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EFFECTS OF SURFACE MODIFICATION OF POLYPROPYLENE FOIL ON POLYMER - OIL INTERACTION

J. Pospieszna, A. Tymań, J. Juchniewicz

Institute of Electrotechnics and Electrotechnology,
Technical University of Wrocław,
POLAND

Abstract

Physico-chemical interactions between the impregnating oil and the polypropylene foil (PP) are of great concern. Such phenomena like swelling and dissolution of polymer films into the oil can deteriorate the electrical properties of the oil - polymer system.

It was found that the swelling control could be attained by the application of the effective diffusion barrier at the PP surface. Formation of thin polymer coatings in radio-frequency discharges was used for this purpose. The effect of surface modification of the PP film on the polymer-oil interaction phenomena were investigated.

1. Introduction

The growing need for higher energy density capacitors of reduced size has led to replacement of the paper by polymer film. Actually, biaxially oriented polypropylene (PP) films are extensively used due to their good electrical and mechanical properties as compared to unoriented PP films.

For oil-impregnated all polypropylene film system the oil-polymer interaction is of great concern. When the foils are impregnated with insulating oils, absorption of the oil by the film and dissolution of the atactic fractions of the polymer into the liquid occur simultaneously, resulting in swelling of the film. The impurities and additives dissolved from the PP films into the oil have been pointed out to play important role in the electrical properties of oil-impregnated PP systems. Generally, dielectric behaviour of these systems have been explained as an interfacial phenomena in terms of trapping or absorption effects and the movement of ion impurities in the solid-liquid system [1, 2].

Studies by G. J. Fitz Patrik, J. Laghari at all [3] have shown that the swelling process definitely involves the surfaces of the film while the electrical and mechanical properties are all related to the bulk of the film.

We pointed out, that the swelling control could be attained by application of an effective diffusion barrier at the PP interface [4].

The plasma-polymerized styrene films have been prepared by high frequency glow discharge. Plasma polymerization, due to its unique mechanism of film deposition from the vapour phase, gives rise to low pin-hole density and highly cross-linked

polymers [5]. Hence, the plasma polymerization technique was chosen for depositing the barrier films.

In the present study the effect of surface modification of the PP foil on the PP-oil interactions was studied. The swelling of the PP and dielectric properties of the polymer foil as well of the layer oil-impregnated PP system were measured.

2. Experimental procedure

The investigations were carried out on biaxially-oriented polypropylene films 12 μm thick made in Finland by Tervakoski. The samples of PP were freely immersed in previously purified alkylkerylbenezine oil (AKB) made in Poland. They were kept at 60°C for definite time.

The degree of the swelling of PP films was estimated from the change in weight of polymer films after the oil-impregnation. The oil adhering on surface of specimen was wiped away by gauze. Then the specimen was ultrasonic cleaned.

Scanning Electron Microscopy (SEM) was used to examine the microstructure of studied samples.

The modified samples of PP were obtained by two-sided deposition of monomer in a radio frequency glow discharge (27 MHz) using a pair of circular parallel electrode plates mounted in a vacuum vessel.

The dielectric spectroscopy was used to the investigation of the dielectric properties of studied samples. The dielectric response over wide frequency range, from 10^{-4} to 10^4 Hz at very low electric field, were measured by means a Solartron-Model 1172 Frequency Response Analyzer with special I-U converter.

The layer oil-impregnated PP systems with the unmodified and modified swollen foils were studied. Furthermore, the dielectric properties of specimens of the pure oil to be used for investigation and oil extracted from the swollen PP foil as well samples of swollen PP foils were measured.

3. Results and discussion

Figure 1 shows the primary process of the swelling of the unmodified and modified samples of PP foil, respectively. We see, that the degree of swelling is strongly reduced owing to surface modification of PP ($\approx 60\%$). In both cases, the swelling takes place comparatively rapidly and saturates afterwards.

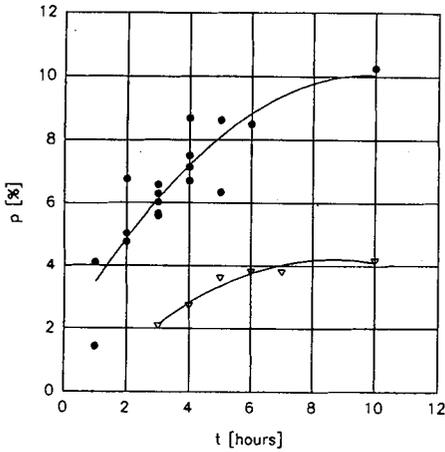


Fig. 1. Swelling of unmodified (●) and modified (○) samples of PP foil in AKB oil at 60°C.

The SEM photographs (fig. 2) show that some surface roughness of unmodified PP foil appears after staying in the oil and a disorientation occurred. These imply that dissolution of amorphous phase into the oil takes place here and the crystalline phase is exposed. The surface of modified PP remained smooth and there no change in the structure of polymer.

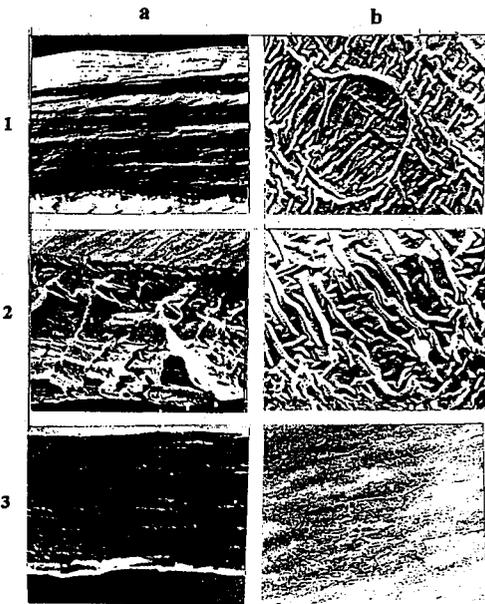


Fig. 2. Scanning electron micrographs of a cross section (1) and a surface (2) of PP samples: a) virgin. b) unmodified swollen, c) modified swollen.

Fig. 3 shows the changes of relationships between a frequency and dissipation factor of the oil-impregnated PP system with the time for which the unmodified film was immersed in oil at 60°C.

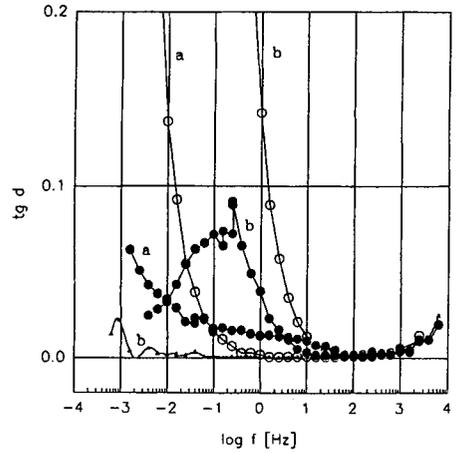


Fig. 3. Relationships between dissipation factor and frequency of oil-impregnated PP system (●), oil (○) and PP foil (▽) a) after making, b) placed for 100 days at 60°C.

It is clearly seen that the interaction between the PP film and the oil lead to a increase of the dissipation factor of the oil. As pointed out previously it can be related to dissolution of the impurities contained in the PP film into the oil.

The typical polarization spectrum due to the swelling is seen in the low frequency region, which is related to the interface / space charge polarization phenomena of long time constant.

Fig. 4 shows the frequency dependence of the dissipation factor due to the swelling of the modified PP films immersed in oil at 60°C for 100 hours.

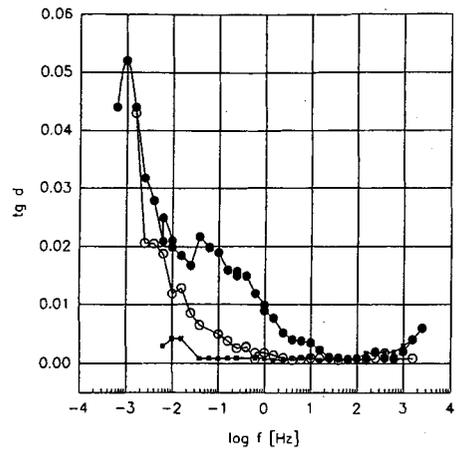


Fig. 4. Relationships between dissipation factor and frequency due to the swelling of modified PP foils for : oil-PP system (●), PP foil (○) and virgin modified PP foil (▽).

As it is shown, the frequency spectrum of dissipation factor of swollen oil-modified polymer system is closely connected with the oil absorption of the film immersed in the fluid. Probably, the swelling of the modified foil takes place without the extraction of impurities dissolved into the oil from amorphous phase of PP foil. By this means modified foils impregnated with oil are plasticized by oil.

In addition, cross-linkings near a PP surface of PP can act as carrier traps and reduce the dissipation factor of an oil-polymer system [6]

4. Conclusions

- The results of our investigations confirm that
- the swelling control could be attained by application of the thin polymer coating on the foil,
 - The plasma polymerization is very effective method to generate the diffusion barrier,
 - such surface modification can improve the electrical properties of the PP-oil system.

Acknowledgements

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References

- /1/ D. Shaw, "AC conduction in impregnated polymer insulating systems", *IEEE Trans. on Electrical Insulation*, vol.-21, No., pp.62-68, 1975.
- /2/ T. Umemura, K. Akiyama, T. Kashiwazaki, "Dielectric behaviour of solid/liquid insulation system", *IEEE Trans. on Electrical Insulation*, Vol.EI-17, No. 3, pp.276-280, 1982.
- /3/ G. J. Fitz Patric, J. Laghari, E. O. Forster, "The effect of impregnants on the morphology and dielectric properties of polypropylene films", *Proc. 2nd Inter. Conf. on Conduction and Breakdown in Solid Dielectrics*, Erlangen, pp.211-216, 1986.
- /4/ S. Głuchowski, J. Pospieszna, J. Juchniewicz, "Attempt to the control of the properties of polypropylene-oil system", *Proc. 3rd Inter. Conf. on Properties and Applications of Dielectric Materials*, Tokyo, pp.871-874, 1991.
- /5/ R. K. Sathir, H. E. Saunders, "Plasma processes for electrical and electronics applications", *IEEE Electrical Insulation Magazine*, Vol. 2, No. 6, pp.8-14, 1986.
- /6/ S. Ochiai, K. Kojima, A. Ohashi, M. Ieda, T. Mizutani, "Conduction current and loss tangent of oil/polymer composite system with polypropylene films sputtered in oxygen", *8th Inter. Symp. on High Voltage Engineering*, Yokohama, pp.103-106, 1993.

Address of authors:

Dr J. Pospieszna
 Dr A. Tymań
 Prof. J. Juchniewicz

Technical University of Wrocław (I-7),
 Wybrzeże Wyspiańskiego 27,
 POLAND