

INTERNAL HEATING EFFECT DURING ELECTROCHEMICAL ETCHING OF LEXAN POLYCARBONATE

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An internal heating effect has been observed during electrochemical etching of Lexan polycarbonate even at low frequencies ($< \text{kHz}$). There is a correlation between the registration efficiency or detector response and the heating rate in Lexan and the two maxima occur at the same a.c. field frequency. The detector response also depends upon temperature, and at higher temperatures the maxima shifts towards higher frequencies. The study reveals that dielectric polarization exhibits a resonance type phenomenon with applied a.c. field.

1. Introduction

The electrochemical etching (ECE) technique was introduced by Tommasino [1] and it has been exploited for neutron fluence measurements [2–4]. However, the mechanism is not yet fully understood and the detector response depends upon some hidden parameters. The internal heating effect during ECE of polymeric detectors is one such hidden parameter recently discovered by Sohrabi [5] at various field strengths (7–42 kV/cm) and frequencies (10–100 kHz). In the present study, the phenomenon of internal heating is investigated in the domain of low frequencies, and its relation with the registration efficiency during ECE of PC foil is established. As pointed out by Sohrabi [5], the temperature rise is appreciable even at low frequencies ($< 10 \text{ kHz}$) if a small volume of etchant is used during ECE. To achieve this, a multichamber ECE system designed by Sohrabi was further modified for our experiment.

2. Experimental procedure

In the present work, Lexan polycarbonate foils irradiated with a fission fragment source (^{252}Cf) for 60 s were selected for ECE. The polymer foils having a thickness of $80 \mu\text{m}$ and an effective diameter of 8 mm were subjected to ECE for 40 min and an a.c. field of 94 kV/cm was applied. A small quantity (0.4 ml) of 6.25 M NaOH was used as the etchant. As the temperature rise was small it was measured with a K-type (Cr/Al) thermocouple. It was ensured that NaOH did not react with the thermocouple material. The experiment was carried out at a temperature of 30°C (room temperature). The temperature rise both for irradiated and

unexposed PC foil was observed at different frequencies, ranging from 100 Hz to 1.5 kHz during the ECE process. The track density was also recorded in the case of the irradiated foils.

The ECE experiment was repeated at various temperatures (16, 30, 35 and 40°C) for varying intervals of time (60, 40, 30 and 20 min, respectively). The frequency was varied from 100 Hz to 1 kHz for each set of observations. The temperature was controlled using a Haake N3 constant temperature bath.

3. Theory

According to the theory, the main heat generating mechanism responsible for dielectric heating is “dielectric loss” although Joule heating might also play some role. In polar polymers such as PC different effects are involved in dielectric loss, such as dipole polarization and Maxwell–Wagner interfacial polarization [6]. Interfacial polarization is important only in the case of inhomogeneous dielectrics [7] and only at extremely low frequencies. Very often it is so slow that it takes place at frequencies which are very much lower than 1 Hz [8]. All these effects lead to a general formula of heat dissipation per unit volume in a dielectric (insulator) under an a.c. field given by the following equation [9]:

$$P = 0.5555 f E^2 \epsilon' \tan \delta, \quad (1)$$

where P is the energy absorbed per unit volume in W/cm^3 (dielectric loss), f is the frequency in Hz, E is the field strength in V/cm , ϵ' is the dielectric constant and $\tan \delta$ is then loss tangent also known as the dissipation factor. The product of $\epsilon' \tan \delta = \epsilon''$ is known as the loss-index. This energy absorbed results in heat

generation and thus rise in temperature. According to relation (1) P is directly proportional to the frequency, the square of field strength and the loss-index which is further frequency dependent.

4. Results and discussion

Polymer carbonate (PC) during ECE itself serves as a heater, thereby it raises the temperature of the etchant. The rise in temperature depends on the physical and chemical characteristics of the polymer, field strength, frequency and etchant conditions. The rate of temperature rise also depends on the volume of etchant used, ECE cell geometry, insulation and also starting temperature.

Sohrabi [5] failed to detect any temperature rise in the etchant up to a threshold frequency of 10 kHz. This may be due to the effect of geometry of the cell and the large volume of the etchant (12 ml) used. In the present study these parameters are taken care of. The design and geometry of the cell is such that a small quantity (as small as 0.2 ml) of etchant can be used. In this paper some of the effects of frequency on heat responses and registration efficiency of PC, as well as the effects of temperature on registration efficiency, are discussed.

In fig. 1, curve A shows etchant temperature as a function of frequency for an unirradiated PC foil. It is evident that the heating rate is not proportional to frequency from 100 Hz to 1 kHz, as it is observed in the

higher frequency range from 10 kHz onwards [5]. There is a sharp peak near 200 Hz and there is again a rise in the heating rate around 1 kHz. A similar effect is observed for the irradiated PC foil (curve B) with only the difference that the heating rate is more pronounced and the peak at 200 Hz is sharper. In curve C, the ECE spot density (spot/cm²) is plotted as a function of frequency. Interestingly, the registration efficiency for the track response and the heating rate maxima for PC foil are at the same frequency, i.e. 200 Hz. The registration efficiency is proportional to the heating rate till 900 Hz and afterwards the curve C does not follow the curves A and B.

In the frequency region below 1 kHz, factor f does not play a significant role for heating of PC [5]. Besides E , the maximum contribution in the heat generation is of loss-index. The maxima at 200 Hz (fig. 1, curves A and B) represent a higher rate of heating and as a consequence, a higher loss-index. The curves A and B level down after the sharp peak and the sudden temperature rise at frequencies ≥ 1 kHz might be due to the effective contribution of f or some combined effects of f and $\epsilon' \tan \delta$.

The temperature rise in the case of the irradiated PC foil (fig. 1, curve B) is higher than that of the unirradiated foil (curve A). For both foils, all the electrical, physical and chemical parameters are the same. A possible reason for the higher heating rate in the irradiated foil might be the rapid thinning of the foil which has become chemically more reactive after irradiation, thus

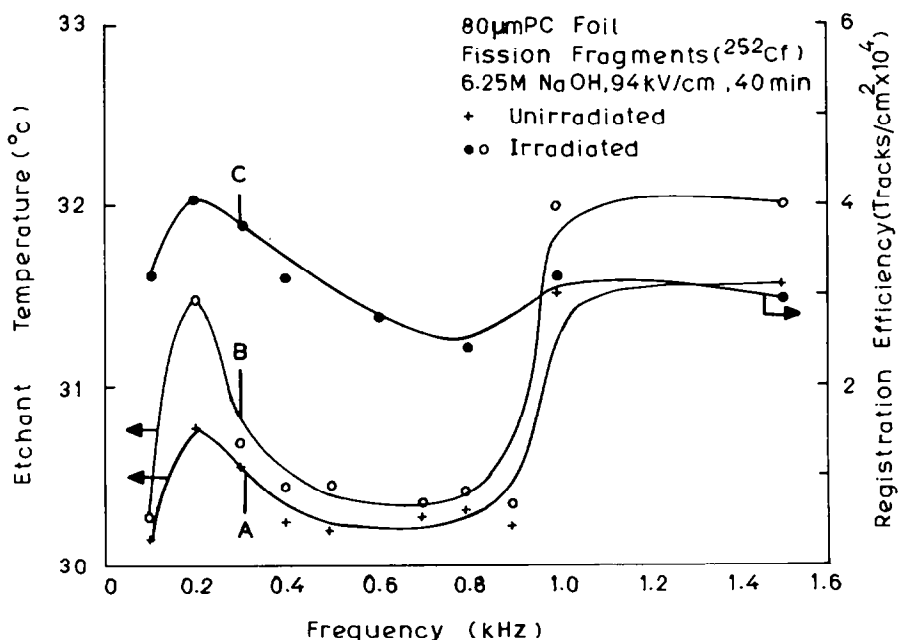


Fig. 1. The etchant temperature (left) and registration efficiency as functions of frequency in Lexan polycarbonate at 30°C (room temperature).

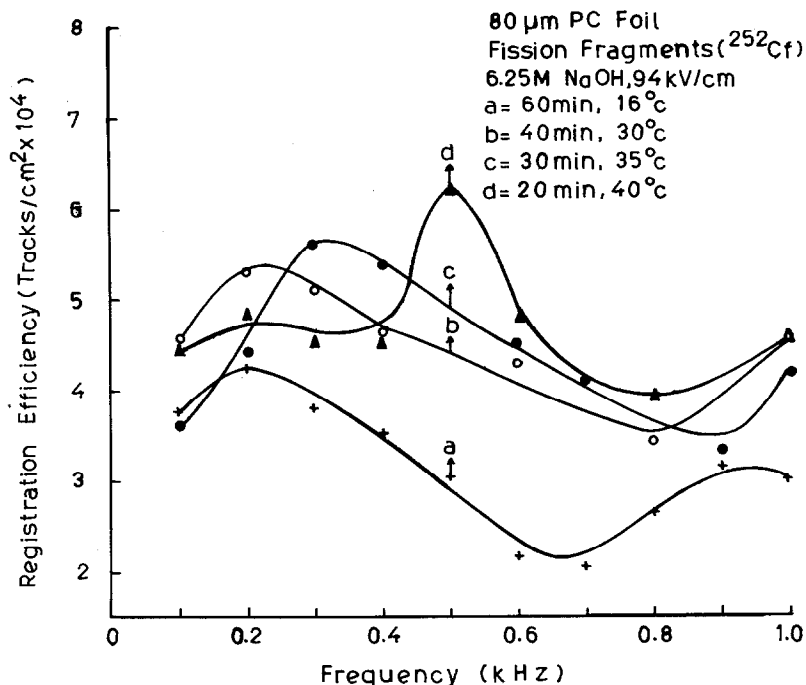


Fig. 2. The registration efficiency as a function of frequency in Lexan polycarbonate for different temperatures.

increasing the field strength E which has an E^2 factor in the dissipation relation. Another factor might be the high charge density at the tip of tracks leading to more heat generation due to electrical stresses. The registration efficiency of the dielectric has a direct relation to the heating rate as is clear from fig. 1 (curves A, B and C), and thus loss-index. At the registration efficiency

maximum the loss-index has high value. This might be due to a resonance type phenomenon of the dielectric polarization (polar and/or interfacial) with the a.c. field frequency.

The different curves in fig. 2(a-d) represent ECE spot density as a function of frequency at 16, 30, 35 and 40°C, respectively. The registration efficiency or track

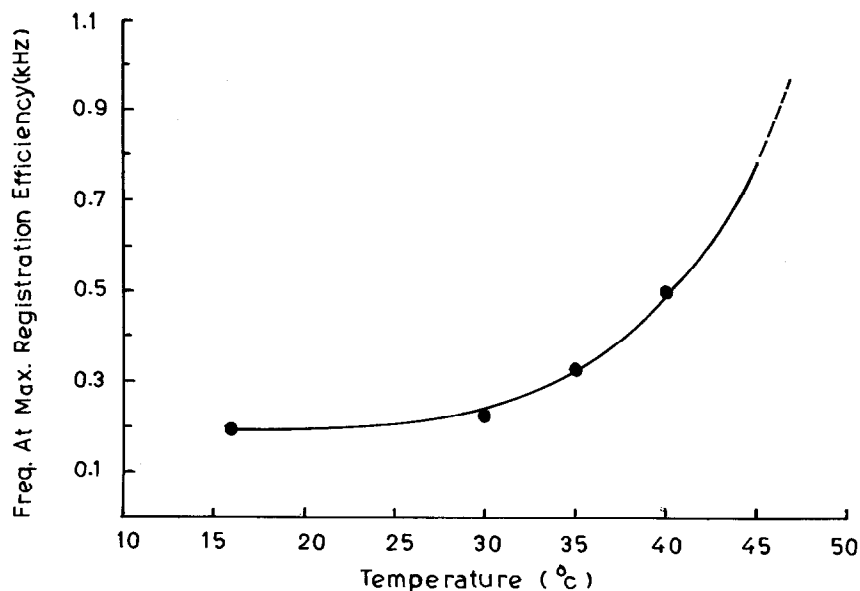


Fig. 3. The registration efficiency maximum as a function of temperature in Lexan polycarbonate.

response maxima for these curves occur at frequencies 200, 225 325 and 500 Hz, respectively.

It is evident from fig. 3 that the registration efficiency is an increasing function of temperature. Up to 30 °C, the registration efficiency is almost constant and with further rise in temperature it shows an exponential rise. As discussed above, at a given temperature the maxima of the registration efficiency and the loss-index occur at the same field frequency. In other words, $\epsilon' \tan \delta$ is an increasing function of temperature [9] or, the establishment time of the dielectric polarization goes on reducing with the rise of temperature.

5. Conclusions

From the above discussion we conclude that:

- (1) An internal heating effect during ECE of polymer exists even at low frequencies.
- (2) Registration efficiency of a polymer has a relation with $\epsilon' \tan \delta$.
- (3) Registration efficiency is an increasing function of temperature.

We also conclude that to get better results from ECE studies the polymer selected should have a high $\epsilon' \tan \delta$;

ECE should be done at a field frequency where $\epsilon' \tan \delta$ is a maximum and the experiment should be performed at a higher temperature. A detailed study is being undertaken on different materials in order to understand the relation between $\epsilon' \tan \delta$ and the registration efficiency and to establish a general theory.

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