

## LONG-TERM STRENGTH OF POLYMER BLENDS FROM RECYCLED MATERIALS\*

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**ABSTRACT.** This report presents data on the long-term strength of five composites made of plastic waste. They contain low density polyethylene, high density polyethylene, polypropylene and polystyrene (LDPE, HDPE, PP and PS). Long-term strength is determined experimentally by tensile creep to fracture. The experimentally determined long-term strength is compared to predictions for its probabilistic boundaries. The calculation method of these predictions uses data from short-term experiments. The calculated predictions are true for four compositions which exhibit ductile fracture. The composite containing 50 wt.% PS has the greatest strength (of the tested specimens) and has brittle fracture. Its calculated estimate of long-term strength is not consistent with the experimental one.

**KEY WORDS:** long-term strength, creep, rupture, prediction.

### 1. Introduction

Polymeric materials gain a growing segment on the market for functional and engineering applications. The increasing use of composite materials and polymer blends can be seen in a wide range of applications demanding lifetimes of 15 to 50 years. However, the mechanical properties of these composites have a time dependent nature, i.e. strength and stiffness are time-dependent due to the hereditary nature (viscoelasticity) of polymers. Investigations on long-term strength (LTS) of polymers are very important, because they contribute to meeting the demands of customer interests for better understanding and predicting their long-term behaviour in practical applications [1]. To avoid

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failures due to unexpected strength loss after long periods of time, it is imperative to develop accelerated tests for simulating the damages that might occur over long periods of time, [2]. In this context, damage and fracture mechanics based models and methods for prediction of long-term strength of viscoelastic materials are developed, to predict the lifetime of materials under a special case of creep to rupture under constant load.

From economic and ecological point of view, blended recycled materials can be also used for different applications but it is necessary to acquire knowledge on long-term behaviour of the obtained material. In this case, the situation is more complicated because recycled polymers possess their own “pre-history” that can be thermo-mechanical or/and thermo-oxidative which defines the possibility for reusing. The statistic data for polymer waste show that the most essential volumetric part is that of polyethylene and polypropylene and hence, the focus in the research studies is supposed to be on these polymers [3, 4]. Usually, these plastics are mixed within the polymer waste and their utilization together as blends is a good solution from ecological point of view.

This study is focused on the long-term behaviour of five polymer blends made of recycled plastics. The LTS limits were predicted fast by the known method [5, 6, 7]. Thereafter, the long-term strength of the same blends were determined experimentally through experiments of creep to rupture under tension.

## 2. A method for predicting the material long-term strength

Using this method [5, 6, 7], a zone is predicted (calculated), in which experimental points of long-term strength, i.e. of time to rupture in creep under constant load, are likely to be. The boundaries of this zone are calculated on the basis of data obtained from short-term (static) experiments and the corresponding mathematical apparatus. The mechanical tests are carried with two or three velocities. The following parameters are obtained from the  $\sigma - \varepsilon$  diagrams:

- Initial modulus of elasticity  $E$  [MPa], respectively  $M$  [MPa/%];
- Strength (for brittle materials) or yield strength (for ductile materials). It is stress at a point “ $M$ ”,  $\sigma_M$  [MPa], where the stress has a maximum;
- The “elastic” strain,  $\varepsilon_e = \sigma_M/E$ ;
- Factor  $K^*$ , which characterizes the presence of inelastic deformations. The way for its determination is described in [6, 7];
- Average speed of elastic deformation  $\varepsilon_e/t_M$ , where  $t_M$  [s] is time from the beginning until reaching the maximum stress.

The dependence of the strength  $\sigma_M$  on the speed is approximated by Eq. (1).

$$(1) \quad \sigma_M = A_K + B_K \ln(\varepsilon_e/t_M).$$

The regression coefficients  $A_K$ ,  $B_K$  and tripled maximum standard deviation  $\Delta$  are used to calculate the coefficients of the prediction:

$$(2) \quad A_L^+ = A_K + B_K \ln(B_K/M) + \Delta,$$

$$(3) \quad A_L^- = A_K + B_K \ln(B_K/M) - \Delta,$$

$$(4) \quad B_L = B_K/K^*.$$

For accurate calculations using Eqs (2) and (3), dimension of the module  $M$  must be MPa/%, if “MPa” and “%” have been used in Eq. (1), during the linear regression. This method for prediction of the area, where long-term strength might fall within, has two variants: According to “stripe prediction”, long-term strength will probably be within the area enclosed by graphs of Eqs. (5) and (6); According to “fork prediction”, long-term strength will probably be within the area enclosed by Eqs. (5) and (7).

$$(5) \quad \sigma_L^+ = A_L^+ - B_L \ln(t_L),$$

$$(6) \quad \sigma_L^- = A_L^- - B_L \ln(t_L),$$

$$(7) \quad \sigma_L^- = A_L^- - B_K \ln(t_L),$$

where  $t_L$  is time to rupture for creep under constant load  $\sigma_L$ .

### 3. Experimental

#### 3.1. Materials and specimens

The samples are made from blends containing recycled plastics: low-density polyethylene (LDPE), high-density polyethylene (HDPE) and polypropylene (PP). The base polyolefin blend consisting of LDPE, HDPE and PP in proportion 1:1:1 was mixed with 10, 15, 25 and 50 wt.% of virgin polystyrene. These formulations, denoted as PO, POPS10, POPS15, POPS25 and POPS50, were melt compounded in a Brabender twin screw extruder.

The samples are dumb-bell shaped for tensile tests, type 1BA (BDS EN ISO 527-2: 2012). The initial distance between the clamping grip edges is 58 mm. The specimen cross-section is about 1x5 mm. Ten specimens were tested at each velocity in short-term tests and up to 4 specimens had crept under chosen stresses (26, 23, 20, 17 MPa and others).

### 3.2. Equipment

For application of the method described above, mechanical tests at few velocities have to be performed. These, short-term experiments, were conducted on a TIRA TEST 2300 test machine, at preset velocities 0.1, 1 and 10 mm/min. The tensile elongation was evaluated as change of the distance between clamping grips, taking into account deformation of the loaded machine parts. The method for data processing is described briefly in [8]. Force - time dependences were recorded using a PROTEK D470 digital multimeter.

Long-term experiments of creep under constant load, in tension, were carried out on lever devices with 1 kN load capacity. Deformation - time dependence was recorded by laboratory extensometers with glued wire strain gauges, and a HBM UPM 60 amplifier.

## 4. Results and discussion

Data for mechanical properties are required, for application of aforementioned method. They are obtained through static (short-term) experiments with two or three speeds.

### 4.1. Short-term (static) strength and data

At loading of 1 mm/min, the PO, POPS10 and POPS15 blends exhibit plastic fracture, the POPS50 blend – brittle rupture and the POPS25 blend is with intermediate behaviour, Fig. 1.

The short-term (static) strength of the studied materials depends on the speed of deformation. Figure 2 shows this dependence for the PO blend. The three groups of points correspond to the three testing speeds. These experimental dependences have been fit by regression Eq. (1), the middle line in Fig. 2, for the five tested blends.

Table 1 shows the regression coefficients ( $A_K$ ,  $B_K$ ) for each blend. The strength has a standard deviation for each testing velocity.  $\Delta$  is the tripled maximum standard deviation. The results were calculated with data from two test speeds: 0.1 mm/min and 1 mm/min. At the speed of 10 mm/min, the step of force recording is  $\sim 10\%$  of the maximum force, which leads to inaccurate recording of the maximum force. Therefore, data obtained at 10 mm/min were not used in calculating the estimate for the long-term strength boundaries.

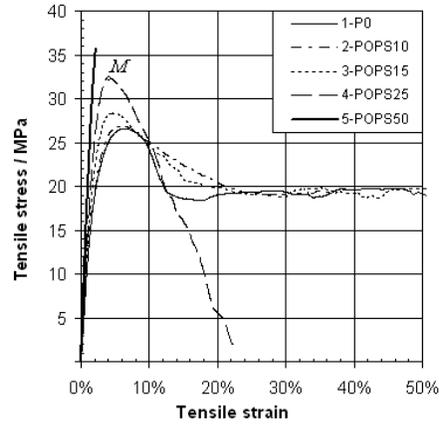


Fig. 1. Stress-strain diagrams of tested blends at 1 mm/min

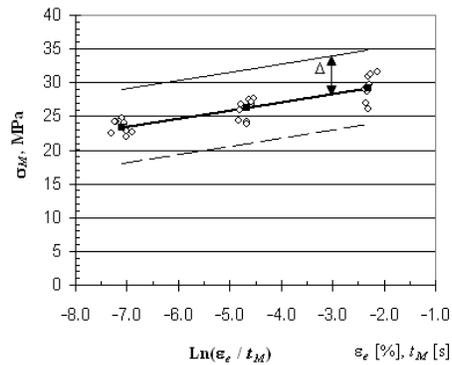


Fig. 2. Short-term (static) strength of the PO blend

Table 1 also contains data for values of coefficient  $K^*$  and Young's modulus  $M$ , assessed at speed of 1 mm/min. All this data is necessary for the calculations according to formulas (2), (3) and (4).

From Table 1 is visible, the blend PO has lower strength and is softest, but blend POPS50 has highest strength and is stiffest.

Table 1. Results from short-term tests, which were used for calculating the predicted boundaries of the long-term strength.

	Property	PO	POPS10	POPS15	POPS25	POPS50	Dimension
1 mm/min	$\sigma_M$	26.1	26.8	28.7	32.6	35.2	MPa
	$K^*$	1.82	1.85	1.64	1.48	1.22	–
	$M$	12.4	14.0	15.4	17.0	20.0	MPa/%
	$A_K$	31.0	36.7	37.5	42.1	36.1	MPa
	$B_K$	1.06	2.11	1.99	2.20	0.22	MPa
	$\Delta$	4.3	2.5	1.9	5.7	9.8	MPa

#### 4.2. Experimental and predicted long-term (creep-rupture) strength

For blend POPS25, the limits, which were predicted by Eqs. (5) and (6), fit to experimental long-term strength the best, Fig. 3. For blends POPS10 and POPS15, predicted stripe limits fit well to experimental LTS, but experimental points lie closer to the lower limit line and there is one experimental point which lies below the lower stripe line. For blend PO, the experimental LTS has slope which corresponds to the slope of lower fork limit, Eq. (7), and it is in the boundaries of the predicted fork limits, Fig. 4. For blend POPS50, the slope of predicted limits is too small and does not correspond to the slope of experimental LTS, Fig. 5.

All experimental LTS' have been compared in Fig. 6. It is visible, the

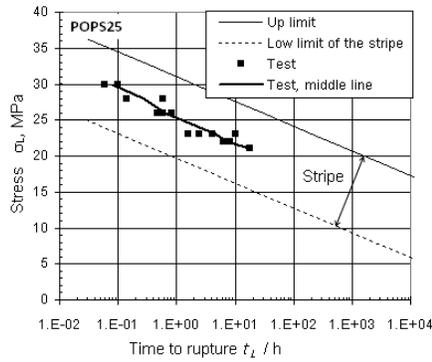


Fig. 3. Long-term strength of blend POPS25. Experimental one and predicted stripe limits

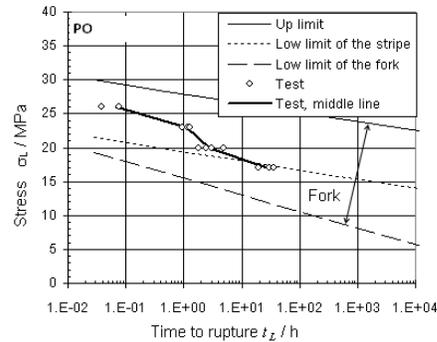


Fig. 4. Long-term strength of blend PO. Experimental one and predicted fork limits

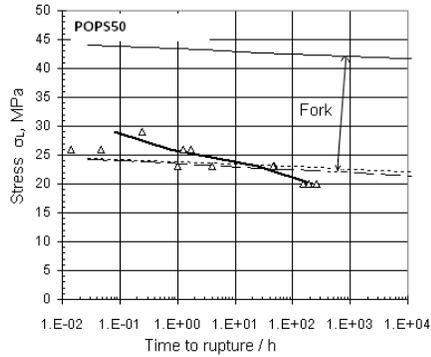


Fig. 5. Long-term strength of blend POPS50. Experimental one and predicted limits

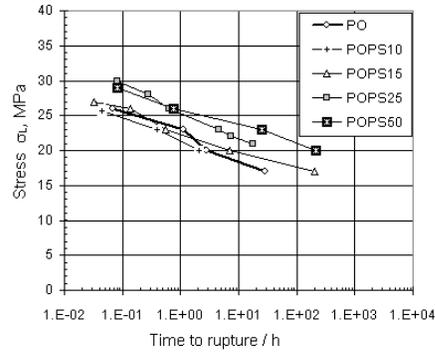


Fig. 6. Experimental long-term strength of all tested blends

blends PO and POPS10 possess smaller LTS than blends POPS25 and POPS50, but all graphs have relatively equal slope.

## 5. Conclusions

Short-term (static) mechanical properties and long-term strength of five composites made of waste plastics were studied. The PO blend has the lowest tensile strength and is most ductile. The POPS50 blend has the biggest strength and is stiff, with brittle fracture. According to the experimentally obtained diagrams of long-term strength for the five studied materials, the POPS10 and PO blends have the lowest long-term strength, while the POPS25 and POPS50 blends have the greatest long-term strength. The experimentally determined long-term strength of the POPS 10, POPS15 and POPS25 blends is within the predicted stripe. The experimental long-term strength is within the wider predictive fork, for the PO composite.

The POPS50 blend exhibits the highest static strength and the greatest long-term strength, but it has also the largest dispersion of sizes and results. The calculated estimation for the boundaries of its long-term strength does not cover the experimental one. This discrepancy is probably due to an inadequate method of workmanship of sample preparation and consequently their dimensions and mechanical properties have shown abnormally large dispersion. Another possible explanation of this result is that the method used for prediction is applicable for ductile materials, but is not suitable for brittle materials as POPS50.

## REFERENCES

- [1] DUDIĆ, D., D. KOSTOSKI, V. DJOKOVIĆ, Z. STOJANOVIĆ. Recrystallization Processes induced by Accelerated Ageing in Isotactic Polypropylene of Different Morphologies (PP). *Polymer Degradation and Stability*, **67** (2000), 233–237.
- [2] REEDER, J. R. Prediction of Long-Term Strength of Thermoplastic Composites Using Time-Temperature Superposition, Report NASA/TM-2002-211781, Virginia, Hampton, Langley Research Center.
- [3] GALLI, P., G. VECELLIO. Polyolefins: The Most Promising Large-Volume Materials for the 21st Century. *J. Appl. Polym. Sci.: Part A: Polym. Chem.*, **42** (2004), 396–415.
- [4] DJOUMALISKY, S., T. DOBREVA, T. RADEVA, E. KRUSTEVA, G. KOTZEV, I. BOROVANSKA, R. KRASTEVA, S. DJUNOVA. Recycling of Mixed Plastic Waste from Households, Proc. of 12<sup>th</sup> Int. Conf. on the Mech. and Tech. of Composite Mater., Varna, 2009, 177–182.
- [5] ZACHARIEV, G. Vorhersage der Lebensdauer und der Zeitstandfestigkeit von Verbundwerkstoff CF/PPS im Retardations-Zugversuch aufgrund kurzzeitiger Zugversuche. *Forschung im Ingenieurwesen*, **62** (1996), No. 3, 42–46, (in German).
- [6] ZACHARIEV, G., H.-V. RUDOLPH, R. KRASTEVA. Prediction of the Lifetime of a Bending Loaded GF/POM Composite in Retardation. In: Proc. of 9th Int. Conf. on Mechanics and Technology of Composite Mater., Sofia, CLPHCHM-BAS, 2000, 432–435.
- [7] KRASTEVA, R. K., G. ZACHARIEV, J. HRISTOVA, J. MINSTER. Creep-rupture Strength Prediction of an Epoxy Composite under Tension. *Mech Time-Depend Mater*, **13** (2009), 207–214.
- [8] KRASTEVA, R. K. A Contemporary Application of the Archimedes' Method, In: Proc. of 12th Int. Conf. on Mechanics and Technology of Composite Materials, Bulgaria, Varna, ed. N. Barovsky, CLPCM-BAS, ISSN 1313-8693, 2009, 409–414.