Research report:

Investigating recyclability: Reprocessability of aluminium coated polypropylene films

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Introduction

Plastic films are used in a wide variety of applications, for example in packaging, in the construction sector or in electronics applications. In case of labels, these films are often modified, for example by coating or printing, to give them additional functionality. Here, however, the question arises whether such films can then be recycled, i.e. whether they can be reprocessed as post-industrial material (i.e. as production waste such as edge trimmings) in such a way that these films can be recycled again into a recyclate or into films of sufficient quality.

Therefore, the objective of this work was to investigate the reprocessability of aluminium-coated and topcoated polypropylene films. To investigate this, recycled material was again extruded into films and the properties of these films were investigated. The summarized results can be found on the following pages.

This work was carried out at TCKT by Roman Wurz as part of a master's thesis project in the period from October to December 2021 and was supported by HUECK FOLIEN GmbH financially and by providing the films.

Experimental

HUECK FOLIEN GmbH provided three different film variants for testing the reprocessability. These were an uncoated PP film, one coated with aluminium and one in which the aluminium coating was additionally provided with a printable topcoat.

These films were first reduced to approximately 5 mm flakes by means of a shredder (slow-running granulator). These flakes were then processed into pellets by means of a laboratory compounder (co-rotating twin-screw extruder) in a temperature range of about 215 °C. In a further step, the pellets were then processed in a slow-running granulator. In a further step, a stabilizer (metal deactivator) was added to some of the materials containing aluminium in order to prevent any acceleration of the degradation of the polymer by the aluminium during the subsequent heat storage.

From the granules thus prepared (PP, PP with aluminium and PP with aluminium and topcoat, the last two with and without metal deactivator MD), films with a thickness of about 70 µm were extruded at about 240 °C using a flat film line.

Strips were cut from these films and characterized by means of film tensile test (ISO 527-3) and tear propagation test before and after aging (5 test specimens each). The films were also evaluated optically and the color change was measured using a colorimeter (Lab color space).

The films were aged for different times at 100°C in a convection oven. The selected times (0, 7, 21 and 35 d) correspond to more than 10 years of storage at room temperature (estimated using the Arrhenius approach from the temperature difference between room and storage temperature), so that any effects on the longevity of such films can also be estimated here.

Results

In the following, selected results from the investigations of the reprocessed films are presented in a compact form. Figure 1 shows the results of the mechanical characterization. Based on the values of the unstored films (0 d), it can be seen that the presence of aluminium or aluminium and a topcoat on the films has no significant effect on the yield strength of the films. This is probably also due to the small amount of aluminium present on the film before reprocessing - the coating has about 40 nm on a PP film of about 50 µm thickness.

Storage increases the yield strength initially (after 7 d) and then it remains at a level. Based on our experience, the increase is probably due to post-crystallization. The addition of a stabilizer was not necessary in this case, since the presence of aluminium shows no effect on the aging behavior of the different films. The deviations are all within the scatter range of the tests.

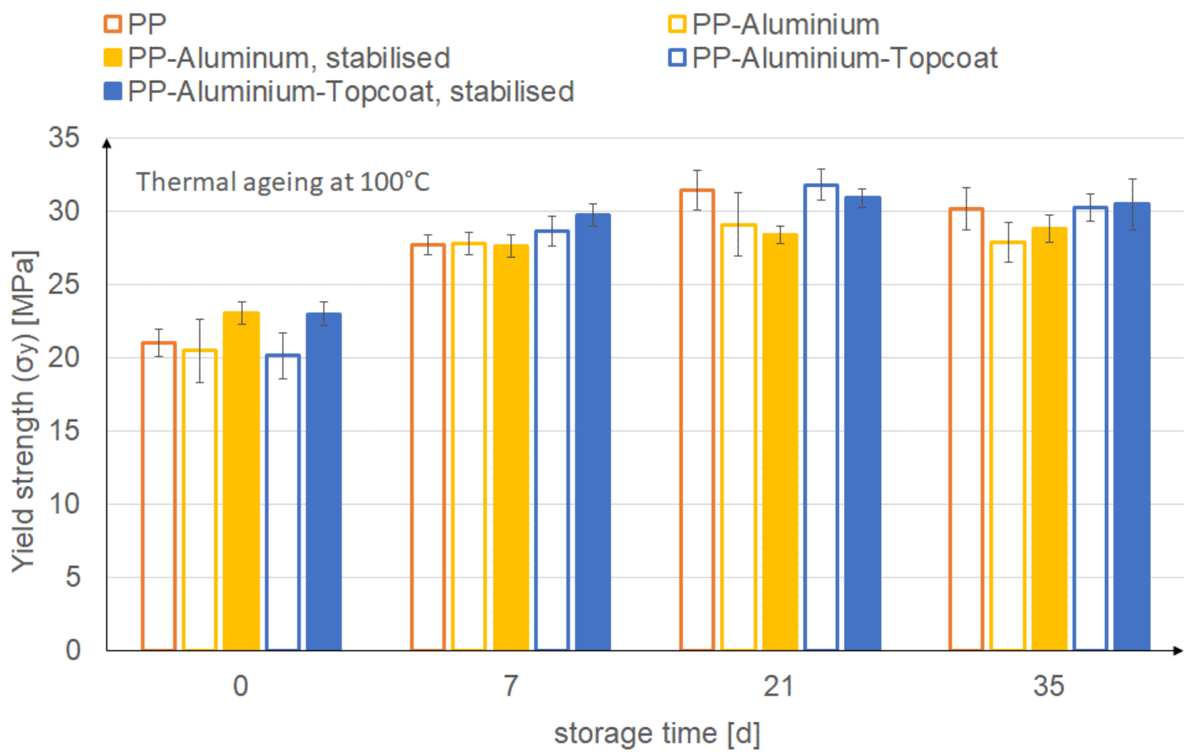

Figure 1: Yield strength of the different film formulations in the initial state (0 d) and after different storage times in the convection oven at 100°C

Comparable effects can be observed for the elongation at stretch (Fig. 2). The difference between the films is small, and after an initial increase in elongation during storage, the values remain approximately at the same level. What is noticeable here is that the results show much wider spreads compared to the yield strength. This is most likely due to the specks (Fig. 3) present in the material, as these represent weak points during the test, and at these points the elongation is then limited by these weak points.

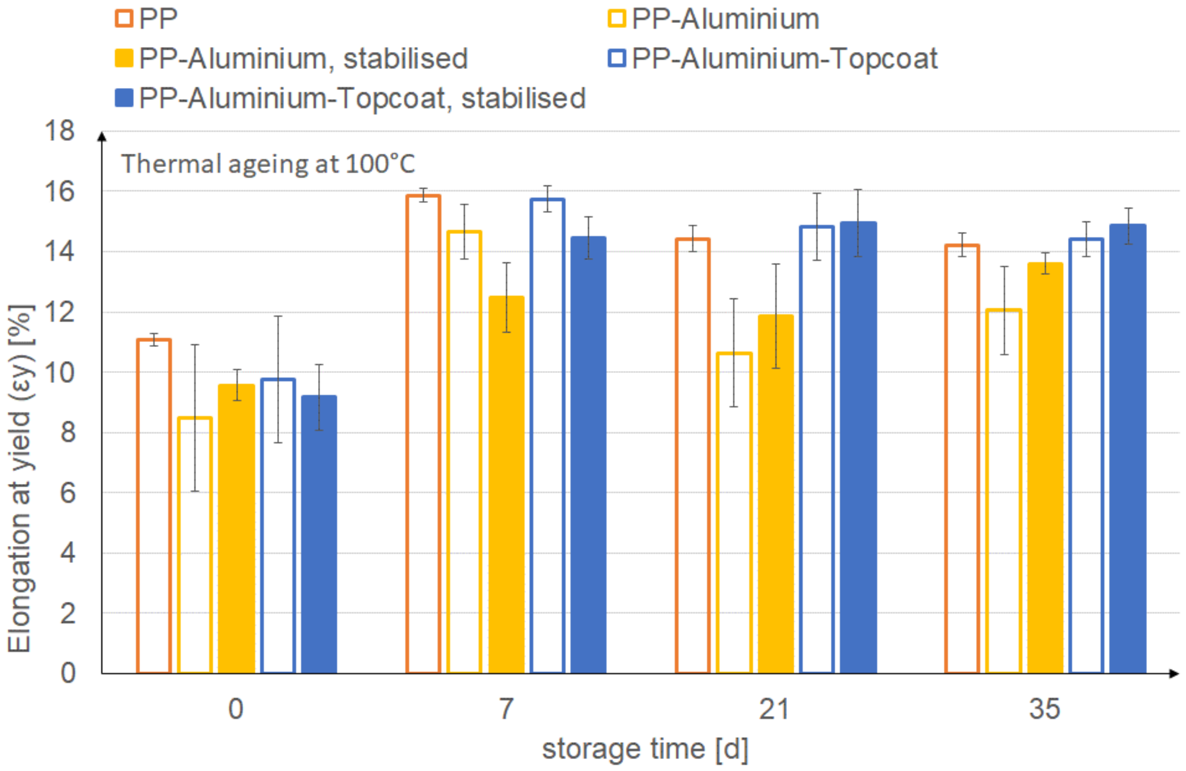

Figure 2: Elongation at yield of the different film formulations in the initial state (0 d) and after different storage times in the convection oven at 100°C

The optical quality of the films is shown in Figure 3. It can be seen that the presence of aluminium here causes the appearance of larger specks. These are most likely due to the process used in these studies, as the low bulk density makes it difficult to feed the films into the twin screw extruder, and therefore the ratio between material feed and shear is not optimal. No melt filter was used in this process either, which would certainly have improved the quality - at least to a certain extent - but the setup was chosen to be as simple as possible in order to determine any influences of the aluminium coating on recycling.

The specks in the films containing aluminium are all about the same in size (up to max. 0.7 mm) and number, i.e. the presence of a topcoat or the addition of the stabilizer does not influence the speck formation here.

Figure 3: Optical appearance of the different film formulations in the initial state (transmitted light images on the light table)

Another mechanical parameter for the quality of films is the tear resistance. This is shown in Figure 4 and shows in the initial state that those films which contain aluminium have somewhat lower values than those for the film without aluminium. However, it must be taken into account here that the values in this test naturally show a higher scatter, which is why the breakage patterns must always be evaluated in the same way. These (Fig. 5) show no significant differences between the materials for the same storage period. At the beginning, the crack is wavy, which suggests a higher energy absorption, which is also seen in the numerical values. Through storage (and the crystallization effect that occurs), the crack becomes smoother, causing the values to be lower, but also causing the differences between the various formulations to disappear.

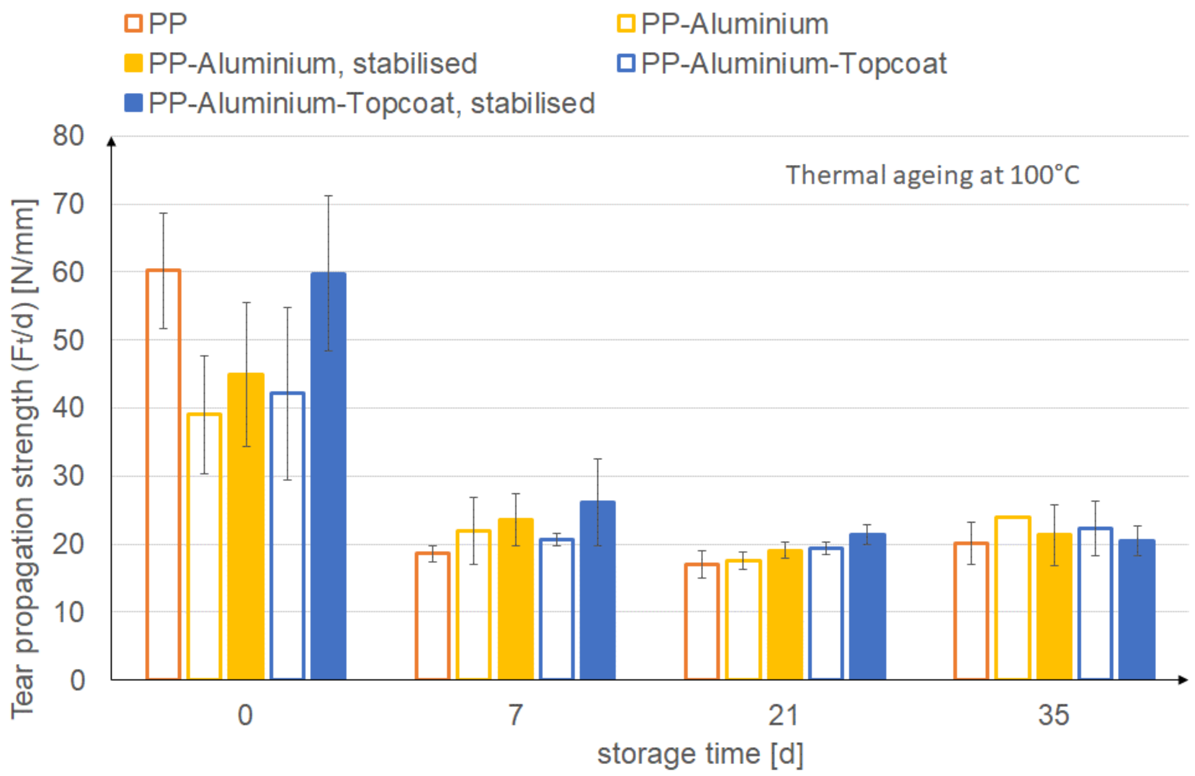

Figure 4: Tear propagation strength of the different film formulations in the initial state (0 d) and after different storage times in the convection oven at 100°C

Figure 5: Specimens after tear propagation testing of the different film formulations in the initial condition (0 d) and after different storage times in the convection oven at 100°C (arrow marks the beginning & direction of the tear).

In addition, the change in the optical appearance of the samples was also investigated by means of color measurement (Lab color space). In fact, there was no difference in the color values over the storage period, e.g. in the b-value (color axis blue-yellow, can be used simplified as a measure of yellowing) the change is less than 1. The difference that is most noticeable is that in the opacity (the opposite of transparency, Fig. 6), which is considerably lower in the films without aluminium than in those with aluminium coating. Over the storage period, however, the changes are again negligible.

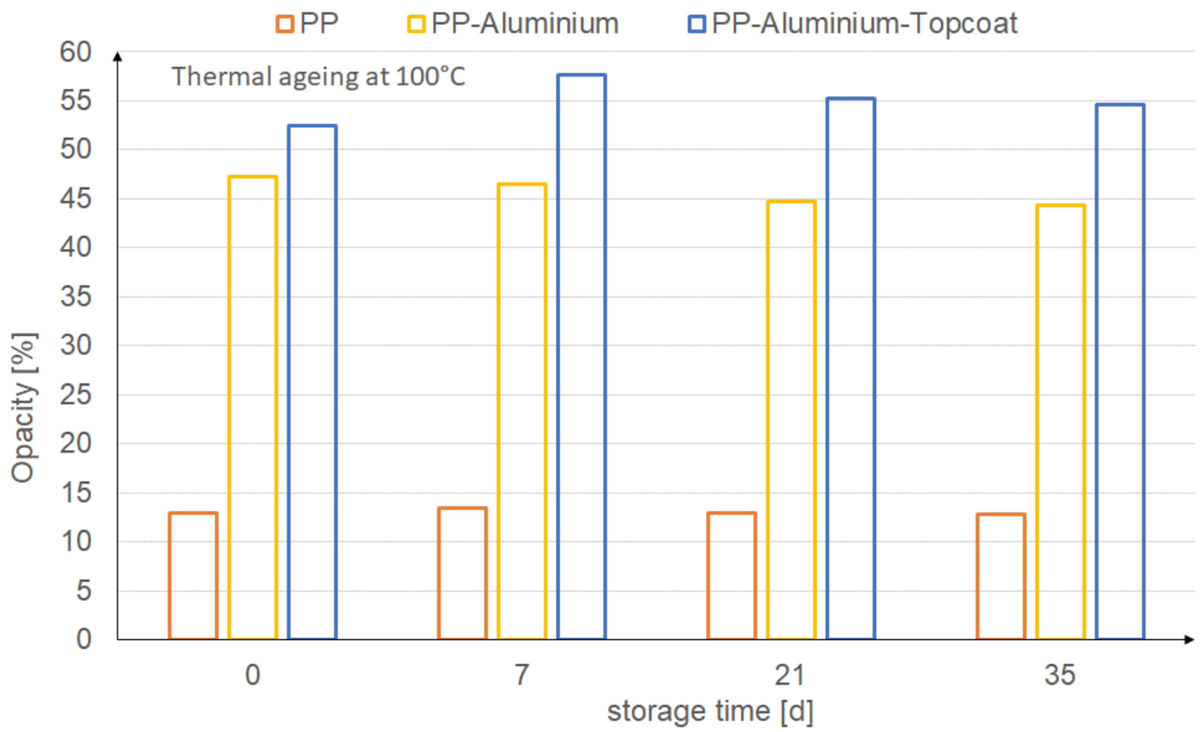

Figure 6: Opacity of the different film formulations without stabilizers in the initial state (0 d) and after different storage times in the convection oven at 100°C

A possible remedy for the discoloration would be to oxidize the aluminium on the surface, for example with sodium hypochlorite. If this is applied to the foil, then the aluminium is oxidized, which also causes the metallic color to disappear, as shown in Figure 7 on the basis of an immersion test with the foils in the as-received condition.

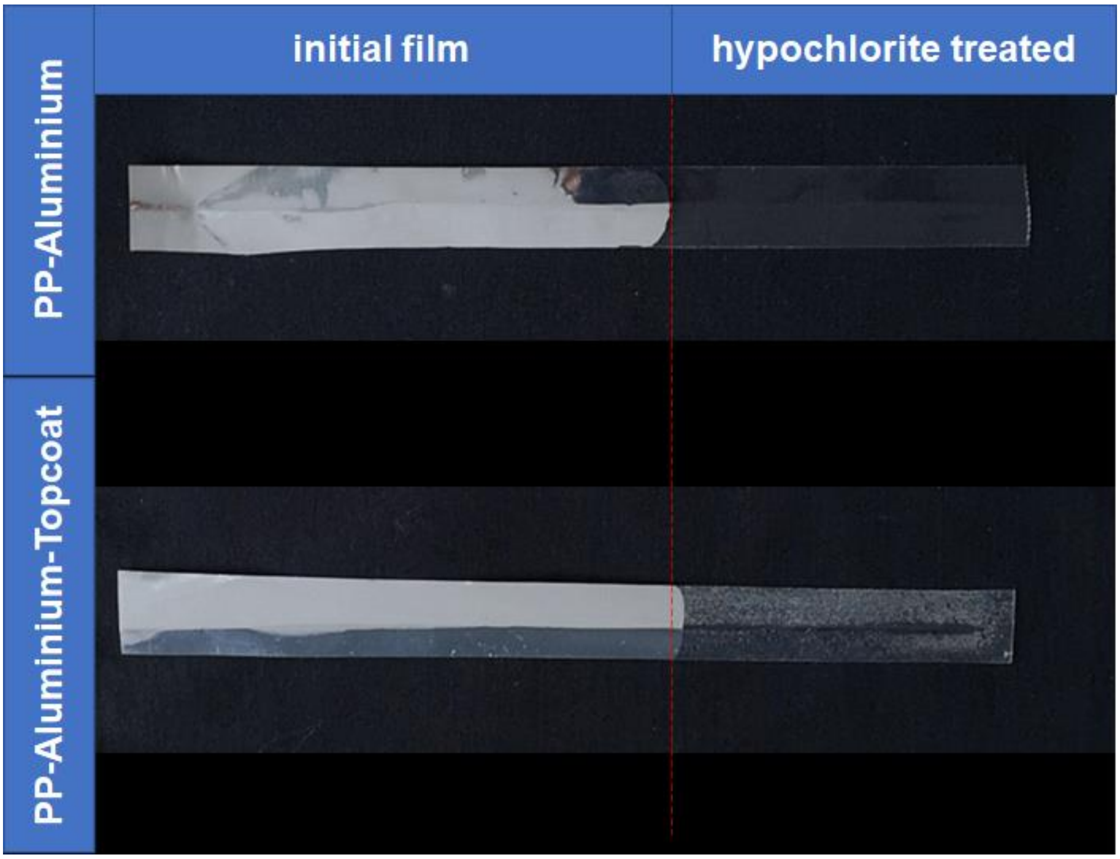

Figure 7: Aluminium-coated films (as supplied) after oxidation by immersion in sodium hypochlorite solution at room temperature.

Conclusions

This work dealt with the question of whether aluminium- and top-coated films can be materially recycled by melting and regranulating them. The results presented here show that such films can be recycled, since the tests of the mechanical properties showed no negative influences in comparison with uncoated PP films. Likewise, thermal storage to evaluate long-term stability showed no significant differences here. The only problematic aspect is the formation of specks, which in the context of this study was certainly also promoted by the process used. Here, it can be assumed that the number and size of flakes can be significantly reduced by a suitable film feeding unit and melt filtration.

In addition, a preliminary test also showed that with the application of an oxidizing agent as a pretreatment, the aluminium is oxidized, and thus the discoloration of the films in the recycling process can be prevented. However, scalability of such processes must be investigated in detail before application.