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Ferroelectric Stability of Annealed PVDF-TrFE Thin Film Incorporated with MgO Nanofillers

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ABSTRACT

Spin coated polyvinlylidenefluoride-trifluoroetylene (PVDF-TrFE 70/30mol%) copolymer thin film were initially produced and annealed at varying temperatures (100°C to 160°C). The morphology, dielectric and ferroelectric analysis showed that PVDF-TrFE film annealed at 120°C produced the highest remnant polarization, P_r of 92 mC/m^2, with orderly and grain-like shaped crystallites. The filled PVDF-TrFE, loaded with various volume percentages (1 – 7%) of Magnesium Oxide (MgO) nanofillers and then, annealed at 120°C, produced homogenous filler distribution with low agglomerates, especially for 3% PVDF-TrFE filled films. Moreover, the annealed PVDF-TrFE/MgO(3%) generated the highest value of Pr in comparison to the other filled nanocomposite thin films. Most importantly, the saturation of hysteresis loop, P_s for annealed PVDF-TrFE/MgO(3%) film was relatively improved by 20% as compared to the unfilled annealed thin film. This study established that, 3% MgO loaded in PVDF-TrFE thin film and annealed at 120 °C demonstrated a stable ferroelectric thin film, closed to an ideal ferroelectric film, in which the ratio P_r/P_s for the film established a value approaching unity (value of 1).

Keywords: Annealing, dielectric, ferroelectric, MgO, PVDF-TrFE, spin coating

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INTRODUCTION

The development of polymeric film with high dielectric properties has sparkedinterest in the application of small electronic devices such as organic capacitors and organic field effect transistors. The utilisation of these

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polymeric materials is the simplest route in fabricating thin film, owing to their flexibility, high strength and ease of processing. Polyvinylidenefluoride (PVDF) copolymerised with trifluoroethylene (TrFE) is a favourable polymer for organic devices due to its high permittivity and ferroelectricity (Bae et al., 2010; Furukawa et al., 2010, Müller et al., 2007; Ismail et al., 2012). The ferroelectricity in PVDF-TrFE originates from the H+ (hydrogen) atoms and F- (fluorine) atoms. The arrangement of these atoms in the PVDF-TrFE chains induced the parallel packing of the all-trans conformation of PVDF-TrFE chains resulting in large spontaneous polarisation (Furukawa, 1989).

In volatile memory devices, data may not be retained when the system is turned off in contrast withnon-volatile memory devices, which have the ability to store charges even after the electric field is removed (Ling et al., 2008). The latter is true for PVDF-TrFE. The performance of PVDF-TrFE as memory device relies mainly on the remnant polarisation (P_r), which is largely affected by the crystallinity of β phase crystals and the degree of dipole orientation in PVDF-TrFE (Furukawa et al., 2006; Ling et al., 2008). In order to achieve high P_r, additional treatments such as annealing, mechanical stretching and poling are widely employed. However, for thin film, annealing process is the most appropriate and practical as it poses minimal damage to the thin film compared with the mechanical stretching or poling at high electric field, which is more suitable for bulk film. Moreover, the distinctive crystal phase transitions during annealing treatment led to an interest in exploring the ferroelectric behaviour of polymeric material (Lee et al., 2010; Prabu et al., 2006; Wahid et al., 2013). In 1980, Furukawa et al. discovered the curie transition in PVDF-TrFE, which indicated the ability of polymeric material to polarise spontaneously (Furukawa et al., 1980). Beyond Curie temperature, the ferroelectric constants of PVDF-TrFE dropped to zero due to the thermal heating, which demote dipole rotational motion and hence, destroy all-trans chain configuration. In addition, this phase transition of PVDF-TrFE was very much affected by the VDF content. Furukawa discovered that 50% to 80% of VDF content in PVDF-TrFE produced high dielectric and ferroelectric properties (Furukawa, 1997). Previous study reported that, annealing at temperature slightly above the Curie temperature, (T_c), but below the melting temperature (T_m) resulted in an improved crystallinity of the PVDF-TrFE film. This condition is favourable due to aligning and packing of C-F dipoles of PVDF-TrFE. This will lead to an increase in the amount of aligned dipoles, hence, increasing the polarisation and dielectric properties of PVDF-TrFE film (Nguyen et al., 2008).

In this research, a batch of spin coated PVDF-TrFE thin films of approximately 250 nm thick were annealed at varying temperatures, in accordance with the thermal transition temperatures attained by differential scanning calorimetri (DSC) analysis. The optimised annealing temperature was predetermined by observing the surface morphology, and subsequent attainment of the dielectric and ferroelectric properties of PVDF-TrFE thin film. The optimised PVDF-TrFE was loaded with MgO nanofillers at various volume percentages. Similar characterisations were noted on the PVDF-TrFE/MgO nanocomposite thin films.

METHOD

PVDF-TrFE Thin Film Preparation

Prior to producing the PVDF-TrFE thin films, aluminum (Al) coated glass substrates were prepared, which represented the bottom electrode for electrical measurements. These glass substrates were cut and cleaned with acetone, methanol and de-ionised water for 10 minutes in an ultrasonic bath, before they were dried with inert nitrogen gas flow. A layer of 30 nm in thick was evaporated on the glass substrates, using Edward AUTO306 high vacuum evaporation system. The PVDF-TrFE (70:30mol %, PiezoTech, France) pallets were dissolved in the solvent (methyl ethyl ketone) with the concentration of 30g/L and stirred for 24 hours. The solution was then spin coated at 1500 rpm for 90 seconds. Finally, PVDF-TrFE film was spin coated as thin film on the Al coated glass substrate with an average film thickness of 250 nm to 300 nm.

Annealing Process

Prior to annealing, the thermogravimetric analysis was carried out on PVDF-TrFE pallets in order to determine the Curie's transition temperature (TC) and the melting temperature (T_m). The two endothermic peaks (during heating) were 113°C (TC) and 154°C (T_m) (Figure 1). These temperatures were set as benchmarks for annealing the PVDF-TrFE thin films.

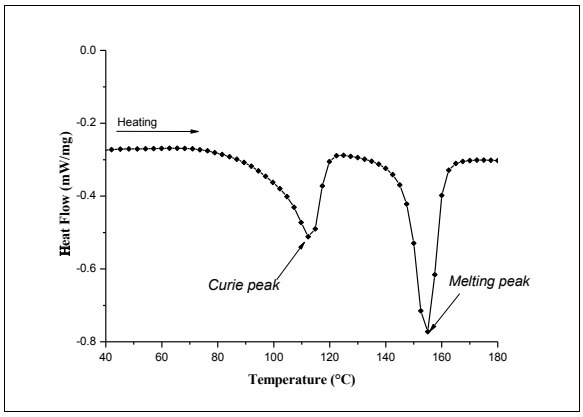


Figure 1. Thermogravimetric spectrum of PVDF-TrFE (70:30mol%)

The annealing was conducted for 2 hours in an oven at temperatures shown in Table 1. All annealed films were allowed to cool at ambient temperature before they were removed from the oven. The unannealed thin film (UN) was utilised as a control film sample in this study.

Table 1
Annealing temperatures of PVDF-TrFE thin films

Acronym	Annealing Temperature (°C)
AN100	100
AN120	120
AN140	140
AN160	160

PVDF-TrFE Loaded with MgO Nanofillers (PVDF-TrFE/MgO Thin Film)

Magnesium oxides (MgO) of particle sizes ≤50 nm were supplied by Sigma-Aldrich. Predetermined MgO volume percentages (1, 3, 5 and 7 vol %) were incorporated in PVDF-TrFE solution individually at concentration of 30 g/L and stirred at room temperature for 24 hours. The PVDF-TrFE/MgO solution was agitated and sonicated in an ultrasonic processor (SONICS Vibracell VCX500, 400 watt) at 60 % amplitude for 12 minutes. A tapered ultrasonic horn was utilised with an end tip of 3 mm in diameter. The sonicated solution was spin coated on the Al-coated glass at 1500 rpm for 90 seconds to produce 250 nm thick nanocomposite films. These nanocomposite thin films were then annealed in the oven at 120°C for 2 hours.

Characterisation

The morphological study of the thin film was carried out using field emission scanning electron microscope (FESEM, JEOL JSM-7800F). All thin films were coated with 10 nm of platinum prior to FESEM imaging in order to reduce electro charging, as well as increasing the image quality. The top electrical contact was produced by evaporating 3 mm^2 of Al above the thin film (Figure 2). Dielectric properties were measured at frequencies of 100 Hz to 10 MHz using high frequency range impedance analyser (Agilent, Impedance Analyzer 4294A), while the ferroelectric measurements were done using the Precision LC system (Radiant Tech. Inc.) at 100 Hz frequency, at an operating voltage of 100 V.

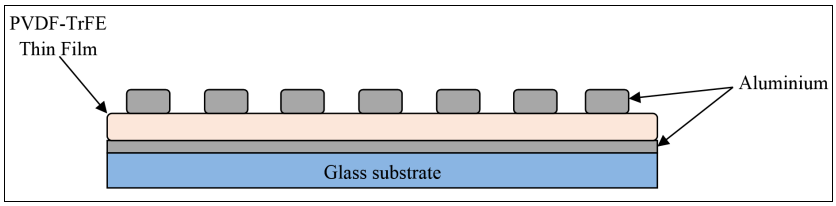


Figure 2. Schematic diagram of PVDF-TrFE thin film with aluminum top and bottom electrodes

RESULTS AND DISCUSSION

Optimisation of the annealing temperature for PVDF-TrFE thin films

First, the morphology of unannealed and annealed PVDF-TrFE thin films was observed at 100k magnification using FESEM (Figure 3). Significant growth of defined crystallite structures was observed in PVDF-TrFE films annealed at 100°C (AN100) (Figure 3(b)). As the annealing temperature was increased to 120°C, the crystallite sizes in the AN120 films were extended in length and width (Figure 3(c)). Previous studies suggested that elongated structure of PVDF-TrFE crystallites signified typical characteristics of ferroelectric crystallites (Lee et al., 2007; Li et al., 2008). In this study, the elongated grain-like crystallites were found to increase in size as the annealing temperature was further increased to 140°C for AN140 film (Figure 3(d)). On the contrary, the PVDF-TrFE thin film annealed at 160°C, temperature above T_m, the crystallites lost its grain-like structure. Instead, most of these crystallites in AN160 film

were observed to merge with the neighbouring crystallites forming fibrous crystallites (Figure 3(e)). This may be due to the recrystallisation process, in which these films were cooled directly from melt. During melting, the increase in thermal energy encouraged mobility of the polymer molecules. This caused high thermal heating, which disorientate most of the PVDF-TrFE crystallites. Upon cooling, these crystallites were unable to re-orientate to form the grain-like crystallites structures, which were formed through annealing, at temperatures below melting temperature. The fibrous-like shaped structures formed in all of the recrystallised films suggested a changed in the crystal phases during recrystallizsation. The splitting of the fibrous surfaces was quite apparent, which may be commonly caused by differential cooling of the film surfaces. The presence of these defects, typically reduced the performance of the AN160 films (Gan & Majid, 2014).

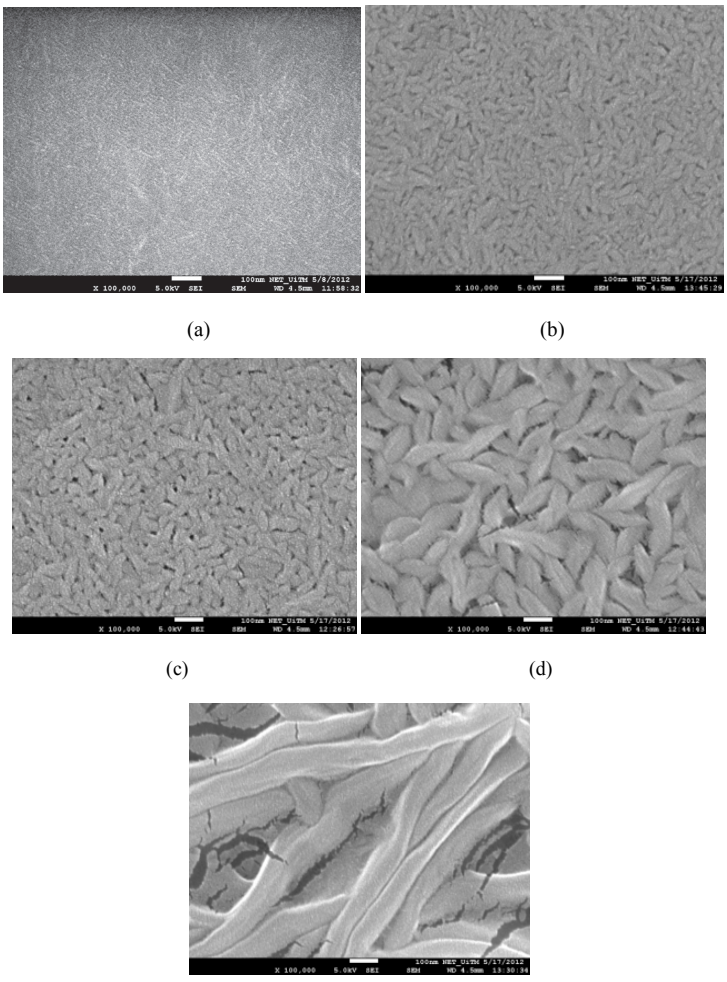
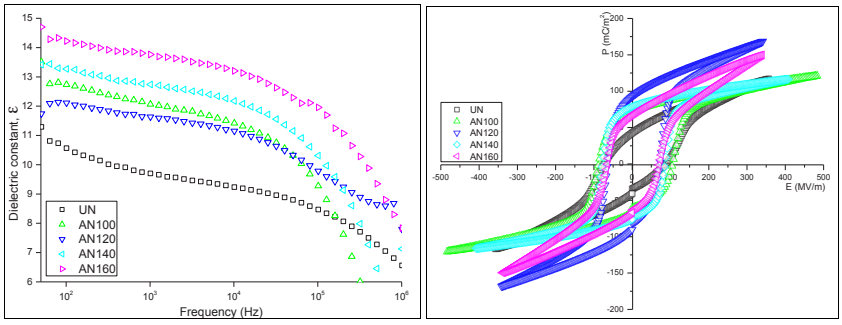


Figure 3. FESEM micrograph at 100K magnification for PVDF-TrFE thin film of (a) UN; (b) AN100; (c) AN120; (d) AN140; and (e) AN160

Figure 4 shows plots of the (a) dielectric constant as a function of frequency; and (b) the ferroelectric hysteresis loops of UN and annealed PVDF-TrFE thin films. The dielectric constant for all annealed PVDF-TrFE thin film increased upon annealing temperatures, at frequency range of between 10^3 to 10^5 Hz. On the contrary, the unannealed thin films showed the lowest dielectric constant of 9.7 at 10^3 Hz frequency. Annealing at 100°C, the dielectric constant increased to 12 measured at the same frequency. However, a insignificant reduction in the value of the dielectric constant to 11.6 for AN120 was observed when the film was annealed at 120°C. When the annealing temperatures were further increased to 140°C and 160°C, the dielectric constant value further increased to 12.7 and 13.8 for AN140 and AN160 films respectively. The increase in dielectric constant with annealing temperatures was consistent with FESEM micrographs, in which the growth and packing of the crystallite structures were significantly improved (Figure 3). This is consistent with previous studies, which reported crystallite growth contributed to an improvement in the electrical properties of PVDF-TrFE (Lee et al., 2010; Li et al., 2008).

However, in ferroelectric analysis, thin film annealed at 120°C (AN120) showed relatively high P_r of 93 mC/m^2. Increasing the annealing temperature to 140°C, caused a reduction in the P_r value to 78 mC/m^2 for AN140 thin film. The P_r continued to decrease as the temperature in the AN160 film was annealed to 160°C. The drop in P_r was consistent with the presence of splits (Figure 3(e)). The splitting may have caused diffusion of Al from the electrodes in the film surface, and hence led to the formation of conductive tracks in the film during electrical measurements (Ismail et al., 2012).

The tightly packed elongated crystallite exhibited by AN120 thin film, coupled with its high P_r value, established that the film demonstrated superior ferroelectric properties (Lee et al, 2010). The AN120 thin film was the preferred film to be utilised for inclusion of MgO. The morphology and ferroelectricity stability of PVDF-TrFE film incorporated with various percentage loading of MgO nanofillers were investigated.

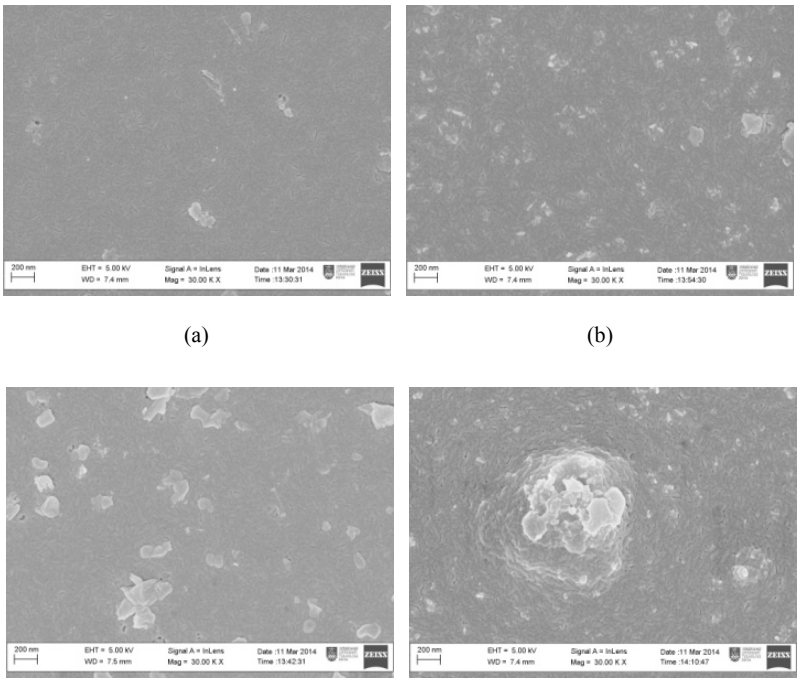


(a) (b)

Figure 4. (a) Dielectric constant, ε'; and (b) ferroelectric hysteresis loops of unannealed and annealed PVDF-TrFE thin films

PVDF-TrFE Incorporated with MgO nanofiller

PVDF-TrFE nanocomposite films (with various MgO nanofiller loadings) produced by spin coating method were then annealed at the optimised temperature of 120°C for 2 hours, and allowed to cool at ambient temperature before they were removed from the oven. The FESEM surface morphology of film for 1%, 3%, 5% and 7% of MgO nanofiller loadings at 30K magnification are shown in Figure 5. These images were observed to be consistent with the MgO imaged by previous authors (Habibah et al., 2013). Due to the low percentage of MgO nanofiller loading, 1%, minimal dispersion of the MgO nanofillers was observed, along with the PVDF-TrFE crystals observed as fine fibrils in the AN120/1%MgO film (Figure 5(a)). At 3% MgO loading, the distribution of the nanofillers in the AN120/3%MgO was apparent throughout the nanocomposite film, with insignificant agglomerations (Figure 5(b)). Specifically, the PVDF-TrFE crystals observed in the AN120/3%MgO were defined and elongated in shaped. This is due to the growth of the crystallites slightly hindered by the presence of nanofillers. These MgO nanofillers acted as nucleation sites that enhanced the formation of elongated packed PVDF-TrFE crystallites. The film formed was smooth, with relatively low porosity in the thin film. These observations are consistent with those of previous reports by other researchers, which suggested smooth non porosity film surfaces correlated to a high dielectric properties in the film (Furukawa et al., 2006; Thomas et al., 2010). However, with increase in nanofillers loading to 5%, agglomerations of MgO were significant on the AN120/5%MgO



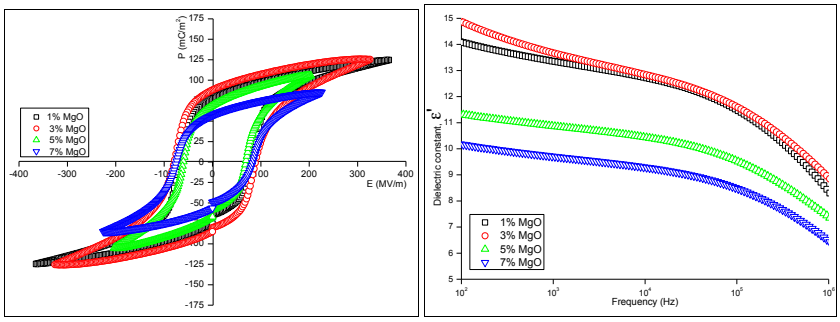
(a) (b)

(c) (d)

Figure 5. FE-SEM micrograph at 30K magnification for sample of (a) 1% (b) 3%, (c) 5%, and (d) 7% MgO nanofiller incorporated into AN120 thin film

film surfaces (Figure 5(c)), and presence of black areas may indicate porosity in the film surface. With further addition of MgO nanofillers to 7%, these agglomerations were observed to develop into massive clusters of MgO, followed by ripples-like structures of PVDF-TrFE in the AN120/7%MgO film (Figure 5(d)). However, it is interesting to note that the presence of these clusters had deterred the formation of PVDF-TrFE crystallites in the film, which were not apparent in the AN120/7%MgO film.

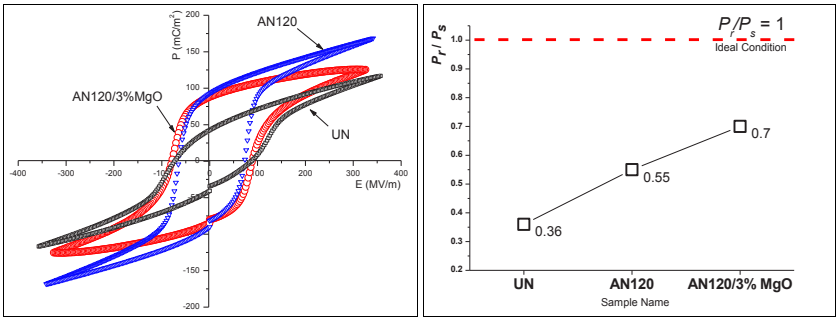
Figure 6 presents the (a) ferroelectric hysteresis loop and dielectric constant, ε'; (b) for AN120 thin films incorporated with 1%, 3%, 5% and 7% of MgO nanofiller. The ferroelectric hysteresis loops were observed in all filled nanocomposite thin films. This confirmed that though MgO nanofiller were present in the PVDF-TrFE film, the AN120/1%MgO, AN120/3%MgO, AN120/5%MgO and AN120/7%MgO nanocomposite thin films sustained their ferroelectric properties. For AN120/1%MgO, the P_r obtained was 79 mC/m^2, which was lower than the unfilled AN120 (P_r = 92 mC/m^2). Nevertheless, the highest P_r value (88 mC/m^2) was obtained from AN120/3%MgO film filled with 3% MgO. As the nanofiller loadings increased between 5% and 7%, significant reduction in P_r values was observed, which were 69 mC/m^2 and 57 mC/m^2 for AN120/5%MgO and AN120/7%MgO films respectively. This was a drop of 22 % and 35% for AN120/5%MgO and AN120/7%MgO films, respectively. This is consistent with the observation by FESEM (Figure 5), in which more than 3% MgO loaded film, agglomerates in the film were obvious. These agglomerations were more significant for 7% MgO loaded thin films. The inconsistency of surface morphology produced from the agglomerations led to the reduction of contact area between electrodes. This may have resulted in restrained ferroelectric performance for the AN120/7%MgO thin film, consistent with previous study (Thomas et al., 2010). This phenomenon was consistent with the trend observed for dielectric measurements shown in Figure 4(b). The 3% MgO loaded film showed the highest dielectric constant of 13.6 at the frequency of 10^3 Hz. An abrupt reduction of dielectric constant was observed as the MgO loadings were increased to 5% and 7%. From the dielectric and ferroelectric results, 3% loading of the MgO nanocomposite thin films (annealed at 120°C) was the optimised percentage nanofillers loading required for a PVDF-TrFE dielectric film suitable to be utilised for storage organic memory device.



(a) (b)

Figure 6. (a) Ferroelectric hysteresis loops; and (b) dielectric constant, ε' of AN120/1%MgO, AN120/3%Mgo, AN120/5% and AN120/7%MgO thin films

Figure 7(a) represents the ferroelectric hysteresis loops for unannealed (UN), PVDF-TrFE annealed at 120°C and annealed PVDF-TrFE thin film incorporated with 3% MgO nanofillers. The loops showed a significant improvement in the remnant polarisation (P_r) and saturation polarisation (P_s) upon inclusion of MgO nanofillers. However, the coercive fields (E_c) were insignificant in these films. Interestingly enough, the shaped of the hysteresis loops were rectangular-like for both annealed PVDF-TrFE (AN120) and annealed filled PVDF-TrFE (AN120/3%MgO) films. This was associated with an increased in polarisation for both annealed films (unfilled and filled), which improved the crystallinity of these films, hence, producing stability during ferroelectric switching (Furukawa, 1989). This finding was further established by observing the ratio of P_r/P_s as shown by the plot in Figure 7(b). The unannealed (UN) thin film obtained the lowest P_r/P_s ratio of 0.36. Annealed thin film without MgO (AN120), produced well saturated hysteresis loop with improved P_r/P_s ratio of 0.55, which is 53% higher in value compared with unannealed thin film (UN). Meanwhile, the P_r/P_s ratio continued to increase to 0.7 for AN120/3%MgO, which is 94% higher than annealed thin film without MgO nanofillers. This suggests that, annealed MgO nanofiller thin film produced enhanced saturation polarisation, which was evident by the improved ferroelectric stability of AN120/3%MgO. An ideal ferroelectric film should be able to produce P_r/P_s value close to unity ($P_r/P_s = 1$), thus suggesting AN120/3% MgO thin film as the favorable film developed in this study.



(a) (b)

Figure 7. a) Comparison of ferroelectric loop; and (b) P_r/P_s ratio of the UN, AN120 and AN120/3%MgO films

CONCLUSION

The study has successfully produced a stable ferroelectric thin film of P_r/P_s close to unity (value of 1). The PVDF-TrFE thin film was loaded with 3% MgO nanofiller and annealed at 120°C (AN120/3%MgO). The morphology of the 3% MgO loaded in PVDF-TrFE thin film was found to produce favourable dispersion of nanofiller in comparison to the other loading percentage films. Hence, annealing the films at temperature of 120°C was found to produce elongated crystallites of defined structures, which contributed to outstanding ferroelectric properties of the AN120/3%MgO nanocomposite film.

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