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Investigation of the structure, magnetic, rheological and mechanical properties of EPDM rubber/Cu-Al-Zn alloy composites

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Abstract

The current research presents the development of EPDM rubber including shape memory alloy. The Cu-Al-Zn alloy was modified with the coupling agent **3**-(Trimethoxysilyl) propyl methacrylate (TMSPM**)** and then incorporated into Ethylene propylene diene rubber (EPDM) by a 2-roll mill technique. The Cu-alloy behaved as superelasticity capability which can be used in various engineering approaches to smart materials. Microstructure, mechanical, rheological and magnetic properties of Cu-Al-Zn/EPDM composites was studied. The rheometeric results indicated that the incorporation of Cu-Al-Zn as a filler enhance the curing process. However the curing time of Cu-Al-Zn/EPDM composites was reduced compared to the control sample without Cu-alloy. For the mechanical properties, increasing Cu-alloy loading in the EPDM composites resulted in an increment of elongation at break. The addition of Cu based alloy as fillers improved the magnetization of composites with coupling agent when compared to EPDM rubber and Cu-Al-Zn/EPDM composites without coupling agent.

***Keywords*:** EPDM rubber composite; Cu-Al-Zn alloy; tensile strength; elongation at break; magnetic moment and SEM.

Introduction

Rubber is one of the commercially used polymeric matrixes mainly due to its good energy absorbing properties. In addition, it can undergo much more elastic deformations under stress than other materials and still return to its original shape without permanent deformation after the releasing the stress. This unique property gives rubber an extensive variety of applications. However rubber itself is a relatively weak material and is therefore reinforced before use, the effective reinforcements include metals, inorganic materials, plastics and fibers [1].

Rubber materials filled with metal powders are widely used in a variety of industries because of their specific and often unique properties. The choice of metal filler is governed by the functional purpose of the composite material and by economic considerations. The reason for the extensive use of metal powders as fillers for polymers is most commonly the potential they provide to adjust the density of the materials, their appearance and their magnetic and other physical properties [2].

On the other hand, it is well known that many copper alloys such as Cu-Al, Cu-Zn-Al, Cu-Al-Ni and Cu-Al-Mn exhibit shape memory properties. Cu-based SMAs (shape memory alloy) have attracted significant attention and interest in recent years due to their good shape memory capacity, narrow temperature region of transformation, simple fabrication and low production cost. Copper-based shape-memory alloys are very sensitive to thermal effects, and it is possible that in thermal cycles its properties change (e.g., shape-recovery ratio, transformation temperatures, crystal structures, hysteresis and mechanical behavior) [3].

The martensite in shape memory alloys is soft in contrast to martensite of steels deformation of these alloys is not slip, twinning or grain boundary sliding but by growth or shrinkage of self-accommodating, multi-oriented martensitic plates/variant [4]. However of the exceptional and controllable properties such materials like: piezoelectrics, electro and

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magnetorheological fluids, and most of all shapes memory alloys (SMAs) are of the highest interest. Cu-based shape memory materials mostly used in different applications like catalysis, electronics and production of metal matrix composites [5].

It is known that rubbers are inherently non-magnetic. The impregnation of magnetic filler in rubber matrix imparts magnetic natures, and considerably modifies the physical properties of the matrix as well. The advantages of rubber bonded magnets (RBMs) over their metallic and ceramic counterparts include low weight, resistance to abrasion, ease of machining and forming process and capability for high production rates [6].

In last years, magnetic polymer composite was first fabricated by Baermann through mixing alnicomagnets with phenolic resins [7]. Since then, several types of composites have been successfully fabricated and implemented in order to combine the outstanding mechanical properties of polymers with the unique properties of the magnetic materials

However EPDM is a rubber of saturated material with unsaturated side chains which is widely used for electrical insulation and automotive equipment [8]. Rubber composites filled with metal alloy powders are widely used in a variety of industries as aerospace, building materials, tubes seals, wire, cable terminations and coverings because of their specific and often unique properties.

The present work reveals evaluation of the influence of magnetic filler on the physicomechanical properties of EPDM composites, aiming to improve the mechanical and magnetic properties.

Experimental part

Materials

Ethylene propylene diene rubber (EPDM). Vistalon 650S, produced by ESSO Chemie Germany, Diene (ethylidenenorbornene) content 9%, ethylene content 55%; Mooney viscosity ML (1± 8) at 127°C 48–52; density 0.86.

N-Cyclohexyl-2-benzothiazole sulphenamide (CBS) is used as accelerator of trade name: Rhenogran® CBS-80 and Vulkacit® CZ. It was supplied from Rheinehemie Germany.

Zinc oxide and stearic acid are used as activators with specific gravity 5.55–5.61 and 0.9–0.97, respectively at 15°C. They were supplied by Aldrich Company, Germany.

Elemental sulfur is used as a vulcanizing agent. It is a fine powder with pale yellow color; its density is 2.0-2.06 g cm^{-3} at room temperature (25°C±1) and it was supplied by Aldrich Company, Germany.

Polymerized 2, 2, 4-trimethyl-1, 2-dihydroquinoline (TMQ) as antioxidant is commercial grade product.

Cu-based alloy (Cu-Al-Zn alloy) DEVARD`s Alloy Extra pure, LOBA CHEMIE PVT.LTD.

Preparation

- **Preparation of coupling agent**

Solution polymerization of 3-(Trimethoxysilyl) propyl methacrylate (TMSPM):

Solution polymerization of 3-(Trimethoxysilyl)propyl methacrylate (TMSPM) using acetone solvent to obtain poly 3-(Trimethoxysilyl)propyl methacrylate with solid content 20% was performed s follow: 20 ml 3-(Trimethoxysilyl)propyl methacrylate monomer and 75 ml acetone solvent were introduced in 250 ml Two necked flask.

The solution was bubbled with nitrogen for 30 min before heating at 80 ºC. Finally, 0.041 gm of AIBN (2, 2′-Azobis(2-methylpropionitrile) in 5 ml acetone was added to the reaction to form milky white liquid state. poly 3-(Trimethoxysilyl) propyl methacrylate was formed with yield 20% after 4 hours polymerization reaction.

Preparation of ethylene propylene diene monomer composites

Different concentrations of Cu-Al-Zn alloy at 2.5, 5, 10, 15, 20 phr were incorporated into EPDM then all ingredients applied according to ASTM D3182-07(2012) standard, in a two-roll mill as the formulations shown in table 1. The rolling mill had the rolls of 470 mm in diameter, 300 mm in width, speed of slow rolls, 24 rpm and gear ratio (1:1.4). The compounded rubbers were left overnight before vulcanization, and then the vulcanization of the composites was carried out in a compression type hydraulic press under 40 kg/cm^2 and a temperature of 152 ± 1°C. The rheological characteristics of the mixes containing Cu-Al-Zn alloy were determined using an oscillating disc rheometer R-100 (MDR one Moving Die Rheometer, TA) at 152±1°C according to ASTMD2084-11, 2011.

Methods of Testing

The microstructural evolution and elemental composition analysis were carried out by using. **Scanning Electron Microscopy** (SEM) (Quanta FEG 250; FEI Co., Hillsboro, OR, USA), with attached energy dispersive X-ray spectroscopy (EDS) attachment. The surface morphology of EPDM composites and the surface of the specimens were coated with gold paste as a contact material for the SEM measurements.

X-ray diffraction measurements of Cu-Al-Zn alloy and composites were performed on the wide-angle goniometer of Empyrean diffractometer operated at 45 kV and 30 mA. The x-ray wavelength is Ni filtered Cu Kα (1.54056 Å). The obtained diffractogram at ambient condition in the 2θ range (4-80) with step size 0.02 2theta and step time 20S/step. Instrumental broadening was corrected using quartz standard sample.

Table 1. Formulations and rheological characteristics of the EPDM/Cu-Al-Zn composites

Formula o./Ingredient phr	E_0	E_1	E_2	E_3	E_4	E_5
Cu-Al-Zn	--	2.5	5	10	15	20
Rheometric characteristic at 152 ±1°C						
M_H, dNm	16.044	16.7	14.58	14.16	13.46	13.04
M_L, dNm	2.07	2.71	2.59	2.51	2.48	2.4
ΔM, dNm	13.97	13.99	11.99	11.65	10.98	10.64
CRI, min $^{-1}$	4.931	6.24	9.24	7.73	7.15	7.14

Notes: Base recipe (in phr): Ethylene propylene diene monomer (EPDM) 100; stearic acid 2; zinc oxide 5; CBS (N-cyclohexyl-2-benzothiazole sulfenamide) 1; Paraffin oil 2; S sulfur 3. ΔM is the difference between maximum and minimum torque; TMQ – polymerized 2, 2, 4-trimethyl-1, 2-dihydroquinoline; CRI – cure rate index,where phr ispart per hundred parts of rubber

Fourier transform infrared spectroscopy was performed using a JASCO FTIR-6100E FTIR spectrometer (Japan) operated in the absorption mode, in the wavenumber range 4,000 to 400 cm^{-1} after mixing with potassium bromide and pressed in the form of discs. The spectra were collected at a resolution of 4 cm^{-1}

The **magnetic** properties of the Cu-Al-Zn alloy and composites were evaluated at room temperature by using a Vaibrating Sample Magnetameter (VSM) lake shore [model 7410] USA.

The **rheological** characteristics of the EPDM composites containing different content Cu-alloy in the presence and absence of coupling agents was performed using an oscillating disc rheometer R-100 MDR one (Moving Die Rheometer), TA Instruments, New Castle, DE, USA) at 152 6 1°C in accordance with ASTM D2084-11(2011).

For the **mechanical** properties the tensile strength, stress at 100%strain, and elongation at break of the obtained vulcanizates were tested according to ASTM D412-06a (2013) with a Zwick testing machine. Five measurements were performed for each sample and the average was taken.

Results and Discussion

Characterization of Cu-based alloy

Fig.1 shows typical SEM microstructure of as-received Cu-Al-Zn alloy. The received. Cu-Al-Zn alloy had a grain size of order 25 µm and containing mainly dendritic structure**.** Energy-dispersive X-ray spectroscopy analyses (EDAX) were performed in order to obtain the chemical composition of the investigated samples are listed in Fig.2.

X-ray diffraction (XRD) high score plus software used to extract the peaks positions and relative intensity to identify the collected pattern, which assured that the sample, could be indexed as malti-phase within a wide 2θ range. A High score plus software used to extract the peaks positions and relative intensity to identify the XRD collected pattern of the metal alloy shown in Fig.3a, which assured that the sample, could be indexed as bi-phases within a wide 2θ range. The major phase reflection at 2θ= 20.76°, 29.44°, 36.95°, 37.94°, 42.11°, 42.63° , 47.34°, 47.84°, 57.17°, 61.47°, 67.09° , 69.22°, 77.54°, congruous to the reflections 110, 020, 002, 121, 220, 112, 130, 022, 222, 332, 123, 240, 332 of the tetragonal aluminum copper theta Al_2Cu phase, with space group I4/mcm specific with the ICDD no. 98-015-1372. Peaks at 2θ= 44.02°,44.19°, , 64.3° congruous to the reflections 2 -1 0, 1 0 10, 0 2 10, of the minor hexagonal phase aluminum copper zinc $Al_{4.2}Cu_{3.2}Zn_{0.7}$, space group R-3 specific with the ICDD no 98-005-7730 [9, 10]. The resulting structural and microstructural parameters presented in Table 3. Fig.3b shows the resulting profile fitting for the metal alloy. From table 3 it's clearly shown that the low value of x^2 = 1.61 obtained from refinement confirm the good fitting quality. The crystallite size value of Al_2Cu tabulated in table was in the nanometer range. It is fully known that the crystallographic glitch like dislocation has an outstanding impact on the physical properties of materials. The dislocation density calculated using refined values of crystallite size is based on the following equation [11]

$$\text{The dislocation density } (\delta) = (1/D^2) \qquad (1)$$

Where D is the crystallite size nm

The calculated values of the dislocation density seen in table3 declare that the maximum dislocation density corresponding to the major phase of Al_2Cu phase.

Magnetic measurement of alloy

Magnetic Cu-based alloy can play a very important role as active materials in many applications [12]. Fig.4 showed the hysteresis loop that confirmed the ferro-magnetization materials. This is maybe the increased number of the elementary cells having tetragonal geometry that exposed to the magnetic field.

Table 2. Formulations and rheological characteristics of the EPDM/Cu-Al-Zn in presence of coupling agent

Formula no./Ingredient phr	E_0	E_6	E_7	E_8	E_9	E_{10}
Coupling agent	--	4	4	4	4	4
Cu-Al-Zn	--	2.5	5	10	15	20
Rheometric characteristic at 152 ±1°C						
M_H, dNm	16.044	16.29	16.48	16.52	16.54	15.67
M_L, dNm	2.07	2.39	2.62	2.65	2.75	2.48
ΔM, dNm	13.97	13.9	13.86	13.87	13.79	13.19
CRI, min -1	4.931	6.33	6.601	7.21	7.17	7.05

Table 3. Lattice parameters, unit cell volume, microstrain, size, dislocation density

Structure parameter	Cu Al_2-θ	$Al_{4.2}Cu_{3.2}Zn_{0.7}$
Space group	I 4/m c m	R-3
a Å	6.0600 (5)	4.1105(6)
c Å	4.8714 (6)	24.7606(4)
α=β	90	90
γ	90	120
Volume $Å^3$	178.895(3)	362.30(7)
Microstrain	0.4803(5)	0.4745(1)
Crystallite size nm	74(2)	157(4)
Dislocation density	1.826 $X10^{-4}$	4.057 x 10^{-5}
R_{exp}	25.78	
R_{wp}	32.7	
Rp	50.7	
χ2	1.61	
Weight %	79.7	17.5

R_{wp} is weighted pattern, R_{exp} is the expected pattern and χ2 is the goodness of fit index $\chi2=R_{wp}/R_{exp}$

FIG.1. SEM micrograph of Cu-Al-Zn alloy

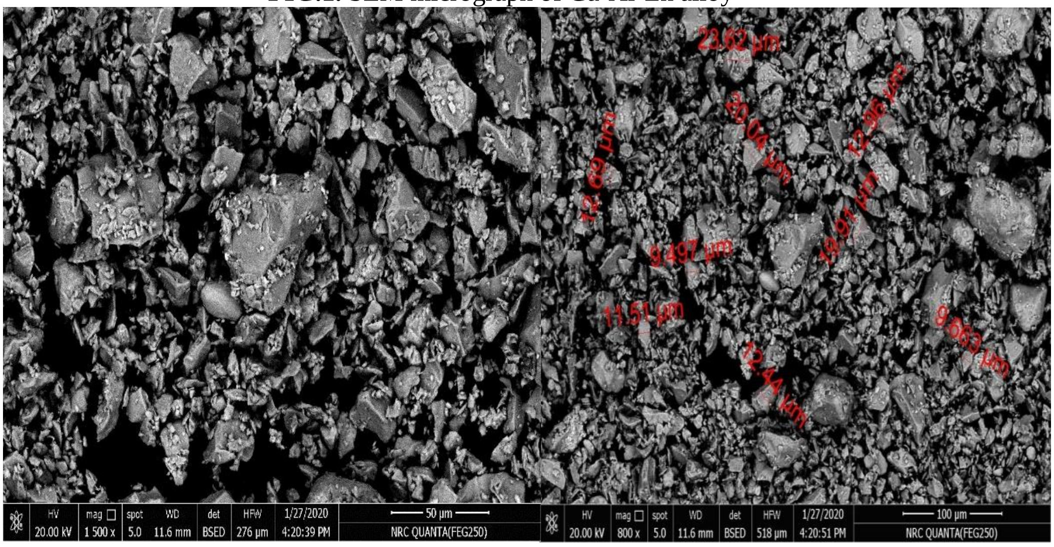
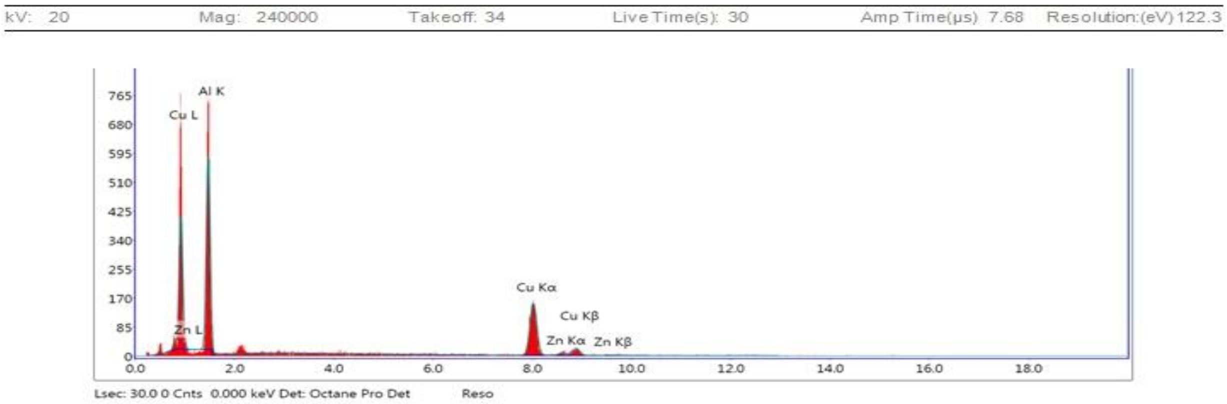
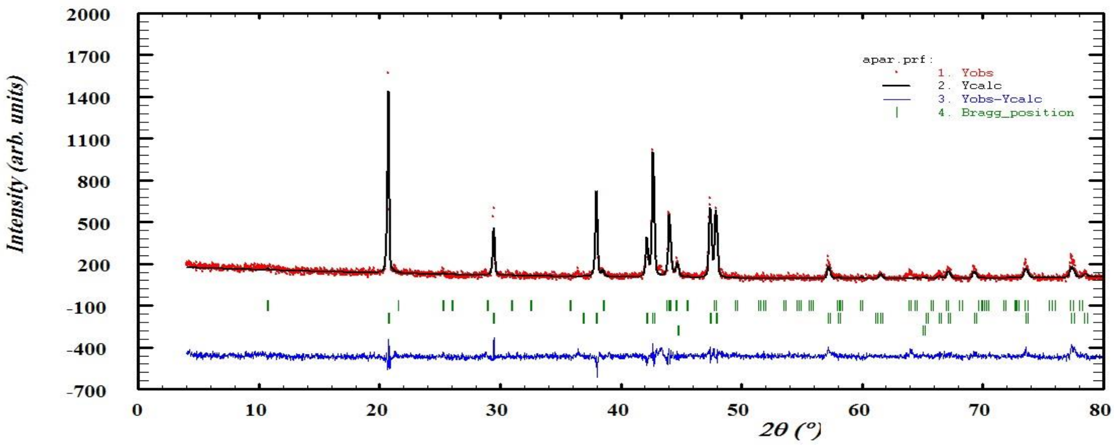


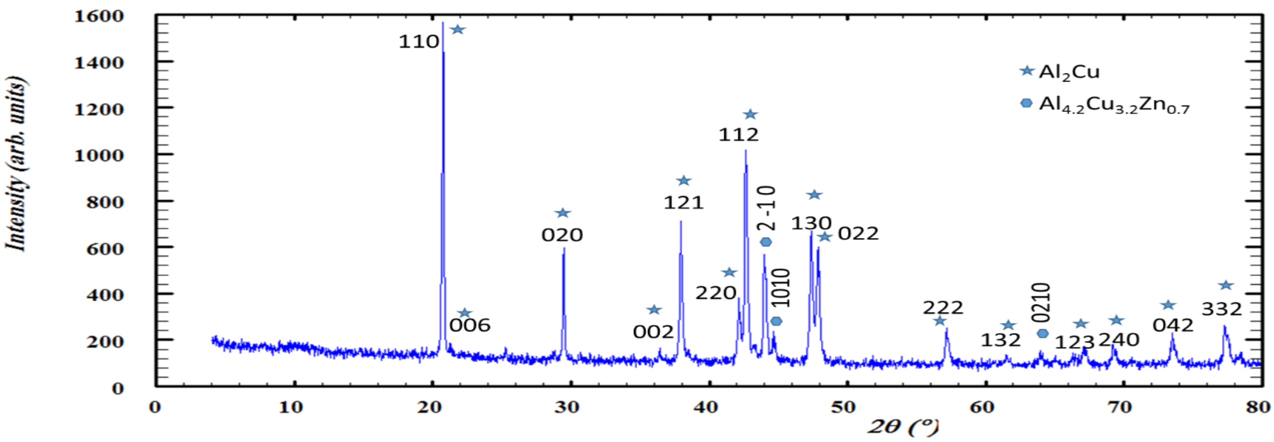
FIG.2. EDAX analyses of the Cu-Al-Zn alloy



Element	Weight %	Atomic %	Net Int.	Error %
AlK	41.86	62.95	182.78	8.76
CuK	54.02	34.5	84.9	4.51
ZnK	4.12	2.56	5.3	27.52

FIG.3a. X-Ray diffraction pattern of the Cu-Al-Zn alloy
FIG.3b. Rietveled refinement for the bi-phasic metal alloy





Characterization of the prepared poly 3-(Trimethoxysilyl)propyl methacrylate (TMSPM)

Fig.5 shows **SEM microstructure** of the investigated prepared compound which has homogenous morphological structure with laminar shape which is a characteristic of this type of material resins and results in decreasing the surface areas [13].

Fig.6 illustrated **Infrared spectroscopy** the structural formula of the prepared polymer was confirmed by observation of (C-Si-O) at 1178 cm^{-1}, the strong absorption band at 1709 cm-1 is attributed to ester (C=O) bond where, stretching absorption band of aliphatic (C-H) bond was observed at 2925 cm^{-1} and its bending vibration at 1393 cm^{-1} [14].

- **Characterization of rubber-alloy composites**

XRD of the pure and loaded EPDM with different percentage of metal alloy in absence of coupling agent are shown in Fig.7. From this figure it is observed that the pure EPDM pattern composed of two halos superimposed on it several crystalline peaks differ in its intensity, the halos at 2θ= 7.2°, 18.74° indicate the semi- crystalline nature of EPDM rubber. The other peaks at 11.47, 21.89, 23.07, 26.71, 27.39, 27.72, 28.62, and 54.28 are related to EPDM. The superimposed peaks at 31.66°, 34.37°, 36.11°, 47.52°, 56.55°, 62.79°, 67.91°, 69.0° congruous to the reflections 010, 200, 011, 012,110, 013, 211, 120 of hexagonal ZnO (card no 98-006-5121) which is added for vulcanization. The appearance of ZnO peaks may be linked to the upshot related to particle surface activity, particularly at the boundary of rubber and ZnO, which has some sulfur and stearic acid on the surface, the same behavior was obtained before by G. Heideman [15].

Other Peaks were shown at 2θ=20.71, 29.373, 37.89, 42.02, 42.59, 47.35, 47.78, 57.12° congruous to the reflections 110, 020, 121, 220, 112, 130, 022, 222 of metal alloys Cu-Al_2 phase 98-015-1372 (98-004-2518) and peak at 44 congruous to the hexagonal phase of $Al_{4.2}Cu_{3.2}Zn_{0.7}$ phase 98-65-4952. It is noticed that the 012 peak of ZnO in the composite in combination (overlapped with) 130 peak of Al_2Cu. From Fig. 7 it is distinctly shown that, the intensity of the metal alloy peaks increases with increasing its content due to its agglomerations while the intensity of the EPDM peak and halos decreases as shown in Fig.8. This increase in the intensity of the metal alloy confirms its existence in the composite or into the bulk polymer matrix. The effect of increasing the intensity of the metal alloy in the composite may be reflected on the strength of the elastic modulus. Besides, no change observed in the peak positions of EPDM instead of decrease in its peak intensity which an indication to that no changes in the chemical structure of EPDM rubber and the blend consists of composite of metal alloy and EPDM. The same behavior was obtained before by Ahmed M. Khalil et al [16] during their studies on the effect of short polyethylene terephthalate fibers on properties of ethylene-propylene diene rubber composites.

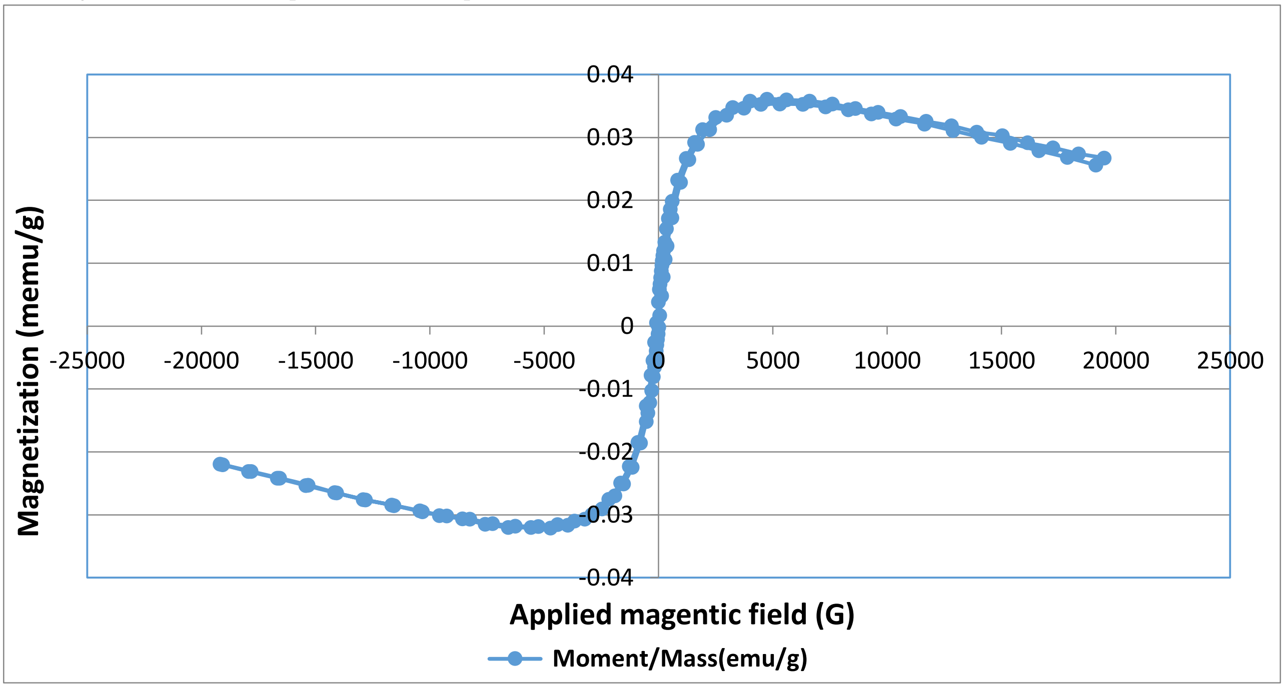


FIG.4. Magnetization curves at room temperature of Cu-Al-Zn alloy

X-ray diffraction pattern of the pure and loaded EPDM with different percentage of metal alloy in presence of coupling agent are shown in Fig. 8. Nearly the same diffraction pattern like as Fig.7 without coupling agent. No more peaks corresponding to the coupling agent alone or in combination with any compound of the composites were observed; this may be due to lower content of coupling agent i.e below the detection limit of X-ray diffraction. Fig.9 is a representative for the effect of coupling agent on the EPDM loaded with 20% of metal alloy, it is clearly shown that a little shift at all EPDM peaks positions accompanied by decreasing of its intensity due to the coupling agent.

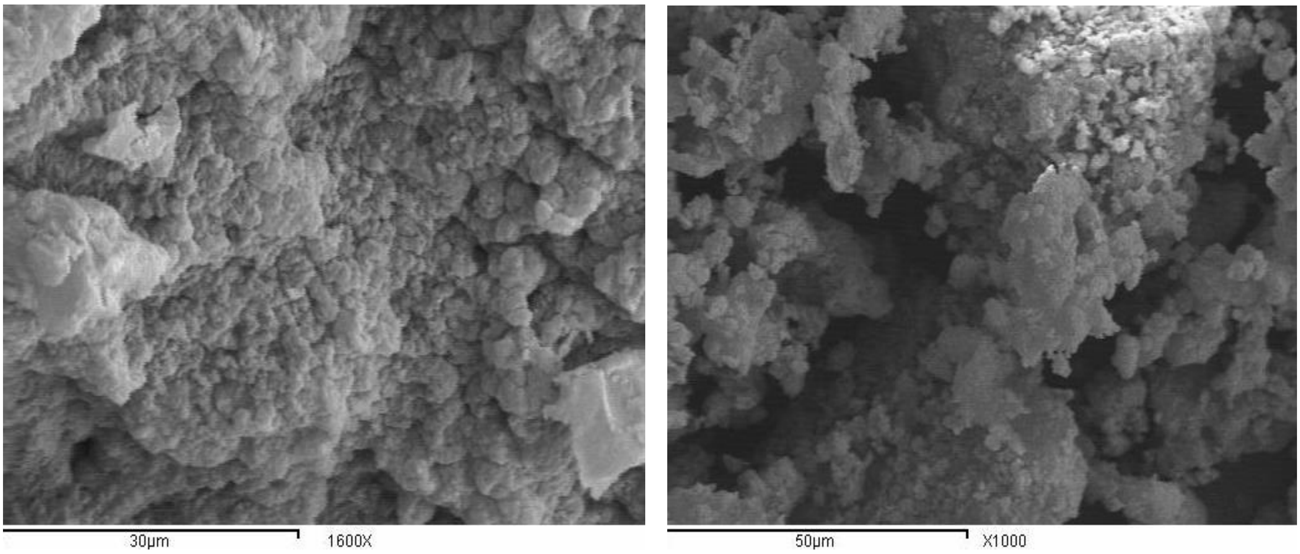


FIG.5 represents SEM micrographs of the prepared poly 3-(Trimethoxysilyl) propyl methacrylate using two different magnifications 1000 and 1600 X.

- **Magnetic properties of EPDM/Cu-Al-Zn composites**

Fig.10 showed the magnetization, M(emu/g) as a function of the applied magnetic field (G) of /EPDM/Cu-Al-Zn alloy composites at different concentrations of alloy (5 phr and 20 phr) with and without coupling agent. It is shown that the magnetization of composites increases with increasing the concentrations of alloy (from 5phr to20phr) in presence of coupling agent than in absence of coupling agent.

Rheological properties of EPDM/ Cu-Al-Zn rubber composites

In this work, a Cu-Al-Zn alloy was added into nonpolar EPDM rubber formulations with different loadings (2.5-20 phr) to study the effect of different content of Cu-based alloy on the rheological properties of EPDM composites (in both cases presence and absence of coupling agent). The minimum torque (M_L) and maximum torque (M_H) from the curing curves showed direct relationship with the loadings of the different content of alloy as in Table 1. M_L is a determination of the viscosity at a very low shear rate, while M_H is an estimate of the dynamic modulus at comparatively low strain amplitudes [17]. At higher loadings of Cu-Al-Zn alloy in presence coupling agent; higher investigated alloy networking and more compounded viscosities of rubber can be observed due to the presence of more adsorbed polymer. This adsorbed polymer can be inactive, besides agglomeration of Cu-based alloy that leads to restricted rubber movement, which causes loss of its identity and make it behave as filler in terms of stress-strain properties [18]. The attachment of rubber chains on the alloy surface may serve as extra cross-links, leading to the increase of the modulus.

It was also observed that the torque difference ΔM which is typically related to the crosslink density of EPDM vulcanizates, decreased with addition of Cu-Al-Zn alloy due to the decrease in the overall rigidity of rubber composites, which not permitted to increase in crosslink density of rubber in composites due to the absence of linkages between Cu-Al-Zn alloy particles and EPDM rubber molecules in the absence of coupling agent TMSPM. As well, the value of ΔM of composites in presence TMSPM is higher than in absence. This means that, in presence of coupling agent TMSPM, caused an improvement in interfacial adhesion makes Cu-Al-Zn alloy and EPDM less repulsive toward each other and hence, results in better dispersion of Cu-Al-Zn alloy particle within in EPDM domains. This result confirms with SEM morphology where the Cu-Al-Zn alloy in presence TMSPM, is the best dispersed in EPDM matrix and also confirms from the increase in value of elongation at break [18].

Table 4 Magnetic parameters of EPDM/ Cu-Al-Zn alloy.

EPDM composites	Ms (emu)	Mr (emu/g)	Hci (G)
Cu-Al-Zn alloy	34.112×10^{-3}	2.2175×10^{-3}	53.905
EPDM	9.5370×10^{-3}	38.525×10^{-6}	227.86
5phr of Cu-Al-Zn alloy without coupling agent	8.6399×10^{-3}	165.24×10^{-6}	1031.8
5phr of Cu-Al-Zn alloy with coupling agent	10.776×10^{-3}	260.55×10^{-6}	2344.8
20phr of Cu-Al-Zn alloy without coupling agent	10.318×10^{-3}	723.89×10^{-6}	3269.2
20phr of Cu-Al-Zn alloy with coupling agent	4.7378×10^{-3}	273.43×10^{-6}	4384.9

Coercivity (Hci) is defined as the minimum value of magnetising intensity that is required to bring the material to its original state. Retentivity (Mr) is a material's ability to retain a certain amount of residual magnetic field when the magnetizing force is removed after achieving saturation. Magnetization (Ms) is the density of magnetic dipole moments that are induced in a magnetic material when it is placed near a magnet.

The curing time (tc_{90}) either in presence or absence coupling agent decreased with increasing the content of alloy as obtained in Fig.11a. The enhancement in the cure rate indicated that Cu-based alloy act as a co-curing agent (secondary accelerator) in the curing process, i.e. Cu-Al-Zn alloy tends to slightly improve the cure rate. The presence of coupling agent minimizes the optimum cure time on adding TMSPM as compatibilizer, this is due to the presence of an additional functional group of acrylate facilitates on polymer surfaces TMSPMA.

Also, the presence of coupling agent TMSPM lead to increase the thermal transition which elevated the vulcanization process and improved interactions between Cu-Al-Zn alloy and rubber matrix [19].

The scorch time (t_{S2}) increased for initial loading of Cu-alloy (2.5 phr) compared to the control, and then there was decrease on additional loadings as shown in Fig. 11b. The increase in t_{S2} in presence coupling agent was due to the reaction between acrylate group (TMSPM) and alloy surface that has reacted with the activator as well as the accelerator. Consequently, the number of sulfurating agents lowered which is important for vulcanization reactions [20].

This increase in scorch time was due to the presence of Cu-Al-Zn alloy which interacted with ZnO during the curing process and makes it incapable to activate the accelerator. Consequently, Zinc Oxide activity was decreased and the curing was inhibited. In the absence of coupling agent $\mathbf{t_{S2}}$ of EPDM/Cu-Al-Zn composites remained at the same level, and then it was slightly decreased with further loadings of Cu-Al-Zn alloy.

Mechanical properties

The effect of addition Cu-Al-Zn alloy on the tensile strength of EPDM composites is presented in Fig. 12a. In the regions of the low Cu-alloy loading, the tensile strength slightly increased with increasing copper alloy content in presence or absence coupling agent. With increasing Cu-alloy content, the tensile strength remained almost constant. The reduction in the tensile strength can be observed in presence coupling agent (TMSPM) which may be due to the agglomeration and the increased Cu-alloy contents. On the other hand the using of coupling agent (TMSPM) with Cu-alloy leads to the formation of a continuous layer between the EPDM matrix and the copper alloy particles [21, 22].

The variation of the elongation at break with Cu-alloy loading for EPDM composites is shown in Fig. 12b. One can see that higher loading of Cu-alloy effectively increases the elongation at break. When the content of alloy exceeds which can completely react with ZnO, the residual Cu-alloy is cross-linked into rubber with a linear structure, which leads to a high elongation at break. In addition, the improvement in elongation at break is explained in terms of non-adherence of the filler to the rubber matrix leading to the softening of the rubber chain and hence allowing to stretch when the strain is applied.

The values of elongation at break decrease in presence of coupling agent with the increase in loading for all content. This behavior is explained in terms of the addition of alloy to the polymer matrix reduces chain mobility, giving rise to a rapidly decrease in the elongation at break [23, 24].

Scanning electron microscopy

Fig.13 shows the SEM images for selected content of Cu-Al-Zn/EPDM, with and without coupling agent. From these figure, it is seen that fine distribution of both fillers was detected at 10 and 20 phr Cu-alloy loading. This fine distribution was the cause of the performance in both magnetic and the mechanical properties of such composites. The surface of Cu-Al-Zn/EPDM composites with and without coupling agent (TMSPM) is characterized with the existence of tiny white particles, which due to ZnO ingredient. In addition, it can be seen that the compatibility between Cu-Al-Zn alloy and EPDM matrix in which there is no phase separation and continuous matrix. A good distribution of Cu-alloy within the EPDM composite and co-continuous structure was observed in the sample containing 20 phr Cu-alloy with or without coupling agent. The rubber-filler adhesion is increased and in high filler content due to the decrease in the tendency of the filler particles to agglomerate [25]. Otherwise Fig.13d-13e displayed the effect of adding coupling agent on Cu-Al-Zn/EPDM composites, which indicate enhancement the interaction between rubber matrix and Cu-alloy in presence coupling agent. This reason can explain of the high value of tensile strength for EPDM composite containing 20 phr metal Cu-alloy.

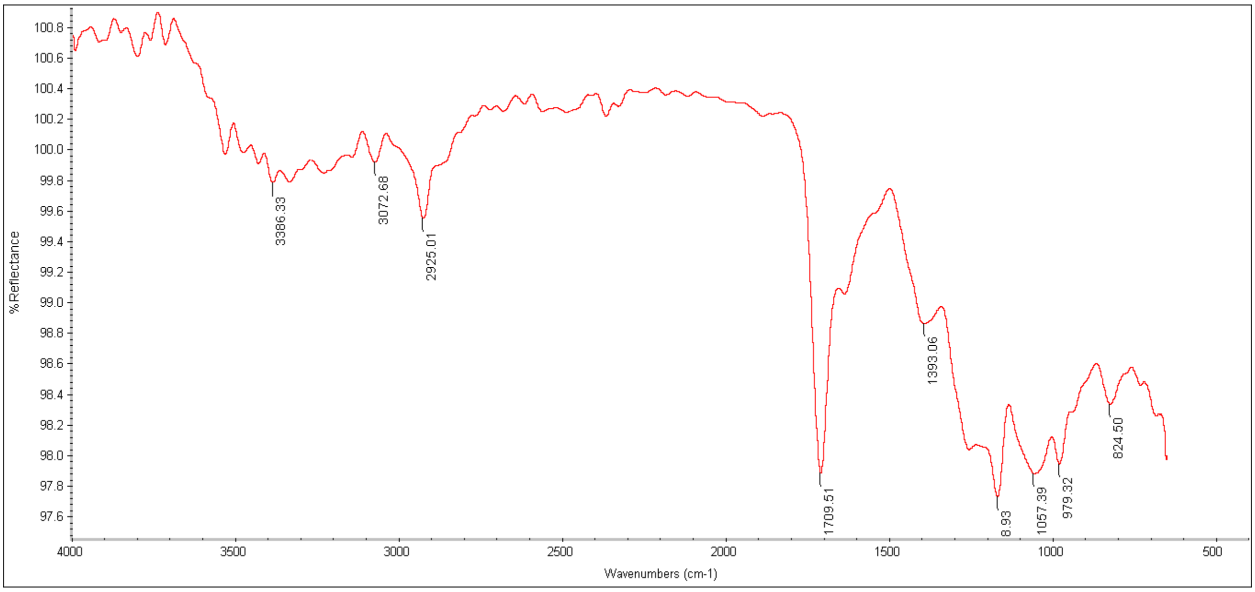


FIG.6. FTIR analysis of the prepared poly 3-(Trimethoxysilyl) propyl methacrylate,

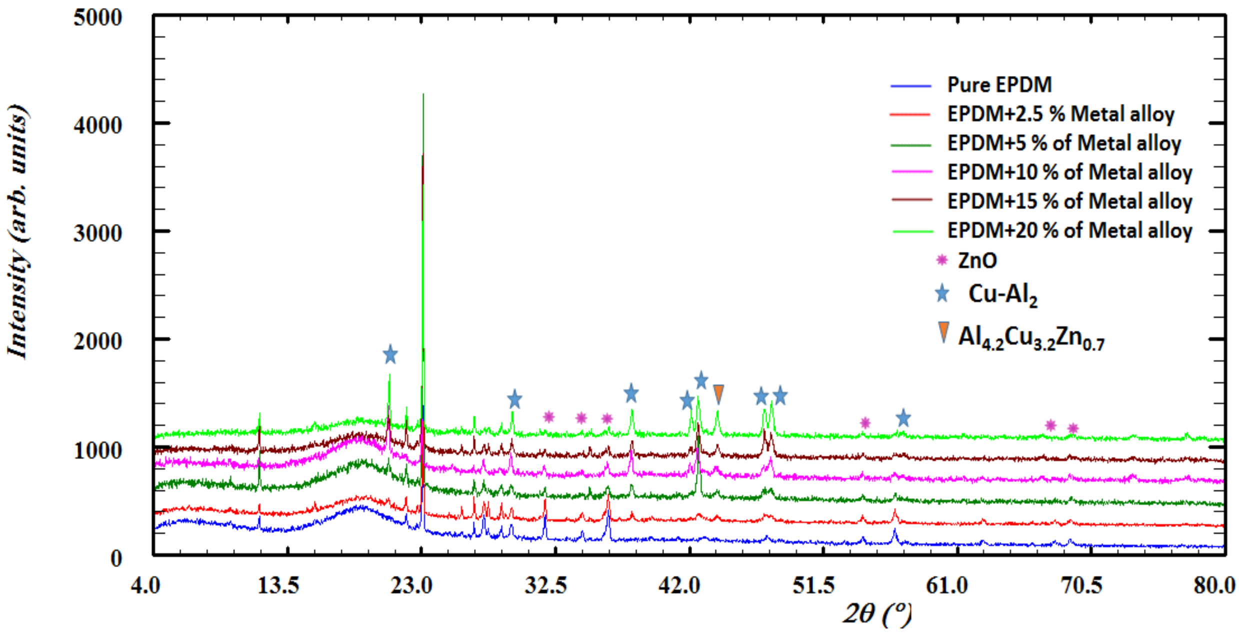


FIG.7. XRD of Pure and loaded EPDM with different percentage of the metal alloy in the absence of coupling agent

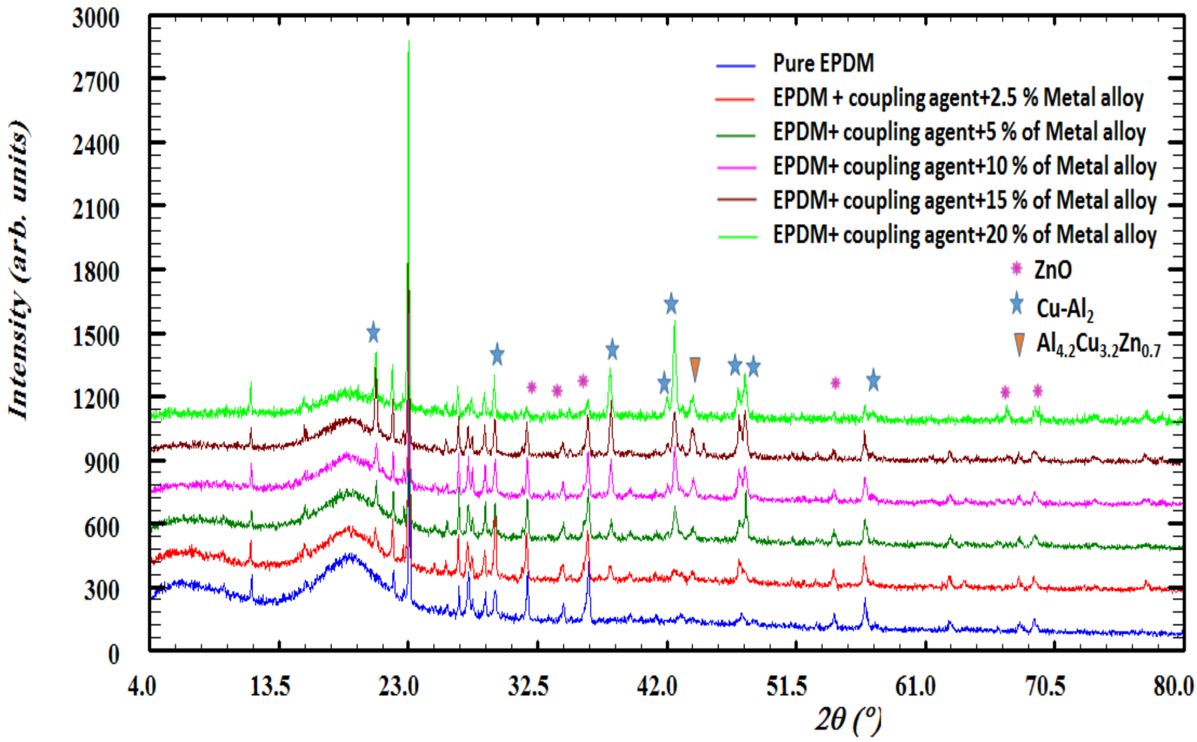


FIG.8. XRD of Pure and loaded EPDM with different percentage of the metal alloy in the presence of coupling agent

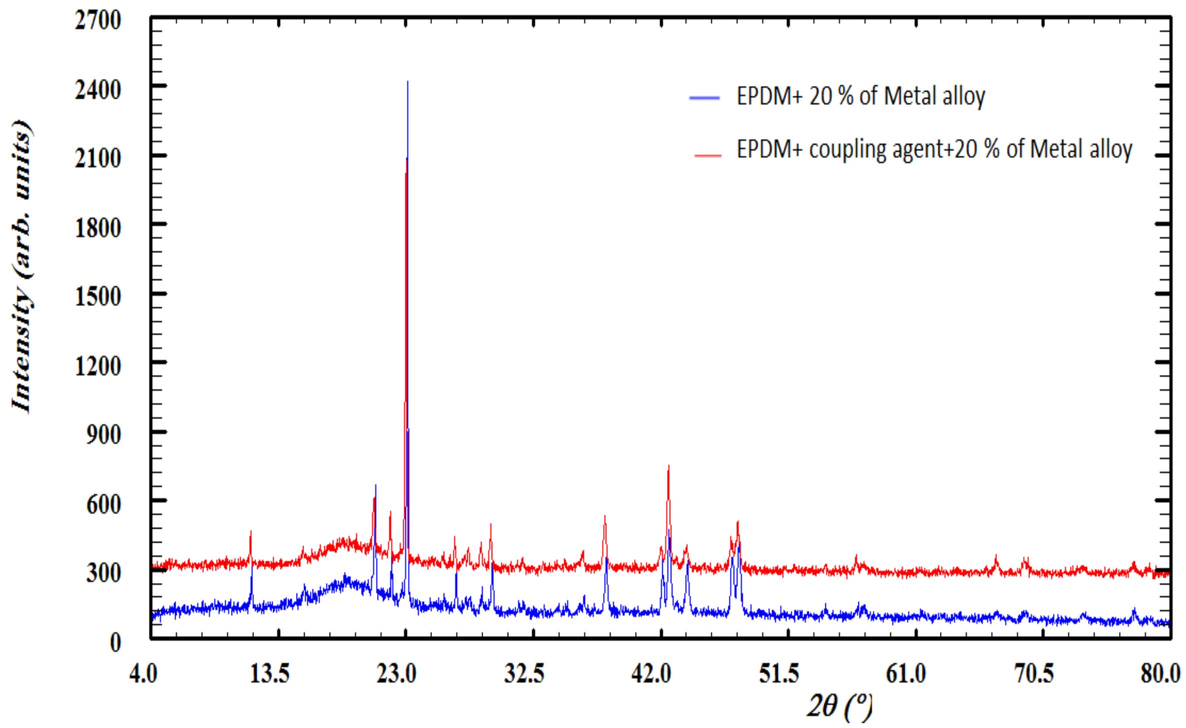


Fig.9. XRD of EPDM loaded of 20% of metal alloy with and without coupling agent

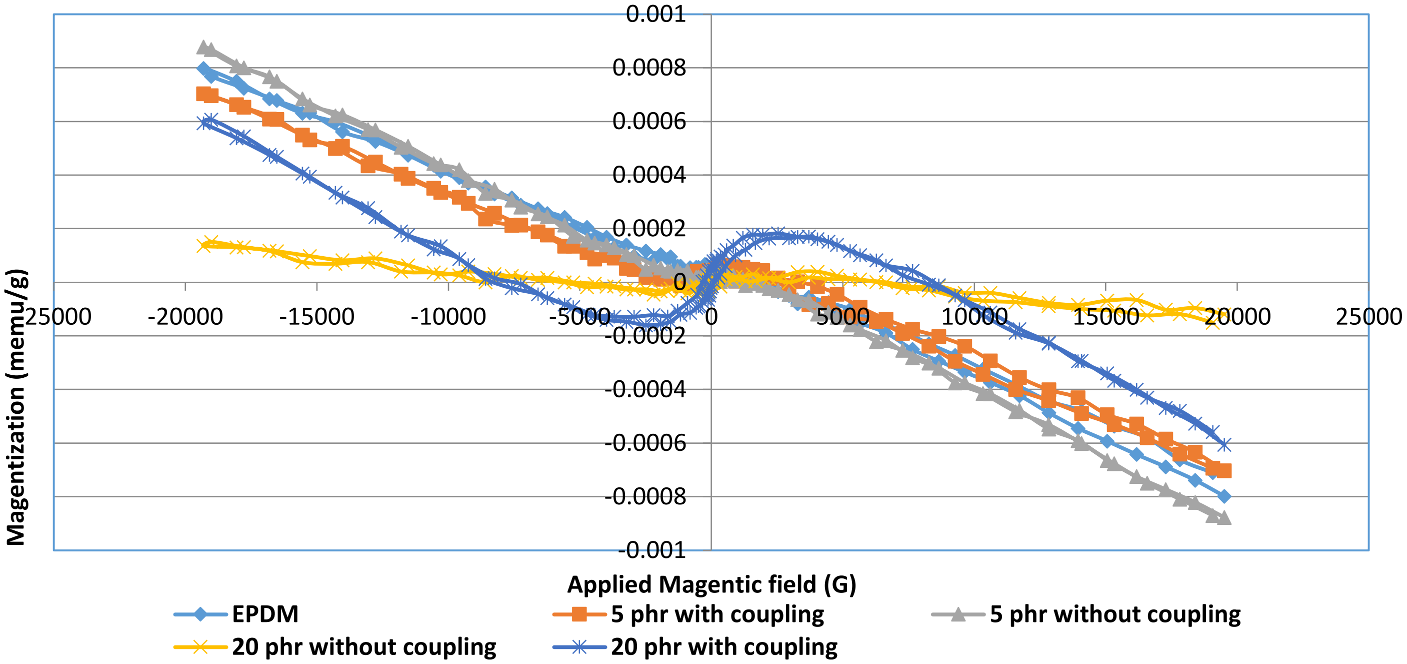
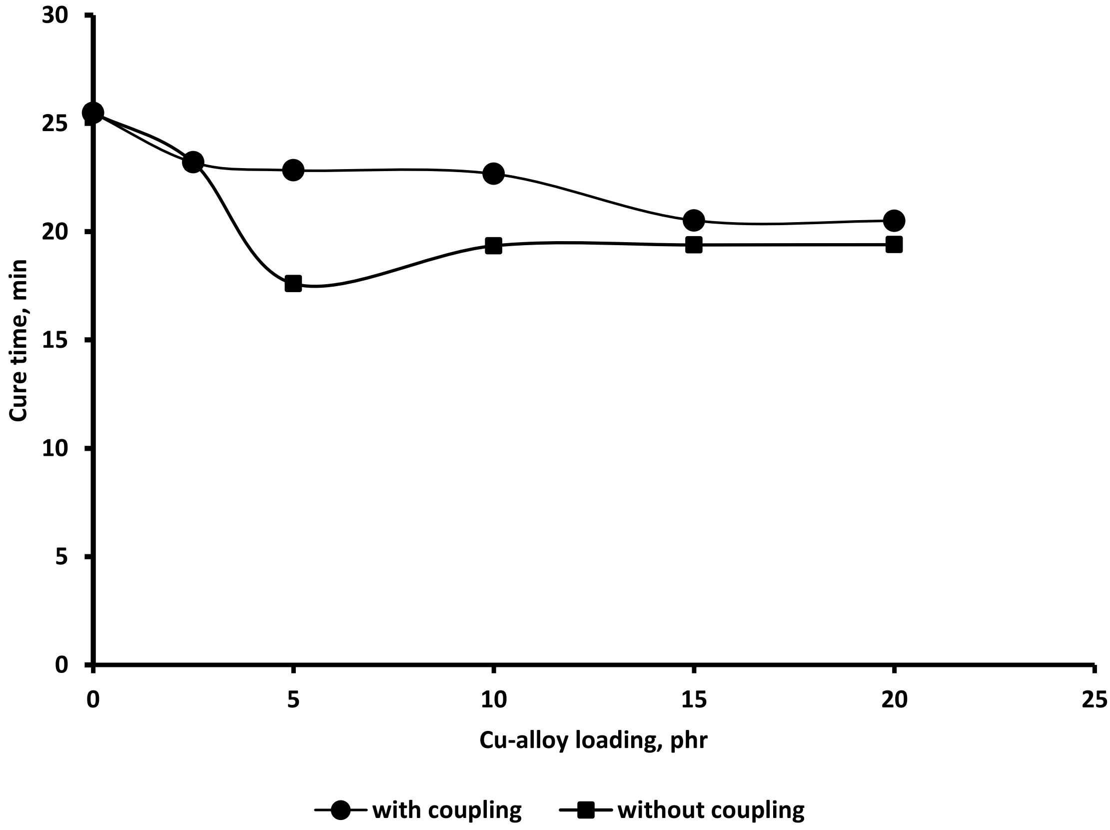


FIG.10. Hysteresis loop performed at room temperature for EPDM/ Cu-Al-Zn/with concentration 10 and 20 phr Cu-Al-Zn alloy with and without coupling agent



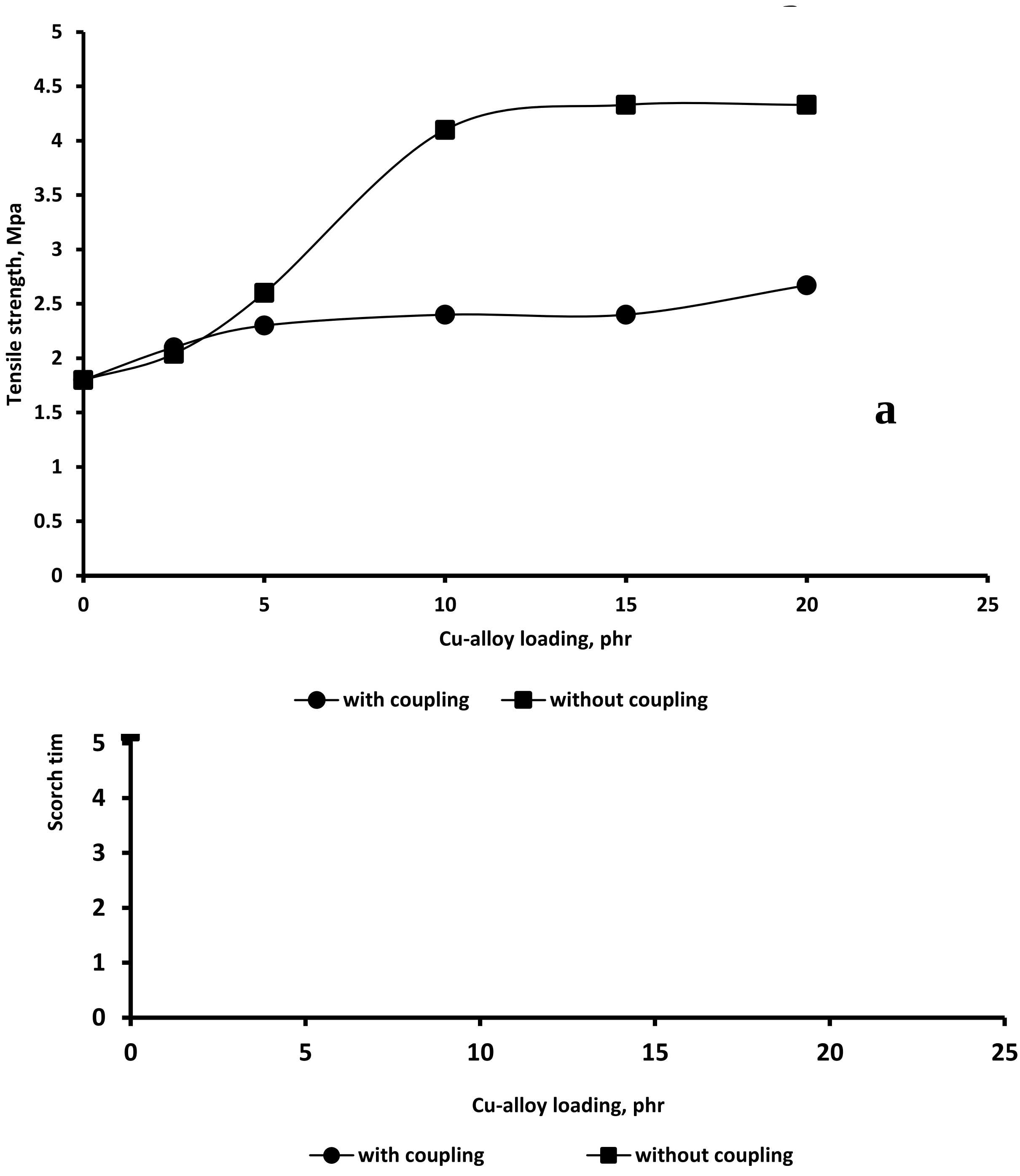


FIG.11. Cure and scorch time of Cu-Al-Zn/EPDM composites

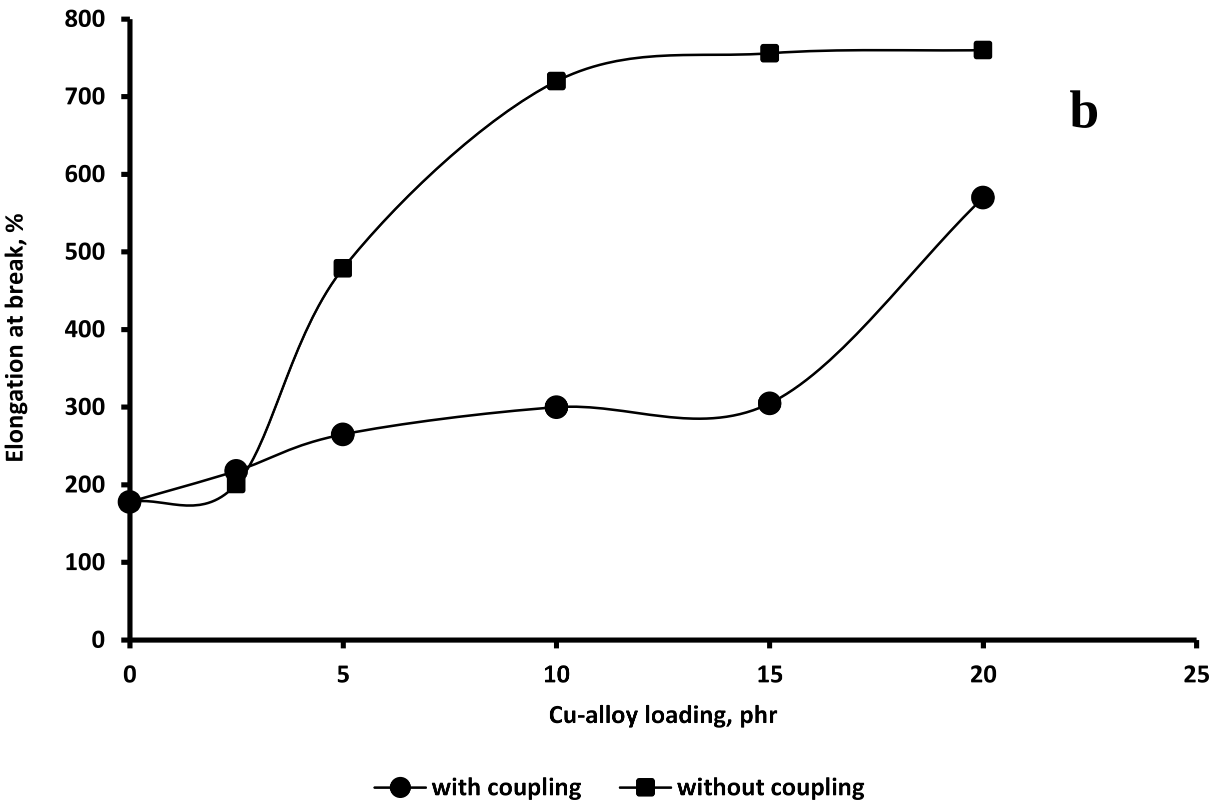
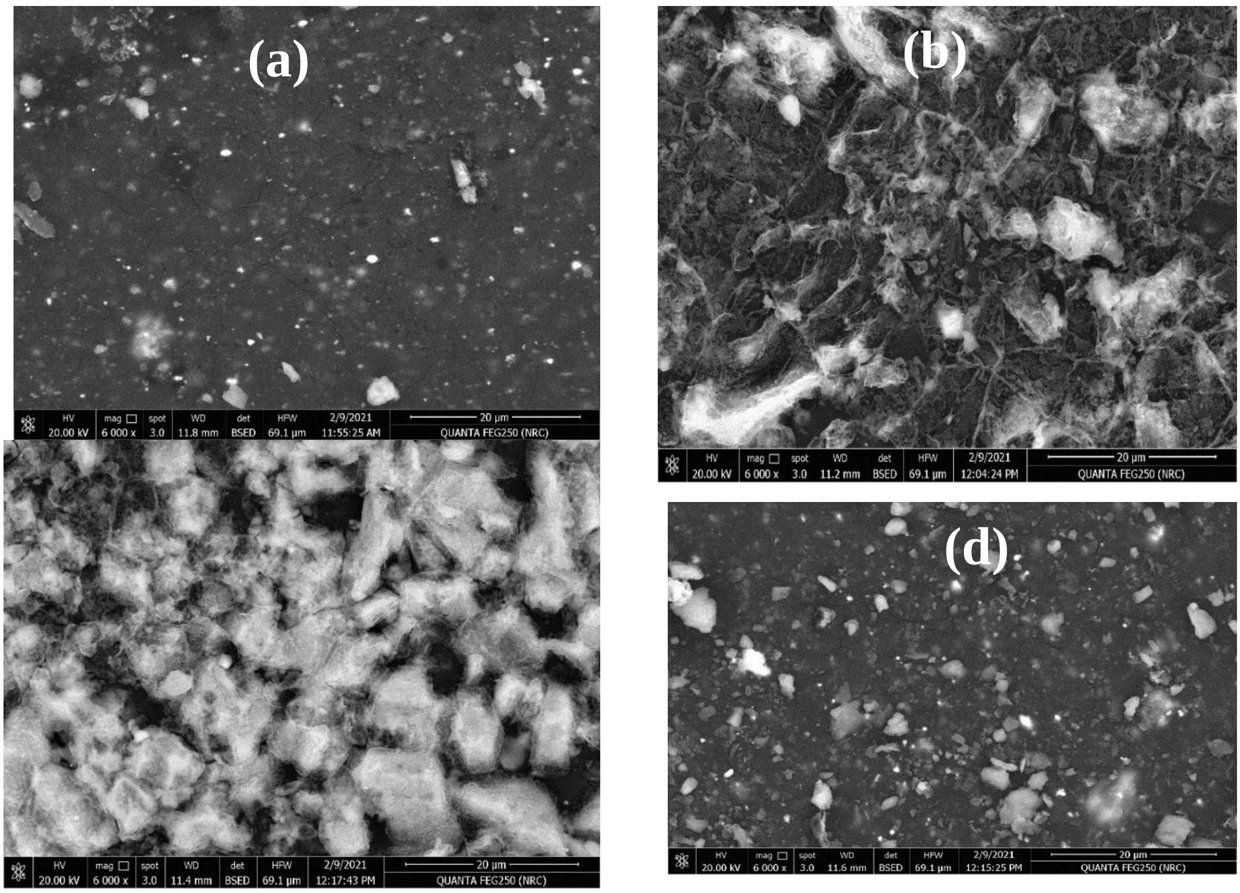


FIG.12. The change of tensile strength and elongation at break of investigated EPDM composites



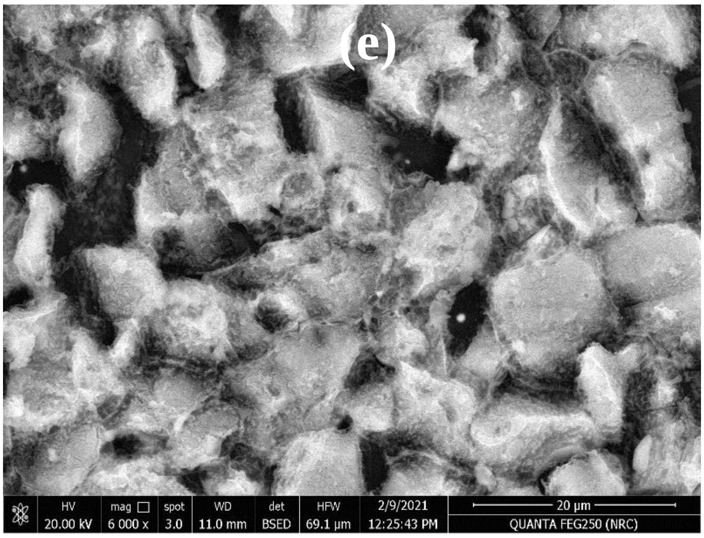


FIG.13. SEM for EPDM composite loaded with (a) 0 phr Cu-alloy, (b) 10 phr & (c) 20 phr Cu-alloy without coupling agent and (d) 10 phr & (e) 20 phr Cu-alloy with coupling agent

Conclusions

- The composition of Cu-Al-Zn alloy was characterized by XRD and EDX.
- The prepared EPDM composites containing coupling agent showed an improvement of the magnetization rather than without coupling agent. This was confirmed by XRD results.
- In presence of coupling agent with Cu-Al-Zn alloy revealed increased maximum and minimum torque. While in absence coupling agent reduced scorch and curing time.
- Generally, two extreme ends of Cu-Al-Zn alloy content i.e. very low (2.5 phr) and very high filler loadings (20 phr) resulted slightly improve in mechanical properties of composites (especially in elongation at break).
- Consequently, the presence of cu-based alloy lead to unchanged in tensile strength of composite while the values of elongation increase.
- Morphology was affected by incorporation of Cu-Al-Zn alloy in EPDM matrix, at very high alloy loadings showed good dispersion of Cu-Al-Zn alloy particles which was evidenced by the absence of agglomerates on SEM micrographs

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