### The effect of addition of the Filler (Zeolite) to the Polyethylene

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الملخص:

استهدفت هذه الدراسة تأثير إضافة المواد المالئة (الزيوليت) إلى بوليمر البولي ايثيلين, واستخدمت هذه المادة كحشو مع البولي ايثيلين المحلي والمنتج من قبل شركة رأس لانوف لتصنيع النفط والغاز والمصنف بالبولي ايثيلين الخطي منخفض الكثافة (LLDPE) النوع LLF-181N. تم طحن وغربلة الزيوليت وتحديد حجم الحبيبات وتكوين خلطات من البولي ايثيلين مع الزيوليت بنسب مختلفة (5, 10, 15, 10, 20)%. تمت عملية الخلط لهذه النسب على جهاز الخلط (Two Roll Machine). وكانت النتائج كالآتي :

- أثبتت نتائج الخواص الميكانيكية أن أفضل نسب إضافة للزيوليت مع البولي ايثيلين الخطي منخفض الكثافة تبدأ من 10% وحتى 20%.

- من خلال نتائج التحليل الحراري للمواد المالئة (الزيوليت) مع البولي ايثيلين كانت أعلى نقطة لدرجة الانصبهار البلورية للزيوليت عند 15%.

- نتيجة لتعرض المادة البلاستيكية للظروف البيئية المتغيرة تم إجراء اختبار مقاومة التكسير البيئي (Environmental Stress Cracking) لخلطات البولي ايثيلين الخطي منخفض الكثافة مع الزيوليت أوضحت النتائج أن جميع نسب الخلط لم يحدث لها أي تكسير.

#### Abstract

This study targeted the effect of the addition of filler (zeolite) to polyethylene polymer. This filler was used as a filler with local polyethylene and produced by Ras Lanuf Oil, Gas Manufacturing Company and LLDPE type LLF-181N. The zeolite was grinded and quantified and the granule size and composition of polyethylene mixtures with zeolite were determined by different percentages (5, 10, 15, 20, 30)%. The mixing process was done by the Two Roll Machine.

- Mechanical properties showed that the best ratios of addition of zeolite with linear low density polyethylene are from 10% to 20%.

- The results of thermal analysis of filler (zeolite) with polyethylene were the highest point of crystalline melting point of zeolite at 15%.

- As a result of exposure of plastic material to changing environmental conditions, the environmental stress cracking test was conducted for low density linear polyethylene mixtures with zeolite. The results showed that all mixing ratios did not break.

## Introduction

In order to study the effect of the addition of fillers specifically zeolite to polyethylene, the local polyethylene polymer and product was used by Ras Lanuf Oil and Gas Manufacturing Company and was classified under LLDPE Type LLF-181N as described in the following table:

| Property                  | The standard<br>American method | Unit   | Value |  |
|---------------------------|---------------------------------|--------|-------|--|
| D 1                       | (ASTM)                          |        |       |  |
| Brand                     | LLF - 181 N                     |        |       |  |
| Continuous heat           | $93 C^0$                        |        |       |  |
| resistance                | 93 C                            |        |       |  |
| Elongation                | 90-800 %                        |        |       |  |
| Fusion coefficient 190    | D1238                           | g /10  | 1.0   |  |
| $C^{0}$ ,2.16 kg          | D1238                           | min    | 1.0   |  |
| Density 23 C <sup>0</sup> | D1505                           | g /cm³ | 0.918 |  |

Table(1).Specification for low density polyethylene (LLF - 181 N)

Zeolite has been used as a filler with low density linear polyethylene and its impact on polymer properties such as mechanical properties, hardness strength, shock strength, resistance to environmental conditions by studying ESCR as a result of the continuous use of heat and cold material at the same time, Thermal analysis test (DSC) to track their tolerance to heat through the degree of crystalline fusion to be supported in the industrial field.

Zeolite is one of the silicate aluminum compounds that are similar in composition to the feldspar, except for the amount of water stored with sodium and calcium as basic minerals. Rarely there is barium in zeolite ore. There are different types of zeolite ore, about 100 types: Heulandite, Chabazite, Clinoptilolite Mesolite, Scolecite, Laumontite, Analcite, Erionite, Mordenite, Phillipsite and Stilbite. One of the studied species is the analsite metal, where this crude is found through continental formations in the region called the formation of a dembaba from the Upper

Carbonic period. The formation of dembaba contains mineralization of the analysmolite which is the result of the decomposition of volcanic ash in the shallow navigators. Zeolite is found in sandstone at a rate of 51 to 55%, one of the sites of the presence of zeolite ore is in the western Awainat in Libya. The components of this crude have been determined as follows:

| Ingredients                    | Average<br>analysis% |  |
|--------------------------------|----------------------|--|
| SiO <sub>2</sub>               | 61.36                |  |
| Fe <sub>2</sub> O <sub>3</sub> | 10.16                |  |
| Al <sub>2</sub> O <sub>3</sub> | 11.08                |  |
| CaO                            | 2.8                  |  |
| MgO                            | very weak            |  |
| Na <sub>2</sub> O              | 3.89                 |  |
| Ti <sub>2</sub> O              | 0.40                 |  |
| SO <sub>3</sub>                | very weak            |  |
| Loss of combustion of          | 9.33                 |  |
| organic matter L.O.I           | 7.55                 |  |
| The total percentage           | 99.02                |  |
| T 11 (0) 7 1'                  |                      |  |

Table(2). Zeolite ore components

The chemical composition of zeolite metal was identified using X-ray diffraction:

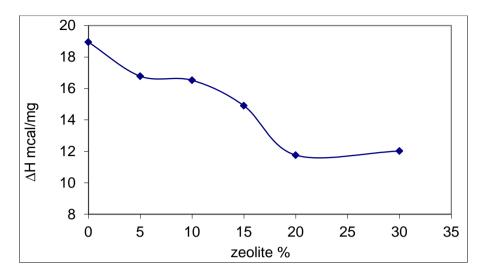
(Na Al (SiO<sub>3</sub>)  $_2$ .H<sub>2</sub>O), this type of zeolite is called Analcite (ANA)

# Results and discussion: Polymer Character Testing 1- Thermal analysis test.

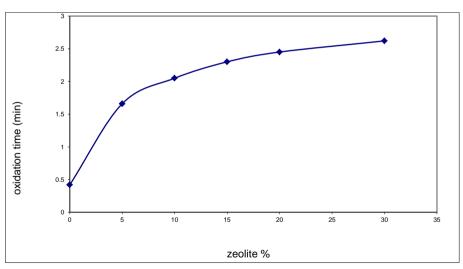
Thermal analysis means the thermodynamic properties of a polymer to determine the polymer's validity for different uses. In this test, the Differential Scanning Calorimeter (DSC) method was used for the samples of polyethylene mixtures with zeolite to measure the change in ( $\Delta$ H), oxidation time (OIT) and the degree of crystalline fusion ( $T_m$ )

| (%)<br>zeolit<br>e | (%)polyethylen<br>e | Antibiotic<br>s (ΔH) m<br>Cal / mg | Time of<br>oxidatio<br>n (min) | End of<br>fusion<br>C°(Tm<br>) | Start<br>fusion<br>°C |
|--------------------|---------------------|------------------------------------|--------------------------------|--------------------------------|-----------------------|
| 0                  | 100                 | 18.95                              | 0.42                           | 129.58                         | 121.4<br>5            |
| 5                  | 95                  | 16.78                              | 1.66                           | 130.77                         | 118.5<br>9            |
| 10                 | 90                  | 16.52                              | 2.05                           | 130.63                         | 119.8<br>7            |
| 15                 | 85                  | 14.90                              | 2.30                           | 131.84                         | 120.1<br>5            |
| 20                 | 80                  | 11.76                              | 2.45                           | 130.45                         | 121.9<br>9            |
| 30                 | 70                  | 12.02                              | 2.62                           | 130.00                         | 119.8<br>9            |

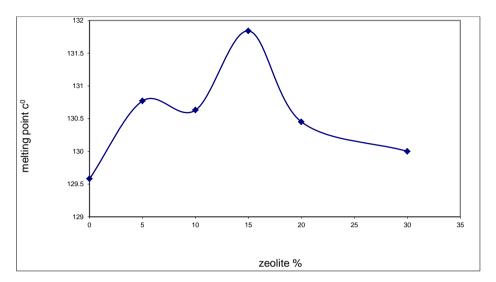
Table(3). The results of the differential analysis of calorimeters for samples of polyethylene with zeolite

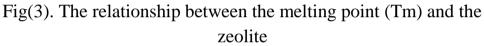


Fig(1). The relationship between the change in the anthaly ( $\Delta H$ ) and the zeolite



Fig(2). The relationship between oxidation time and the ratio of zeolite





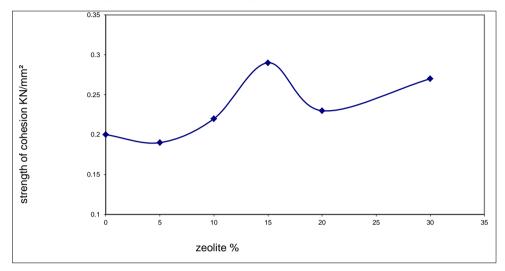
#### 2- Tensile and elongation test

In this test, LLDPE and polymerization mixtures were measured with zeolite and different ratios, in terms of strength, elongation, maximum elasticity, and fracture point, using the ASTM D638 tensile strain gauge for a series of samples at Each ratio is added to the filler.

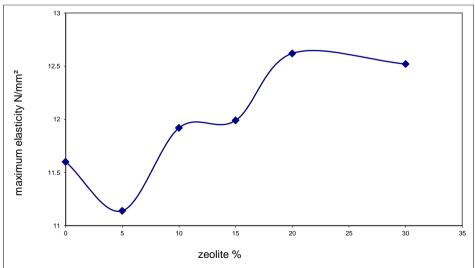
| Zeolit<br>e<br>ratio<br>(%) | Polyethyle<br>ne<br>percentage<br>(%) | Brea<br>k<br>Point<br>N /<br>mm <sup>2</sup> | Maximu<br>m<br>flexibilit<br>y<br>N / mm <sup>2</sup> | Elongatio<br>n<br>percentag<br>e(%) | Coheren<br>ce<br>strength<br>KN/mm <sup>2</sup> |
|-----------------------------|---------------------------------------|--|---|-------------------------------------|---|
| 0                           | 100                                   | 19.02  | 11.60   | 602.84                              | 0.20  |
| 5                           | 95                                    | 15.11  | 11.14   | 469.78                              | 0.19  |
| 10                          | 90                                    | 12.32  | 11.92   | 430.00                              | 0.22  |
| 15                          | 85                                    | 12.09  | 11.99   | 342.79                              | 0.29  |
| 20                          | 80                                    | 12.70  | 12.62   | 83.06                               | 0.23  |

| 30 | 70 | 12.55 | 12.52 | 45.78 | 0.27 |
|----|----|-------|-------|-------|------|
|----|----|-------|-------|-------|------|

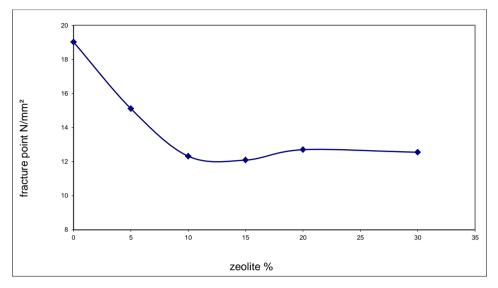
Table(4). Characteristics of tensile strength and elongation of samples of low density polyethylene with zeolite.



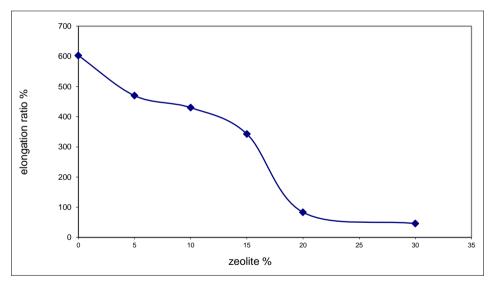
Fig(4). The relationship between the strength of cohesion and the addition ratios of zeolite



Fig(5). The relationship between the maximum elasticity and addition ratios of zeolite



Fig(6). The relationship between the fracture point and the addition ratios of zeolite



Fig(7). The relationship between the elongation ratio and the addition ratios of zeolite

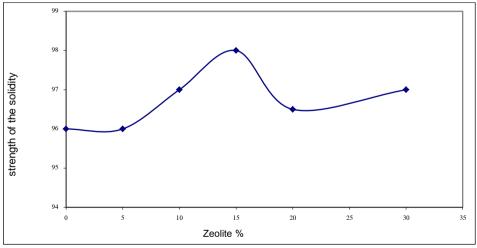
## **3- Tough Strength Test**

The rigidity test was performed using the Shore-A device according to ASTM D2240 for mixtures made of polyethylene with zeolite. Results obtained as follows :

| Polymer % | Fillers % | The strength<br>(Shore-A) of<br>zeolite |
|-----------|-----------|---|
| 100       | 0         | 96.00                                   |
| 95        | 5         | 96.00                                   |
| 90        | 10        | 97.00                                   |
| 85        | 15        | 98.00                                   |
| 80        | 20        | 96.50                                   |
| 70        | 30        | 97.00                                   |

Table(5). Strength strength of polyethylene mix samples withfree straw and zeolite.

The strength of the zeolite hardness with polyethylene is shown that it has the highest strength at 15% and 98%.



Fig(8). The relationship between the strength of the solidity (Shore-A) and the addition ratios of zeolite

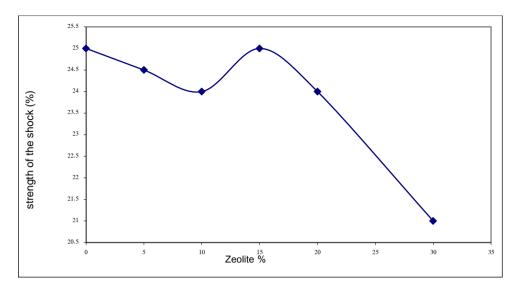
## 4- Test the power of collision

The shock strength measurements of LLDPE mixtures with zeolite were conducted by ZWICK & Co. KG Einsingen bei ULM to identify how much these mixtures could afford the sudden shock.

| Polymer % | Fillers % | The shock power of zeolite(%) |
|-----------|-----------|-------------------------------|
| 100       | 0         | 25.00                         |
| 95        | 5         | 24.50                         |
| 90        | 10        | 24.00                         |
| 85        | 15        | 25.00                         |
| 80        | 20        | 24.00                         |
| 70        | 30        | 21.00                         |

Table(6). Shock power for samples of polyethylene blends with zeolite

When the results of the shock strength of low density linear polyethylene mixtures are comparing with the fillers used, we note that the addition ratios of 5% and 10% are mostly equal, and the difference is observed when the ratio of filler to polymer is increased. The shock strength values for the zeolite additive ratios at one level are between 24% - 25%, meaning that the mixture has the same resistance and that the minimum strength of the shock to zeolite was 30% for the zeolite in the mixture.



Fig(9). The relationship between the strength of the shock (%) and the addition ratios of zeolite

## 5- Environmental Cracking Resistance Test (ESCR)

The environmental resistance of PE samples with zeolite was measured by the Environmental Cracking System (ESCR) according to standard method (ASTM-1693) under special conditions at 50  $^{\circ}$  C with a concentrated liquid of glycol (10% Igepal). The results have showed that samples of LDPE mixtures with zeolite during this study their tolerance to environmental conditions as raw material within the manufacturing range and all mixing ratios did not break for 240 hours, which encourage the industrial use of these materials.

#### Conclusions

From what has been studying the effect of addition of zeolite to the low-density linear polyethylene polymer and tracking the results of the polyethylene tests, some tests were found to have positive and negative results where the positive ones were correspondent with the results of used polymer polyethylene. The best ratios of the study were ranged from 10% to 20% when the zeolite added to low density polyethylene. This encourages the use of natural fillers with any polymers other than polyethylene and multiple studies on various types of fillers.

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