0 0 0 0 3 9 0 0 2 3 2

LBL-1817

c. 1

MEMORY SWITCHING EFFECTS IN As-Te-I CHALCOGENIDE GLASS

RECEIVED
LAWRENCE
RADIATION LABORATORY

OCT 22 1973

LIBRARY AND
DOCUMENTS SECTION

Robert M. Jecmen

(M. S. Thesis)

July 1973

Prepared for the U. S. Atomic Energy Commission
under Contract W-7405-ENG-48

For Reference

Not to be taken from this room

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY

LBL-1817
c. 1

DISCLAIMER

This document was prepared as an account of work sponsored by the United States Government. While this document is believed to contain correct information, neither the United States Government nor any agency thereof, nor the Regents of the University of California, nor any of their employees, makes any warranty, express or implied, or assumes any legal responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by its trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof, or the Regents of the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof or the Regents of the University of California.

0 0 0 0 3 9 0 0 2 0 3

Table of Contents

ABSTRACT v

I. INTRODUCTION 1

II. SAMPLE PREPARATION 3

III. POWER APPLICATION 6

IV. SWITCHING OBSERVATIONS 7

A. Mechanism of Switching to the High Resistance State . . . 7

B. Switching Negative Electrode Filaments 9

C. Filament Broadening Observation 15

V. CYCLING OBSERVATIONS 18

VI. GROWTH RATE STUDY 31

VII. REVERSE POLARITY STUDY 33

VIII. CONCLUSIONS 36

ACKNOWLEDGEMENTS 37

REFERENCES 38

0 0 0 0 3 9 0 0 2 0 4

MEMORY SWITCHING EFFECTS IN As-Te-I CHALCOGENIDE GLASS

Robert M. Jecmen

Inorganic Materials Research Division, Lawrence Berkeley Laboratory and Department of Materials Science and Engineering, College of Engineering; University of California, Berkeley, California

ABSTRACT

The mechanism of switching an $As_{0.53}Te_{0.43}I_{0.04}$ glass from the memory to the high resistance state and the effect of cycled switching on the chemical composition and electrical properties of the glass was studied. Switching to the high resistance state was found to occur as a result of Joule heating and subsequent melting of the crystalline filamentary path. Of the two filamentary segments which form upon completion of the memory state, the negative electrode segment is more stable upon switching to the high resistance state. The negative electrode segment is believed do be crystalline As_2Te_3. Cycled switching can be carried to high repetitions without substantially changing the chemical composition or electrical properties of the glass and its memory state provided the proper choice of switching and growth current pulses are made.

0 0 0 0 3 9 0 0 2 0 3

I. INTRODUCTION

Ovonic memory switching (OMS) occurs in a large number of semiconducting glasses.[1,2,3] The switching process is characterized by a high resistance state when a voltage less than a critical value ($V_{threshold}$) is applied across a sample. When the voltage is increased to the threshold value, the glass switches to a low resistance state. This low resistance state remains stable as the voltage is reduced to zero provided the voltage pulse was maintained at the threshold value for a critical length of time. This memory state is characterized by a visible filamentary path in the glass matrix. It is generally accepted that this filamentary path is a crystalline phase of much higher conductivity than the glass matrix. Switching from the conductive state back to the original high resistance state can be accomplished by passing a current pulse greater than a critical value (I_c) which is in turn greater than the current magnitude used in forming the memory state (I_f).

This switching process from the conductive memory state back to the original high resistance state has not been studied in great detail. It has been proposed that this portion of the switching cycle occurs as a result of Joule heating and subsequent melting of the crystalline filament.[4] The cessation of the switching pulse (I_s) quenches the filamentary region in the glassy high resistance state. The observations, however, have been limited to the visual disappearance of the filamentary path upon switching to the nonconductive state.

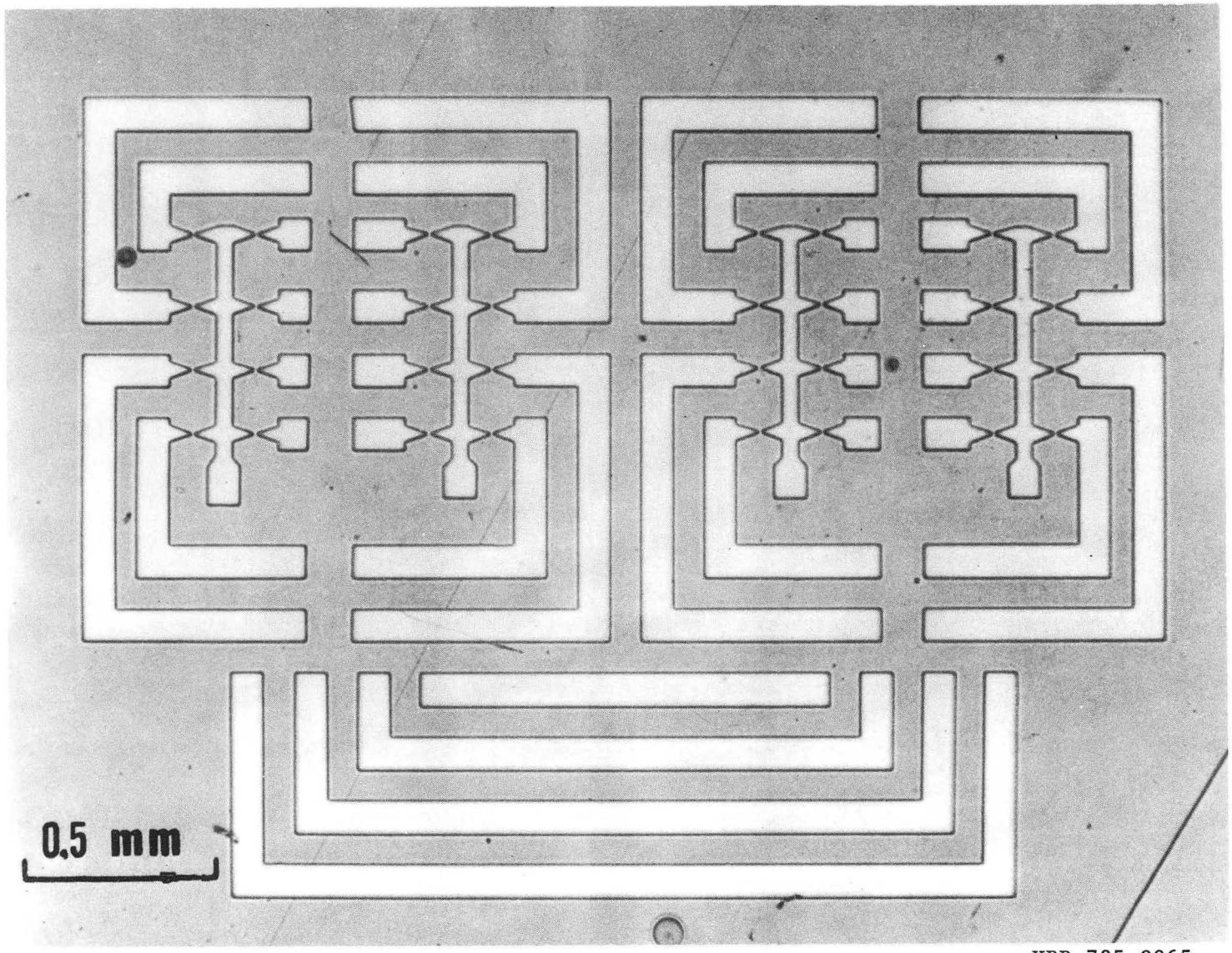
Successful application of this memory switching phenomena is contingent upon stable performance during repeated switching. The effect of repeated cycling on the composition, crystallinity, and electrical properties of the memory state as well as its effect on the critical electrical parameters of switching must be determined.

0 0 0 0 3 9 0 6 2 0 6

II. SAMPLE PREPARATION

A chalcogenide glass with composition $As_{0.53}Te_{0.43}I_{0.04}$ was selected for study. The glass composition has been reported to display memory switching by Pearson in 1970.[5] The components were weighed under a nitrogen atmosphere and placed in a quartz capsule. The capsule was evacuated to 10^{-3} Torr, cooled in liquid nitrogen to prevent vaporization, and sealed with an acetylene torch. The sealed capsule was then placed in a protective molybdenum cylinder which was rotated in a furnace at 1000°C for 72 hours. The melt was cooled initially to 500°C and finally quenched in water while still protected by the molybdenum cylinder. This provided a slower quench and avoided severe cracking due to thermal shock. Small pieces of glass were chipped from the resulting ingot, mounted in an epoxy resin and polished.

An electrode mask assembly was designed so that metallic electrodes could be vapor deposited on the surface of the polished glass sample. The design was such that any of 32 electrode gaps could be independently activated by applying an external power source to the proper points on the assembly. (See Fig. 1) A photolithographic process was used in which the electrode design was cut out of a transparent ruby lith paper, photographed and reduced in size 117 times. The glass sample was then covered by a Shipley photoresist solution, dried under an infrared lamp, placed in contact with the photoreduced electrode mask, and exposed to an intense light source. The exposed sample was placed



XBB 735-3365

Fig. 1. Vapor deposited electrode assembly.

0 0 0 0 3 9 0 0 2 0 7

in a developer for several seconds, washed, and dried in nitrogen. This left the dried photoresist solution only in those areas which were protected from the exposure by the mask. The sample was then placed under a vacuum of 10^{-6} Torr and Sn was vapor deposited over its surface. Finally, the sample was washed in acetone which dissolved the photoresist solution layer, lifting the deposited metal off with it, and leaving the well defined electrode pattern on the glass surface.

III. POWER APPLICATION

The thirty two electrode gaps on the glass sample were activated one at a time by placing two tungsten wire probes in mechanical contact with the appropriate areas on the deposited metal electrode. The probes were positioned through the use of an optical microscope and two translatable stages. One stage positioned the positive tungsten probe with respect to the optic axis of the microscope. The second stage translated the sample.

The power was applied in pulses by a constant current source. The circuitry was designed such that current pulses from 0.1 ma to 6.0 ma could be chosen. The pulse shape could also be regulated from 0.5 ma to 1 second for each of the rise, hold, and fall segments of the current pulse.

0 0 0 0 3 9 0 0 2 0 8

IV. SWITCHING OBSERVATIONS

A. Mechanism of Switching to the High Resistance State

It has been found that a current pulse of greater magnitude than that used in forming the memory state is necessary to switch back to the high resistance state. The switching process is accomplished by passing a single pulse of current through the memory state filament. The magnitude of the pulse (I_s) necessary to switch to the high resistance state is a function of the current of formation, I_f. In general, $I_s \simeq 1.5\ I_f$. This is a rough relationship and is seen to vary ± 0.4 ma upon repeated testing. In all cases, however, $I_s > I_f$.

The switching process from the memory state to the high resistance state is characterized by a visual disappearance of the memory state filament. Figure 2a shows the memory state filamentary path connecting the two vapor deposited metal electrodes on the surface of the glass sample. The formation current magnetude was 0.8 ma, and the pulse rise, hold, and fall times were 5 ms, 20 ms, and 5 ms respectively. In Fig. 2b, the memory filament is seen to disappear after a switching pulse I_s = 2.0 ma had been applied.

An electron microprobe experiment was carried out to determine the changes in composition which occurred in the filamentary region as a result of switching to the high resistance state. The original memory filament shows a dramatic increase in Te concentration compared to the matrix glass level. This is accompanied by a corresponding decrease in As and I in the same region. This finding is consistent

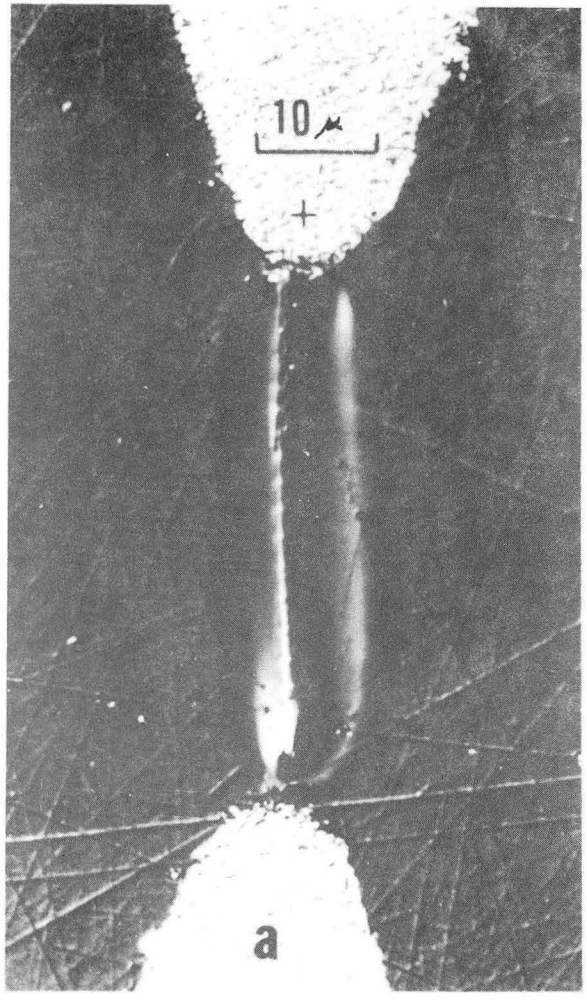
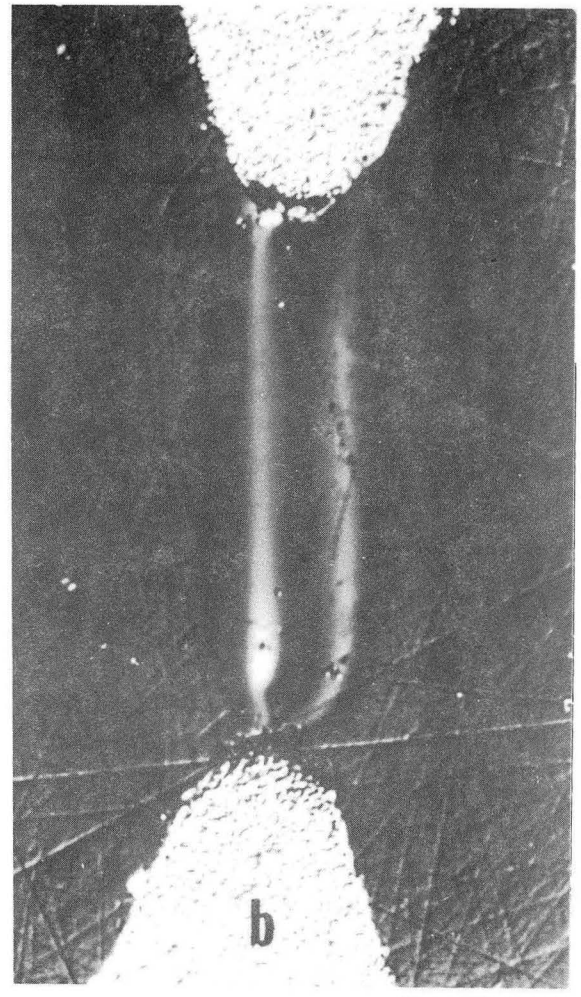


Fig. 2(a) Completed memory state filament.



XBB 735-3366

Fig. 2(b) Loss of filament upon switching to the high resistance state.

with Roberts'[6] determination of the memory state as pure crystalline Te. The remaining As and I counts arise as a result of the electron beam penetrating through the filament and producing counts from the glassy region underneath the memory path. The filamentary region, after switching to the high resistance state, also shows an increase in Te concentration over the matrix glass level. However the increase is not as dramatic as in the memory state. Therefore, switching to the high resistance state caused a decrease in the Te concentration and a resulting dispersion of the Te concentration profile. (Fig. 3)

The observations of $I_s \sim 1.5\ I_f$, the visual loss of the filamentary path upon switching, and the decrease in magnitude and dispersion of the Te concentration profile indicate that a Joule heating and melting mechanism is responsible for the switching process from the memory to the high resistance state. Calculations show that the energy input from a switching pulse of 2.0 ma with rise, hold, and fall times of 5 ms, 20 ms, and 5 ms respectively is far in excess of that necessary to melt a pure Te crystalline filament of the size found in this experiment. Thus a melting mechanism is likely which is a sensitive function of the thermal conductivity properties of the surrounding region.

B. Switching Negative Electrode Filaments

Often a memory filamentary path is composed of two segments. One grows from the positive electrode and is characterized by a segmented structure caused by the current pulsing during formation.

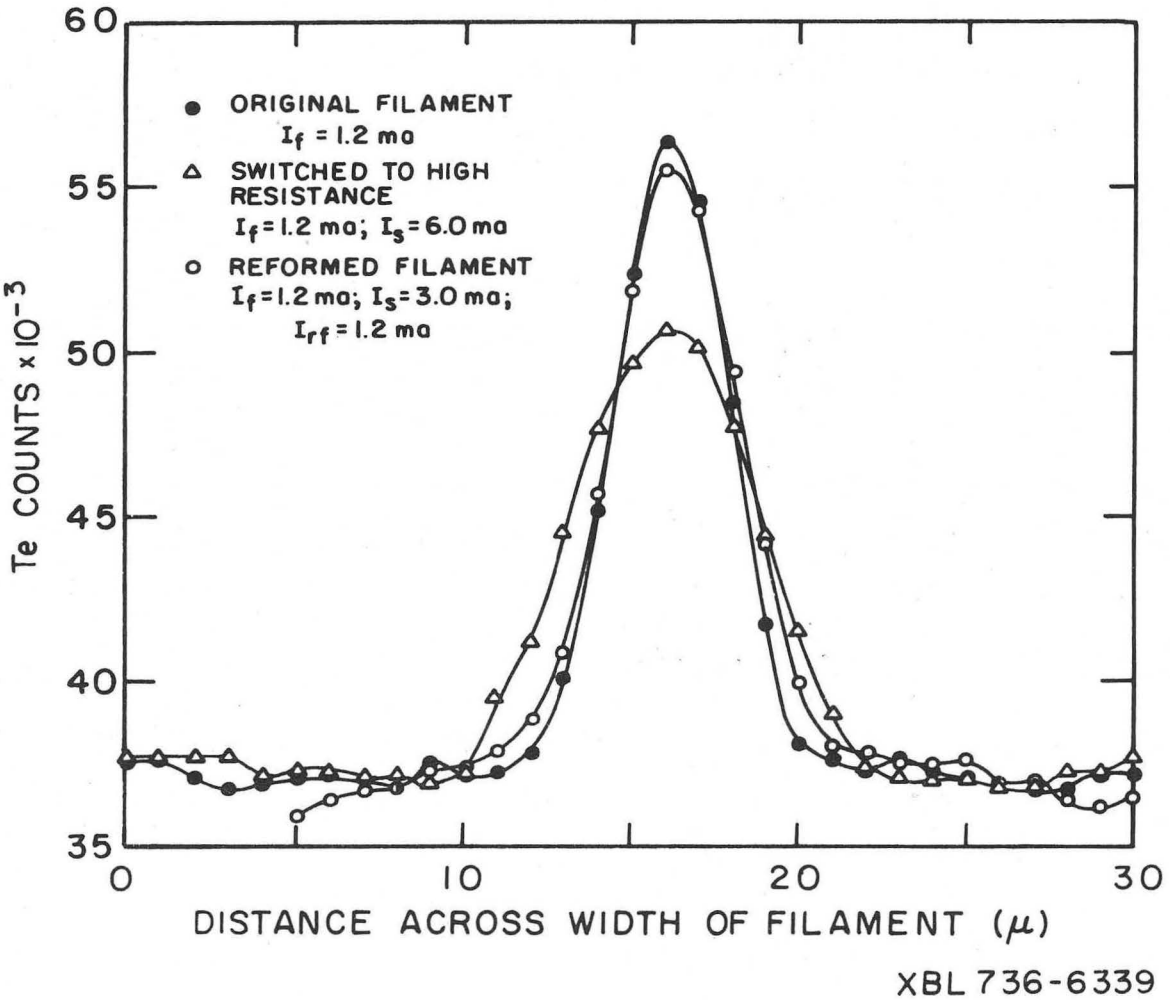
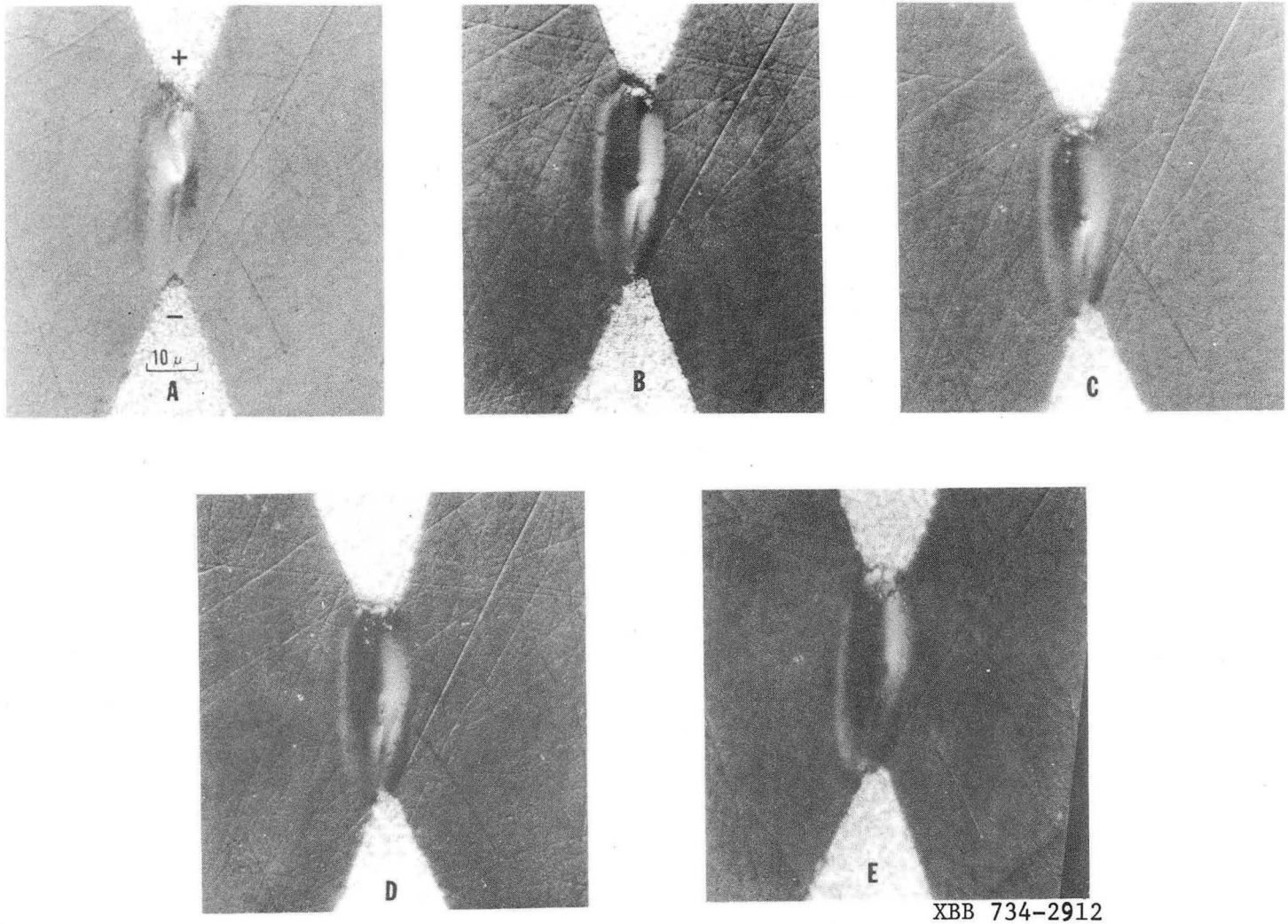


Fig. 3. Electron microprobe analyses of Te concentration in filamentary region of an original filament, a filamentary region switched to the OFF state, and a reformed filament.

A less common phenomena sometimes occurs in which a filamentary path grows from the negative electrode as well. This part of the filament is usually very straight, subsurface, and lacking detailed segmented structure. When the two different filamentary paths meet, the memory state is achieved. These two filamentary segments, referred to as positive and negative electrode filaments, exhibit different properties upon switching to the high resistance state. In Fig. 4a, a completed memory state can be seen composed of both positive and negative electrode segments. Upon passing a current pulse of 2.0 ma (Fig. 4b), the positive electrode filament is seen to disappear whereas the negative electrode segment remains stable. Figures 4c, 4d, and 4e show the negative electrode filament to be stable upon further switching pulses of the same current magnitude and pulse geometry.

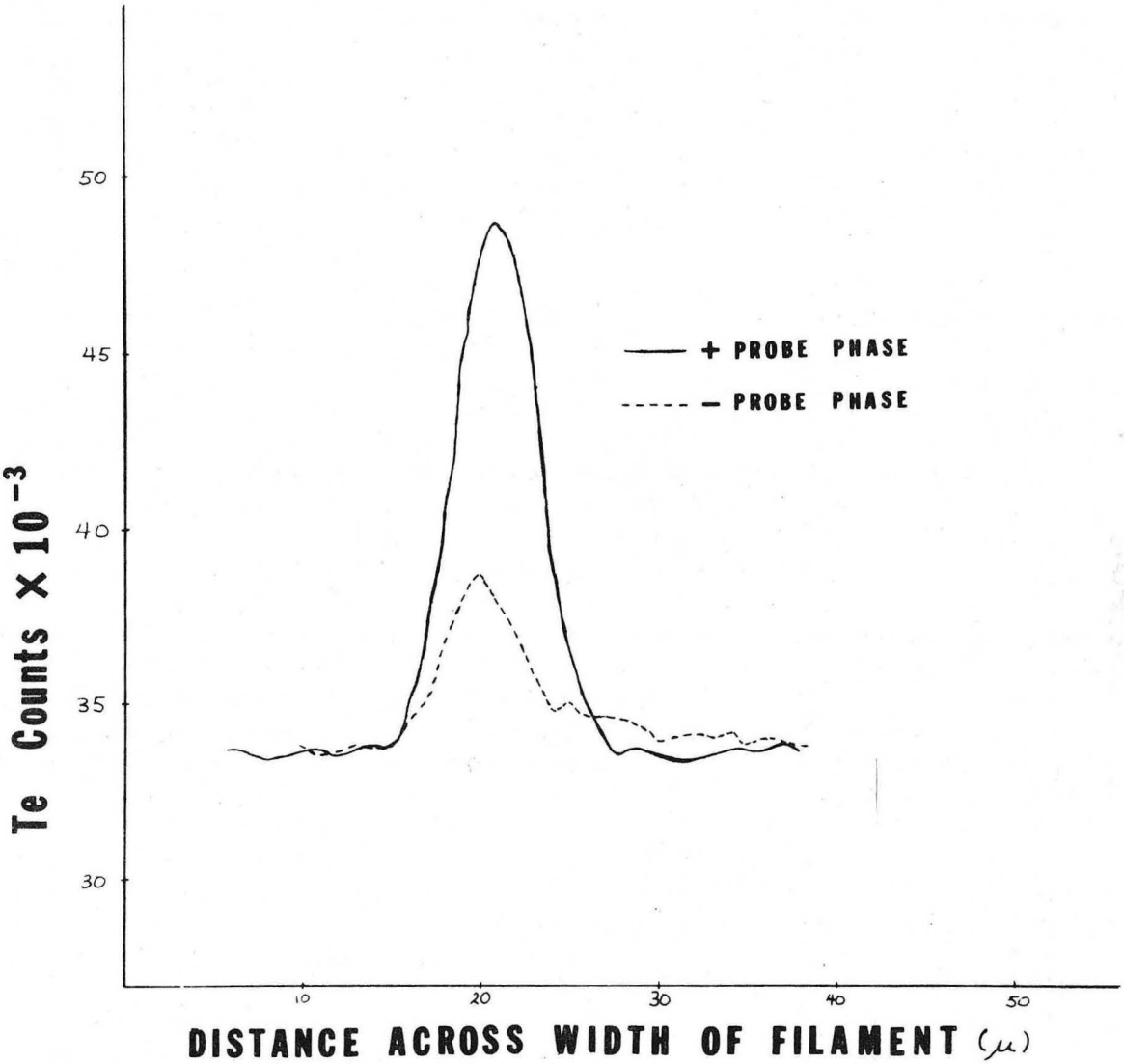
An electron microprobe analysis of the Te concentration in the negative electrode filament shows it to be higher than the matrix glass level but substantially less than the positive electrode filament (Fig. 5). This cannot be taken as immediate proof of a different composition because of the beam penetration phenomena mentioned before. Thus variation in filament thickness could cause the difference in Te counts observed using the electron microprobe. However, this experiment is completely reproduceable, so chance thickness variations are extremely unlikely.

These findings indicate that the negative electrode filaments may be a different phase than the crystalline Te positive electrode filament. The increased stability upon switching can be explained



XBB 734-2912

Fig. 4(a). Original memory state exhibiting positive and negative electrode filamentary segments.
4(b-c) Negative electrode remains stable upon subsequent switching pulses of 2.0 ma.



XBL 734-5998

Fig. 5. Electron microprobe analysis of Te concentrations of positive and negative electrode filamentary segments.

if the negative electrode filament was crystalline As_2Te_3. It is reasonable to assume that crystalline As_2Te_3 well have a substantially higher conductivity than crystalline Te because of arsenics' relatively high conductivity. Thus, for a similar cross section, less Joule heating will result in an As_2Te_3 negative electrode filament than in a Te positive electrode filament. Lower temperatures will result and this will cause the observed increased stability upon switching to the high resistance state.

The subsurface nature of negative electrode filaments also improves the heat conduction properties away from the current carrying filamentary path. As was mentioned before, the heat conduction properties are important factors in determining the current magnitude necessary for switching to the high resistance state. Thus higher currents will be necessary to build up the same temperatures as that found in a surface filament. This is undoubtedly another important factor causing increased stability of the negative electrode filaments.

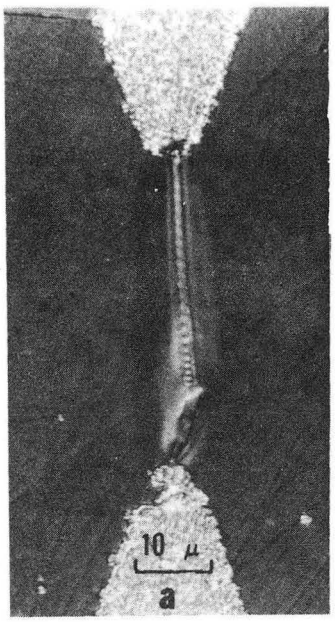
The melting point of As_2Te_3 is approximately 50°C lower than Te. However, the combined effects of its increased conductivity and subsurface nature should more than compensate for its lower melting point.

The As_2Te_3 negative electrode phase model is also necessary to explain the electron microprobe data. The microprobe analysis indicated the negative electrode has a higher Te concentration than the 53 atomic percent found in the matrix glass but less than the 100% Te found in the positive electrode phase. The 60 atomic percent Te found in As_2Te_3 lies in the proper range.

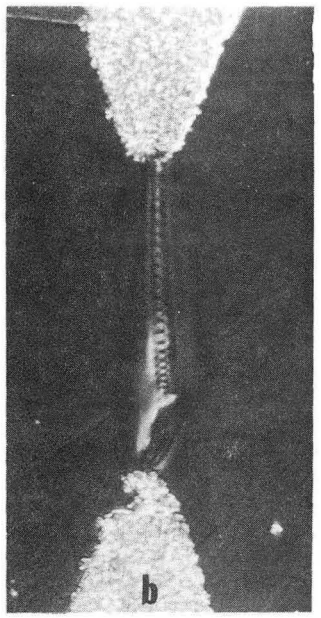
0 0 0 0 3 9 0 0 2 1 1

C. Filament Broadening Observation

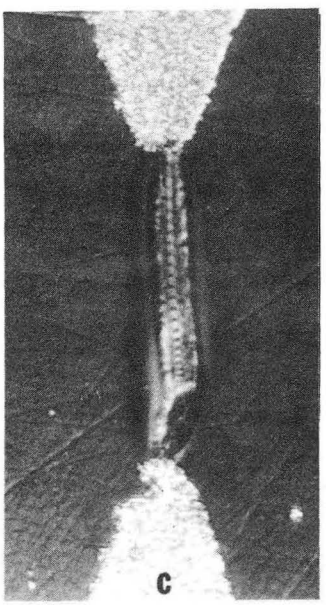
The switching process from the memory state to the high resistance state is dependent upon a history of pulses passed through the memory state as well as the current of memory formation I_f. It was observed that if the current being passed through the memory state was increased gradually rather than abruptly, then the memory state would remain stable at much higher current levels than normally needed to switch. The resistance of the memory state would decrease simultaneously as the current being passed through the filament was gradually increased. The effect of increasing the current being passed through the filament was to increase the width of the filamentary path (Fig. 6). As the current is increased, the surrounding matrix glassy region heats up to the point where it melts and an additional width of crystalline Te memory filament forms. The filament widening mechanism occurs so as to slightly reduce the voltage across the memory path as the testing current magnitude is increased (Fig. 7). This occurs as a result of the cross sectional area of the filament increasing such that the current density actually decreases slightly as the testing current is increased.



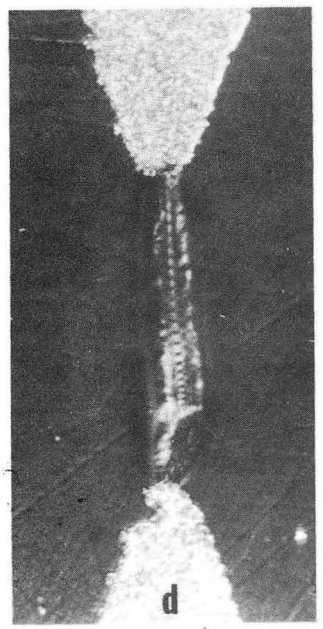
I_f = 0.65 ma



I_f = 0.80 ma



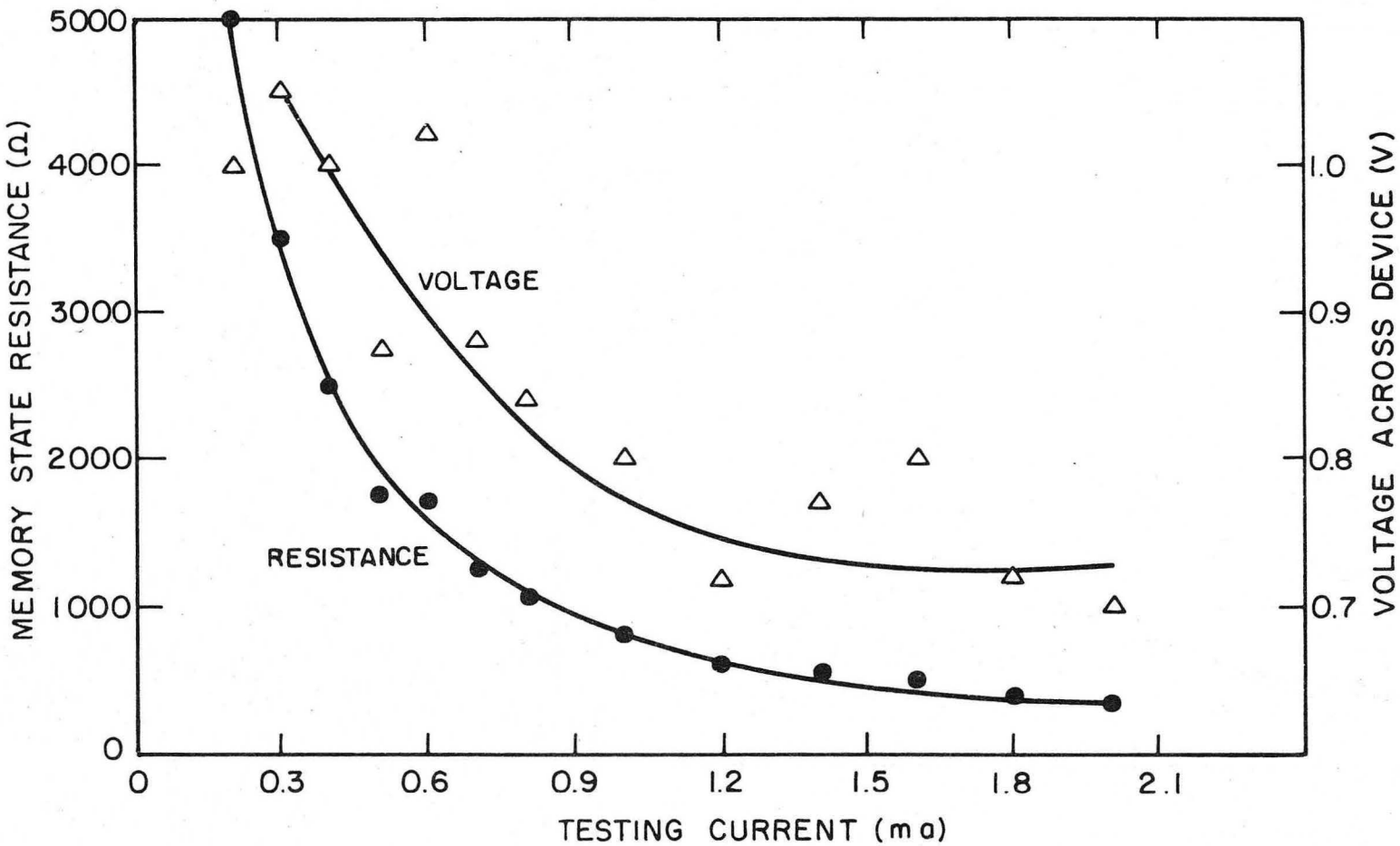
I_f = 1.0 ma



XBB 735-3222

I_f = 1.2 ma

Fig. 6. Filament broadening due to gradual increase in testing current.



XBL 736-6336

Fig. 7. Variation in the electrical properties of the memory state as a function of the testing current.

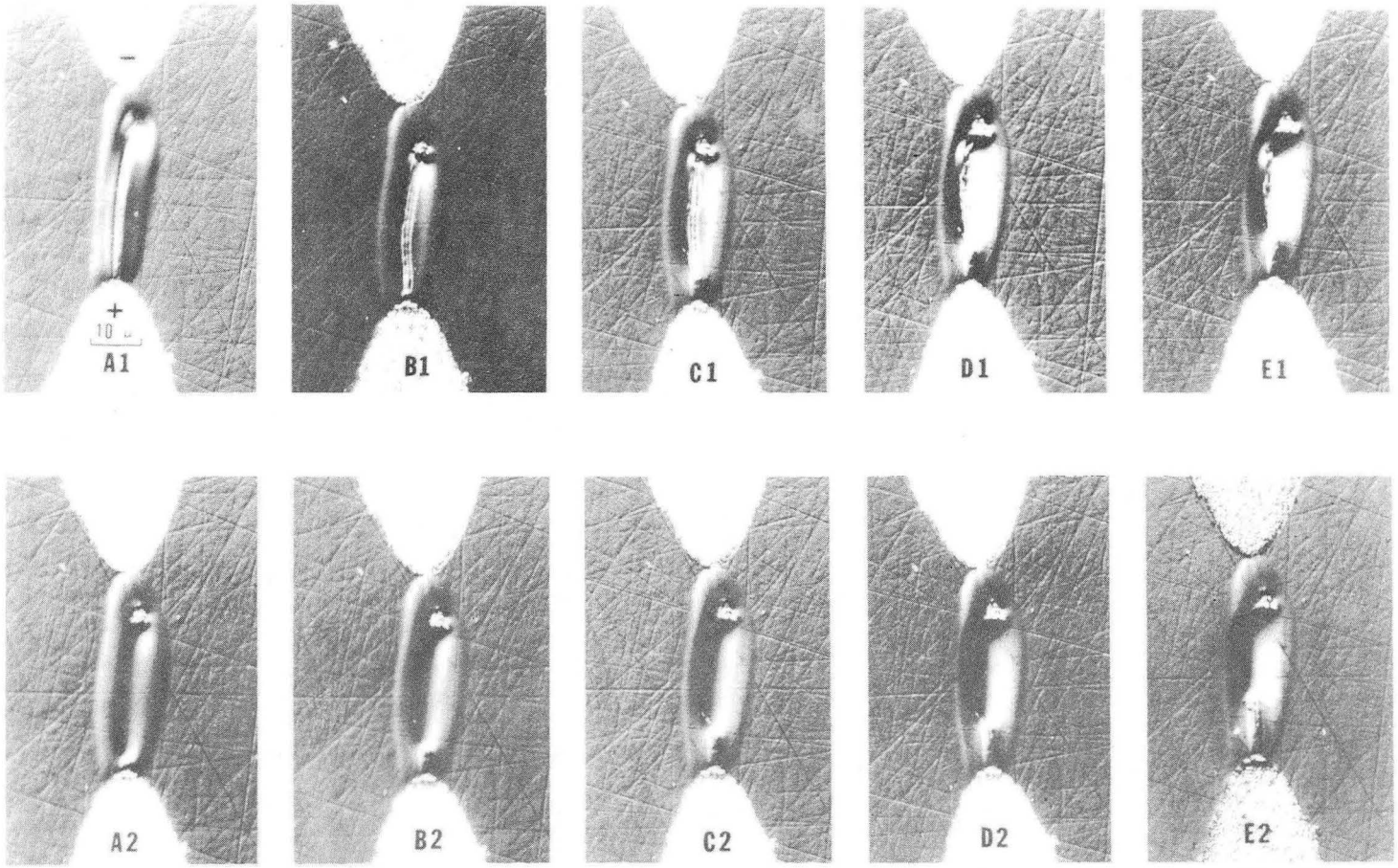
V. CYCLING OBSERVATIONS

The effects of repeated switching from the low to high resistance state and back again were studied to try and determine the stability of a potential device using this type of switching mechanism.

A series of photographs is shown in Fig. 8 depicting the visual changes in the electrode gap upon repeated switching. It should be noticed that by using a switching pulse of 1.4 ma with rise-hold-and fall times of 5 ms - 20 ms and 5 ms respectively the filament grown at 0.8 ma was not only destroyed but a second filament was being formed one segment at a time with each successive switching cycle. After 10 cycles, the 1.4 ma switching pulse actually completed its own higher current filament shown in Fig. 8(J2). Note the section of subsurface negative probe filament completes the path between the electrodes. The resistances of conductive and non-conductive states were kept for each cycle for these experiments. The data is given in Table 1.

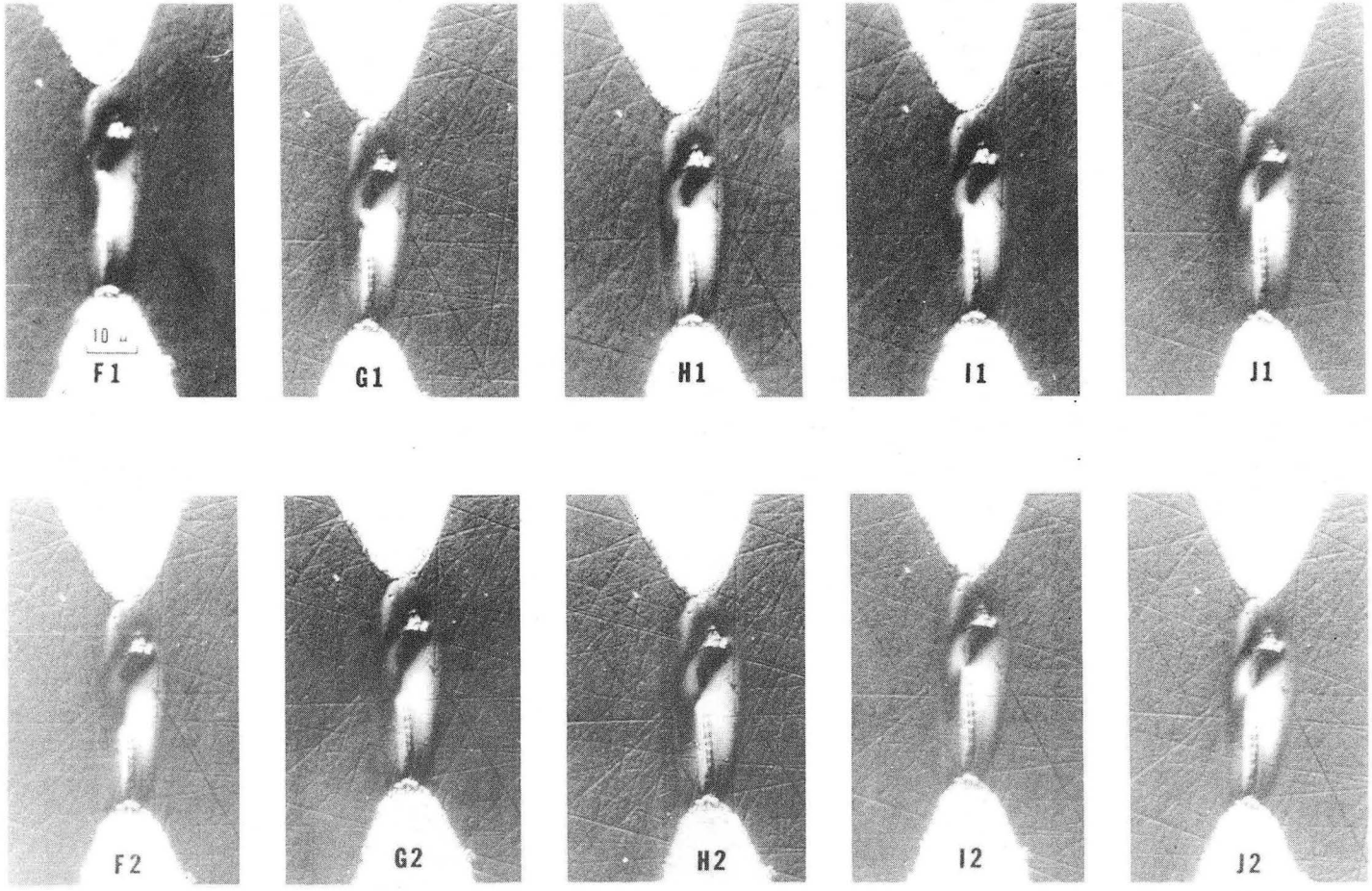
Subsequent cycling experiments were carried out in an attempt to prevent the secondary filamentary growth from occurring. It had been previously observed that a conductive filament would not grow if the current pulse parameters (rise-hold-fall times) were not in a proper range. Specifically, growth would not occur if the hold time was very short. Thus the switching pulse was changed to 5 ms rise 1 ms hold and 5 ms fall while the growth pulse (non-conductive to conductive switching) was maintained at 5 ms rise 20 ms hold and 5 ms fall times. Photographs of this cycling experiment are shown in

0 0 0 0 6 9 0 0 2 1 3



XBB 734-2909

Fig. 8. Successive switching cycles of one filamentary region. I_f = 0.8 ma, I_s = 1.4 ma, 5 ms rise, 20 ms hold, 5 ms fall times. Row 1 is ON state and row 2 is OFF state. Specific cycle in the series is indicated by the letters A through J.



XBB 734-2910

Fig. 8. Successive switching cycles of one filamentary region. I_f = 0.8 ma, I_s = 1.4 ma, 5 ms rise, 20 ms hold, 5 ms fall times. Row 1 is ON state and row 2 is OFF state. Specific cycle in the series is indicated by the letters A through J.

0 0 0 0 3 9 0 3 2 1 4

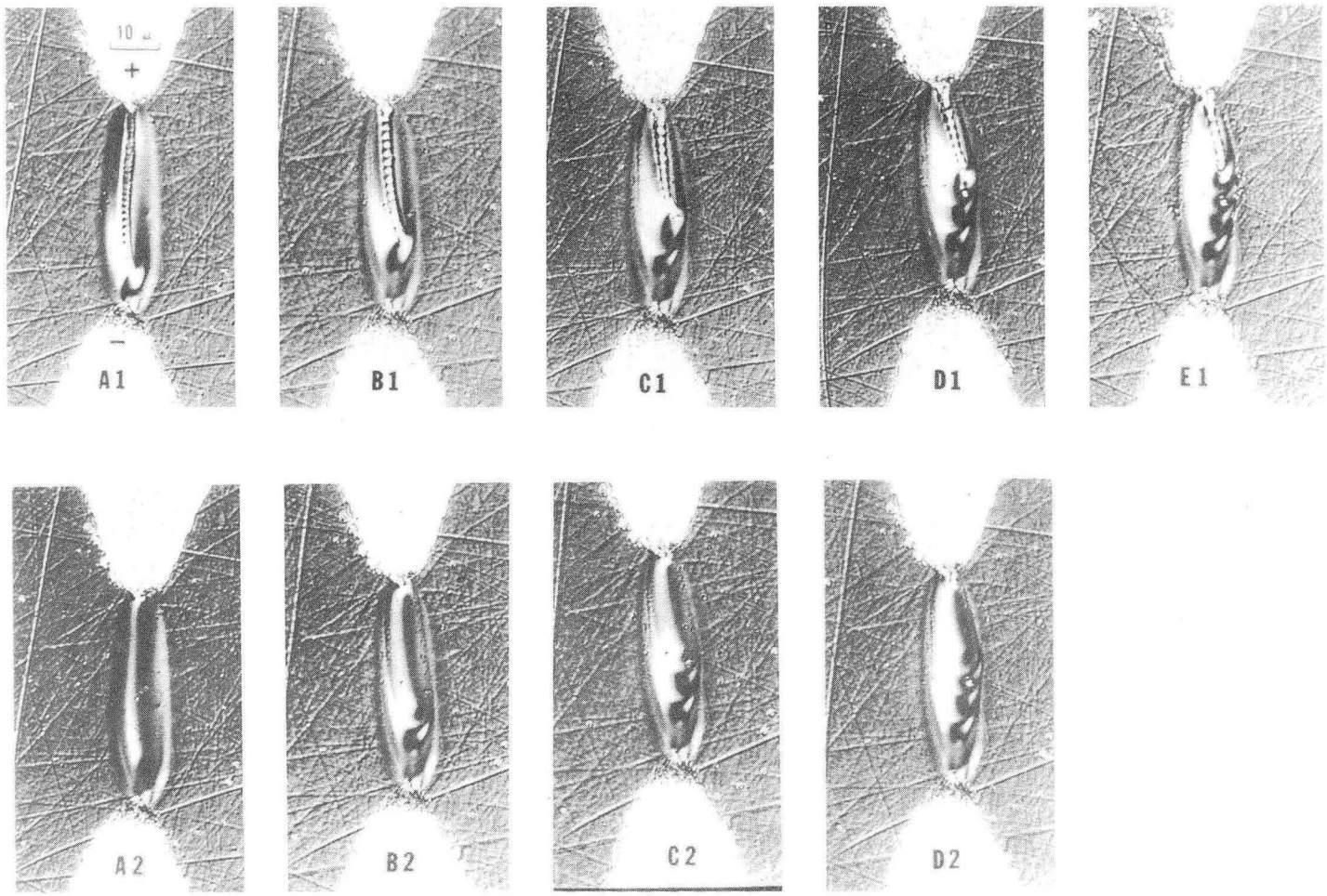
Table 1

Effect of repeated switching on ON and OFF state resistances.

Cycle #	Exp. 1		Exp. 2		Exp. 3	
	ON Ω	OFF Ω	ON Ω	OFF Ω	ON Ω	OFF Ω
1	2000 Ω	29000 Ω	1750 Ω	41000 Ω	1750	35000
2	1875	28000	1575	40000	1750	37000
3	1625	29000	1750	40000	2250	36000
4	1550	28000	1750	40000	2250	36000
5	1550	28000	1625	40000	2100	34000
6	2125	32000	1600	39000	2100	35000
7	2250	30000	1400	conductive at 1750 Ω	1875	35000
8	1950	29000			2250	36000
9	1825	27000			1875	35000
10	1375	conductive at 1000 Ω			2100	43000
11					1750	40000
12					2100	39000
13					2400	38000
14					2300	37000
15					2500	36000
16					2300	36000
17					2500	35000
18					2000	35000
19					2250	34000
20					2250	35000
21					2500	35000
22						
23						
24						
25						

Fig. 9 and it is evident that a secondary filament is not forming due to the conductive to non-conductive switching pulse. However it should be noted that with each growth cycle a short segment of negative probe filament is forming which does not get destroyed by the ON (conductive)to OFF (non-conductive) switching pulse. The negative probe segment cannot be directly seen in the photographs since it is subsurface but it can be evidenced by the position where the positive probe section of the filament stops in the conductive state and the regions of undulations in the "hillock" region which surrounds the filaments. With repealed switching, this negative probe filament section increases to the point where it itself forms nearly the entire conductive path and ON to OFF switching ceased to function at the 1.5 ma current level employed on all previous cycles. The data of the resistances per cycle is also given in Table 1 for this cycling experiment.

This cycling experiment was repeated once more in an attempt to prevent not only the secondary filament formation upon ON to OFF switching but also negative probe filament growth during the OFF to ON filamentary growth process. If this can be achieved, cycling may be possible for extremely high repetition rates without necessary changes in the switching current magnitudes to compensate for the secondary or negative probe filament formation. The problem is that prevention of negative probe growth seems to be based on electrode geometry, pulsing parameters and possibly some unknown additional parameters. Thus the negative probe growth results change from electrode gap to gap.



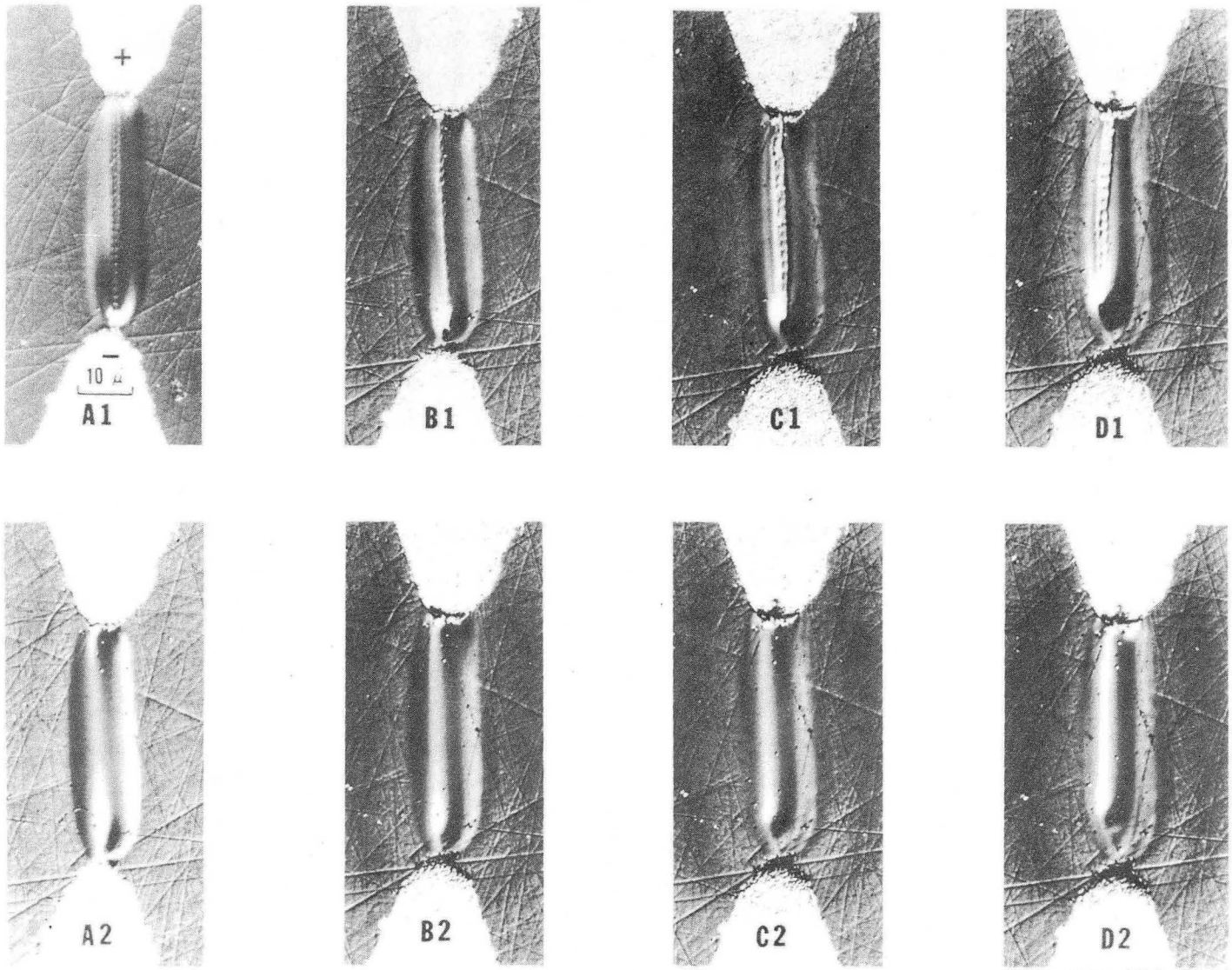
XBB 734-2911

Fig. 9. Successive switching cycles of one filamentary region I_f = 0.8 ma, I_s = 1.5 ma, 5 ms rise, 1 ms hold, 5 ms fall times.

In order to carry out this experiment, a gap was chosen which originally showed no negative probe phase upon completion of the conductive state of the first cycle. The OFF to ON current pulse was 0.8 ma and the switching ON to OFF current pulse was 2.0 ma with parameters of 5 ms rise 2 ms hold and 5 ms fall. In all three cycling experiment the OFF to ON parameters were 5 ms rise 20 ms hold and 5 ms fall time. Photographs of this cycling experiment are shown in Fig. 10. As can be seen, even in this case a negative probe phase began to grow after cycling began. However, the rate of negative probe filament growth per cycle was far less than the previous experiment and this particular gap was switched for 25 times and still remained functioning. However at the later stages of cycling, the filament became irregular in shape and not all of it would be destroyed by the 2.0 ma ON to OFF switching current. Thus it was assumed that eventually, the cycling process would fail to function when due to the irregularities of filament formation in the later cycling stages, a filamentary path would be formed which would not switch from ON to OFF from the 2.0 ma pulse.

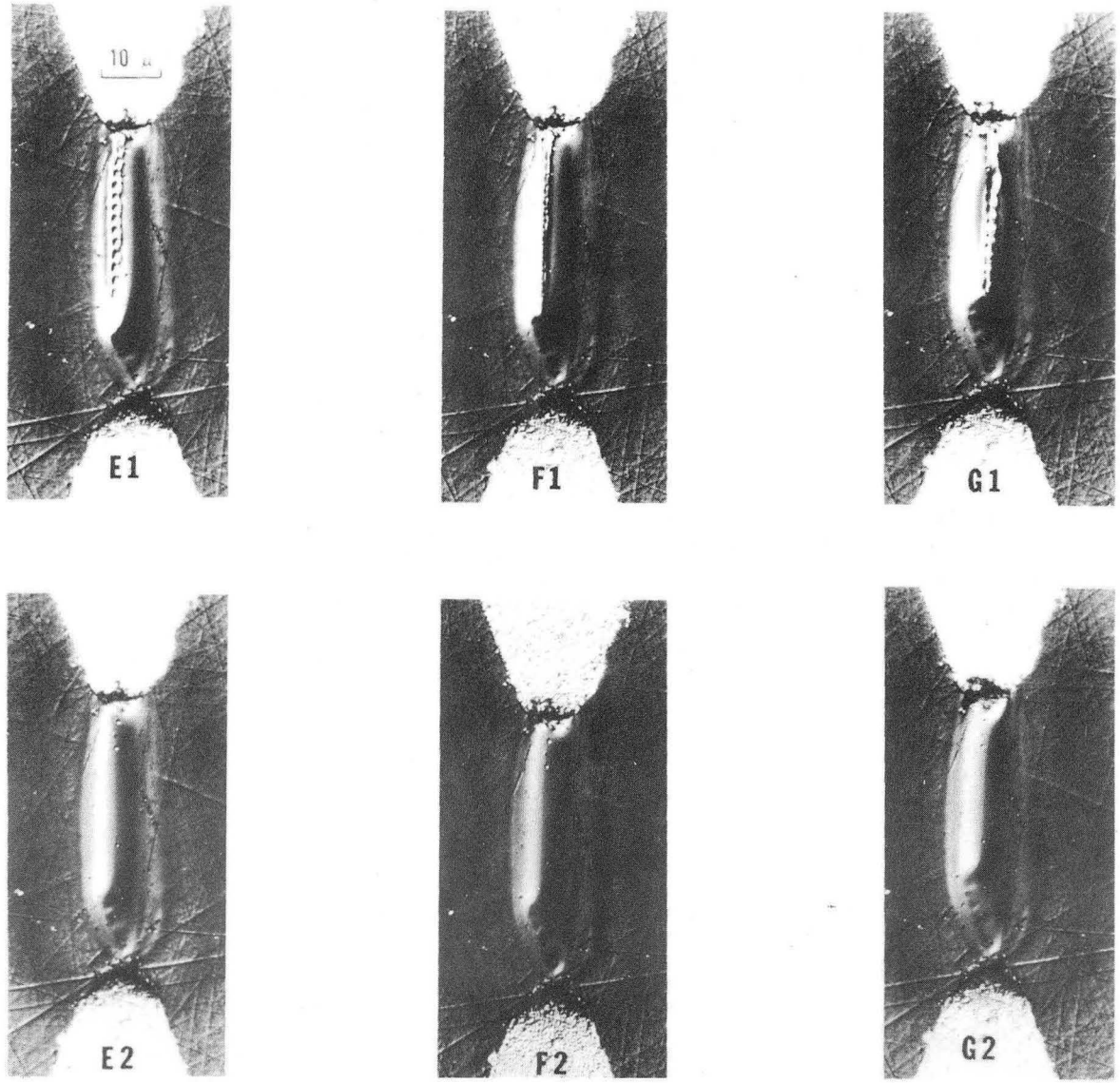
A 4.0 ma pulse was used at cycle 21 and the entire filament, positive and negative probe sections as well was seen to disappear leaving a hillock empty of any filamentary definition. Thus it may be possible with suitable choice of switching (ON → OFF) to growth (OFF → ON) current ratios, the switching process may be cycled to far greater repetition rates with the problems of negative probe filamentary phases or unswitched positive probe phases being eliminated.

0 0 0 0 5 9 0 0 2 1 6



XBB 734-2908

Fig. 10. Successive switching cycles of one filamentary region. I_f = 0.8 ma I_s = 2.0 ma 5 ms rise 2 ms hold 5 ms fall times.



XBB 734-2913

Fig. 10 (Cont.)

A final cycling experiment was designed to avoid both positive and negative electrode filamentary segments based on the results of the first three cycling experiments. A switching pulse current magnitude of 6.0 ma was used to avoid the stable negative electrode segment from shorting out the electrode gap and a short duration current pulse of 0.5 ms rise, 2.5 ms hold, and 0.5 ms fall times was used to avoid the growth of a secondary filament. Figure 11a through 11p shows the memory states during cycling up to cycle no. 30. (11p) Notice in Fig. 11m, the memory state is composed of nearly all negative electrode filament. Figure 11n shows the switching pulse has completely destroyed both the positive and negative filamentary segments of the previous memory state since the new memory state exhbits a much greater proportion of positive electrode filament. The cycling was carried out to 50 cycles with no consistent change in the visual or electrical properties of the filamentary memory state or those parameters necessary to carry out the switching from the memory to the high resistance state. (See Fig. 12.)

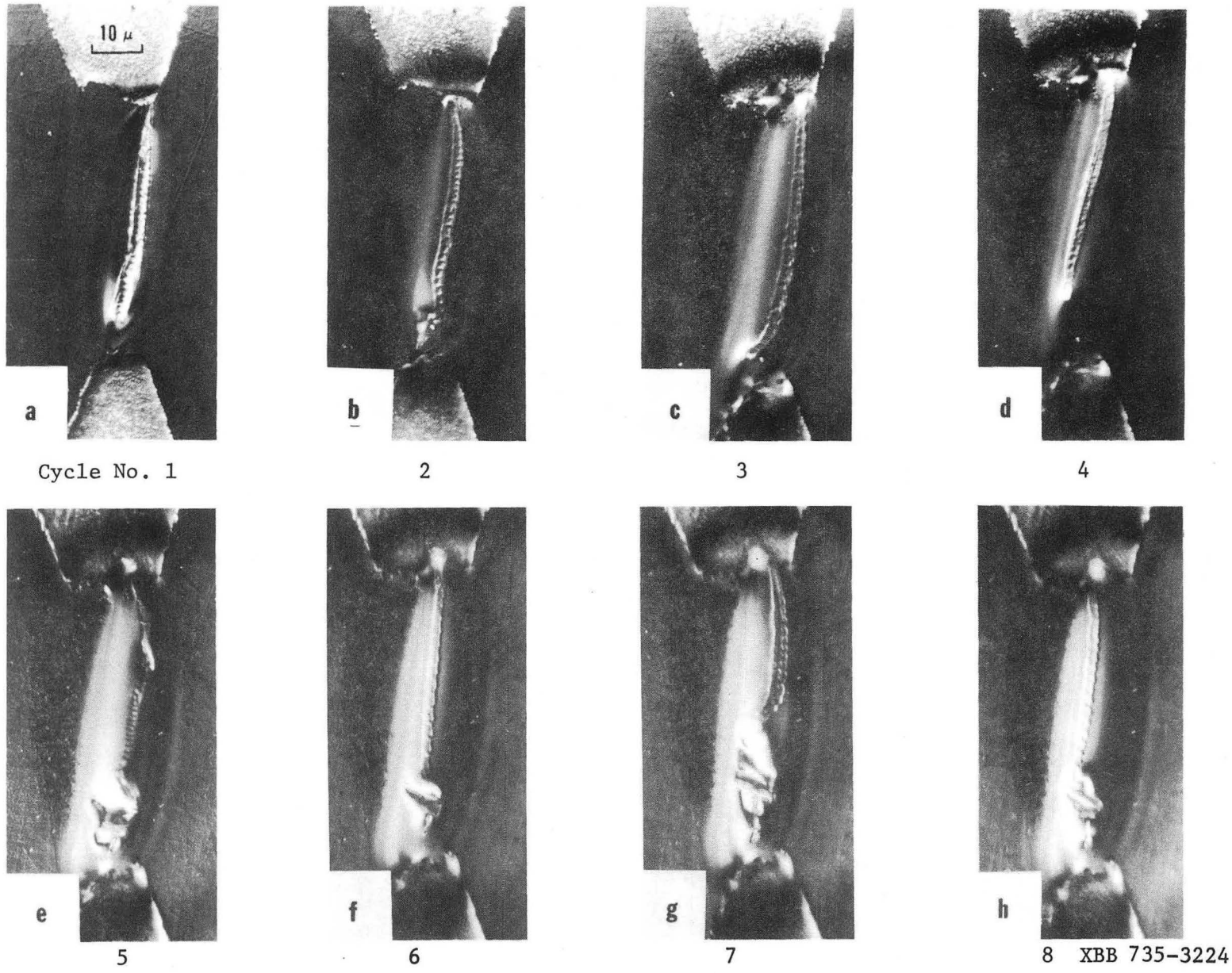
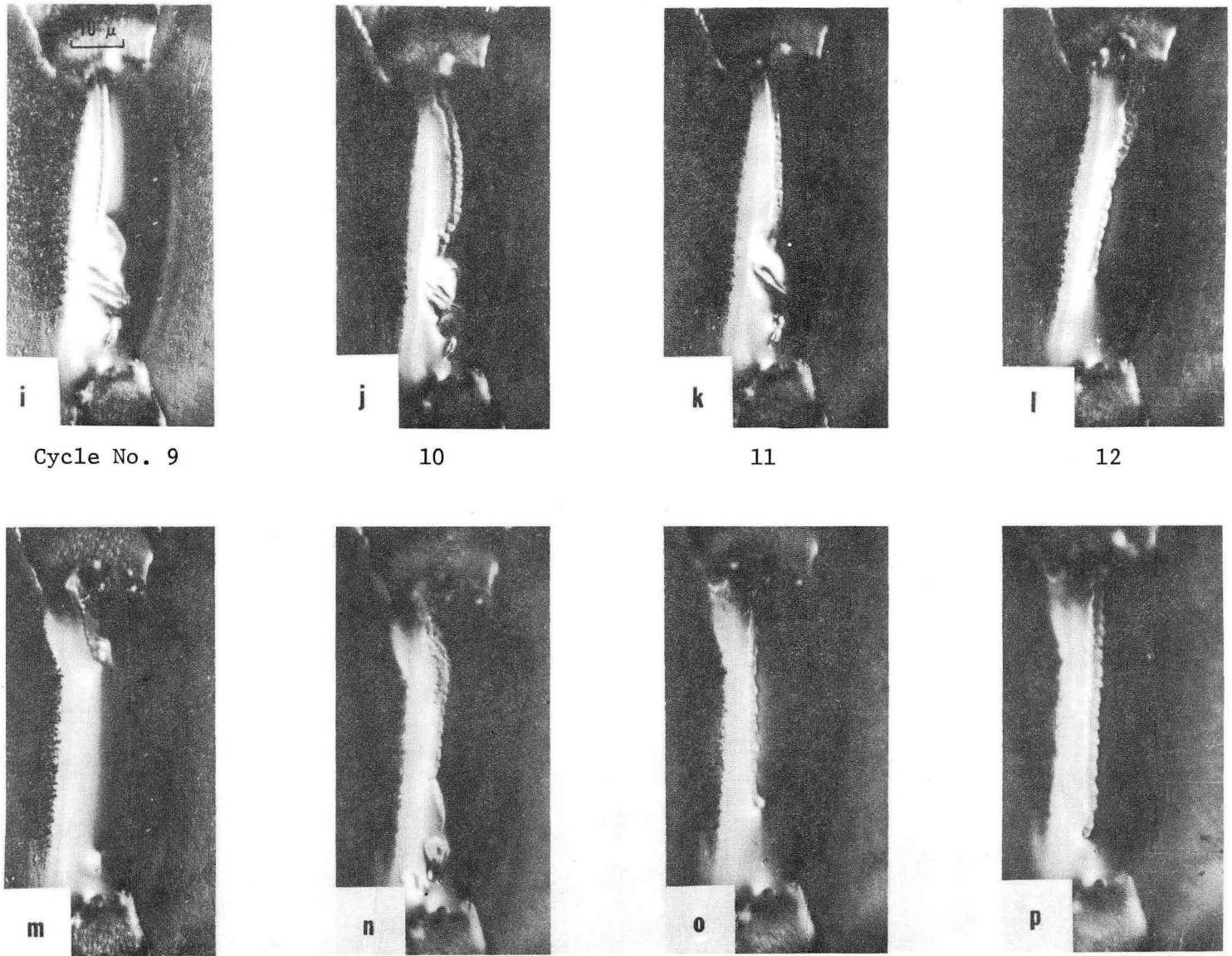


Fig. 11. Memory states of one filamentary region during cycling. I_f = 0.9 ma, I_s = 6.0 ma, 0.5 ms rise, 2.5 ms hold, 0.5 ms fall times.

0 0 0 0 5 9 0 0 2 1 8



Cycle No. 9 | 10 | 11 | 12

13 | 14 | 20 | 30

XBB 735-3223

Fig. 11 (Cont.)

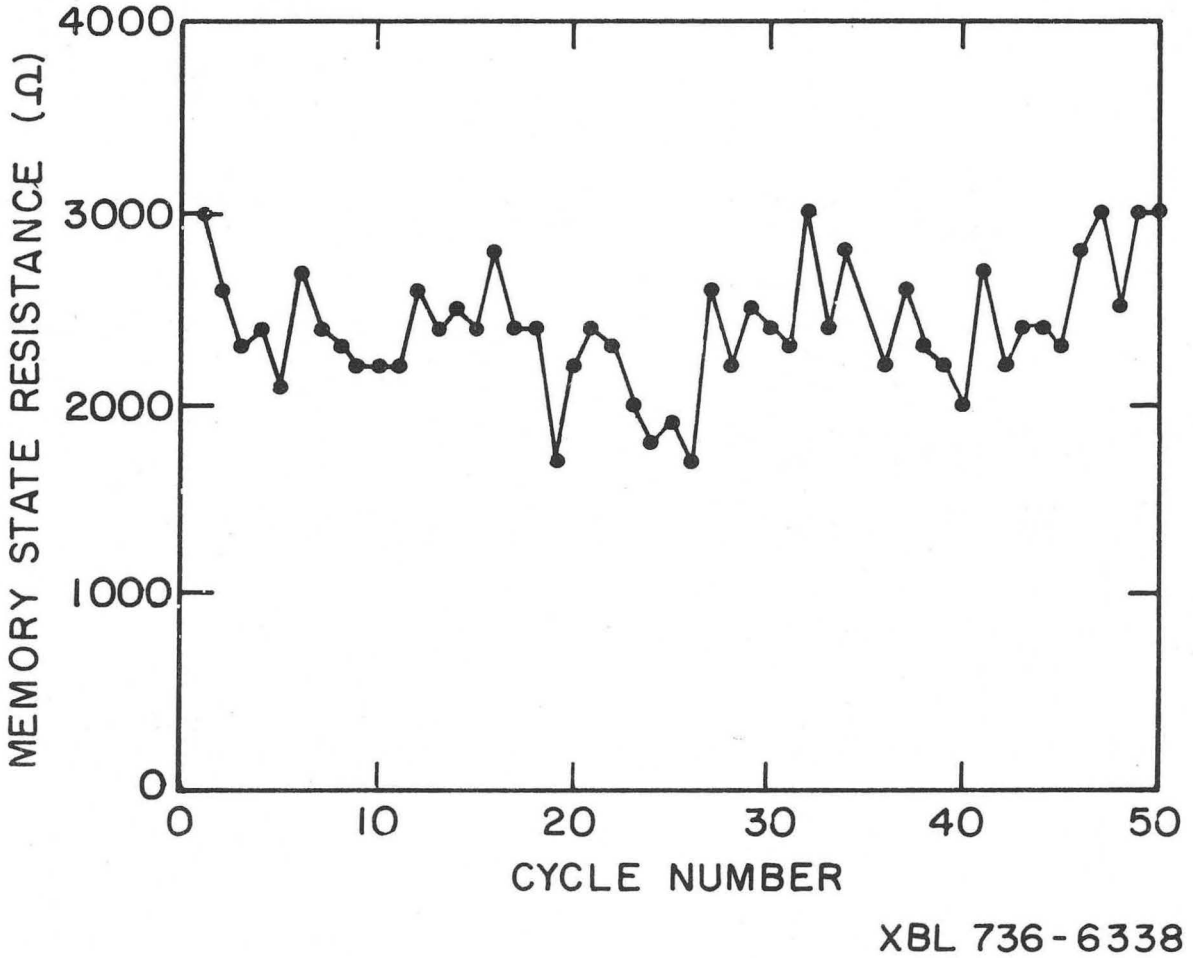


Fig. 12. Variation in memory state resistance during cycled switching.

0 0 0 0 3 9 0 6 2 1 9

VI. GROWTH RATE STUDY

The photographs shown in Fig. 9 were used to get a measure of the filamentary crystal growth rate as a function of cycling repetition. Since the switching mechanism from the memory to the high resistance state is believed to be a melting process which leaves a high Te content in the region, the reformation filamentary growth rate could be expected to increase over that of the original growth rate because long range Te diffusion would not be necessary to build up the Te concentration in the filamentary region. This hypotheses is born out by inspecting the photographs in Fig. 9 . It should be noted that the filament structure consists of narrow segments normal to the filament which crystallizes during one growth pulse. Thus a measure of the segment width will give an indication of the filament growth rate for any specific repetition in the switching cycle. It can be seen that the growth rate is not even constant within one filament. Thus to compare any two filaments, the same area of the filament must be used for growth rate analysis. The growth rate data derived from these cycling experiments is summarized in Fig. 13. In all cases, the growth time per segment is assumed to be 20 ms, the hold time. It may indeed be somewhat longer if growth occurs during a segment of the rise and fall times. However, this would just add a constant to the growth time and not affect the growth rate dependance on switching cycle.

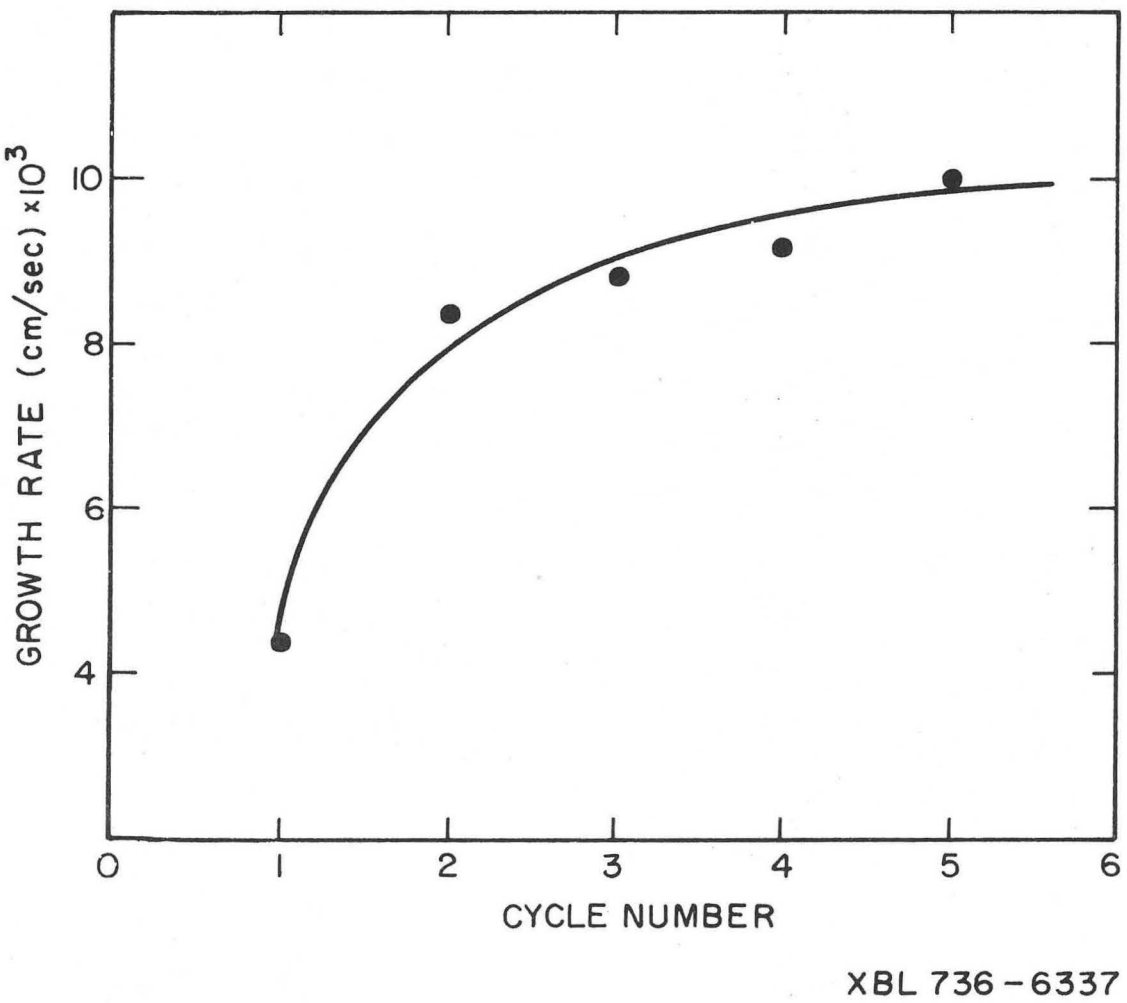


Fig. 13. Functional relationship of crystal growth rate of filament with switching history of filamentary region.

0 0 0 0 3 9 0 6 2 2 0

VII. REVERSE POLARITY STUDY

The effect of reversing the polarity of the current pulse was tested on the growth of a memory filament. Figure 14 shows that one pulse (5 ms - 20 ms - 5 ms) was sufficient to completely destroy that portion of the memory filament already grown. When the duration of the reverse polarity pulse was reduced, (same current magnitude as original growth pulses) the filament was seen to grow back to the new negative electrode and then begin growing out of the new positive electrode upon repeated pulsing. Often the original partially grown filament would begin growing back to the new negative electrode and then stop when a crystallite of irregular shape formed on the receding tip. (Fig. 15) The filamentary tip would also seem to widen once it has stopped receding because of the formation of a crystallite. This experiment exemplifies the importance of the electric field in the mechanism of memory state formation. The receding filament can be explained as a result of Te ions being pulled off of a molten interface between the glass and the crystalline Te filament. The reverse polarity electric field pulls them toward the new positive electrode which then begins to crystallize out its own positive electrode filament (Fig. 15e). This process indicates the original mechanism of memory state formation to be a melting of the matrix glass due to Joule heating, and then a subsequent migration of Te ions toward the positive electrode where they build up in concentration until they crystallize out as pure Te.

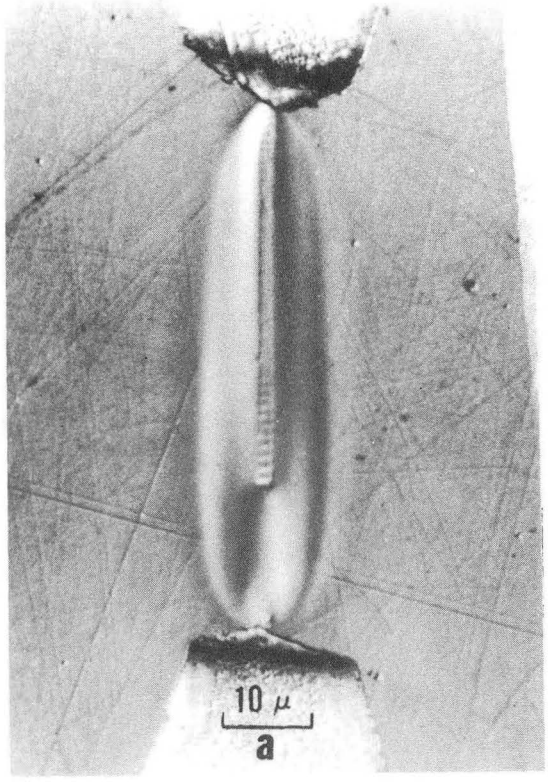
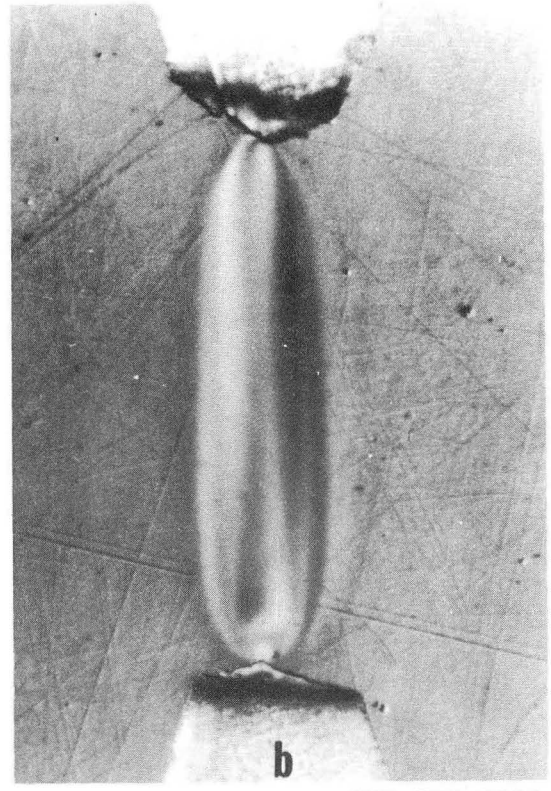
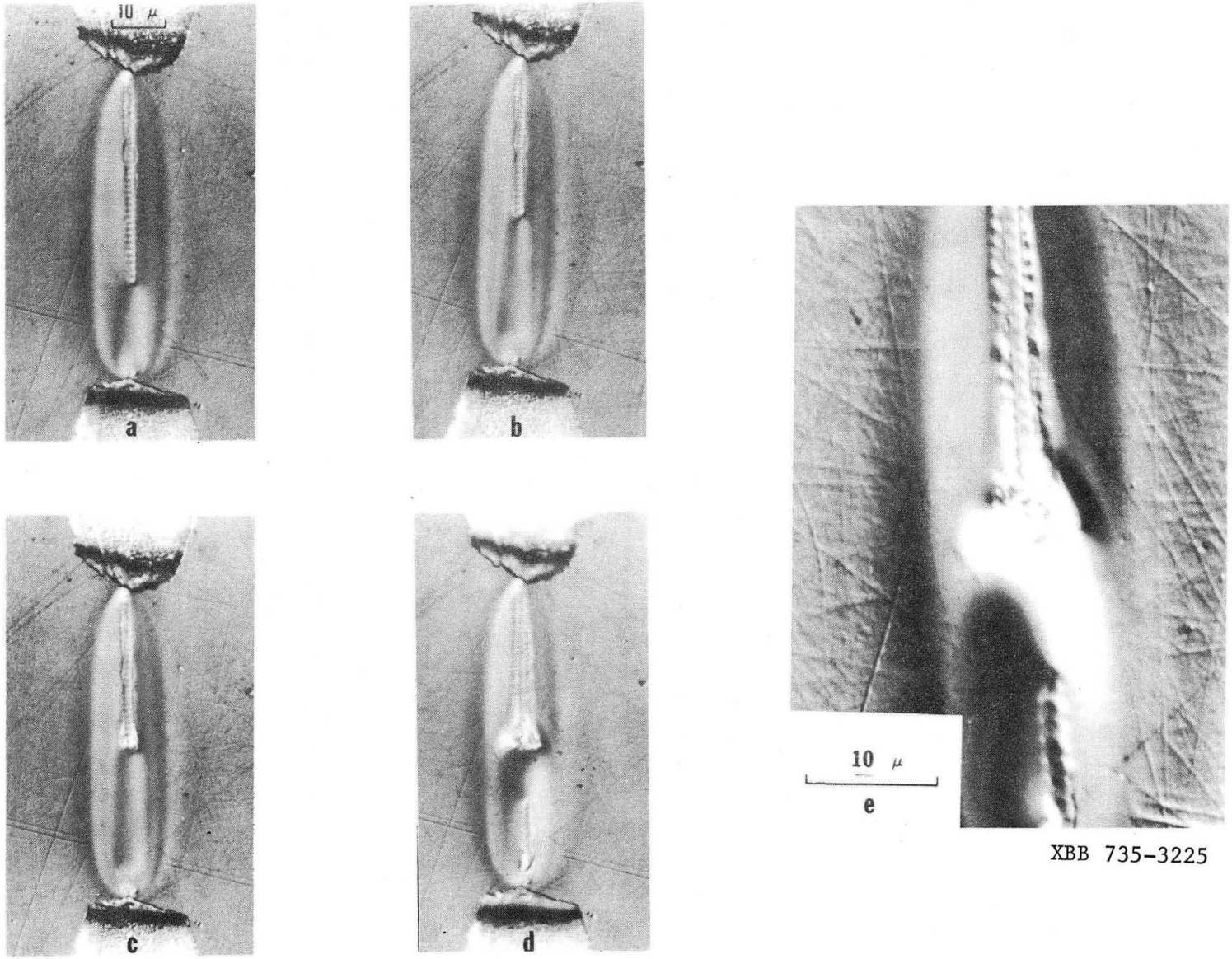


Fig. 14(a). Partially grown memory filament.



XBB 735-3226

Fig. 14(b). Destruction of partially grown filament with one 5 ms rise, 20 ms hold, 5 ms fall reverse polarity pulse.



XBB 735-3225

Fig. 15(a). Partially grown memory filament. 15(b-e) Receeding filament and new positive electrode filament growth due to reversing polarity.

0 0 0 0 5 9 0 0 0 2 2 1

VIII. CONCLUSIONS

The switching process from the memory to the high resistance state resistance state seems to be governed by a Joule heating and subsequent melting of the crystalline filamentary path. Upon the cessation of the switching pulse, the filamentary region is quenched into a Te rich glass of high resistivity. The switching process is a sensitive function of the heat conduction properties of the surrounding region.

The memory filamentary path is often composed of two segments; one growing from the positive electrode and one growing from the negative electrode. The negative electrode segment is more stable than the positive electrode filament during switching. This is probably due to the better heat conduction properties of a subsurface filament in addition to the increased conductivity of crystalline As_2Te_3 over the crystalline Te phase of the positive electrode filaments.

A filamentary region can be cycled to high repetitions with no change in composition or electrical properties if a suitable choice of a switching pulse is made. A current spike $\leqslant$ 3 ms hold time is necessary to prevent secondary filament growth. A current magnitude $\geqslant$ 6 ma is necessary to melt both the positive and the negative electrode filamentary segments.

The filamentary growth mechanism is based on migration of Te ions to the positive electrode where the concentration builds up until the Te crystallizes and forms the memory state. Reversing the polarity before completion of the memory path strips off Te ions from the molten interface between the filament tip and matrix glass.

ACKNOWLEDGEMENTS

I wish to express my gratitude to Professor Jack Washburn and Jon Roberts for their helpful comments and suggestions.

This work has been carried out under the auspices of the U. S. Atomic Energy Commission.

REFERENCES

1. D. Shanefield, J. Non-Crystalline Solids 2, 382 (1970).
2. R. Uttecht, H. Stevenson, C. H. Sie, J. D. Grienes, K. S. Raghavan, J. Non-Crystalline Solids 2, 358 (1960).
3. P. O. Silva, G. Dir, C. Griffiths, J. Non-Crystalline Solids 2, 316 (1970).
4. H. J. Stocker, J. Non-Crystalline Solids 2, 371 (1970).
5. A. D. Pearson, IBM Journal of Research and Development 13, 510 (1969).
6. J. Roberts, M. S. thesis, University of California, Berkeley, 1973.

LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

TECHNICAL INFORMATION DIVISION
LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
BERKELEY, CALIFORNIA 94720