependent on viscometer

P: density of the solution that flows into the capillary

t: flow time

Finally, we redefined the viscosities in view of the flow times.

Relative viscosity  $\mu_{rel} = t/t_0$

Specific viscosity  $\mu_{sp} = (t-t_0)/t_0$

Reduced viscosity  $\mu_{red} = (t-t_0)/t_0 C = \mu_{sp}/C$

Intrinsic viscosity  $[\mu] = \lim (t-t_0)/t_0 C = \lim \mu_{sp}/C (c \text{ to } 0)$

With:

t: flow time of the volume V of the solution (resin + solvent)

t0: flow time of the same volume V of the pure solvent.

**HUGGING equation:**

$$\mu_{sp}/C = [\mu] + K [\mu^2] C + K [\mu^3] C^2 + \dots$$

Since we have worked at high dilution, the former equation becomes

$$\mu_{sp}/C = [\mu] + K [\mu^2] C$$

$$[\mu] = \lim (t-t_0)/t_0 C = \lim \mu_{sp}/C$$

K: constant of HUGGING

K  $[\mu^2]$ : coefficient of VIRIEL slope of the curve

We plotted the specific viscosity curve in view of the concentration  $(t-t_0)/t_0 C = \mu_{sp}/C = f(C)$ :

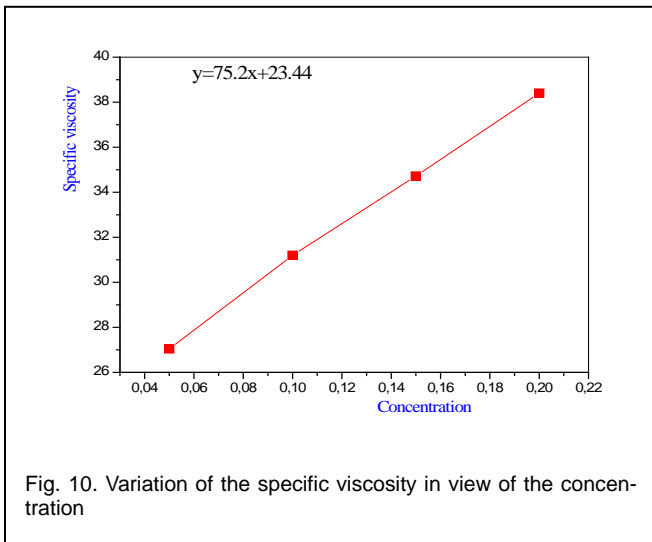


Fig. 10. Variation of the specific viscosity in view of the concentration

From the figure above, we obtained a line whose intercept is the intrinsic viscosity  $[\mu]$ .

We have determined the molecular weight of the epoxide prepolymer nanoglycidyl trihydrazine tripropoxy tribisphenol A of ethylene synthesized from the following equation:

$$[\mu] = KM^a$$

$[\mu]$ : intrinsic viscosity of polymer measured experimentally

Mv: viscosimetric average molecular mass

From the linear equation, we have a = 75.2 and b =  $[\mu] = 23.44$

With: k = 0.051493 and alpha = 0.72 (in toluene)

We thus obtain: Mv = 4953 g/mol

### 3.3.2. Determination of polymerization's degree

$$Dp_n = \frac{M_v}{Mass(NGTHPTBAE)} = \frac{4953}{1486} = 3.34$$

### 3.4. Scanning Electron Microscopy (SEM)

In order to identify the morphology of the good dispersion of the immersed charge in the thermosetting technological nanocomposite macromolecular matrix (NGTHPTBAE/MDA/PN) and to confirm the result, we used the surface analysis by using the scanning electron microscope. The obtained morphological results are presented in the photos of the SEM figure11.

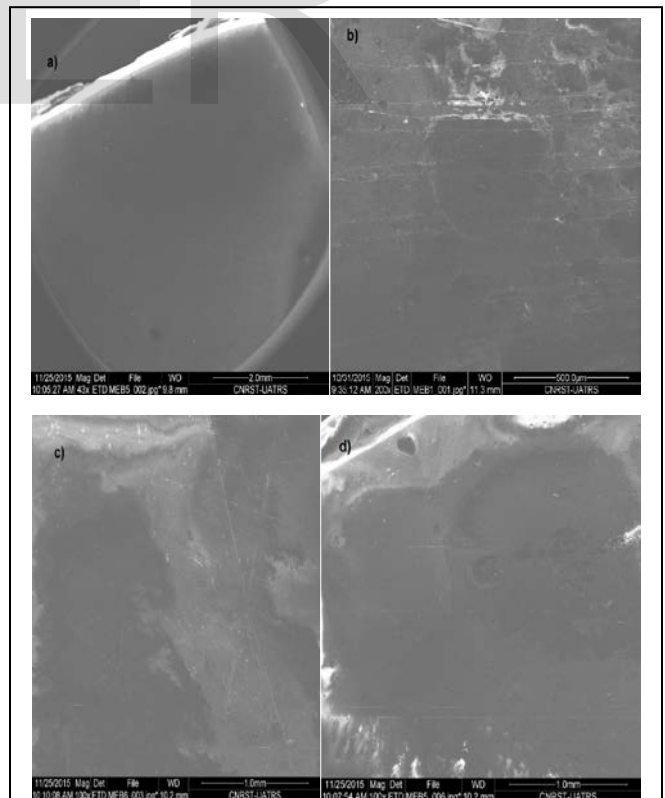


Fig. 11. The morphology of the synthesized epoxide prepolymer crosslinked with methylene dianiline and formulated at 0% (a), 5% (b), 10% (c) and 15% (d) of the natural phosphate filler.

The morphology of the various crosslinked and formulated nanocomposite samples (NGTHPTBAE/MDA/PN) were observed by the field emission scanning electron microscope (SEM-EC). The comparison of the different morphologies based on four different formulations showed that the type of formulation is a key factor in controlling the morphology of the nanocomposite (NGTHPTBAE/MDA/PN). The micrographs (SEM-EC) of various samples of the nanocomposite prepared in the absence and in the presence of the natural phosphate charge are shown in figure 11. The morphology of the thermosetting nanocomposite varied greatly with the different percentages of the natural phosphate charge. According to these observations by using the scanning electron microscope of the morphology of the thermosetting nanocomposite (NGTHPTBAE/MDA/PN), we have clearly shown the good dispersion of the charge of the natural phosphate in the macromolecular epoxy matrix, which is newly synthesized by the chemical modification and this is shown through the photos a, b, c, d of figure 11.

#### 4 CONCLUSION

We synthesized, in this work, a nanofunctional polyepoxide resin by chemical modification in the presence of hydrazine as nucleophile. The structure of this synthesized multifunctional macromolecular matrix was characterized by Fourier Transform Infrared Spectroscopy (FTIR), its structure confirmed by proton nuclear magnetic resonance ( $^1\text{H}$  NMR) and carbon 13 ( $^{13}\text{C}$  NMR). After the synthesis of the nanofunctional polyepoxide resin, we studied on the one hand the viscosimetric behavior of the macromolecular matrix by using a capillary viscometer of the Ubbelohd type, and we determined the molecular weight  $M_v$  and the degree of polymerization  $DP_n$  of this macromolecular matrix; we have crosslinked and formulated, on the other hand, the thermosetting technological nanocomposite material, in the presence of methylene dianiline as hardener and the charge of natural phosphate (PN) at different percentages (0%, 5%, 10% and 15%). We finally monitored the dispersion of the natural phosphate charge in the composite material (NGTHPTBAE/MDA/PN) by using a scanning electron microscope.

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