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Experimental study of influence factors on compression stress relaxation of ACM

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Materials

ABSTRACT

Purpose: Purpose: of this paper is to investigate the influence of electron beam (EB) radiation, temperature and deformation on the compression stress relaxation behavior of UV curing polyacrylate rubber (ACM).
Design/methodology/approach: ACM plugs were obtained by UV radiation curing. Some samples were post cured by EB dose of 250 kGy, providing two material conditions. The plugs were submitted to compression stress relaxation experiments (CSR) for 5.000 min on two temperature levels, 23°C and 90°C, and deformation levels of 10% and 15%. Results were analyzed using 2k Factorial Design, quantifying the influence of these factors on the maintained compression force.
Findings: The results have shown an increase in the maintained compression force, respectively a decrease in the compression stress relaxation, for all EB irradiated ACM specimens along the entire measured period of time. The increase of the three analyzed factors has caused an increase of the maintained compression force. Through the 2k Factorial Design was possible to find out the deformation as the most relevant influence factor, followed by irradiation, as second influence factor and temperature.
Research limitations/implications: ACM molecular structures were not physical-chemically investigated. Therefore, possible molecular structural changes, caused by EB radiation, were not described.
Practical implications: Decreasing of compression stress relaxation implicates on increasing working life of sealing components made from the studied material, besides the improvement of sealing capacity of these components, even under reduced deformation conditions.
Originality/value: The use of EB radiation as successful method to decrease compression stress relaxation of UV curing ACM rubber, has opened opportunities to industrial applications. Thus, provide knowledge about influence factors on ACM relaxation behavior.
Keywords: Engineering polymers; Mechanical; Compression stress relaxation

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1. Introduction

Elastomers are largely used as sealing components because of their large deformation properties and capacity of structural recovering after deformation. The compression stress relaxation behaviour, inherent to such materials, can be a limiting characteristic when applying them as sealing medium. It can influence the useful life of components made from such a polymer system [1]. Because of their viscoelasticity, elastomers usually present a compression force decreasing, consequently sealing force also decreases, in function of time, when is kept at constant deformations. Time dependent or viscoelastic behavior of elastomers is linked to complicated molecular adjustments resulting from macroscopic mechanical deformations. Such behavior is peculiar to elastomers because they consist of flexible, long chain molecules that intertwine with each other [2].

The stress relaxation phenomena has its causes on chemical and physical mechanisms. Sperling [3] presents five compression stress relaxation causes: chain scission, bond interchange, viscous flow, thirion relaxation and molecular relaxation.

Maxwell based models are usually applied to describe relaxation process. The proposed model, where dashpot and spring are associated in series, allows verifying the stress on both components, since the applied stress is identical on the elements. Complex process cannot have their behavior usually described by such simple Maxwell based models, in this case a combination between Maxwell and Kevin models, called four elements model, may be used to represent its behavior. For two phase materials, such as polymer blends composed by plastic and rubber materials, Sperling [3] suggests the use of a called Takayanagi model, which indicates arrays of rubbery (R) and plastic (P) phases, instead of arrays of dashpots and springs. The quantity λ or its indicated multiplications indicates volume fractions of the materials. As with spring and dashpots, the Takayanagi model can also be expressed analytically. For parallel model, the horizontal bars connecting the two elements must remain parallel and horizontal, yielding and isostrain condition [3]. The Equation 1 shows the Takayanagi series model.

$$E = (1-\lambda)E_P + \lambda E_R \quad (1)$$

For compression stress relaxation problems, Maxwell based models are used in associated series elements. Since the deformation is fixed, at compress stress relaxation, the sealing force is determined by the modulus of the polymer, thus a knowledge how modulus varies with time under service conditions will provide a basis for the design of sealing materials which is most likely to give the required service life [4].

Many authors proposed the electron beam radiation (EB) as method to increase the modulus of polymeric materials like PP [5], FKM [6] and ACM [7]. Dos Santos and Batalha [8] have shown that besides being used as a modulus increasing method, EB can also provide a decreasing of compression stress relaxation of ACM. Placek et al [9] has showed the sealing capacity effects on EPDM seals, when exposed to low dose ionizing irradiation for long periods.

ACM are acrylic base elastomers, largely applied on sealing component manufacturing because of their high thermal and chemical resistances. Such properties make the material able to resist to environmental conditions present on powertrain applications. Vijayabaskar et al [10] proposed the EB radiation as method of improving the mechanical performance of standard curing ACM rubber.

Ageing usually affects polymer performance in different ways. Gnatowski [11] has presented the effects of electrochemically aged PP and respective structure changes. Another kind of ageing and a major influence factor concerning rubber compression stress relaxation is the temperature. An increase of the temperature will accelerate the relaxation process, but it will also increase the compression force for specific deformation level. Treloar [12] explains such peculiar effect affirming that it arises from the normal thermal expansivity of the unstrained rubber, as a result of unstrained length varies with change in temperature. Treloar [12] examines deeply the phenomena and presents an explanation based on the first and second thermodynamic laws. He assumes that the internal energy of the system keeps constant during deformation, while an entropy change occurs.

This paper has the purpose of investigating the influence of EB, temperature and deformation on the compression stress relaxation behavior of UV curing ACM. Stress relaxation behavior can be investigated in uniaxial tensile or in compression conditions [13]. Usually the rubbery sealing elements are applied in compression conditions, where the generated compression force is responsible for the element sealing properties.

Experimental results were applied to Factorial Design 2k, in order to provide quantified data about possible changes on the compression stress relaxation behavior. Such changes may be caused by investigated influence factors.

2. Experimental methodology

2.1. Material

ACM rubber plugs, with diameter of 6.3 mm and length of 13 mm, were obtained by manual process. They were injected in liquid form at room temperature of 25±2°C into a silicone mold, which was exposed to UV radiation, according to the supplier specification, promoting the material cure. The silicone mold was obtained curing RTV silicone in high temperature and humidity conditions.

2.2. Specimens irradiation

The electron beam irradiation of the injected ACM plugs was carried out at room temperature of 25±2°C, at the Nuclear and Energy Research Institute in Sao Paulo, by an electron beam accelerator Model DC 1500/25/4 of the Radiation Dynamics, Inc. Dose of 250 kGy was applied, this dose has been defined based on the work of Dos Santos and Batalha [7]. Table 1 shows the EB accelerator and process specifications.

Table 1.
EB accelerator and irradiation process specification

EB accelerator specification	
Energy range	0.5-1.5 MeV
Current range	0.3-25 mA
Beam power	15 kW
Irradiation process specification	
Electron energy (selected)	1.013 MeV
Beam current (selected)	4.54 mA
Process	Conveyor
Conveyor speed	6.72 m/min
Dose rate	22.42 kGy/s
Dose by cycle under the beam	5 kGy

2.3. Mechanical testing facilities

The influence of the three selected factors (EB, temperature and deformation) on the compression stress relaxation of ACM rubber was investigated using a mechanical experiment. Each influence factor was analyzed in two levels, as shown in Table 2.

Table 2.
Influence factors and their analyzed levels

Factor	Inferior level	Superior level
Temperature	23° C	90° C
EB radiation	0 kGy	250 kGy
Deformation level	10%	15%

The association of such factors, according to factorial design 2k, has generated eight test conditions, which considers all involved factors and their respective levels. Three samples of each condition were tested. The Table 3 shows the experimental design test, representing the maximum level factor by the signal (+) and the minimum level by (-).

Table 3.
Obtained experimental conditions

Condition	Temperature (Temperatura)	Deformation (Deformação)	Radiation (Radiação)
1	-	-	-
2	-	-	+
3	-	+	-
4	-	+	+
5	+	-	-
6	+	-	+
7	+	+	-
8	+	+	+

Table 4.
Obtained experimental conditions (results in N)

time	1	2	3	4
500	23,78	29,02	32,90	42,64
1000	23,34	28,48	32,06	41,86
1500	22,60	28,33	31,67	40,93
2000	22,45	27,65	30,93	40,58
2500	22,36	27,40	30,64	39,80
3000	21,48	27,26	30,64	39,90
3500	21,13	26,96	30,05	39,85
4000	21,14	27,01	29,95	39,41
5000	20,60	26,32	29,41	39,11
time	5	6	7	8
500	26,22	30,05	34,17	44,46
1000	25,78	29,26	33,53	43,09
1500	25,53	28,62	33,09	42,50
2000	25,00	28,19	32,90	41,81
2500	24,90	28,14	32,56	41,66
3000	24,65	27,74	32,36	41,42
3500	24,55	27,70	32,21	41,27
4000	24,75	27,59	32,11	41,27
5000	24,21	27,30	31,82	40,78

ACM plugs were submitted to pre-defined deformations at testing temperature for period of 5.000 min. The deformation was kept constant and a cell load has provided the compression force variation during the defined period of time. Compression force x time curves were obtained for the eight test conditions, comparing directly the non-irradiated against the 250 kGy irradiated condition.

The experiments were carried out using an Elastocon EB02 Compression Stress Relaxation equipment, Figure 1, at the ACM supplier in Germany. This equipment model is able to test three samples simultaneously. Tests conditions on 90°C temperature level has required a sample pre-heating before straining at 10% or 15% level, in order to bring the samples to test condition before starting the measurements.

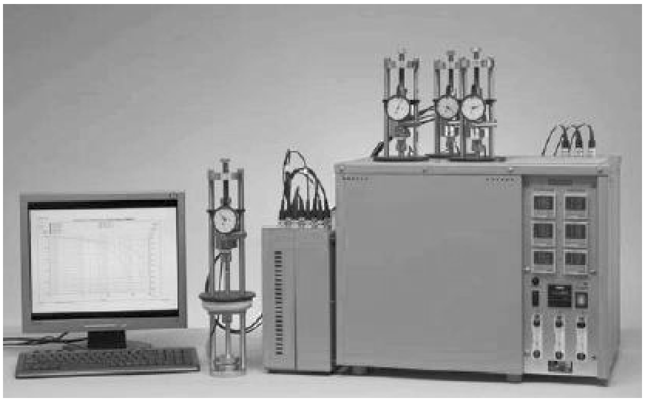

Fig. 1. Elastocon EB02 CSR equipment

Experimental results were applied to factorial design 2k method, making possible to quantify the influence of each factor, of their interactions and their relevance. Uniquely the measurements carried out after 5.000 min were taken in consideration on the factorial design 2k

3. Results and discussion

3.1. Compression stress relaxation

Experimental data obtained from compression stress relaxation is important in predicting the work life of the sealing elements made from ACM. The Table 4 shows the maintained sealing, or compression, force after specific periods. For each condition the measured compression force decreases in function of time, it is caused by the partial viscous behavior of the ACM. Comparing the Table 4 data is possible to verify the influence of EB radiation, which has caused an increasing of sealing forces, during all measured period of the irradiated specimens in comparison to non irradiated conditions.

The partial elastic behavior contributes to ACM sealing performance maintaining the applied force. Dos Santos and Batalha showed an increasing of the ACM elastic behavior storage through EB radiation. Using dose of 250 kGy the UV curing irradiated ACM has presented a higher capacity of keeping elastic properties.

In case of like rubber materials, such elastic behavior should be treated as non linear elastic behavior, some models like Ogden, Boyce and Gent are largely and successfully used to describe non linear elastic behavior of rubbery materials.

The Figure 2 shows the experimental curves concerning to the conditions 1 and 2. The only difference between both material conditions is the irradiation level. These results show the superior performance of the irradiated condition, where the compression force curve, of the irradiated material, presents higher values in comparison to non irradiated material. Such superior performance can be verified during all period. The EB radiation has caused an increase of approximately 27% of the compression force after 5.000 min.

Considering the 15% deformation condition the same trend was noted. The EB radiated samples have presented better performance, it means a sealing force approximately 33% higher, after 5.000 min, as shown in the Figure 3, where the behavior of conditions 3 and 4 are plotted. Comparing to the conditions 1 and 3, as the conditions 2 and 4, where the deformation level is the unique varying factor, it is possible to quantify the force increase caused by increasing the deformation. For the non irradiated condition, the compression force has increased almost 43% by increasing the deformation, while the irradiated condition the increasing was 49%, both values after 5.000 min.

The Figure 4 is related to condition 5 and 6, where the temperature condition was introduced into the tests. The curves presented on the Figure 4 are comparable to the curves of the Figure 2, having the temperature level as varying factor. In this case, the curves related to EB irradiated and non irradiated conditions present an overlapping region. Besides the overlapping, the average compression force was increased by the EB radiation in approximately 13%, after 5.000 min.

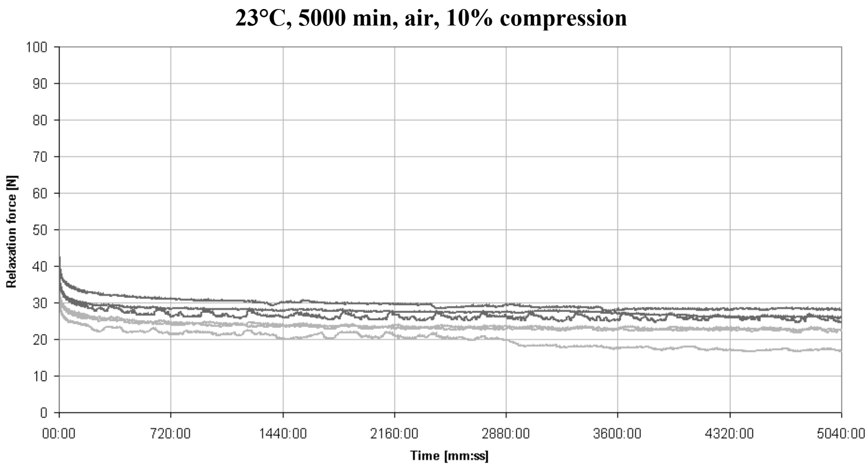

Fig. 2. CSR curves of conditions 1 (green) and 2 (blue)

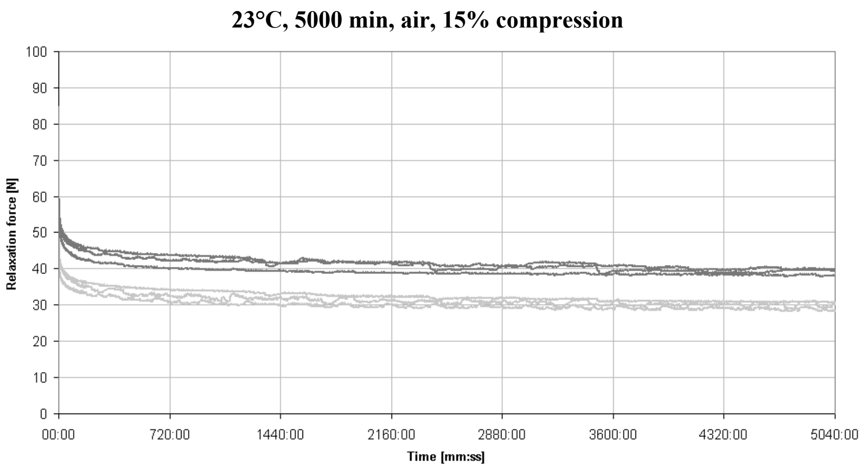

Fig. 3. CSR curves of conditions 3 (yellow) and 4 (red)

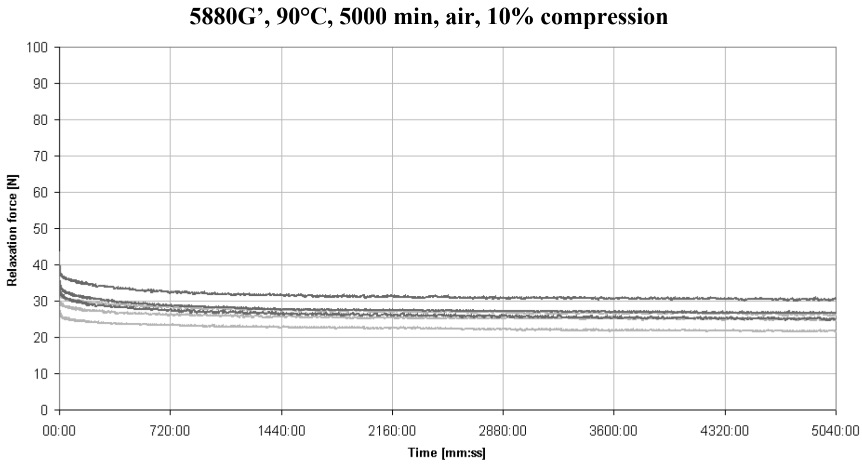

Fig. 4. CSR curves of conditions 4 (green) and 6 (blue)

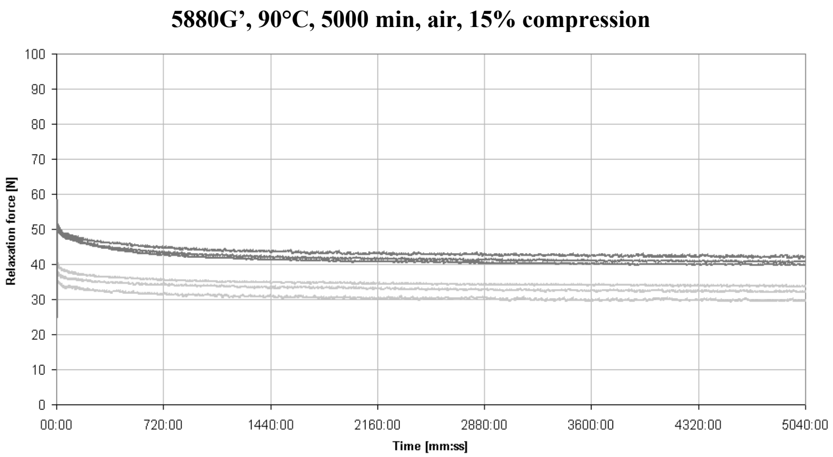

Fig. 5. CSR curves of conditions 7 (yellow) and 8 (red)

Taking temperature and deformation factors at high level and varying the EB radiation dose, the curves presented on Figure 5 were obtained. Maintaining the behavior presented on Figures 2, 3, and 5, the EB radiation has increased the compression force after 5.000 min at constant deformation. The compression force for the non irradiated condition after 5.000 min was 31,82 N, being increased to 40,78 N for the irradiated condition.

Dos Santos and Batalha showed through DMA analysis that a EB radiation dose of 250 kGy, applied to UV curing ACM elastomer, can increase the elastic storage modulus of the material. This modulus increasing could be a feasible reason for the higher sealing force presented by all irradiated samples, after 5.000 min. Associated to superior elastic modulus storage, the 250 kGy dose also increase the crosslinking level of the material, explaining the initial compression force increasing for the irradiated samples.

3.2. Factorial Design 2k

Experimental results measured at 5.000 min of constant deformation were applied to Factorial Design 2k. Figure 6 shows quantitatively the mean influence of each factor on the compression force after 5.000 min. As shown, deformation is the most influent factor, which has caused the highest increasing of the sealing force in comparison to the other two factors. Comparing the Table 4 data it is possible to verify an increasing of approximately 31% up to 49%, which were caused by the deformation level increasing.

The EB radiation was the second most influent factor. By increasing the post cure EB radiation dose, from 0 kGy to 250 kGy, the sealing force has strongly increased on levels of around 12% up to levels of almost 33%.

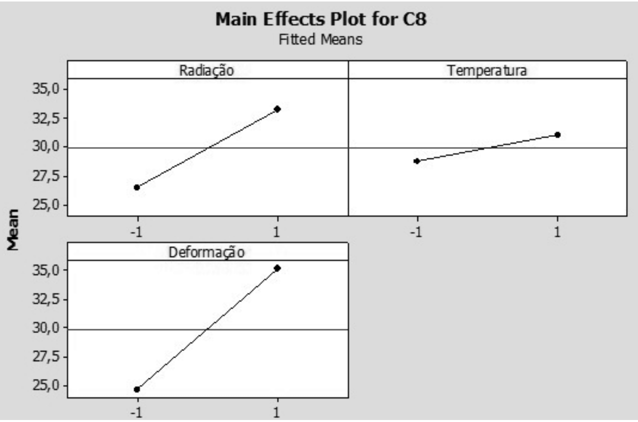

Fig. 6. Main influences of analyzed factors

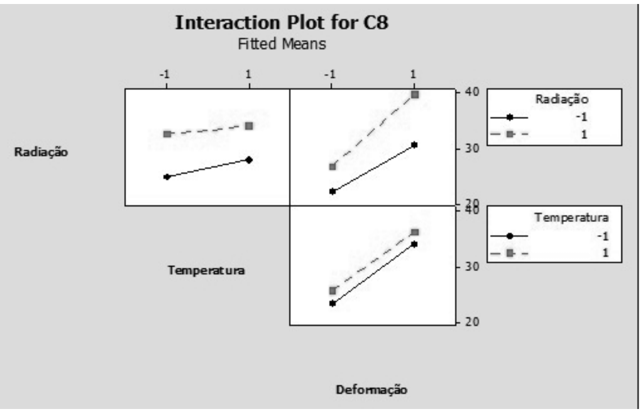

Fig. 7. Effects caused by interactions of factors

Temperature appears as the less influent factor, the temperature increasing has responsible for increasing the sealing force on approximated levels of 3% up to 17%. Because of the high material entropy at 90°C, consequence of high molecular agitation, a higher level of energy is needed to strain the samples up to defined deformation level.

Besides the main effects, the Factorial Design has provided data concerning the influence of the factor interactions, it means, the effects which were caused by the combination of factors, presented on Figure 7. The influence of the interaction between factors can be verified comparing the tangent of the curves. Different tangents indicate influences of factor combinations.

Figures 7 and 8 show, as the unique statistically significant, the interaction between radiation and deformation. The curves show that by keeping the deformation at low level (10%) and varying the radiation, the compression force will increase approximately 19%. While by keeping the deformation at high level (15%) and increasing the radiation, the compression force will increase almost 30%. The Pareto chart, presented on Figure 8, confirms statistically the relevance of three main factors and describes interaction based on the standardized effect.

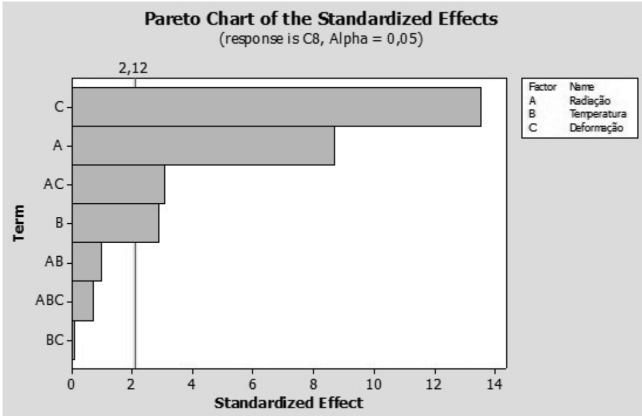

Fig. 8. Main influences of analyzed factors

4. Conclusions

Based on experimental results is it possible to conclude:

Deformation level, temperature and EB radiation influences the compression stress relaxation behaviour of ACM rubber.

Deformation is the most influent factor, it reduces the compression stress relaxation and consequently it increases the compression force after 5.000 min up to 49%.

EB radiation can be used as method to improve the compression stress relaxation of ACM sealing elements. The 250 kGy dose has increased the compression force up to 33%.

The interaction between the factors EB radiation and deformation has decreased the compression stress relaxation after 5.000 min.

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