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Crack Formation on a Drying Droplet in the Presence of Static Electric Field#

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Abstract — The formation of crack patterns in a drying droplet of Laponite is studied under a DC (direct current) field. Though in the absence of a field, the droplet produces no crack on drying, it gives remarkable and reproducible results in the presence of a radial field. Cracks always appear from positive electrode and propagate towards the negative electrode and water seeps out from the negative electrode. When the centre electrode is negative, small cracks appear from outer periphery and the central portion remains almost free of crack. The behavior of cracks depends on the strength, direction and duration of application of field. The patterns show a typical memory effect different from 'Nakahara effect'. We name this as 'persistence memory'. From SEM (Scanning Electron Microscopy) images, it is seen that there are some striation marks left on the cracked surface, similar to cracks in larger systems.

Keywords : *Gel droplet, electric field, crack formation, scaling relation.*

INTRODUCTION

Formation of crack patterns in the clay mineral Laponite is of practical interest, not only due to its novel nano-structure but also due to the commercial application, like cosmetics, toothpaste, paint etc. Crack formation in a material is an inherent property of that material. If energetically profitable, it will form and we cannot resist it. Recently it has been realized that crack patterns can be controlled by introducing different types of environments and tailored to produce 'designer cracks' which may be useful in nanofabrication [1] and manufacture of surfaces with special texture. Application of DC (direct current) electric field in drying droplet of Laponite is one

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of the methods of controlling crack patterns. As the clay mineral Laponite develops surface charge in aqueous solution [2], response to an electric field must be expected. Crack pattern in aqueous solution of Laponite is strongly affected by the electric field, both DC [3, 4] and AC i.e. alternating current field [5]. A 'memory' effect is also observed. On applying a field and switching off it off, the cracks formed and found to retain the effect of the field.

MATERIALS AND METHODS

Aqueous solution of clay mineral Laponite is used. To apply electric field two electrodes made of aluminium wire of diameter ~ 0.45 mm are formed. One electrode in the form of a ring of diameter ~ 18 mm is placed on an acrylic sheet. Preparation of Laponite solution is temperature sensitive. At high concentration of solution (4 g or more Laponite in 100 ml water) as in our case the solution should be prepared below 26–27°C, above 27°C it forms clots and we cannot get uniform solution. 0.625 g of Laponite is added with 10 ml of de-ionised water while it is on a magnetic stirrer. After stirring for 15–20 sec, a droplet is deposited inside the ring and another rod like electrode is placed inside the droplet. After waiting ~ 3-4 minutes for even spreading and gelation of the solution, a field is switched on from a constant voltage power supply. Application of field before gelation may lead to excessively high current through water, effectively short-circuiting the electrodes and may damage the circuit permanently. The experimental setup is same as in [5] shown in Fig. 1a1), a2). We apply the voltage V_a (5V - 12V in most of the experiments, in very few cases the lowest voltage ~ 2V and highest ~ 20V) to the droplet for both centre positive (abbreviated as CP) and centre negative (abbreviated as CN) cases for different magnitudes. Also we study the patterns by varying the duration of application of field (τ) for each magnitude and direction of fields. Scanning Electron Microscopy (SEM) images are analyzed to get the micro-structure on the surface of the Laponite film and on the cracked surface of the film.

RESULTS

When we apply an electric field to droplet, drying starts and cracks always appear first from positive electrode for both CP and CN conditions and radially propagate towards the negative electrode. It is always observed that water seeps out from the droplet at the negative electrode after a few seconds of the application of field. This water forms near the cathode because of the reaction of Laponite clay with aluminum (or in general electro-positive elements) ions coming from cathode. This water dries up later. The patterns are more or less same as in large circular geometry [3], but there are no branches in CP and in the cross-radial crack in CN. In fact for a droplet

in CN configuration, cracks have never been seen in our experiments near the center and we may conclude that central portion is free of cracks. The nature of cracks in both CP and CN are shown in Fig. 1 b1), b2).

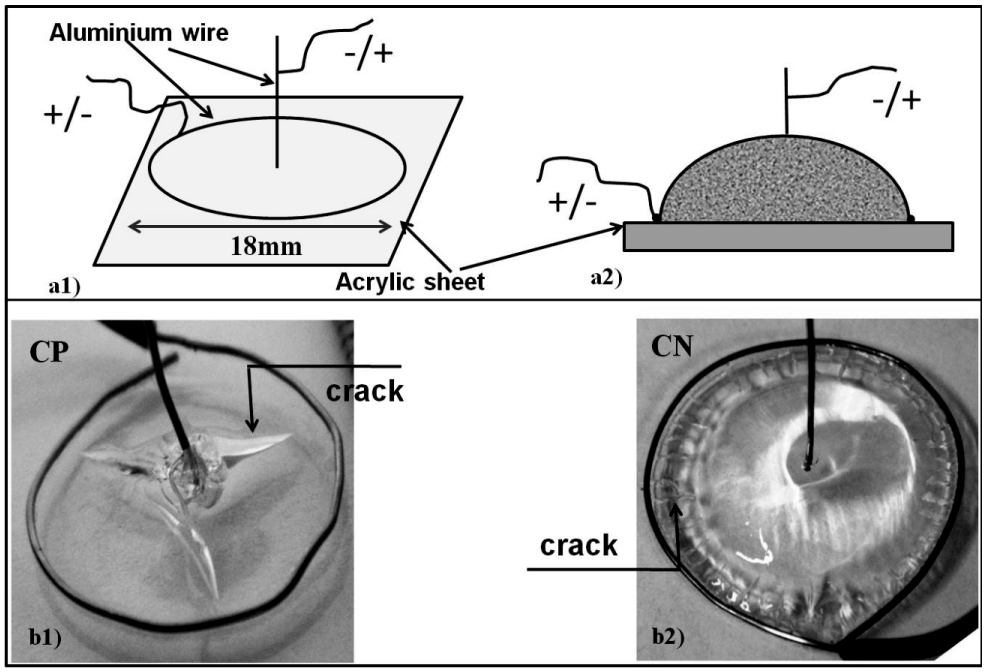

Fig. 1. a1) Experimental set up, a2) profile of droplet. b1) and b2) show the crack patterns for CP and CN respectively. The diameter of the ring (i.e. droplet) in this set up is ~ 18 mm.

For a continuous application of any fixed V_a, first crack appears at time t_c after the application and then crack number N(t) grows up to time t_{sat} while N(t) reaches N_{sat}, which is the maximum value of N(t). After t_{sat}, N(t) remains constant at N_{sat}. When we apply V_a for a short duration τ (less than t_c), the first crack appears at time t_a which is greater than t_c. For all τ for each voltage V_a, the final number of cracks (n_f) is noted. It is seen that n_f always less than or equal to N_{sat}. The quantification of results in this experiment is as follows.

1) t_c decreases as V_a increases for both CP and CN. t_c for CN is less than that for CP for a fixed V_a. The data for t_c vs. V_a for both CP and CN are listed in Table 1.

TABLE 1.

Data for t_c with V_a for both CP and CN

For CP		For CN	
V_a (V)	t_c (minute)	V_a (V)	t_c (minute)
3	60.0	2	29.0
5	17.0	3	11.0
8	9.0	5	3.0
10	5.0	8	2.0
12	3.0	10	1.0
14	1.0	12	0.9
16	0.8	14	0.7
18	0.7	16	0.5
20	0.5	18	0.4
		20	0.3

2) Current (I(t)) through the sample falls from a maximum value towards zero with time (t) for CP. For CN, it increases first to a maximum peak and then falls to zero with t. The data is given in Table 2.

3) Table 3 gives the data from which it is said that t_a decreases with both V_a and τ for both CP and CN.

4) The final number of cracks n_f is more or less constant (~ 3 or rarely 4) for all V_a and all τ in CP.

5) In CN, n_f increases with V_a and τ and reaches to saturation value which is different for different V_a. The data are given in Table 4.

6) N_{sat} increases with V_a (for CN) and it shows exponential growth behavior shown in Table 5.

7) t_{sat} rapidly deceases with V_a (for CN). This is shown in Table 6.

8) From I(t) data, the energy E_{dis} (may be assumed as heat energy) assumed for formation of first crack for all V_a in both CP an CN are calculated and it seen that E_{dis} is approximately constant for all V_a in both CP and CN. The data is given in Table 7.

TABLE 2.

Data for current I(t) vs. time t.

5V				8V				10V				12V			
CP		CN		CP		CN		CP		CN		CP		CN	
Time (sec)	Current (mA)	Time (sec)	Current (mA)	Time (sec)	Current (mA)	Time (sec)	Current (mA)	Time (sec)	Current (mA)	Time (sec)	Current (mA)	Time (sec)	Current (mA)	Time (sec)	Current (mA)
10	2.3	1	2.1	2	3.6	1	3.8	1	4.8	1	5.8	1	6.2	1	6.2
15	2.2	2	2.1	5	3.4	2	4.1	2	4.6	2	6.2	2	6.1	2	6.5
20	2.2	5	2.2	10	2.8	5	4.4	5	4.4	5	6.5	5	5.9	5	7.2
25	2.0	10	2.3	15	2.6	10	4.6	10	4.3	10	6.9	10	5.6	10	7.7
30	2.0	15	2.4	20	2.6	15	4.9	15	4.2	15	7.5	15	5.4	15	8.2
35	1.9	20	2.5	25	2.6	20	5.3	20	4.1	20	7.8	20	5.1	20	8.4
40	1.7	25	2.6	30	2.5	30	5.5	25	3.9	25	7.9	25	4.9	25	8.6
45	1.6	30	2.7	35	2.5	35	5.7	30	3.7	30	8.2	30	4.6	30	9.0
50	1.4	40	2.8	40	2.4	45	5.9	35	3.7	40	8.6	35	4.3	35	9.4
55	1.3	45	2.8	45	2.3	55	6.1	40	3.6	50	8.8	40	4.1	40	9.5
60	1.2	50	2.9	50	2.3	65	6.3	45	3.5	55	8.7	45	3.9	45	9.5
65	1.1	60	2.9	55	2.3	75	6.4	50	3.4	60	8.6	50	3.7	50	9.9
70	1.0	65	3.0	60	2.2	85	6.6	55	3.4	80	8.4	55	3.6	55	10.0
75	1.1	70	3.0	65	2.2	90	6.5	60	3.3	90	8.3	60	3.4	60	10.1
80	1.0	75	3.1	70	2.1	95	6.6	65	3.2	95	8.4	65	3.2	65	9.9
100	1.0	90	3.1	75	2.1	100	6.5	70	3.1	100	8.5	70	3.0	70	9.8

(Table-2 Contd.)

105	0.9	95	3.2	80	2.2	105	6.6	75	3.1	105	8.4	75	2.9	75	9.9
210	0.9	100	3.2	85	2.1	115	6.5	80	3.0	115	8.5	80	2.7	80	10.0
215	0.8	105	3.3	90	2.1	125	6.4	85	2.8	120	8.4	85	2.7	85	9.8
220	0.9	120	3.3	95	2.0	135	6.3	90	2.7	125	8.8	90	2.6	90	9.7
240	0.8	125	3.4	100	2.1	140	6.2	95	2.5	130	8.7	95	2.6	95	9.6
250	0.9	135	3.4	105	2.0	145	6.0	100	2.3	140	8.9	100	2.6	110	9.7
260	0.8	140	3.5	110	2.0	155	5.8	105	2.1	145	8.7	105	2.5	135	9.6
270	0.9	160	3.6	115	1.9	160	5.7	110	2.0	155	8.8	110	2.5	145	9.1
280	0.8	215	3.7	120	1.9	170	5.6	115	1.9	180	8.5	115	2.4	155	9.2
325	0.8	220	3.7	125	1.9	180	5.4	120	1.9	185	9.3	120	2.3	165	9.1
330	0.7	225	3.8	130	1.8	185	5.3	125	1.8	195	9.2	125	2.3	175	8.8
335	0.8	310	3.8	135	1.9	205	5.1	130	1.7	200	9.0	130	2.3	185	8.6
340	0.8	315	3.7	140	1.9	207	4.8	135	1.7	205	9.1	135	2.3	195	8.5
345	0.7	320	3.8	145	1.8	209	5.4	140	1.6	210	9.0	140	2.2	205	8.4
350	0.8	330	3.7	150	1.8	211	5.5	145	1.6	225	8.8	145	2.2	215	8.2
360	0.7	335	3.8	155	1.8	213	5.8	150	1.6	230	8.6	150	2.2	225	8.1
385	0.7	355	3.7	160	1.7	215	6.0	155	1.5	240	8.4	155	2.1	255	7.9
390	0.6	405	3.7	165	1.7	221	5.9	160	1.5	250	8.2	160	2.0	285	7.2
425	0.6	410	3.6	170	1.6	227	5.8	165	1.4	265	7.8	165	2.0	315	6.3
430	0.5	440	3.5	185	1.6	240	5.7	170	1.4	270	7.9	170	1.9	345	6.2
440	0.6	470	3.3	190	1.5	250	5.5	180	1.4	280	7.4	175	1.8	375	5.7

(Table-2 Contd.)

480	0.6	500	3.1	195	1.5	265	5.3	195	1.4	285	7.3	190	1.7	405	5.1
490	0.5	530	3.0	200	1.4	280	5.2	215	1.3	290	7.1	210	1.6	435	4.8
550	0.5	560	2.7	265	1.5	295	5.0	235	1.3	305	6.8	250	1.4	465	4.7
555	0.4	590	2.5	270	1.1	355	4.6	255	1.2	310	7.0	280	1.2	495	4.6
560	0.4	620	2.3	275	1.0	415	4.2	275	1.1	325	6.7	295	1.0	555	3.9
590	0.5	680	2.0	280	1.0	475	3.6	335	1.1	340	6.3	310	0.9	615	3.2
620	0.4	740	1.7	285	0.9	535	2.8	365	1.1	350	5.7	340	0.8	675	2.5
710	0.4	800	1.5	290	0.8	595	2.0	425	0.9	380	5.0	400	0.7	735	2.9
770	0.5	860	1.3	295	0.7	655	1.1	485	0.8	410	4.7	460	0.6	795	1.7
830	0.3	920	1.1	305	0.5	715	0.9	545	0.7	470	4.1	520	0.5	855	1.4
950	0.3	980	1.0	315	0.4	775	0.8	605	0.6	500	3.9	580	0.4	915	1.2
1010	0.4	985	1.1	330	0.3	835	0.6	665	0.5	620	2.7	820	0.3	975	1.1
1070	0.4	995	1.0	340	0.2	955	0.5	725	0.5	800	2.4	1000	0.2	1035	1.0
1130	0.3	1055	0.9	760	0.2	1075	0.6	785	0.4	980	1.6	1060	0.3	1095	0.9
1190	0.2	1175	0.8	820	0.1	1135	0.3	965	0.4	1100	1.2	1120	0.2	1155	0.8
1200	0.2	1200	0.8	1200	0.1	1200	0.4	1200	0.3	1200	1.1	1200	0.2	1200	0.7

TABLE 3.

Data for t_a with τ for different V_a for both CP and CN

5V				8V				10V				12V			
CP		CN		CP		CN		CP		CN		CP		CN	
τ (sec)	t_a (min)	τ (sec)	t_a (min)	τ (sec)	t_a (min)	τ (sec)	t_a (min)	τ (sec)	t_a (min)	τ (sec)	t_a (min)	τ (sec)	t_a (min)	τ (sec)	t_a (min)
15	237	15	240	15	195	15	141	15	193	15	78	15	190	15	30
30	180	30	117	30	173	30	82	30	171	30	45	30	168	20	15
60	160	60	90	60	148	60	51	60	144	45	20	60	140	25	2
180	151	180	3	180	130	120	2	180	110	60	1	180	80	30	0.5
300	134	300	3	300	116	300	2	300	50	300	1	300	24	300	0.5
4800	80	4800	3	3120	52	4800	2	1500	25	4800	1	720	12	4800	0.5
8000	80	8000	3	6000	52	8000	2	5000	25	8000	1	5000	12	8000	0.5
10000	80	10000	3	10000	52	10000	2	10000	25	10000	1	10000	12	10000	0.5
14400	80	14400	3	14400	52	14400	2	14400	25	14400	1	14400	12	14400	0.5

TABLE 4.

Data for n_f with τ for different V_a

5V		8V		10V		12V	
τ(sec)	n_f	τ(sec)	n_f	τ(sec)	n_f	τ(sec)	n_f
15	1	15	2	15	3	15	3
30	4	30	5	30	8	30	10
60	6	60	9	60	13	60	14
180	12	180	20	180	28	180	30
300	18	300	32	300	36	300	39
1560	30	720	40	480	43	360	45

TABLE 5.

Data for N_{sat} with applied voltage V_a

V_a (V)	N_{sat}
2	13
3	21
4	26
5	30
6	33
7	37
8	40
10	43
12	45

9) SEM images show that there are micro cracks in the film of Laponite and the length and density of micro cracks depend on the strength and polarity of field.

10) Striation marks form on the cracked surface.

11) Laponite shows a ‘memory’ in electric field.

TABLE 6.

Data for t_{sat} with applied voltage V_a

V_a (V)	t_{sat} (minute)
2	60
3	45
4	41
5	21
6	19
7	17
8	13
10	10
12	6

TABLE 7.

Data for energy dissipated E_{dis} with applied voltage V_a

V_a (V)	E_{dis} (Joule)
–12	4.5
–10	4.7
–8	4.9
–5	4.8
+5	3.4
+8	4.7
+10	4.9
+12	4.4

Variation of t_{sat} with V_a :

The time (t_{sat}) at which the number of cracks N(t) saturates to N_{sat} for continuous field in CN condition depends on the magnitude of applied voltage V_a (Table 6). We find that t_{sat} follows approximately the exponential relation.

$$t_{sat}(V_a) = t_{s0} * \mathrm{Exp}\left(-\frac{V_a}{V_{s0}}\right) \quad (1)$$

Where, t_{s0} = 100 minutes and V_{s0} = 4V. Fig. 2 shows the variation of t_{sat} with V_a. The uncertainty in the time is ~30 sec which is smaller than the size of the symbols.

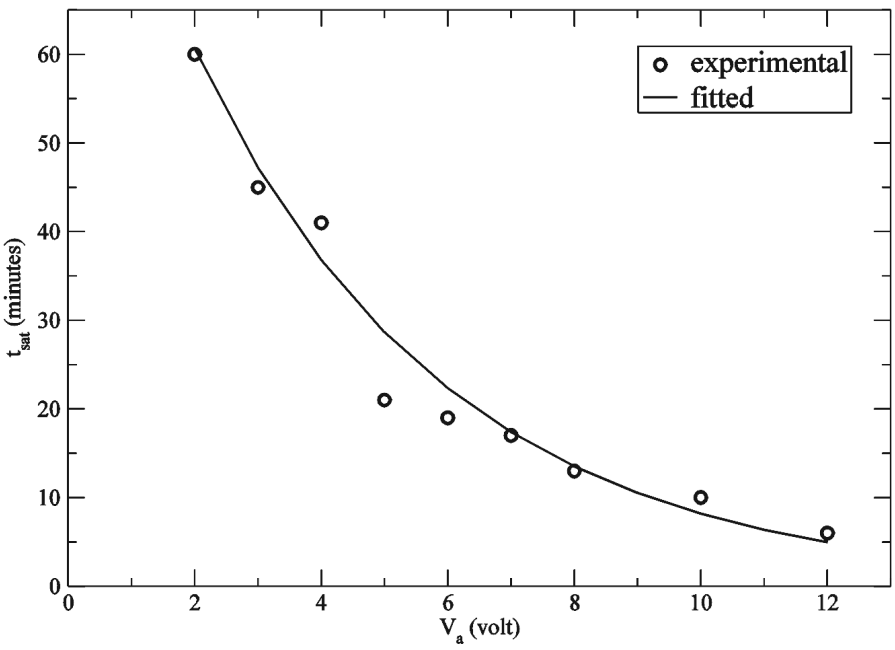

Fig. 2. Plot of t_{sat} vs. V_a. Open circles represents the experimental points and solid line represents the fitted one. The maximum error in measuring t_{sat} is ~ 30 sec which is smaller than the size of the symbol.

Energy Dissipated E_{dis} to Produce First Crack :

Application of electric field increases the temperature of the sample due to the heat energy produce because of flow of current through the sample. This heat energy is dissipated to form crack. Energy dissipated E_{dis} (assumed as heat energy) for the appearance of first crack is calculated from the I(t) vs. t data by using rule

(2)

From experimental data E_{dis} is calculated numerically by using Trapezoidal rule for different voltages for both CP and CN conditions. For all voltages for both CP and CN conditions, E_{dis} remains more or less constant (Table 7) as shown in Fig. 3, but only the point for 5V in CP deviates somewhat. If we consider the theoretical value of t_c for 5V in CP to calculate E_{dis}, the value will be closer to the average constant value which is ~ 4.624 Joule. Repeated experiments also show the above mentioned deviation. This might be an error in determining t_c for 5V in CP. In Fig. 3, the solid line represents the average value of E_{dis}.

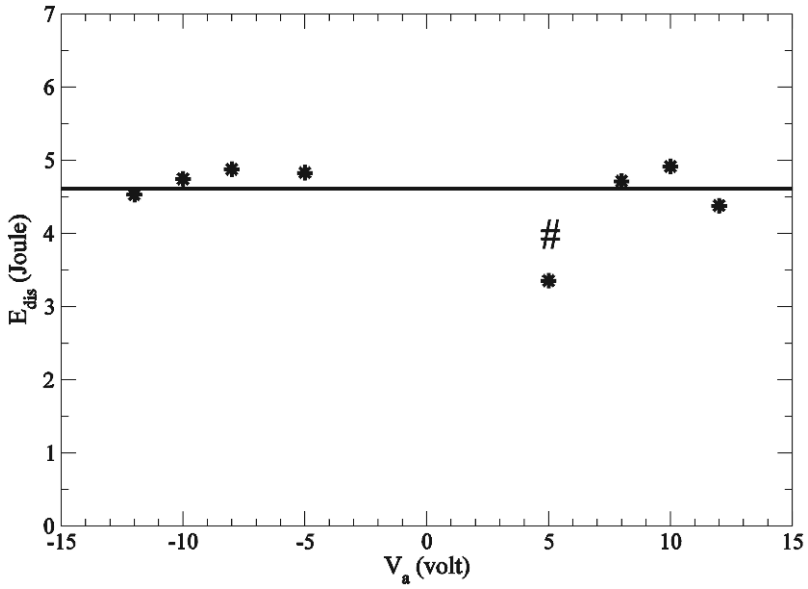

Fig. 3. Energy dissipated E_{dis} upto appearance of first crack with applied voltage V. # represents the value of E_{dis} for 5V in CP, calculated by taking the theoretical value of t_c. Solid line represents the average value of E_{dis} (~ 4.624J).

SEM Images and Striation Marks on the Cracked Surface :

When a droplet of Laponite is deposited on an acrylic sheet without any electrode, no visible cracks appear at all. But if we see the dried film under Scanning Electron

Microscope (SEM), there are huge numbers of micro cracks present on the film. The SEM images of the dried films, produced in the presence of field, show that as magnitude of V_a increases, length and density of micro cracks decrease (Fig. 4 b), c)). The number density and length of micro cracks are greatest for the film without field shown in Fig. 4 a). Comparisons of Fig. 4 b) with d) and Fig. 4 c) with e) say that the number of micro cracks near negative electrode is greater than that near positive electrode.

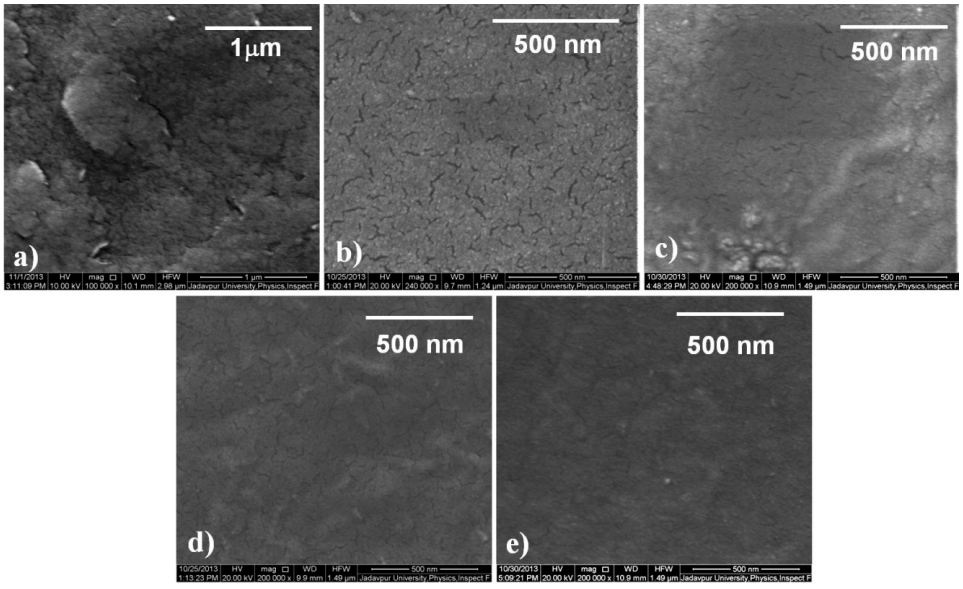

Fig. 4. SEM Images for film of Laponite a) without electric field, b) near negative electrode with 15V DC, c) near negative electrode with 20V DC, d) near positive electrode with 15V DC and e) near positive electrode with 20V DC. Note that a) has much lower resolution, so cracks seen here are much larger than in other cases.

Fig. 5 a) and b) show the SEM images of striation marks on a cracked surface for 20V DC near positive electrode and near negative electrode respectively. In Fig. 5, the striation marks curve down to the lower surface of the film and arrow marks represents the direction of propagation of cracks. By drawing normal to the striation marks, we get the crack front [6] along the upper surface of the film. These striation marks are not visible by naked eye.

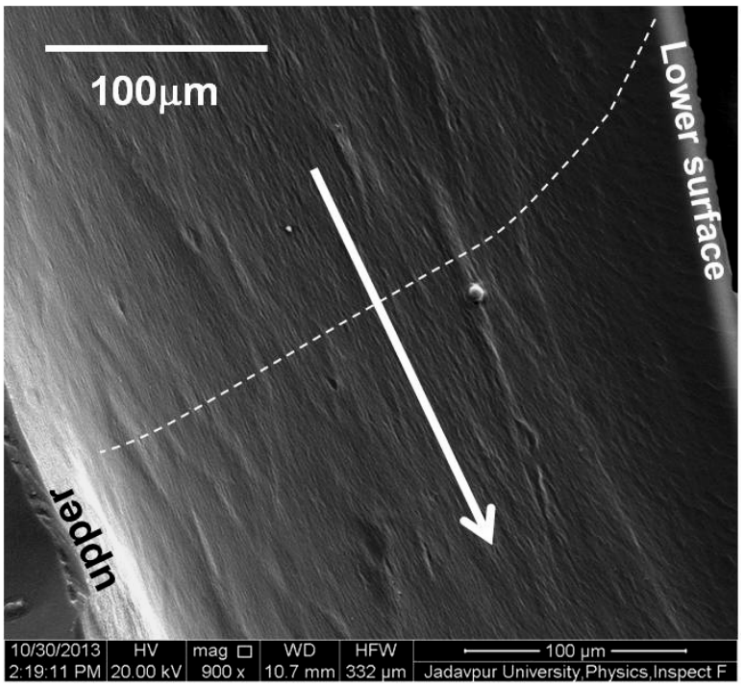

Fig. 5. This figure represents the cracked surfaces of a crack near negative electrode for 20V DC. The arrow represents the direction of propagation of crack. Striation marks curve down to the lower surface of film. The dotted line represents the normal drawn on the striation marks, which is the crack front.

τVariation of Field :

If the field is switched off before appearance of first crack i.e. before t_c, the final crack patterns are more or less similar as that for continuous field, but here the crack appearance time t_a is different from t_c. In CP, the number of cracks (n_f) for different τs and for all V_a, is almost constant (~ 3). In CN, n_f increases with increase of τ for any fixed V_a. All the results related to τ variation have been reported in the earlier publication [5]. It seems that there is some 'memory' in the Laponite film.

DISCUSSIONS

The formation of crack patterns is the result of competition of drying rate and relaxation rate [7]. Adhesion between substrate or electrode with the sample is also

important for crack formation. The significant feature of the results of the Laponite droplet is that most of properties of the droplet of Laponite can be described by some simple laws as reported in [5]. Here t_{sat} for CN follows the exponential decay rule. The average value of energy needed to create first crack is almost constant for all voltages in both CP and CN conditions. From the constant value of E_{dis}, we can find out the time of appearance of first crack (t_c) for any voltage either in CP or in CN condition, provided the current distribution with time is known. Greater the temperature increase rate in the sample due to current flow, faster is the crack appearance (i.e. heat energy produced due to current flow reaches to E_{dis} fast) and propagation.

The 'memory' of the system of clay mineral Laponite i.e. the response of electric field to the system long after the field is switched off, is a quite remarkable effect.

One tentative explanation reported in the earlier publication [5] is as follows. When the voltage is applied, negatively charged laponite particles are attracted to the positive electrode and start to repel each other. This results in initiation of incipient micro cracks, not visible to the naked eye. These micro cracks act as micro notches which set the direction for crack propagation on drying. This is similar to the intentional creation of a small notch, which is a well-known process for guiding cracks when studying mechanical fracture. If the applied voltage is switched off at this point, there is no further perturbation to the initiator (i.e., the micro notch) and the subsequent macro crack grows as its continuation. On the other hand, if the voltage is kept on, perturbation to the crack initiator may result from potential redistribution due to cracks already formed. We call this 'memory' as 'persistence memory'. The word 'persistence' implies that the material behaves like a continuum, so that the micro-crack initiator formed when the field was on *persists* in its original direction of propagation after switching off of the field.

The memory effect in the presence of electric field is somewhat different from 'Nakahara effect' in the presence of mechanical perturbation [8]. In 'Nakahara effect' after applying the perturbation, if a portion of the large sample is cut and isolated from the rest, the isolated portion also retains its memory as well as the whole sample. But in our case, the whole sample shows memory effect but if we cut out and isolate a portion (this has been tried on a large sample, not on a small droplet) of the sample from the rest after the removal of application of an electric field, the isolated portion does not show any signature of the application of field. So the 'memory effect' in our case is a global effect for the whole system, not a local effect.

SEM images show that as voltage increases, the number and length of micro

cracks decrease. Again for a fixed voltage, the number of micro cracks is greater near negative electrode than that near positive electrode. In the presence of field, stress developed inside the film releases to form large cracks and no more stress left to create more number of micro cracks. As field strength increases, the probability for the formation of large cracks increases reducing the possibility of forming micro cracks. So the greatest number of micro cracks is obtained for 0V i.e. without field. The same is true for more micro cracks near negative electrode than that near positive electrode. Field induced large cracks form from positive electrode and hence there is less probability to form micro cracks. Note that resolutions are different in different SEM images as in Fig. 4 and corresponding scales are shown in the images. In 0V, average crack length is ~ 300–400 nm whereas at 15V, the average crack length is ~ 100–150 nm.

Temperature and humidity affect the drying rate significantly. As the temperature increases or humidity decreases, drying rate increases. The experiment should ideally be done under fixed temperature and humidity. In our experiment the temperature ranges from 23°C to 26°C and humidity ranges from 36% to 56% as we could not control the ambient conditions exactly.

CONCLUSIONS

From the above study we can conclude the following :

1) Cracks always appear from positive electrode and propagate towards the negative electrode. Crack patterns depend on the strength and direction of the field.
2) Field effect is also present in the microscopic level. Enhancement of field strength reduces the micro cracks. Application of electric field concentrates stress and rapidly creates the large cracks, the stress being thus released, much less micro-cracks form. Again in the presence of field, micro cracks are more crowded near negative electrode than that near positive electrode. This is also because of the large cracks appearing from positive electrode which release the stress precluding micro-cracks, but more micro-cracks are obtained near negative electrode, where there are no large cracks. From this study it may be possible to use an electric field to produce crack-free regions at certain positions on a film.
3) There is some ‘memory’ in Laponite clay that may be named as ‘persistence memory’ somewhat different from ‘Nakahara effect’ [8] for mechanical perturbation, or from the memory in the presence of magnetic field [9].

In our case, an isolated portion of the sample does not have memory, so the memory effect is a global effect for the whole system, rather than a local effect.

4) Crack opening propagates generally along the upper surface of the film. This may be concluded from the striation marks left on the cracked surface.

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