SHORT COMMUNICATIONS

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Study of deformation process of stored polymethacrylate free films

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In the present study the effects of the additives and of storage on the deformation of films prepared from methacrylic acid/ethyl acrylate copolymer with and without polysorbate 80 were evaluated. The films containing polysorbate 80 revealed a longer deformation process with a different deformation curve shape and higher force and work of deformation. Storage induced very significant changes in the characteristics of the films. The decrease in the deformation work was more relevant than the reduction in the deformation force. Increase of the storage time increased the changes in these parameters. The presence of polysorbate 80 in the films reduced the change in behaviour of the films at lower relative humidity.

Different types of acrylic polymers are widely used in pharmaceutical technology (Lehmann 1999). Various additives are applied in coating fluids containing acrylic derivatives. Film formation from an aqueous polymeric dispersion of acrylic polymer is a complex matter. Additives can change this process and hence the properties of the films formed (Felton and McGinity 2002). The widely used additives include plasticizers, which decrease the film-forming temperature and the temperature of the glass transition and increase the mechanical properties of the films formed (Zelkó et al 2002; Lippold and Pagés 2001). The mechanical property of films can be examined by several methods (Siew et al. 2000; Schultz et al. 1997). Most studies use the deformation force measured at the deformation point, the tensile strength or the elasticity to examine the mechanical properties of films (Crowley et al. 2004; Repka and McGinity 2000). Certain properties of films are less accessible with these methods.

The changes in the properties of films during storage (aging of the films) are well known from the literature. Several methods are used to examine the alterations in the film structure during this process (Süvegh and Zelkó 2002; Lippold and Pagés 2001; Bajdik et al. 2003). The additives used in the polymer films can change the behaviour of the films during storage.

In the present study, the mechanical properties of acrylic polymer films with and without polysorbate 80 were compared. Polysorbate 80 in a well-defined concentration can act as a plasticizer (Cole 1995). The process of film deformation was evaluated with an apparatus developed in our institute. This work was supplemented by a stability test. This work deals with a less well-known aspects of the aging of films containing polysorbate 80.

It was observed, that the characteristics of the deformation curves (force-time) changed because of the presence of the additive and during storage (Fig.). A fresh film containing polysorbate 80 displays mainly elastic behaviour and does not break during these experiments, but undergoes a springing with decreasing amplitude during which the structure of the film is irreversibly modified. The forces at which the springing started were regarded as the deformation forces of these films. The springing was absent for fresh films without polysorbate 80. Storage changed the characteristics of the deformation. The breaking of films occurred sharply for both samples.

Larger deformation forces were necessary for the breaking of films containing polysorbate 80 (Table). The presence of this additive caused a lesser sensitivity to storage at the lower temperature and RH. The higher temperature and humidity did not induce different behaviour for the films with different compositions.

The evaluation of the deformation time showed that this parameter of fresh films containing plasticizer was significantly ($p < 0.05$) longer. This time became significantly shorter during storage. Lower temperature and RH caused a greater change. Increase of the storage time led to a decrease in this parameter for both storage conditions. In every case, higher values were detected for the samples containing polysorbate 80. Storage caused more relevant changes in this parameter than in the deformation force.

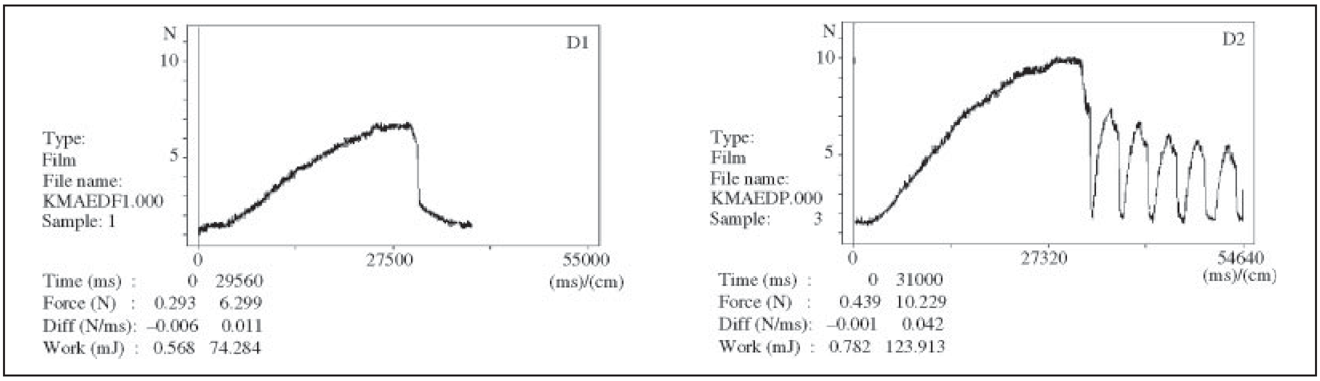

Fig.: Deformation curve of fresh samples

Table: Deformation parameters of films

Parameter	Sample	Fresh	25 °C/60% RH		40 °C/75% RH	
			1 week	1 month	1 week	1 month
Force (N)	D1	6.10 ± 0.36	4.07 ± 0.34	2.75 ± 0.35	7.13 ± 0.97	2.97 ± 0.10
Force (N)	D2	10.61 ± 0.72	10.34 ± 0.62	8.26 ± 0.80	9.93 ± 0.64	5.25 ± 0.42
Time (s)	D1	25.61 ± 4.15	5.17 ± 1.12	2.99 ± 0.25	12.65 ± 1.21	4.78 ± 1.68
Time (s)	D2	36.59 ± 2.40	6.15 ± 0.36	5.33 ± 0.53	13.40 ± 1.01	5.06 ± 0.12
Work (mJ)	D1	52.34 ± 0.92	7.03 ± 1.79	2.76 ± 0.44	30.33 ± 6.65	5.79 ± 0.58
Work (mJ)	D2	129.29 ± 1.55	21.19 ± .26	14.67 ± 1.82	42.72 ± 4.26	8.85 ± 0.84

Larger differences between the films with different compositions were detected as concerns the deformation work calculated force-displacement curve. Its value for films containing polysorbate 80 was 2.5 times higher than that for the other films. The decrease in this parameter during storage was the most significant. In every case, a higher deformation work was detected for the films containing polysorbate 80. Storage at the lower temperature and RH induced less relevant changes in these films. This phenomenon can be supported by the binding of vapour of the polysorbate 80. In the films, the bound water acts as a plasticizer. It can concluded that the plasticizer effect of the polysorbate 80 was more effective at lower RH.

It can be concluded that it is not sufficient to detect the deformation force for the evaluation of the mechanical properties of polymer films. A 5-fold higher deformation work can be detected for samples with the same deformation force. Finally, it can be stated that methods for examination of the deformation process must be developed for the better recognition of the properties of the film-forming agent and additives and their changes during storage.

Experimental

Kollicoat MAE 30 D (D1) and Kollicoat MAE 30 DP (D2) (BASF Ludwigshafen, Germany) aqueous dispersions were applied. The difference between the dispersions is that D1 contains a 2.3% polysorbate 80 as a surface-active ingredient. The coating fluids were prepared according to the manufacturers recommendations. Compositions of coating fluids: film former: 200.0 g, propylene glycol 9.0 g, dimethicon E 1049 2.0 g, micronized talc 16.0 g, water-soluble dye: 0.1 g, distilled water: 172.9 g.

The coating fluid was sprayed onto a rotating teflon surface and the resulting films were examined. In every case the film thickness was in the range 250–300 μm. All the films were stored in a closed and thermostated desiccator (Hereaus Instruments, Hanau, Germany). The storage conditions were: 40 ± 2 °C/75 ± 5% RH and 25 ± 2 °C/< 20% RH. The storage times were 1 week and 1 month.

The strength tester and the software were developed in our institute. This device contains a special specimen holder and a hemisphere stamp, and is connected to a computer via an interface; thus, not only can the ultimate deformation force be measured, but the process (force-time and force-displacement curves) can be followed. The round specimen is located horizontally and the stamp moves vertically. Film thickness was measured at the middle of the specimen with a screw micrometer (Mitutoyo, Japan).

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