

## THE EFFECT OF PROLONGED RELEASE MINERAL FERTILIZERS COATED WITH CO-POLYESTER FILMS FROM PET WASTE RECYCLING ON MAIZE PLANTS

Amelia ANGHEL<sup>1</sup>, Anca-Rovena LĂCĂTUȘU<sup>1</sup>, Radu LĂCĂTUȘU<sup>1,2</sup>, Stela IANCU<sup>3</sup>, Mihaela LUNGU<sup>1</sup>, Rodica LAZĂR<sup>1</sup>, Andrei VRÎNCEANU<sup>1</sup>

<sup>1</sup>National Research & Development Institute for Soil Science, Agrochemistry and Environment Protection, blvd. Mărăști 61, sector 1, 011464, Bucharest, Romania, E-mail: amelutu@yahoo.com

<sup>2</sup>“Al. I. Cuza” University, Bulevardul Carol I, Nr.11, 700506, Iasi, Romania, E-mail: radu.lacatusu@icpa.ro

<sup>3</sup>National Institute for Research & Development in Chemistry and Petrochemistry – ICECHIM, Spl. Independentei no. 202, sector 6, Bucuresti, 060021 Bucharest, Romania

**Corresponding author e-mail:** anca.lacatusu@gmail.com

### Abstract

The paper presents researches carried out in order to test complex mineral fertilizers coated with biodegradable co-polyester films using as raw material the polyethylene terephthalate (PET) waste, such as plastic bottles for food. Achieving of biodegradable coatings, based on polyethylene terephthalate, requires modifying the chemical structure of aromatic polyester (PET) with dicarboxylic acids (aromatic and / or aliphatic), and / or other polyols, to create a biodegradable capsule for prolonged release mineral fertilizers. In order to highlight capacity of complex mineral fertilizers coated with co-polyester films, to releasing nutrients in the soil in an extended time period, unlike common complex mineral fertilizers, a green house experiment was organized. Soil material used was collected from two contrasting soil types in terms of physico-chemical characteristics, namely Luvic Phaeozems, and Calcaric Fluvisols. The experimental variants were set up in vegetation pots of about 20 kg soils / pot capacity, in which fertilizer materials were introduced, namely co-polyester films coated complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ ) and regular granular complex mineral fertilizers. The crop was very early PR39D81 hybrid corn with excellent resistance to drought (hybrids belonging to the group FAO 200). For all measured phenological parameters: plant height, plant weight, root weight, cobs weight, grain weight and grain number, were recorded very significant increases in the values of the fertilized variants as compared to unfertilized, controls, variants of both soil types. The best results were obtained on Luvic Phaeozems soil where average size of maize plants at flowering stage was 29 cm higher in the variant fertilized with co polyester film coating complex mineral fertilizers, and 6cm higher in the variant fertilized with usual formula mineral fertilizers. Data obtained in this experiment showed that the effects generated by the use of complex mineral fertilizers coated with co polyester films in the proposed formula has been beneficial for the development of maize plants in a measure at least as high as in the classical variant of fertilization.

**Keywords:** co-polyester films, mineral fertilizers, phenological parameters, polyethylene terephthalate, slow-release, soil.

### INTRODUCTION

To avoid problems caused by dissolving of fertilizers in soil moisture at a speed higher than its absorption in the plant, many types of slow-release active substance fertilizers were designed. One method to obtain such products involves creation of "core / shell" type micro-capsule, where *shell* consists in a polymeric film with low water permeability and the *core* is the active substances. If the polymer used for encapsulation is biodegradable, fertilizers are released slowly as a result of polymeric film biodegradation [9].

The physicochemical flexibility of branched polyesters achieved through simple modification of the polymer systems, making them very usefully for the development of a variety of drug delivery vehicles in biomedical industry. The polymers' modular design is advantageous for the encapsulation of a wide range of drug compounds [6].

Aromatic polyesters such as polyethylene terephthalate, so-called PET shows excellent properties, which caused him to be marketed worldwide, often as packaging for liquids. However, until now, these polymers are considered resistant to microbial attack, so

there is biodegradable, which is a huge disadvantage [1, 7, 8]. Starting from a desire to meet both biodegradability and superior properties of plastic materials, tests were performed on biodegradability co polyester monomers containing both aliphatic and aromatic [15]. Biodegradability of a plastic is implying the possibilities of living organisms to use it as a food source, by transforming its chemical structure within a reasonable period of time. The organisms which having this metabolic ability are microorganisms. Primary (or partial) biodegradability is altering the chemical structure resulting in loss of specific properties of polymers while the final (or total) biodegradability is total mineralization and assimilation of the resulting material by microorganisms [3, 4, 5]. The material is fully degraded by microorganisms to produce carbon dioxide or methane, water, mineral salts and biomass [12, 2]. The time period involved is usually several weeks to several months.

To obtain biodegradable coating materials, from polyethylene terephthalate, was performed modifying of the chemical structure of aromatic polyester (PET) by including of dicarboxylic acids and / or other polyols, thus resulting a co polyester structure. This co-polyester presents molecular weight high enough to ensure formation of a flexible and resistant to moisture continuous coating film proper for producing encapsulated fertilizers; able to

provide controlled release of fertilizers, mainly by biodegradation, and which can being decayed to a convenient and measurable period of time under the influence of environmental factors.

Soil parameters such as humidity, temperature, pH, salinity, presence or absence of oxygen and nutrient supply level exerts a powerful effect on microbial degradation of polymers, so these conditions must be taken into account when testing biodegradability of polymers [11, 13, 14, 10].

## MATERIAL AND METHOD

In order to highlight capacity of complex mineral fertilizers coated with co-polyester films, to releasing nutrients in the soil in an extended time period, unlike common complex mineral fertilizers, a green house experiment was organized, using soil material collected from two contrasting soil types in terms of physical-chemical characteristics, especially argyle content, namely Luvic Phaeozems, and Calcaric Fluvisols.

Experiment carried out in greenhouse, in vegetation pots with capacity of 20 kg of soil material / pot, with 5 repetitions for each variant, with those two different types of fertilizers: regular complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ ), and complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ ) coated with co-polyesters films, according to following experimental scheme:

Treatment code	Soil type	Treatment
V1	Luvic Phaeozems	Control - unfertilized
V2		Regular complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ )
V3		Complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ ) coated with co-polyesters films
V4	Calcaric Fluvisols	Control - unfertilized
V5		Regular complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ )
V6		Complex mineral fertilizers ( $N_{15}P_{15}K_{15}$ ) coated with co-polyesters films

The crop was very early PR39D81 hybrid corn with excellent resistance to drought (hybrids belonging to the group FAO 200). At the final of the plants vegetation period (at harvest), all phenological parameters were

been measured: plants height. Plants weight, cobs weight, grains number/cobs, grains weight and roots weight. Data were statistically processed using standard analysis of variance (ANOVA), and differences

between experimental variants were considered according to Tukey test (5%) values.

## RESULT AND DISCUSSIONS

**The plants height** recorded significant differences between experimental variants, thus in both soil and fertilization types maize plants were been higher than control variants (Table 1, Fig. 1).

Whatever the type of fertilization, plant height of corn grown on Luvic Phaeozems was higher and statistically assured, than those grown on Calcaric Fluvisols. This result is normal because of natural properties of the first soil, well-known as more favorable for plants growing than those of the other soil type.

Table 1. Significance of biometrics data measured at maize plants in different fertilizing variants

Soil type	Fertilization	Plants height (cm)		Plants weight (g)		Cobs weight (g)		Grains number/cob		Grains weight (g)		Roots weight (g)	
		values	significance	values	significance	values	significance	values	significance	values	significance	values	significance
Luvic Phaeozems	Control - unfertilized	190	Ct	87.64	Ct	51.74	Ct	128	Ct	33.36	Ct	14.26	Ct
	Regular complex mineral fertilizers (N <sub>15</sub> P <sub>15</sub> K <sub>15</sub> )	227	***	86.5		101.5	***	375	***	78.43	***	22.55	
	Complex mineral fertilizers (N <sub>15</sub> P <sub>15</sub> K <sub>15</sub> ) coated with co-polyesters films	216	**	84.47		95.2	***	328	***	74.75	***	12.96	
Calcaric Fluvisols	Control - unfertilized	163	Ct	91.91	Ct	55.86	Ct	123	Ct	37.36	Ct	21.34	Ct
	Regular complex mineral fertilizers (N <sub>15</sub> P <sub>15</sub> K <sub>15</sub> )	202	***	77.86		83.68	**	279	***	62.18	**	18.41	
	Complex mineral fertilizers (N <sub>15</sub> P <sub>15</sub> K <sub>15</sub> ) coated with co-polyesters films	212	***	75.94		81.73	**	276	***	58.71	**	25.52	
	DL 5%	14.1		17.769		15.676		70.4		14.187		9.852	
	DL 1%	19.3		24.35		21.481		96.5		19.441		13.5	
	DL 0,1%	26.3		33.171		29.263		131.4		26.484		18.391	

### The plants height

On Luvic Phaeozems, the best results concerning the maize **plant height** were been obtained under fertilization with regular complex mineral fertilizer, while on Calcaric Fluvisols the higher plants have grown under fertilization with complex mineral fertilizers coated with co-polyesters films.

### The plants weight

Despite the fact that corn plants were shorter in fertilized variants, their weight was higher than that of plants grown in fertilized variants, in both types of soil or fertilization (Table 1, Fig. 2). Differences were more visible in the Calcaric Fluvisols case, while in Luvic Phaeozems values were been very closely.

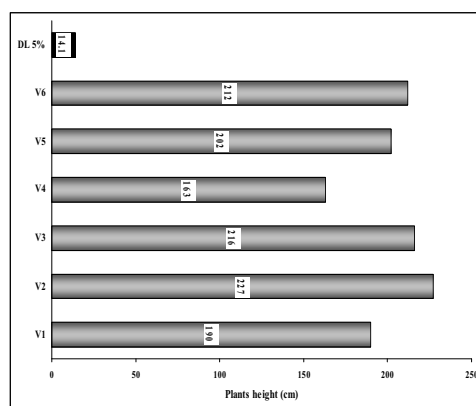


Fig. 1. Effect of fertilizer type on maize plants height

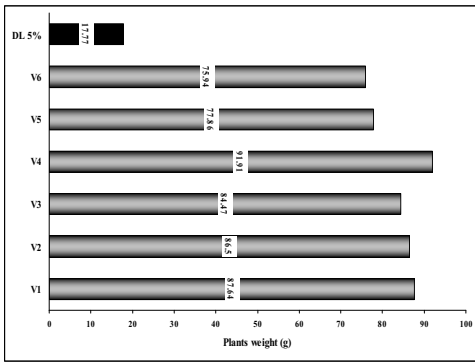


Fig. 2. Effect of fertilizer type on maize plants weight

### The corn cubs weight

Very suggestive are the differences between experimental variants, in terms of **weight of corn cubs** (table 1, fig. 3). Thus both type of mineral fertilizers applied have led to very significant increases in weight of corn cubs, also for plants cultivated on Calcaric Fluvisols and Luvic Phaeozems. The best result was recorded in Calcaric Fluvisols variant fertilized with regular complex mineral fertilizer.

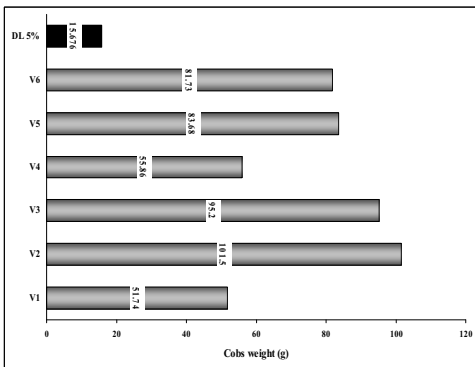


Fig. 3. Effect of fertilizer type on corn cobs weight

However, the two types of fertilizers, regular or coated with co polyester films have not resulted in significant differences between them, cobs weight values being relatively similar, for plants cultivated on the same soil type.

### The grain number / corn cub

Results of measurement of other biometric parameter, **the grain number / corn cub**, are very similar with those described already for corn cubs weight (Table 1, Fig. 4).

The great value for entire experiment was recorded in variant cultivated on Calcaric Fluvisols and fertilized with regular mineral fertilizer. For the same soil type, value of the grain number / corn cub was noticeably reduced, but without statistic significance.

Very significant are remaining the differences between fertilized variants for both soil types and also for both types of mineral fertilizers, regular or coated with co polyester films, and the control variants, without fertilization.

Were been recorded very close values of grain number / corn cub in both control variants.

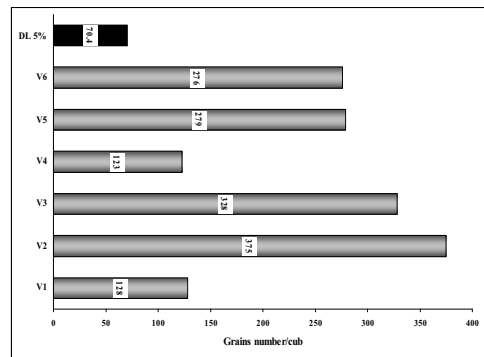


Fig. 4. Effect of fertilizer type on grains number/cubs

### The grain weight

Obviously, the measurement of this biometric parameter furnished results that led to the same pattern as data reported for the corn cubs' weight (Table 1, Fig. 5).

There are significant differences between the variants generated by soil type, the "harvest" recorded on Luvic Phaeozems being higher than that from Calcaric Fluvisols.

But, the most important differences were obtained between experimental variant with and without fertilization.

The higher parameter value was obtained in variant cultivated on Calcaric Fluvisols and fertilized with regular mineral fertilizer.

### The roots weight

Although the values obtained are not statistically different, the measurements showed differences between experimental variants (Table 1, Fig. 6).

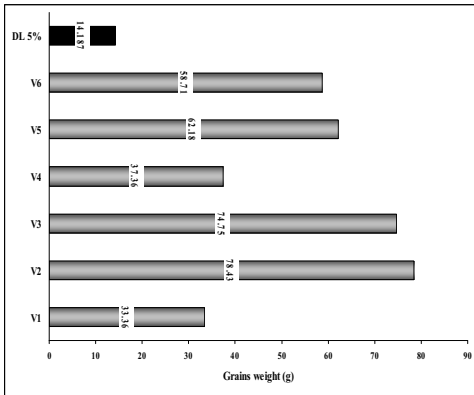


Fig. 5. Effect of fertilizer type on grains weight

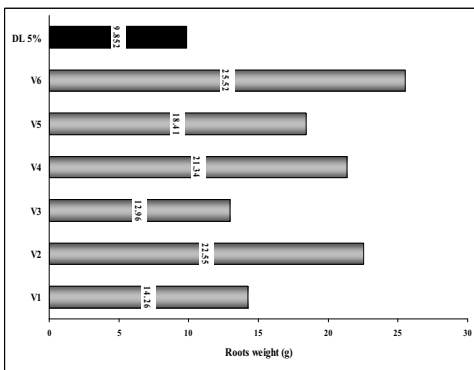


Fig. 6. Effect of fertilizer type on maize roots weight

In case of Luvic Phaeozems, the best development of maize roots have been recorded in the variant fertilized with regular complex mineral fertilizers, while in Calcaric Fluvisols the maize roots have been grown much better in experimental variant fertilized with complex mineral fertilizers coated with

co polyester films. This result may be due to the improvement of soil structure through the application of complex mineral fertilizers coated with co polyester films, so that, in case of such a clayed soil, to promote a better development of roots of cultivated plants.

Data presented in Table 2 clearly showing the beneficial effects of applying the complex mineral fertilizers with NPK, both in regular formula, and coated with co polyester films version.

For all biometric parameters analyzed: plant height, weight of each plant, roots weight, cubs weight, grain weight and grain number, very significant increases in the values of the fertilized variants as compared to unfertilized (controls) variants of both soil types, were recorded

The best results were obtained in variants organized on Luvic Phaeozems soil, where the average size of maize plants at the flowering stage, in the variant fertilized with complex mineral fertilizers coated with co polyester films was 29 cm higher than control, and 6 cm higher than in variant fertilized with complex mineral fertilizers in regular formula.

It can be stated that the data obtained in this experiment have shown that the effects resulting from the use of complex mineral fertilizers coated with co polyester films in formula proposed in this research has been beneficial for development of the plants in a measure at least as good as the conventional fertilization.

Table 2. Significance of fertilization type on biometric parameters of maize plants in different fertilizing variants

Fertilization	Plants height (cm)		Plants weight (g)		Cobs weight (g)		Grains number/cob		Grains weight (g)		Roots weight (g)	
	values	significance	values	significance	values	significance	values	significance	values	significance	values	significance
Control - unfertilized	177	Ct	89.77	Ct	53.8	Ct	125	Ct	35.36	Ct	17.8	Ct
Regular complex mineral fertilizers (N <sub>15</sub> P <sub>15</sub> K <sub>15</sub> )	215	***	82.18		92.59	***	327	***	70.31	***	20.48	
Complex mineral fertilizers (N <sub>15</sub> P <sub>15</sub> K <sub>15</sub> ) coated with co-polyesters films	214	***	80.2		88.47	***	302	***	66.73	***	19.24	
DL 5%	9.9		12.565		11.085		49.8		10.032		6.966	
DL 1%	13.6		17.218		15.189		96.5		13.747		9.546	
DL 0,1%	18.6		23.455		20.692		131.4		18.727		13.005	

## CONCLUSIONS

Experimental data clearly showing the beneficial effects of applying the complex mineral fertilizers with NPK, both in regular formula, and coated with co polyester films version.

For all biometric parameters analyzed: plant height, weight of each plant, roots weight, cubs weight, grain weight and grain number, very significant increases in the values of the fertilized variants as compared to unfertilized (controls) variants of both soil types, were recorded.

The effects resulting from the use of complex mineral fertilizers coated with co polyester films in formula proposed in this research has been beneficial for development of the plants in a measure at least as good as the conventional fertilization

Use of complex mineral fertilizers coated with co polyester films in formula proposed in this research did not induced toxicity aspects for maize plants.

Our research highlighted that the use of PET waste for production of co polyester films with applicability in production of complex mineral fertilizers, in prolonged release formulation, is a real and extremely useful possibility for recycling of this enormous waste accumulated in the environment.

## REFERENCES

- [1] Aminabhavi, T.M., Balandgi, R.H., Cassidy, P.E., 1990. *A review on biodegradable plastics*. Polym. Plast. Technol. Eng. 29 (3), 235-262.
- [2] Augusta, J., Muller, R.-J., Widdecke, H., 1993. *A rapid evaluation plate-test for the biodegradability of plastics*. Appl. Microbiol. Biotechnology. 39, 6733-678.
- [3] Battersby, N.S., Fieldwick, P.A., Ablitt, T., Lee, S.A., Moys, G.R., 1994. *The interpretation of CEC L-33-T-2 biodegradability data*. Chemosphere. 28, 787-800.
- [4] Buchanan, C.M., Gardner, R.M., Komarek, R.J. 1993. *Aerobic biodegradation of cellulose acetate*. J. Appl. Polym. Sci. 47, 1709-1719.

- [5] Calmon, A., Guillaume, S., Bellon-Maurel, V., Feuilloley, P., and Silvestre, F., 1999. *Evaluation of material biodegradability in real conditions – Development of a burial test and analysis methodology based on numerical vision*. J. Environ. Polym. Degrad. 7 (3), 157-166.
- [6] Dailey Lea Ann, Wittmar, M., Kissel, T., 2005. *The role of branched polyesters and their modifications in the development of modern drug delivery vehicles*, Journal of Controlled Release 101, 137-149
- [7] Huang, S.J. 1989. *Biodegradation*, p.597-606. In G. Allen, J.C. Bevington (ed), *Comprehensive Polymer Science: The Synthesis, Characterization, Reactions and Applications of Polymers*, vol. 6, Pergamon Press, Oxford.
- [8] Kawai, F. 1995. *Breakdown of plastics and polymers by microorganisms*. Adv. Biochem. Eng. 52, 151-194.
- [9] Lacatusu Anca-Rovena, Lacatusu, R., Dumitru, M., Iancu Stela, Lungu Mihaela, Balaceanu Claudia, Lazar Rodica, Anghel Amelia, Vrinceanu, A., 2010. *The Effect Of Complex Mineral Fertilizers Coated With Co-Polyester Films From Pet Waste Recycling On Soil Chemical Properties*, The Proceedings of the 15th World Fertilizers Congress of the International Scientific Center for Fertilizers (CIEC), 29 August-2 September 2010, Bucharest Romania, Ed Romanpan Academy, ISBN 978-973-27-2043-1; p 642-654.
- [10] McCartin, S.M., Press, B., Eberiel, D., McCarthy, S.P. 1990. *Simulated landfill study on the Accelerated biodegradability of plastics materials*. Am. Chem. Soc., Polymer Preprints. 31(1), 439-440.
- [11] Ohtaki, A., Sato, N., Nakasaki, K. 1998. *Biodegradation of poly( $\epsilon$ -caprolactone) under controlled composting conditions*. Polym. Degrad. Stab.61(3), 499-505.
- [12] Palmisano, A.C., Pettigrew, C.A. 1992. *Biodegradability of plastics*. Bioscience. 42(9), 680-685.
- [13] Tosin, M., Degli-Innocenti, F., and Bastioli, C. 1996. *Effect of the composting substrate on biodegradation of solid materials under controlled composting conditions*. J. Environm. Polym. Degrad. 4(1), 55-63.
- [14] Tuominen, J., Kylmä, J., Kapanen, A., Vanelampi, O., Itavaara, M., Seppälä, J. 2002. *Biodegradation of lactic acid based polymers under controlled coposting conditions and evaluation on the ecotoxicological impact*. Biomacromolecules, 3(3), 445-455.
- [15] Witt, U., Eining, T., Yamamoto, M., Kleeberg, I., Deckwer, W.-D., Müller, R.-J. 2001. *Biodegradation of aliphatic-aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates*. Chemosphere 44.