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PART II

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THEORETICAL STUDIES ON THE DEGRADATION OF LADDER POLYMERS

PART II. VAPORIZATION STUDIES

MARTIN M. TESSLER, 1/LT., USAF

TECHNICAL REPORT ML-TDR-64-151, PART II

DECEMBER 1965

AIR FORCE MATERIALS LABORATORY
RESEARCH AND TECHNOLOGY DIVISION
AIR FORCE SYSTEMS COMMAND
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FOREWORD

This report was prepared by the Polymer Branch of the Nonmetallic Materials Division. The work was initiated under Project No. 7342, "Fundamental Research on Macromolecular Materials and Lubrication Phenomena," Task No. 734203, "Fundamental Principles Determining the Behavior of Macromolecules." It was administered under the direction of the AF Materials Laboratory, Research and Technology Division, 1stLt Martin M. Tessler, project engineer.

This report covers work conducted from January 1964 to December 1964. The manuscript was released by the author in May 1965 for publication as an RTD technical report.

This technical report has been reviewed and is approved.

William E. Gibbs

WILLIAM E. GIBBS
Chief, Polymer Branch
Nonmetallic Materials Division
Air Force Materials Laboratory

ABSTRACT

The random degradation of four and six-membered ring ladder polymers were investigated by means of a digital computer and the results compared to a single chain polymer undergoing degradation under identical conditions. The percent vaporization versus time and the rate of weight loss versus time was plotted and significant differences were obtained. The results indicate that ladder polymers should have increased stability over single chain polymers undergoing random degradation.

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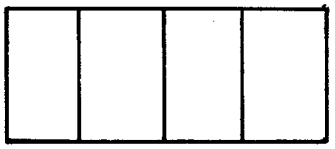
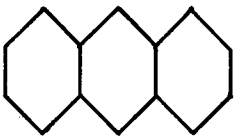
INTRODUCTION

Previously (Reference 1), the thermal stability of a ladder polymer undergoing random degradation was compared with that of a single chain polymer by using a digital computer to set up a statistical (Monte Carlo) model of the degrading system. The change in molecular weight versus time was plotted and significant differences were found in the shapes of the curves for the ladder polymers as compared to a single chain polymer.

A common test for thermal stability of a polymer is to measure its loss of weight upon heating. The previous report (Reference 1) studies the changes in molecular weight of a ladder polymer undergoing random degradation upon heating in a closed system where no weight loss has occurred. The present report discusses the random degradation of a ladder polymer in an open system where, upon heating, small molecular weight fragments of the polymer are vaporized out of the system. Two types of ladder polymers are studied. Type I is a fused four-membered ring and Type II is a fused six-membered ring. All of the results for a four-membered ring are equally applicable to an eight-membered ring.



TYPE I



TYPE II

Initially, a simplified scheme of degradation was considered where a broken bond can break a molecule only if the bond opposite it is broken. The much more realistic scheme of random degradation, where breaks in the crosslinks will result in increased molecule breaks, was also considered. This is called the complex case of degradation.

The following assumptions were made in defining the degrading system:

1. The polymer sample is initially monodisperse; that is, only chains of a single length are present.

2. All bonds in the polymer chain are of equal strength and accessibility, regardless of the length of the chain and the positions of the bonds in the molecule.

3. The rate of bonds breaking is proportional to the number of unbroken bonds in the degrading system.

The simple and complex cases of degradation were studied for Type I and Type II ladder polymers. The percent vaporization at time kt and the rate of weight loss at time kt were calculated and compared with a single chain polymer degrading under identical conditions. The constant, k, is the proportionality constant between the rate of breaking bonds and the number of unbroken bonds in the degrading system. It can only be determined experimentally, so the time factor is kt instead of t, where t is time.

MONTE CARLO MODEL

To construct a Monte Carlo (Reference 2) model for the degradation reaction, a portion of the computer storage is set aside to represent the polymer molecules. Each storage location represents one bond. If a bond is not broken, a zero is placed in the computer storage location corresponding to that bond.

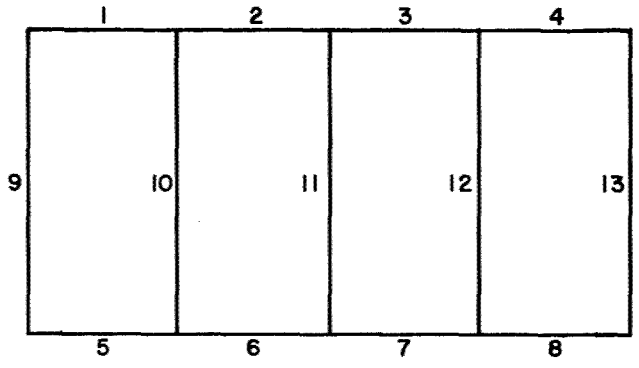


Figure 1. Ladder Polymer Model

In the simple case, when a bond is broken it is set equal to 1 and the opposite bond checked. If the opposite bond is broken, both bonds are set equal to 2. In the complex case, when an isolated bond is broken (bond 3 in Figure 1), it is set equal to 1. If vaporization occurs through the breaking of adjacent side chain bonds only (bonds 1, 10, 11, and 3 in Figure 1 are broken), the side chain bonds are set equal to 2. Since x broken side chain bonds result in x-1 atoms vaporizing, all of the broken side chain bonds are set equal to 2 except the last one so that each bond which is set equal to 2 corresponds to a vaporized atom. If two opposite bonds are broken (bond 2 and 6 are broken in Figure 1), they are set equal to 3 except when a bond is broken opposite a bond which has a number 2 in its storage location. The initial broken bond is set equal to 3, but the opposite bond is changed from a 2 to a 5. If two pairs of opposite broken bonds result in vaporization (bonds 1 and 5 and bonds 4 and 8 in Figure 1 are broken), all of the bonds in the vaporized fragment are set equal to 4. If any of the bonds have numbers 2 or 5 in their storage, they are also set equal to 4, but the number of vaporized atoms is reduced by one. This prevents the same atom from being vaporized twice. If bonds 1, 5, 10, and 6 in Figure 1 are broken and the computer then breaks bond 2, an error will result because only one atom must be vaporized and not two. If the bond opposite the initial broken bond has a number of 3, 4, or 5, the computer checks whether its adjacent bond also has a number of 3, 4, or 5 and if it does, no vaporization of that atom occurs (it has been vaporized previously). If the opposite bond has a number 2 in it, it is set equal to 5 and no vaporization is recorded as described previously.

Initially, all of the storage locations are set to zero. A random number is generated which has the limits of 0 to B_o -1, where B_o is the total number of bonds initially present in the system. Each number then represents a particular bond in a particular molecule and the number of molecules and the number of bonds per molecule are selected by the programmer and can readily be altered. The present study investigated a system of 100 molecules with 199 bonds per molecule for ladder polymer Type 1 and 100 molecules with 196 bonds per molecule for ladder polymer Type II. A single chain polymer with the same number of bonds per molecule was degraded in the computer for comparison purposes.

When a random number is generated, the bond which it represents is checked to see whether or not it is broken. If it is already broken, a duplicate is recorded and a new random number generated. If it is not broken, the bond is set equal to one and the opposite bond checked. If the opposite bond is broken, in the simple case both bonds are set equal to 2. The computer then checks the adjacent bonds in both directions until it comes to another pair of opposite bonds with a value of 2 or the end of the molecule. The size of the fragment is measured and if it contains ten atoms or less, vaporization occurs. All of the vaporized bonds are set equal to 2 and the concentration of unbroken bonds is adjusted to account for the vaporized bonds. The size of the fragments which are volatile is arbitrary and can be varied if desired.

In the complex case, if the opposite bond is broken, both bonds are set equal to 3 except when the opposite bond has a value of 2. The opposite bond is then set equal to 5 and the original broken bond is set equal to 3. The computer then checks the side chain bonds in both directions until it comes to another pair of opposite bonds with a value of 3, 4, or 5 or until it comes to the end of the molecule. The size of the fragment is measured and if it contains ten atoms or less, vaporization occurs. Before the vaporized bonds are set equal to 4, they are checked to see if their value is 0, 2, or 5. The total number of atoms in the fragment minus the number of bonds containing 2 or 5 equals the number of atoms vaporized. The number of bonds which are changed from 0 to 4 is also calculated and the number of unbroken bonds left in the system is reduced by that number of bonds.

If no vaporization occurs or if the opposite bond is not broken, no further work is done in the simple case and a new random number is generated. If the opposite bond is broken and no vaporization occurs in the complex case, the crosslinks are checked to see if broken crosslinks can lead to a broken molecule. The computer checks all of the crosslinks to the left and right until it either comes to an unbroken crosslink or the last crosslink at the end of the molecule. If the computer finds an unbroken crosslink, it then scans all of the side chain bonds on both sides of the ladder polymer from the broken bond produced by the random number generator to the last side chain bond before the unbroken crosslink. If a broken bond is found on the side of the molecule opposite the original broken bond, the computer treats it exactly as described previously for the case of two opposite bonds breaking. If a broken bond is found on the side of the molecule adjacent to the original broken bond or if all of the crosslinks to the end of the molecule are broken, the computer checks to see if the fragment contains ten or less atoms. If it does, all of the bonds are set equal to 2 and the total number of unbroken bonds remaining in the system is reduced by the number of bonds changed from 0 to 2. If the opposite bond is not broken in the complex case, the computer scans the crosslinks and does all of the calculations previously described.

If the random number generator produces a bond which corresponds to a crosslink, no further work is done in the simple case. In the complex case, the computer checks to see if the broken crosslink will result in a broken molecule. If it does, it then checks to see if the fragments are small enough to be vaporized and adjusts the bond numbers accordingly.

The time factor kt is equal to ln (B_o/B) where B_o is the number of bonds present initially and B is the number of unbroken bonds present at time kt. The number of unbroken bonds present at time kt equals the number of bonds present initially minus the number of random numbers generated plus the number of duplicates recorded minus the number of unbroken bonds vaporized out of the system. The computer calculates the number of atoms vaporized out of the system, the percent vaporization, and the rate of vaporization at time kt and prints out the desired data at convenient intervals of kt.

RESULTS

The curves shown for the simple case in Figure 2 and the complex case in Figure 3 are based on computer calculations for the percent vaporization versus kt. As the degrading single chain polymer undergoes a much more rapid decrease in molecular weight when compared to a ladder polymer (Reference 1), we would naturally predict that the single chain polymer would also lose weight at a much faster rate. Examination of Figures 2 and 3 shows quite clearly that this prediction is correct. When 10 percent of the single chain polymer has vaporized, less than 2 percent of the ladder polymer has vaporized in the simple case and less than 4 percent in the complex case. A difference in the shapes of the curves is also readily apparent. The single chain polymer shows a large initial loss in weight during degradation while the ladder polymers have an induction period during which the loss in weight is very small. As degradation proceeds, the shapes of the curves become identical and merge together. The curves for the rate of weight loss versus kt are shown in Figure 4 for the single case and in Figure 5 for the complex case. The very large initial rate of weight loss for a single chain polymer as compared to a ladder polymer is the major point of interest of these curves. These results indicate that the ladder polymers should have increased thermal stability over single chain polymers.

REFERENCES

1. M. M. Tessler, Technical Documentary Report No. ML-TDR-64-151, July 1964. Research and Technology Division, Wright-Patterson Air Force Base, Ohio.

2. A. S. Householder, G. E. Forsythe, and H. H. Germand, Monte Carlo Method, National Bureau of Standards, Applied Mathematics Series, 12 (1951).

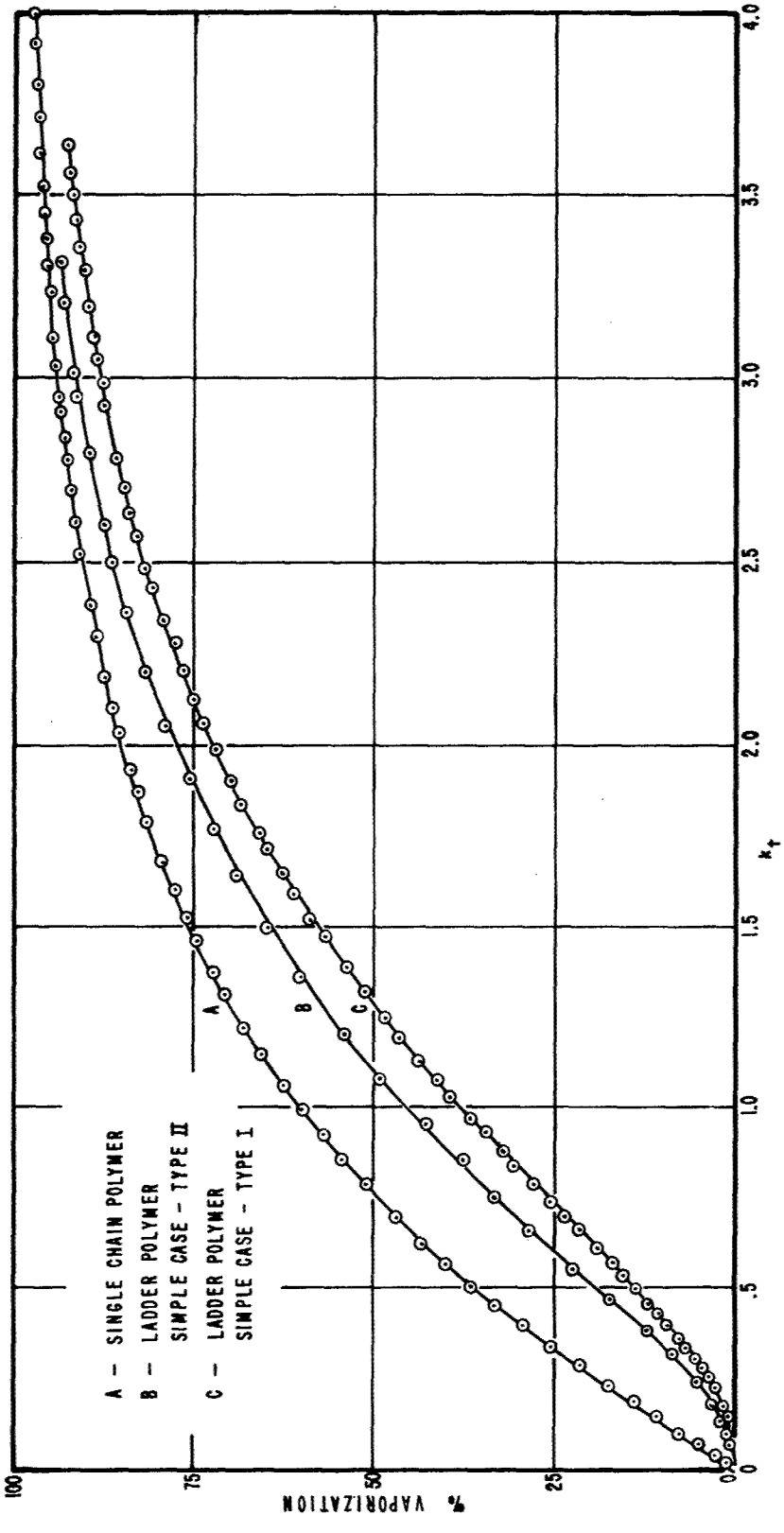


Figure 2. Plot of Percent Vaporization Versus Time for Simple Case Ladder Polymers Undergoing Random Degradation.

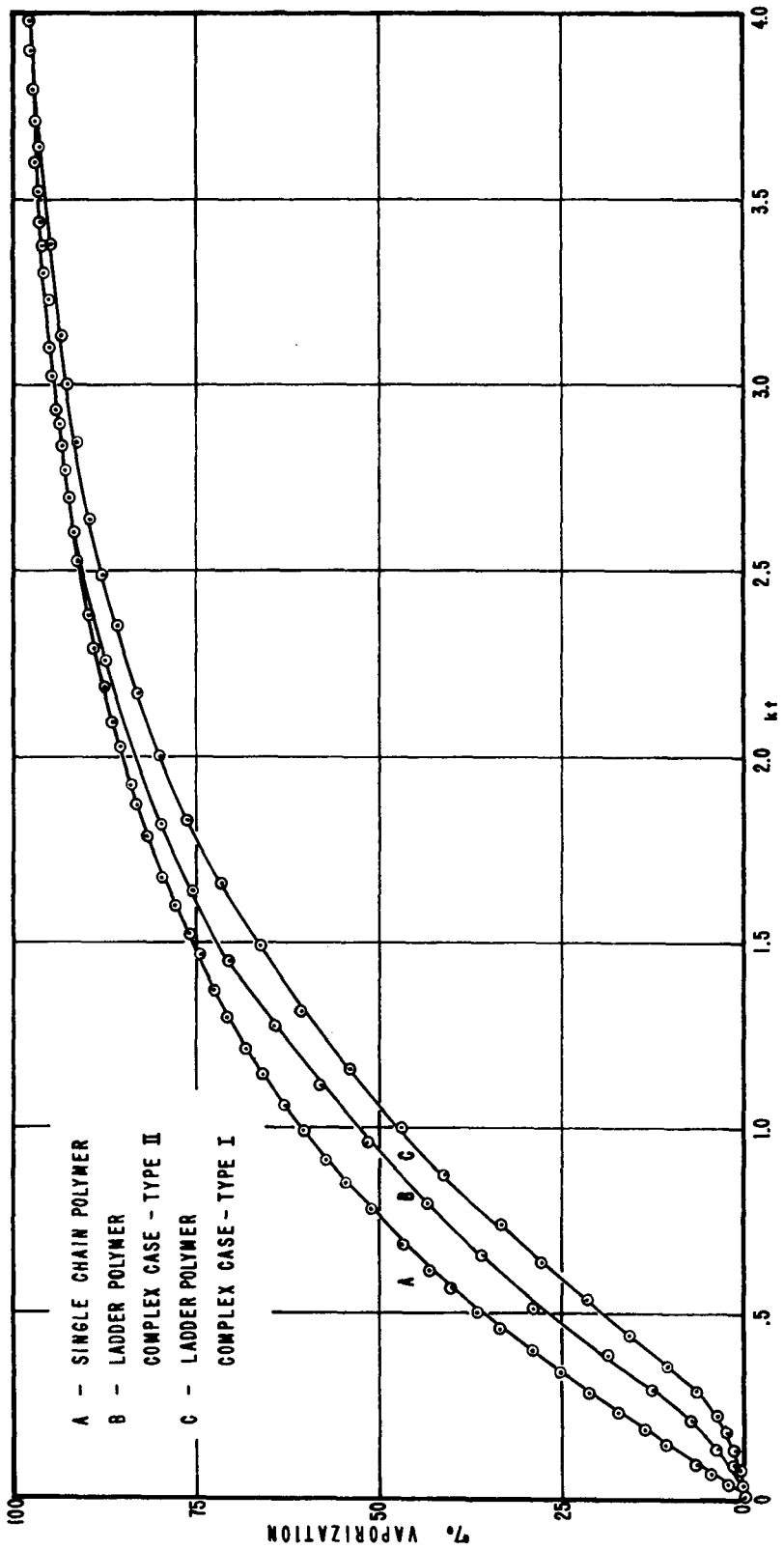


Figure 3. Plot of Percent Vaporization Versus Time for Complex Case Ladder Polymers Undergoing Random Degradation.

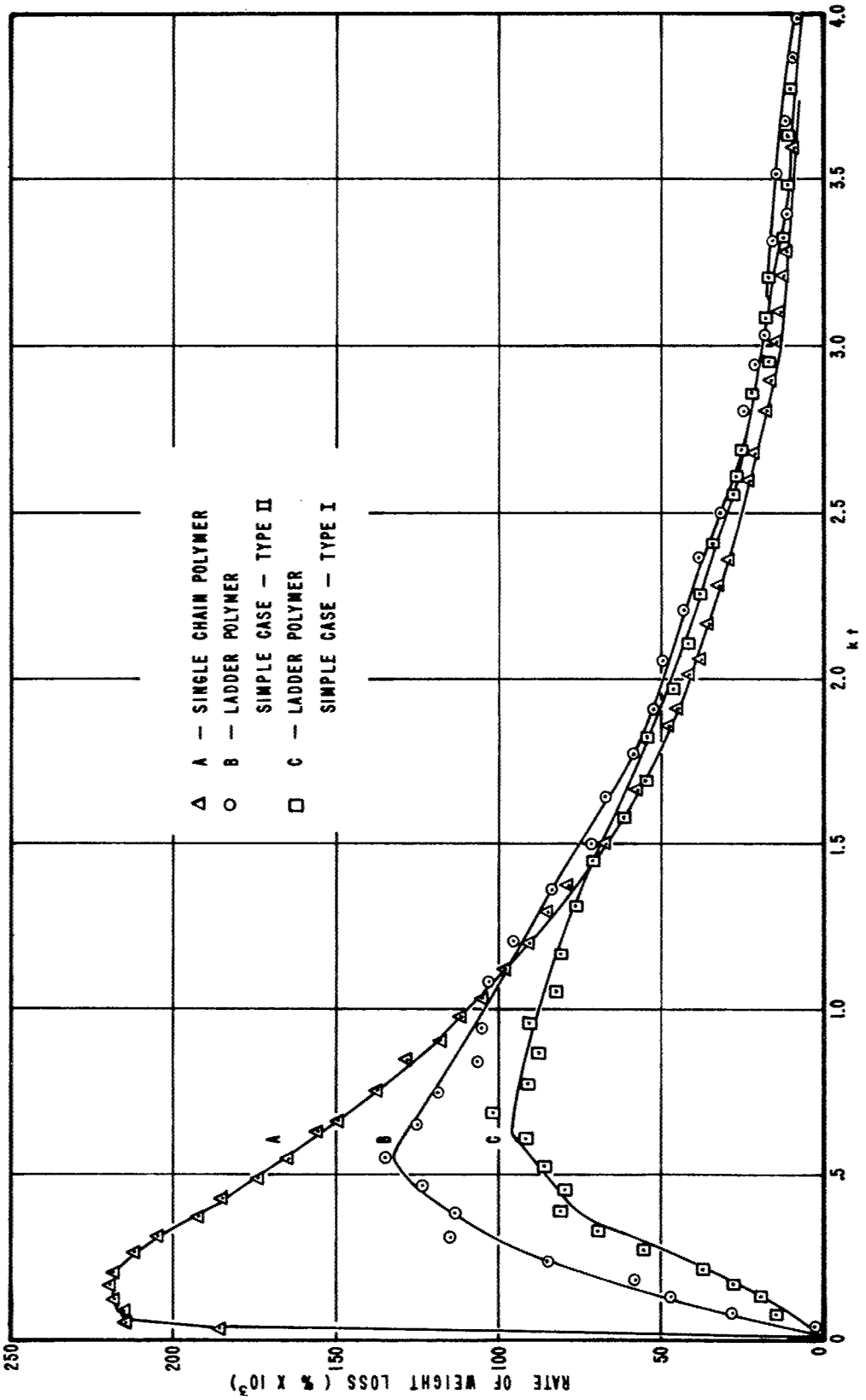


Figure 4. Plot of Rate of Weight Loss Versus Time for Simple Case Ladder Polymers Undergoing Random Degradation.

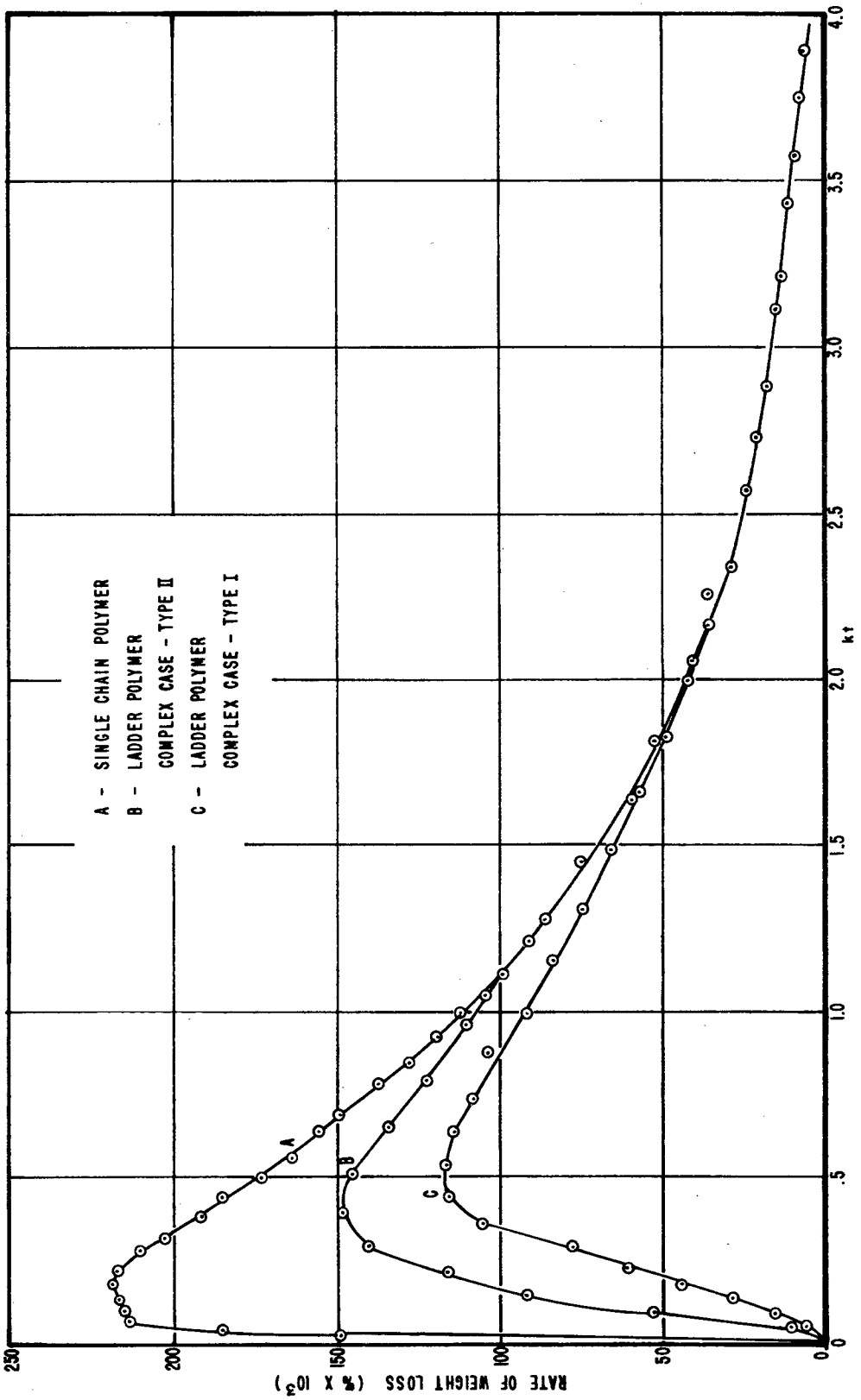


Figure 5. Plot of Rate of Weight Loss Versus Time for Complex Case Ladder Polymers Undergoing Random Degradation.

APPENDIX I

COMPUTER PROGRAM

VAPORIZATION OF SINGLE CHAIN POLYMER

```
C10 VOLATIZATION OF SINGLE CHAIN FROM RANDOM NUMBER GENERATION
      DIMENSION L(100,200),E(10)
      FREQUENCY 7(5,1,0),110(9,1,1),194(5,1,0),118(1,1,9),196(5,1,0)
     1,151(0,1,9),200(0,1,1),204(0,1,1)
      DO 25 IX=1,70
      DO 24 I=1,200
      R=RAND1(Y)*19900.
      NR=R
      ML=NR/199+1
      NB=199*ML-NR
 7    IF(L(ML,NB)-1)28,11,11
 28   L(ML,NB)=1
C VAPORIZATION OCCURS WHEN CHAIN IS TEN ATOMS OR LESS
      DO 103 KW=1,10
 110  IF(NB+KW-199)104,104,105
 104  NBAKW=NB+KW
 194  IF(L(ML,NBAKW)-1)103,105,105
 103  CONTINUE
C CHECKED ALL BREAKS TO THE RIGHT
      GO TO 150
 105  KB=KW
      E(KB)=E(KB)+1.
      DO 125 KK=1,KW
      NBAKK=NB+KK
 200   IF(1-L(ML,NBAKK))125,125,201
 201  BVOL=BVOL+1.
 125  L(ML,NBAKK)=1
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
 150  DO 115 KW=1,10
 118  IF(NB-KW)108,108,116
 116  NBAKW=NB-KW
 196  IF(L(ML,NBAKW)-1)115,108,108
 115  CONTINUE
C CHECKED ALL BREAKS TO THE LEFT
      GO TO 24
 108  KB=KW
      E(KB)=E(KB)+1.
      DO 126 KK=1,KW
      NBAKK=NB-KK
 204   IF(1-L(ML,NBAKK))126,126,205
 205  BVOL=BVOL+1.
 126  L(ML,NBAKK)=1
```

```
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
      GO TO 24
 11   NDUP=NDUP+1
 24   CONTINUE
      XI=IX
      DUPN=NDUP
      TIME=LOGF(19900./(19900.-200.*XI+DUPN-BVOL))
C MOLECULAR WEIGHT OF EACH ATOM IS 14
      VOL=E(1)*14.+E(2)*28.+E(3)*42.+E(4)*56.+E(5)*70.+E(6)*84.
     1 +E(7)*98.+E(8)*112.+E(9)*126.+E(10)*140.
      PCVOL=100.*VOL/(200.*1400.)
      DIMENSION A(70),T(70)
      A(IX)=VOL
      T(IX)=TIME
 151  IF(IX-1)152,152,155
 155  IY=IX-1
      DWT=(A(IX)-A(IY))/(T(IX)-T(IY))
      GO TO 153
 152  DWT=A(IX)/T(IX)
 153  WRITE OUTPUT TAPE 3,30,PCVOL,TIME,DWT,NDUP,VOL
 30   FORMAT(1X,3F20.6,I15,F20.2)
 25   CONTINUE
      CALL EXIT
      END
)1 (X-  -   (G7                                                          070SQU00
*J -9(NZ5GA 7 -2 -E -G  -  3 5-  - 97 95 -- 94 78 -- -5 -5 -7 -- 4E -4 -070SQU01
*U7(P($T    9 -G  5484 -1 -9 -5 -     2(X)  -  9  -                      070SQU02
```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APPENDIX II

COMPUTER PROGRAM

VAPORIZATION OF LADDER POLYMER

SIMPLE CASE - TYPE I

```
C9 TYPE I LADDER POLYMER FOUR OR LESS LADDER UNITS VOLATILE
      DIMENSION L(100,200),E(6)
      FREQUENCY 7(1,0,2),8(5,1,1),9(5,1,1),110(9,1,1),194(5,1,0)
     1,118(1,1,9),196(5,1,0),4(1,0,1),10(5,1,1),13(1,1,5),133(9,1,1)
     2,184(5,1,0),140(1,1,9),192(5,1,0),14(5,1,1),150(0,1,9)
     3,200(1,1,3),203(1,1,3),206(1,1,3),210(1,1,3),213(1,1,3)
     4,216(1,1,3),220(1,1,3),223(1,1,3),226(1,1,3),230(1,1,3)
     5,233(1,1,3),236(1,1,3),320(0,1,9),330(0,1,9),360(0,1,9),390(0,1,9)
     6,500(1,1,5),310(1,1,5),600(1,1,5),340(1,1,5),610(1,1,5),370(1,1,5)
     7,620(1,1,5),400(1,1,5)
      DO 25 IX=1, 40
      DO 24 I=1,800
      R=RAND1(Y)*19900.
      NR=R
      ML=NR/199+1
      NB=199*ML-NR
 7    IF(NB-66)8,8,4
 8    IF(L(ML,NB)-1)28,11,11
 28   L(ML,NB)=1
 9    IF(L(ML,NB+66)-1)24,100,100
 100  L(ML,NB)=2
      L(ML,NB+66)=2
C MOLECULE IS BROKEN
      DO 103 KW=1,5
 110  IF(NB+KW-66)104,104,105
 104  NBAKW=NB+KW
 194  IF(L(ML,NBAKW)-2)103,105,105
 103  CONTINUE
C CHECKED ALL BREAKS TO THE RIGHT
      GO TO 114
 105  KB=KW
      E(KB)=E(KB)+1.
      KW=KB-1
 320  IF(KW)310,310,300
 300  DO 125 KK=1,KW
      NBAKK=NB+KK
 200  IF(1-L(ML,NBAKK))201,201,202
 202  BVOL=BVOL+1.
 201  L(ML,NBAKK)=2
      NN=NB+66+KK
 203  IF(1-L(ML,NN))204,204,205
 205  BVOL=BVOL+1.
```

```
 204  L(ML,NN)=2
      NY=NB+132+KK
 206  IF(1-L(ML,NY))125,125,207
 207  BVOL=BVOL+1.
 125  L(ML,NY)=2
 500  IF(1-L(ML,NY+1))501,501,502
 502  BVOL=BVOL+1.
 501  L(ML,NY+1)=2
 310  IF(1-L(ML,NB+133))503,503,504
 504  BVOL=BVOL+1.
 503  L(ML,NB+133)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
 114  DO 115 KW=1,5
 118  IF(NB-KW)108,108,116
 116  NBAKW=NB-KW
 196  IF(L(ML,NBAKW)-2)115,108,108
 115  CONTINUE
C CHECKED ALL BREAKS TO THE LEFT
      GO TO 24
 108  KB=KW
      E(KB)=E(KB)+1.
      KW=KB-1
 330  IF(KW)340,340,350
 350  DO 131 KK=1,KW
      NBAKK=NB-KK
 210  IF(1-L(ML,NBAKK))211,211,212
 212  BVOL=BVOL+1.
 211  L(ML,NBAKK)=2
      NN=NB+66-KK
 213  IF(1-L(ML,NN))214,214,215
 215  BVOL=BVOL+1.
 214  L(ML,NN)=2
      NY=NB+133-KK
 216  IF(1-L(ML,NY))131,131,217
 217  BVOL=BVOL+1.
 131  L(ML,NY)=2
 600  IF(1-L(ML,NY-1))505,505,506
 506  BVOL=BVOL+1.
 505  L(ML,NY-1)=2
 340  IF(1-L(ML,NB+132))507,507,508
 508  BVOL=BVOL+1.
 507  L(ML,NB+132)=2
```

```
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
      GO TO 24
 4    IF(NB-132)10,10,14
 10   IF(L(ML,NB)-1)17,11,11
 17   L(ML,NB)=1
 13   IF(1-L(ML,NB-66))120,120,24
 120  L(ML,NB)=2
      L(ML,NB-66)=2
C MOLECULE IS BROKEN
      DO 132 KW=1,5
 133  IF(NB+KW-132)134,134,135
 134  NBAKW=NB+KW
 184  IF(L(ML,NBAKW)-2)132,135,135
 132  CONTINUE
C CHECKED ALL BREAKS TO THE RIGHT
      GO TO 138
 135  KB=KW
      E(KB)=E(KB)+1.
      KW=KB-1
 360  IF(KW)370,370,380
 380  DO 136 KK=1,KW
      NBAKK=NB+KK
 220  IF(1-L(ML,NBAKK))221,221,222
 222  BVOL=BVOL+1.
 221  L(ML,NBAKK)=2
      NN=NB-66+KK
 223  IF(1-L(ML,NN))224,224,225
 225  BVOL=BVOL+1.
 224  L(ML,NN)=2
      NY=NB+66+KK
 226  IF(1-L(ML,NY))136,136,227
 227  BVOL=BVOL+1.
 136  L(ML,NY)=2
 610  IF(1-L(ML,NY+1))509,509,510
 510  BVOL=BVOL+1.
 509  L(ML,NY+1)=2
 370  IF(1-L(ML,NB+67))511,511,512
 512  BVOL=BVOL+1.
 511  L(ML,NB+67)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
 138  DO 139 KW=1,5
 140  IF(NB-KW-66)148,148,142
```

```
 142   NBAKW=NB-KW
 192   IF(L(ML,NBAKW)-2)139,148,148
 139   CONTINUE
C CHECKED ALL,BREAKS TO THE LEFT
       GO TO 24
 148   KB=KW
       E(KB)=E(KB)+1.
       KW=KB-1
 390   IF(KW)400,400,410
 410   DO 149 KK=1,KW
       NBAKK=NB-KK
 230   IF(1-L(ML,NBAKK))231,231,232
 232   BVOL=BVOL+1.
 231   L(ML,NBAKK)=2
       NN=NB-66-KK
 233   IF(1-L(ML,NN))234,234,235
 235   BVOL=BVOL+1.
 234   L(ML,NN)=2
       NY=NB+67-KK
 236   IF(1-L(ML,NY))149,149,237
 237   BVOL=BVOL+1.
 149   L(ML,NY)=2
 620   IF(1-L(ML,NY-1))513,513,514
 514   BVOL=BVOL+1.
 513   L(ML,NY-1)=2
 400   IF(1-L(ML,NB+66))515,515,516
 516   BVOL=BVOL+1.
 515   L(ML,NB+66)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
       GO TO 24
 14    IF(L(ML,NB)-1)12,11,11
 12    L(ML,NB)=1
       GO TO 24
 11    NDUP=NDUP+1
 24    CONTINUE
       XI=IX
       DUPN=NDUP
       TIME=LOGF(19900./(19900.-800.*XI+DUPN-BVOL))
C MOLECULAR WEIGHT OF EACH ATOM IS 14
       VOL=E(1)*28.+E(2)*56.+E(3)*84.+E(4)*112.+E(5)*140.
       PCVOL=100.*VOL/(134.*1400.)
       DIMENSION A(70),T(70)
```

```
      A(IX)=VOL
      T(IX)=TIME
 150  IF(IX-1)152,152,155
 155  IY=IX-1
      DWT=(A(IX)-A(IY))/(T(IX)-T(IY))
      GO TO 153
 152  DWT=A(IX)/T(IX)
 153  WRITE OUTPUT TAPE 3,30,PCVOL,TIME,DWT,NDUP,VOL
 30   FORMAT(1X,3F20.6,I15,F20.2)
 25   CONTINUE
      CALL EXIT
      END
)1 (X-  -   (G7                                                                  070SQU00
*J -9(NZ5GA 7 -2 -E -G  -  3 5-  - 97 95 -- 94 78 -- -5 -5 -7 -- 4E -4 -070SQU01
*U7(P($T    9 -G  5484 -1 -9 -5 -     2(X)  -  9  -                              070SQU02
```

APPENDIX III

COMPUTER PROGRAM

VAPORIZATION OF LADDER POLYMER

SIMPLE CASE - TYPE II

```
C14 TYPE II LADDER POLYMER TWO OR LESS LADDER UNITS VOLATILE
      FREQUENCY 7(1,0,2),8(0,1,1),700(1,1,0),40(1,1,5),42(1,1,5)
     1,41(1,1,5),43(1,1,5),110(9,1,1),194(9,1,0),320(0,1,9),200(1,1,5)
     2,702(1,1,0),703(0,1,5),704(0,1,5),711(9,1,1),714(5,1,0),716(0,1,9)
     3,723(1,1,5),731(1,1,5),118(1,1,9),122(5,1,0),330(0,1,9),813(1,1,5)
     4,742(1,1,0),743(0,1,5),744(0,1,5),751(1,1,9),757(5,1,0),758(0,1,9)
     5,762(1,1,5),770(1,1,5),4(1,0,1),10(0,1,1),400(1,1,0),401(1,1,5)
     6,404(1,1,5),402(1,1,5),412(1,1,5),340(9,1,1),343(5,1,0),344(0,1,9)
     7,348(1,1,5),350(1,1,0),351(0,1,5),352(0,1,5),361(9,1,1),365(5,1,0)
     8,371(0,1,9),374(1,1,5),381(1,1,5),384(9,1,1),387(5,1,0),391(0,1,9)
     9,394(1,1,5),600(1,1,0),601(0,1,5),602(0,1,5),611(1,1,9),614(5,1,0)
      FREQUENCY 630(0,1,9),634(1,1,5),661(1,1,5),14(5,1,1),690(0,1,9)
     1,260(0,1,5),270(0,1,5),281(0,1,5),290(0,1,5)
     2,910(1,1,5),900(1,1,0),940(0,1,5),941(0,1,5),947(0,1,1),980(0,1,1)
     3,948(0,1,1),951(0,1,1),981(0,1,1),952(0,1,1),960(3,0,1)
      DIMENSION L(100,200),E(6)
      DO 25 IX=1,60
      DO 24 I=1,800
      R=RAND1(Y)*19600.
      NR=R
      ML=NR/196+1
      NB=196*ML-NR
 960  IF(NB-156)910,910,14
 910  IF(1-L(ML,NB))11,11,911
 911  NA=NB/2
 900  IF(2*NA-NB)940,941,940
 940  IF(2-L(ML,NB+1))942,943,942
 942  LOP=0
      LOPP=0
C LOP=0 MEANS ADJACENT BOND NOT BROKEN///IF LOP=1,ADJACENT BOND BROKEN
C LOPP=0 MEANS NB IS ODD NUMBER///LOOP=1 MEANS NB IS EVEN NUMBER
      GO TO 7
 943  L(ML,NB)=2
      LOP=1
      LOPP=0
      E(1)=E(1)+1.
      GO TO 7
 941  IF(2-L(ML,NB-1))944,945,944
 944  LOPP=1
      LOP=0
      GO TO 7
 945  LOPP=1
```

```
      LOP=1
      L(ML,NB)=2
      E(1)=E(1)+1.
      GO TO 7
 7    IF(NB-78)8,8,4
 8    IF(1-LOP)947,947,28
 947  IF(LOPP)114,114,701
 28   L(ML,NB)=1
      NA=NB/2
 700  IF(2*NA-NB)40,41,40
  40  IF(1-L(ML,NB+78))100,100,42
  42  IF(1-L(ML,NB+79))101,101,24
 101  L(ML,NB+79)=2
      GO TO 105
 100  L(ML,NB+78)=2
 105  L(ML,NB)=2
      GO TO 701
  41  IF(1-L(ML,NB+78))102,102,43
  43  IF(1-L(ML,NB+77))203,203,24
 203  L(ML,NB+77)=2
      GO TO 104
 102  L(ML,NB+78)=2
 104  L(ML,NB)=2
C MOLECULE IS BROKEN
 701  DO 103 KW=1,5
 110  IF(NB+KW-78)106,106,800
 106  NBAKW=NB+KW
 194  IF(L(ML,NBAKW)-2)103,800,800
 103  CONTINUE
C CHECKED ALL BREAKS TO THE RIGHT ON TOP
      GO TO 980
 800  NA=NB/2
      NKW=KW
 702  IF(2*NA-NB)703,704,703
 703  IF(2-L(ML,NB+78))707,705,707
 707  NBA=NB+79
      GO TO 706
 705  NBA=NB+78
      GO TO 706
 704  IF(2-L(ML,NB+78))708,709,708
 708  NBA=NB+77
      GO TO 706
```

```
 709 NBA=NB+78
 706  DO 710 KV=1,5
 711  IF(NBA+KV-156)712,712,715
 712  NBAKV=NBA+KV
 714  IF(L(ML,NBAKV)-2)710,715,715
 710  CONTINUE
      GO TO 980
C CHECKED ALL BREAKS TO THE RIGHT ON BOTTOM
 715  E(KV)=E(KV)+1.
      NBAKV=NBA+KV
      KV=KV-1
 716  IF(KV)108,108,721
 721  DO 722 KK=1,KV
      NBAKK=NBA+KK
  723 IF(1-L(ML,NBAKK))722,722,725
  725 BVOL=BVOL+1.
  722 L(ML,NBAKK)=2
  108 KW=NKW
      E(KW)=E(KW)+1.
      KW=KW-1
  320 IF(KW)310,310,300
  300 DO 125 KK=1,KW
      NBAKK=NB+KK
  200 IF(1-L(ML,NBAKK))125,125,202
  202 BVOL=BVOL+1.
  125 L(ML,NBAKK)=2
  310 NW=NB-NB/2+157
      NX=NBAKV-78-(NBAKV-78)/2+157
      KB=NX-NW
 260  IF(KB)980,980,251
 251  DO 730 KK=1,KB
      NBAKK=NW-1+KK
  731 IF(1-L(ML,NBAKK))730,730,733
  733 BVOL=BVOL+1.
 730  L(ML,NBAKK)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
 980  IF(LOP)114,114,948
 948  IF(LOPP)114,114,24
 114  DO 115 KW=1,5
 118  IF(NB-KW)810,810,121
 121  NBAKW=NB-KW
 122  IF(L(ML,NBAKW)-2)115,810,810
```

```
  115 CONTINUE
C CHECKED ALL BREAKS TO LEFT ON TOP
      GO TO 24
  810 NA=NB/2
      NKW=KW
  742 IF(2*NA-NB)743,744,743
  743 IF(2-L(ML,NB+78))747,745,747
  747 NBA=NB+79
      GO TO 746
  745 NBA=NB+78
      GO TO 746
  744 IF(2-L(ML,NB+78))748,749,748
  748 NBA=NB+77
      GO TO 746
  749 NBA=NB+78
 746  DO 750 KV=1,5
 751  IF(NBA-KV-79)755,756,756
 756  NBAKV=NBA-KV
 757  IF(L(ML,NBAKV)-2)750,755,755
 750  CONTINUE
      GO TO 24
C CHECKED ALL BREAKS TO THE LEFT ON BOTTOM
 755  E(KV)=E(KV)+1.
      NBAKV=NBA-KV
      KV=KV-1
 758  IF(KV)120,120,761
 761  DO 781 KK=1,KV
      NBAKK=NBA-KK
  762 IF(1-L(ML,NBAKK))781,781,764
  764 BVOL=BVOL+1.
  781 L(ML,NBAKK)=2
  120 KW=NKW
      E(KW)=E(KW)+1.
      KW=KW-1
  330 IF(KW)760,760,811
  811 DO 812 KK=1,KW
      NBAKK=NB-KK
  813 IF(1-L(ML,NBAKK))812,812,814
  814 BVOL=BVOL+1.
  812 L(ML,NBAKK)=2
  760 NW=NB-NB/2+157
      NX=NBAKV-78-(NBAKV-78)/2+157
```

```
      KB=NW-NX
 270  IF(KB)24,24,252
 252  DO 765 KK=1,KB
      NBAKK=NW-KK
  770 IF(1-L(ML,NBAKK))765,765,772
  772 BVOL=BVOL+1.
  765 L(ML,NBAKK)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
      GO TO 24
 4    IF(NB-156)10,10,14
 10   IF(LOP)29,29,951
 951  IF(LOPP)339,339,410
 29   L(ML,NB)=1
      NA=NB/2
  400 IF(2*NA-NB)401,402,401
  401 IF(1-L(ML,NB-78))403,403,404
  404 IF(1-L(ML,NB-77))405,405,24
  405 L(ML,NB-77)=2
      GO TO 406
  403 L(ML,NB-78)=2
  406 L(ML,NB)=2
      GO TO 410
  402 IF(1-L(ML,NB-78))411,411,412
  412 IF(1-L(ML,NB-79))413,413,24
  413 L(ML,NB-79)=2
      GO TO 420
  411 L(ML,NB-78)=2
  420 L(ML,NB)=2
C MOLECULE IS BROKEN
  410 DO 421 KW=1,5
 340  IF(NB+KW-156)341,341,345
 341  NBAKW=NB+KW
 343  IF(L(ML,NBAKW)-2)421,345,345
  421 CONTINUE
C CHECKED ALL BREAKS TO THE RIGHT ON BOTTOM
      GO TO 981
  345 NA=NB/2
      NKW=KW
  350 IF(2*NA-NB)351,352,351
  351 IF(2-L(ML,NB-78))353,354,353
  353 NBA=NB-77
      GO TO 360
```

```
 354 NBA=NB-78
     GO TO 360
 352 IF(2-L(ML,NB-78))355,356,355
 355 NBA=NB-79
     GO TO 360
 356 NBA=NB-78
360  DO 370 KV=1,5
361  IF(NBA+KV-78)362,362,363
362  NBAKV=NBA+KV
365  IF(L(ML,NBAKV)-2)370,363,363
370  CONTINUE
     GO TO 981
C CHECKED ALL BREAKS TO THE RIGHT ON TOP
363  E(KV)=E(KV)+1.
     NBAKV=NBA+KV
     KV=KV-1
371  IF(KV)342,342,373
373  DO 380 KK=1,KV
     NBAKK=NBA+KK
 374 IF(1-L(ML,NBAKK))380,380,376
 376 BVOL=BVOL+1.
 380 L(ML,NBAKK)=2
 342 KW=NKW
     E(KW)=E(KW)+1.
     KW=KW-1
 344 IF(KW)372,372;346
 346 DO 347 KK=1,KW
     NBAKK=NB+KK
 348 IF(1-L(ML,NBAKK))347,347,349
 349 BVOL=BVOL+1.
 347 L(ML,NBAKK)=2
372  NW=NB-78-(NB-78)/2+157
     NX=NBAKV-NBAKV/2+157
     KB=NX-NW
281  IF(KB)981,981,358
358  DO 280 KK=1,KB
     NBAKK=NW-1+KK
 381 IF(1-L(ML,NBAKK))280,280,383
 383 BVOL=BVOL+1.
280  L(ML,NBAKK)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
981  IF(LOP)339,339,952
```


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```
952   IF(LOPP)339,339,24
339   DO 390 KW=1,5
384   IF(NB-KW-78)392,392,386
386   NBAKW=NB-KW
387   IF(L(ML,NBAKW)-2)390,392,392
390   CONTINUE
C CHECKED ALL BREAKS TO LEFT ON BOTTOM
      GO TO 24
392   NA=NB/2
      NKW=KW
600   IF(2*NA-NB)601,602,601
601   IF(2-L(ML,NB-78))603,604,603
603   NBA=NB-77
      GO TO 610
604   NBA=NB-78
      GO TO 610
602   IF(2-L(ML,NB-78))605,606,605
605   NBA=NB-79
      GO TO 610
606   NBA=NB-78
610   DO 620 KV=1,5
611   IF(NBA-KV)612,612,613
613   NBAKV=NBA-KV
614   IF(L(ML,NBAKV)-2)620,612,612
620   CONTINUE
      GO TO 24
C CHECKED ALL BREAKS TO THE LEFT ON TOP
612   E(KV)=E(KV)+1.
      NBAKV=NBA-KV
      KV=KV-1
630   IF(KV)388,388,632
632   DO 640 KK=1,KV
      NBAKK=NBA-KK
634   IF(1-L(ML,NBAKK))640,640,635
635   BVOL=BVOL+1.
640   L(ML,NBAKK)=2
388   KW=NKW
      E(KW)=E(KW)+1.
      KW=KW-1
391   IF(KW)631,631,393
393   DO 399 KK=1,KW
      NBAKK=NB-KK
```

```
  394 IF(1-L(ML,NBAKK))399,399,395
  395 BVOL=BVOL+1.
  399 L(ML,NBAKK)=2
 631  NW=NBAKV-NBAKV/2+157
      NX=NB-78-(NB-78)/2+157
      KB=NX-NW
 290  IF(KB)24,24,259
 259  DO 660 KK=1,KB
      NBAKK=NX-KK
  661 IF(1-L(ML,NBAKK))660,660,662
  662 BVOL=BVOL+1.
  660 L(ML,NBAKK)=2
C VAPORIZED SEGMENTS REMOVED FROM SYSTEM
      GO TO 24
   14 IF(L(ML,NB)-1)688,11,11
  688 L(ML,NB)=1
      GO TO 24
   11 NDUP=NDUP+1
   24 CONTINUE
      XI=IX
      DUPN=NDUP
      TIME=LOGF(19600./(19600.-800.*XI+DUPN-BVOL))
C MOLECULAR WEIGHT OF EACH ATOM IS 14
      VOL=E(1)*14.+E(2)*28.+E(3)*42.+E(4)*56.+E(5)*70.
      PCVOL=100.*VOL/(158.*1400.)
      DIMENSION A(70),T(70)
      A(IX)=VOL
      T(IX)=TIME
  690 IF(IX-1)691,691,692
  692 IY=IX-1
      DWT=(A(IX)-A(IY))/(T(IX)-T(IY))
      GO TO 695
  691 DWT=A(IX)/T(IX)
  695 WRITE OUTPUT TAPE 3,30,PCVOL,TIME,DWT,NDUP,VOL
   30 FORMAT(1X,3F20.6,I15,F20.2)
   25 CONTINUE
      CALL EXIT
      END
)1 (X-  -   (G7                                                          070SQU00
*J -9(NZ5GA 7 -2 -E -G  -  3 5-  - 97 95 -- 94 78 -- -5 -5 -7 -- 4E -4 -070SQU01
*U7(P($T    9 -G  5484 -1 -9 -5 -     2(X)  -  9  -                      070SQU02
```

APPENDIX IV

COMPUTER PROGRAM

VAPORIZATION OF LADDER POLYMER

COMPLEX CASE - TYPE I

```
C25 TYPE I COMPLEX-10 ATOMS OR LESS VOLATILE
      WRITE OUTPUT TAPE 3,111
  111  FORMAT(8X,5HPCVOL,12X,4HTIME,11X,3HDWT,11X,4HNDUP,6X,3HVOL,
     1 10X, 4HBVOL,12X,1HE,11X,2HEE,9X,2HXI)
      DIMENSION L(100,200)
      FREQUENCY 1(0,1,5),2(1,0,2),8(9,1,1),400(0,2,1),404(0,2,1),4(1,0,1
     1),480(9,1,1),426(0,2,1),430(0,2,1),482(0,0,9),486(5,1,1),489(5,1,1
     2),497(5,1,1),502(0,1,1),503(0,1,1),702(9,0,0),706(5,1,1),511(5,1,1
     3),531(5,1,1),541(0,1,1),542(0,1,1),600(0,1,9)
      COMMON L,NB,NBA,E,BVOL,IEE,IND,LA,LB,LC,ML,GOG,GAG
      DO 25 IX=1,25
      DO 24 I=1,600
      R=RAND1(Y)*19900.
      NR=R
      ML=NR/199+1
      NB=199*ML-NR
      GAG=0
      GOG=0
      LC=0
C UNBROKEN BOND=0   BROKEN BOND=1   OPPOSITE BROKEN MOLECULE=3
C ADJACENT BROKEN MOLECULE=2   VAPORIZED BOND FROM OPPOS. BROKEN MOL.=4
C  WHEN OPPISITE BOND IS ALREADY 2, IT BECOMES 5 RATHER THAN 3
C IF NBA IS ORIGINALLY 3,4,OR 5 AND NBA+LB-LA IS 3,4, OR 5, THEN GAG=1 AND
C  AND IEE=1.
    1 IF(1-L(ML,NB))11,11,7
    7 L(ML,NB)=1
    2 IF(NB-66)8,8,4
C CHECK OPPOSITE BOND
    8 IF(L(ML,NB+66)-1)12,13,13
   13 L(ML,NB)=3
      IF(L(ML,NB+66)-2)711,712,741
 712  L(ML,NB+66)=5
      GO TO 713
 741  GOG=1.
 711  L(ML,NB+66)=3
C MOLECULEE IS BROKEN
C IND=0 MEANS NO VAPORIZATION HAS OCCURED. IND=1 MEANS VAPORIZ. OCCURED.
 713  NBA=NB+66
C CHECK TO LEFT NEXT
      GAG=GOG
      CALL MVAP(0,1,0)
  400 IF(IND)12,12,402
```

```
   12 NBA=NB+66
      GAG=0
      CALL MLNK(0,1,0)
  402 NBA=NB+66
      IF(L(ML,NB+66)-1)405,900,900
C CHECK TO RIGHT NEXT
  900 GAG=GOG
      CALL MVAP(1,0,67)
  404 IF(IND)405,405,24
  405 GAG=0
      CALL MLNK(1,0,67)
      GO TO 24
    4 IF(NB-132)480,480,481
  480 IF(L(ML,NB-66)-1)420,421,421
C CHECKED OPPOSITE BOND
  421 L(ML,NB)=3
      IF(L(ML,NB-66)-2)714,715,744
 715  L(ML,NB-66)=5
      GO TO 716
 744  GOG=1.
 714  L(ML,NB-66)=3
C MOLECULE IS BROKEN
 716  NBA=NB-66
C CHECK TO LEFT NEXT
      GAG=GOG
      CALL MVAP(0,1,0)
  426 IF(IND)420,420,428
  420 NBA=NB-66
      GAG=0
      CALL MLNK(0,1,0)
  428 NBA=NB-66
C CHECK TO RIGHT NEXT
      IF(L(ML,NB-66)-1)431,901,901
  901 GAG=GOG
      CALL MVAP(1,0,67)
  430 IF(IND)431,431,24
  431 GAG=0
      CALL MLNK(1,0,67)
      GO TO 24
C BROKEN BOND IS CROSSLINK
  481 DO 485 K=1,67
  482 IF(NB-K-133)483,484,484
  484 NBK=NB-K
```

```
  486 IF(L(ML,NBK)-1)487,485,485
  485 CONTINUE
C ALL CROSSLINKS TO THE LEFT CHECKED
  487 DO 488 KK=1,K
      NX=NB-132-KK
  489 IF(L(ML,NX)-1)488,490,490
  488 CONTINUE
      NB1=0
      GO TO 495
  490 NB1=NX
C CHECKED ADJACENT BONDS TO THE LEFT - NB LESS 66
C   MUST ALSO CHECK ASSUMED NB FOR GAG WHEN ORIGINAL NB IS CROSSLINK.
C  LC=1 IF NB IS 3,4, OR 5 AND NB + LB-LA IS 3,4, OR 5
  495 DO 496 KK=1,K
      NX=NB-66-KK
  497 IF(L(ML,NX)-1)496,498,498
  496 CONTINUE
      NB2=0
      GO TO 500
C CHECKED OPPOSITE BONDS TO THE LEFT - NB MORE THAN 66
  498 NB2=NX
  500 NBA1=NB
  502 IF(NB1)503,503,504
  504 NB=NB1
      GAG=0
      IF(L(ML,NB1)-2)640,640,641
  640 LC=0
      GO TO 642
  641  LC=1
  642  CALL MLNK(1,0,67)
  503 IF(NB2)800,800,506
  506 NB=NB2
      GAG=0
      IF(L(ML,NB2)-2)644,644,645
  644  LC=0
      GO TO 646
  645  LC=1
  646  CALL MLNK(1,0,67)
  800 NB=NBA1
      GO TO 24
  483 DO 705 K=1,67
  702 IF(NB+K-199)703,703,704
  703 NX=NB+K
```

```
  706 IF(L(ML,NX)-1)710,705,705
  705 CONTINUE
C ALL CROSSLINKS TO THE RIGHT CHECKED
  704 K=K-1
  710 DO 550 KK=1,K
      NX=NB-133+KK
  511 IF(L(ML,NX)-1)550,513,513
  550 CONTINUE
      NB3=0
      GO TO 530
  513 NB3=NX
C CHECKED ADJACENT BONDS TO THE RIGHT - NB LESS 66
  530 DO 551 KK=1,K
      NX=NB-67+KK
  531 IF(L(ML,NX)-1)551,533,533
  551 CONTINUE
      NB4=0
      GO TO 540
C CHECKED OPPOSITE BOND TO THE RIGHT - NB MORE THAN 66
  533 NB4=NX
  540 NBA1=NB
  541 IF(NB3)542,542,543
  543 NB=NB3
      GAG=0
      IF(L(ML,NB3)-2)780,780,781
  780  LC=0
      GO TO 782
  781  LC=1
  782  CALL MLNK(0,1,0)
  542 IF(NB4)544,544,545
  545 NB=NB4
      IF(L(ML,NB4)-2)790,790,791
  790  LC=0
      GO TO 792
  791  LC=1
  792  CALL MLNK(0,1,0)
  544 NB=NBA1
      GO TO 24
   11 NDUP=NDUP+1
   24 CONTINUE
      XI=IX
      DUPN=NDUP
      TIME=LOGF(19900./(19900.-600.*XI+DUPN-BVOL))
```

```
      EE=IEE
      VOL=14.*(E-EE)
      PCVOL=100.*VOL/(1400.*134.)
      DIMENSION A(70),T(70)
      A(IX)=VOL
      T(IX)=TIME
  600 IF(IX-1)601,601,602
  602 IY=IX-1
      DWT=(A(IX)-A(IY))/(T(IX)-T(IY))
      GO TO 603
  601 DWT=A(IX)/T(IX)
  603 WRITE OUTPUT TAPE 3,30,PCVOL,TIME,DWT,NDUP,VOL,BVOL,E,EE,XI
   30 FORMAT(1X,3F16.6,I10,5F12.1)
   25 CONTINUE
      CALL EXIT
      END
      SUBROUTINE MVAP(LAD,LBD,LCD)
      DIMENSION L(100,200)
      FREQUENCY 30(1,1,9),33(1,0,1),39(1,1,9),44(5,1,1),801(5,1,1),
     152(1,0,1),802(5,1,1),59(1,0,1),363(5,1,1)
      COMMON L,NB,NBA,E,BVOL,IEE,IND,LA,LB,LC,ML,GOG,GAG
      LA=LAD
      LB=LBD
C CHECK IF BROKEN MOLECULE RESULTS IN VAPORIZATION
      DO 29 K=1,7
   30 IF((LB-LA)*NB+(LA-LB)*K*(1-2*LA)+67*LA+66*(NB/67)*(LA-LB))35,35,32
   32 NBK=NB+(LA-LB)*K
   33 IF(L(ML,NBK)-2)29,29,35
   29 CONTINUE
C CHECKED FOR BROKEN MOLECULE ON ADJACENT SIDE
      GO TO 45
   35 LN=K
      DO 38 K=1,7
   39 IF((LB-LA)*NBA+(LA-LB)*K*(1-2*LA)+67*LA+66*(NBA/67)*(LA-LB))
     1 43,43,41
   41 NBK=NBA+(LA-LB)*K
   42 IF(L(ML,NBK)-2)38,38,43
   38 CONTINUE
      GO TO 45
C CHECKED FOR BROKEN MOLECULE ON OPPOSITE SIDE
   43 LN1=K
   44 IF(LN+LN1-10)46,46,45
```

```
   45 IND=0
      RETURN
  46  IF(LN-1)946,901,946
  901  IF(LC)946,946,902
  902 IF((LB-LA)*NB+(LA-LB)*  (1-2*LA)+67*LA+66*(NB/67)*(LA-LB))
     1 920,920,905
  905  NLN=NB+LB-LA
      IF(L(ML,NLN)-2)946,946,920
  946  DO 50 K=1,LN
C MOLECULE HAS VAPORIZED
      NLN=NB+(LA-LB)*K+LB-LA
  801 IF(L(ML,NLN)-1)51,52,52
   51 BVOL=BVOL+1.
 52   IF(L(ML,NLN)-2)54,50,54
 54   IF(L(ML,NLN)-5)800,50,800
 800  E=E+1.
   50 L(ML,NLN)=4
C VAPORIZED ADJACENT BONDS
  920 IF(LN1-1)756,721,756
 721  IF(GAG)756,756,725
 725  IF((LB-LA)*NBA+(LA-LB)*(1-2*LA)+67*LA+66*(NBA/67)*(LA-LB))780,
     1780,758
 758  NLN=NBA+LB-LA
      IF(L(ML,NLN)-2)756,756,780
 756  DO 57 K=1,LN1
      NLN=NBA+(LA-LB)*K+LB-LA
  802 IF(L(ML,NLN)-1)58,59,59
   58 BVOL=BVOL+1.
   59 IF(L(ML,NLN)-2)60,80,60
 60   IF(L(ML,NLN)-5)811,80,811
 811  E=E+1.
 80   L(ML,NLN)=4
C VAPORIZED OPPOSITE BONDS
      NYY=NB+132+LB+(LA-LB)*K-(NB/67)*(66)
  363 IF(L(ML,NYY)-1)63,57,57
   63 BVOL=BVOL+1.
   57 L(ML,NYY)=4
C VAPORIZED CROSSLINKS
  780 IND=1
      RETURN
      END
      SUBROUTINE MLNK(LAD,LBD,LCD)
      DIMENSION L(100,200)
```

```
      FREQUENCY 100(1,1,9),106(5,1,1),107(0,1,1),810(5,1,1),811(0,1,1),
     1105(0,1,2),815(5,1,1),133(5,1,1),156(5,1,1),161(5,1,1),163(6,1,1),
     2152(0,1,1)
      COMMON L,NB,NBA,E,BVOL,IEE,IND,LA,LB,LC,ML,GOG,GAG
      LA=LAD
      LB=LBD
C CHECK IF BROKEN CROSSLINKS GIVE BROKEN MOLECULE
      DO 104 K=1,11
      NY=NB+132+LB+(LA-LB)*K-(NB/67)*(66)
  100 IF(200*LA+(LB-LA)*NY-132*LB)105,105,106
  106 IF(L(ML,NY)-1)107,104,104
  104 CONTINUE
      K=11
      GO TO 109
  107 IF(K-1)108,108,109
  108 RETURN
  109 K=K-1
      DO 110 KK=1,K
      NBA=NB+        66+(LA-LB)*KK-(NB/67)*(132)
  810 IF(L(ML,NBA)-1)110,150,150
  110 CONTINUE
C CHECKED IF OPPOSITE BOND IS BROKEN TO GIVE BROKEN MOLECULE
      GO TO 151
  150 L(ML,NB)=3
      IF(L(ML,NBA)-2)717,718,747
 718  L(ML,NBA)=5
      GO TO 719
 747  GAG=1.
 717  L(ML,NBA)=3
 719  CALL MVAP(LA,LB,LC)
C MOLECULE IS BROKEN
      NBAA=NBA
  811 IF(IND)151,151,152
  105 IF(K-1)130,130,131
  130 RETURN
  131 K=K-1
      DO 120 KK=1,K
      NLN=NB+(LA-LB)*KK+(LB-LA)
  815 IF(L(ML,NLN)-1)132,133,133
  132 BVOL=BVOL+1.
  133 IF(L(ML,NLN)-2)134,135,120
  135 IEE=IEE+1
```

```
  134 E=E+1.
      L(ML,NLN)=2
  120 CONTINUE
      RETURN
C VAPORIZED ADJACENT BONDS
  151 DO 155 KK=1,K
      NBA=NB+(LA-LB)*KK
  156 IF(L(ML,NBA)-1)155,158,158
  155 CONTINUE
      IF(L(ML,NB)-3)650,152,650
C CHECKED IF ADJACENT BOND IS BROKEN TO GIVE BROKEN MOLECULE
  158 DO 168 K=1,KK
      NBA=NB+(LA-LB)*K+LB-LA
  161 IF(L(ML,NBA)-1)162,163,163
  162 BVOL=BVOL+1.
  163 IF(L(ML,NBA)-2)164,165,166
  165 IEE=IEE+1
  164 E=E+1.
      L(ML,NBA)=2
C VAPORIZED ADJACENT BONDS
      GO TO 168
  166 E=E+1.
  168 CONTINUE
      IF(L(ML,NB)-3)650,152,650
C IF MOLECULE IS BROKEN,CHECK BOTH DIRECTIONS
  152 IF(LA)200,200,201
  200 LA=1
      LB=0
      LC=67
      GO TO 210
  201 LA=0
      LB=1
      LC=0
  210 NBA=NBAA
      CALL MVAP(LA,LB,LC)
  650 RETURN
      END
)1 (X-  -   (G7                                                          070SQU00
*J -9(NZ5GA 7 -2 -E -G  -  3 5-  - 97 95 -- 94 78 -- -5 -5 -7 -- 4E -4 -070SQU01
*U7(P($T    9 -G  5484 -1 -9 -5 -     2(X)  -  9  -                      070SQU02
```

APPENDIX V

COMPUTER PROGRAM

VAPORIZATION OF LADDER POLYMER

COMPLEX CASE - TYPE II

```
C26 TYPE II COMPLEX - 10 ATOMS OR LESS VOLATILE
      WRITE OUTPUT TAPE 3,111
 111  FORMAT(8X,5HPCVOL,12X,4HTIME,11X,3HDWT,11X,4HNDUP,6X,3HVOL,10X,
     14HBVOL,12X,1HE,11X,2HEE,9X,2HXI)
      DIMENSION L(100,200)
      FREQUENCY 1(0,1,5),2(1,0,2),700(1,1,0),40(1,1,5),42(1,1,5),701(1,0
     1,1),802(1,0,1),41(1,1,5),721(1,1,5),724(1,0,1),722(1,0,1),400(0,1,
     21),404(0,1,1),4(1,0,1),740(1,1,0),741(1,1,5),743(1,1,5),745(1,0,1)
     3,751(1,0,1),756(1,1,5),758(1,1,5),760(1,0,1),765(1,0,1),770(0,1,1)
     4,773(0,1,1),482(0,1,9),486(5,1,1),489(9,1,1),497(5,1,1),502(0,1,1)
     5,801(1,0,1),503(0,1,1),803(1,0,1),702(9,1,0),706(5,1,1),511(5,1,1)
     6,531(5,1,1),541(0,1,1),805(1,0,1),542(0,1,1),806(1,0,1),600(0,1,9)
      COMMON L,NB,NBA,E,BVOL,IEE,IND,LA,LB,LC,ML,GOG,GAG
      DO 25 IX=1,25
      DO 24 I=1,600
      R=RAND1(Y)*19600.
      NR=R
      ML=NR/196+1
      NB=196*ML-NR
      GAG=0
      GOG=0
      LC=0
 1    IF(1-L(ML,NB))11,11,7
 7    L(ML,NB)=1
 2    IF(NB-78)8,8,4
C CHECK OPPOSITE BOND
 8    NA=NB/2
 700  IF(2*NA-NB)40,41,40
 40   IF(1-L(ML,NB+78))100,100,42
 42   IF(1-L(ML,NB+79))101,101,21
 100  L(ML,NB)=3
 701  IF(L(ML,NB+78)-2)711,712,841
 712  L(ML,NB+78)=5
      GO TO 713
 841  GOG=1.
 711  L(ML,NB+78)=3
C MOLECULE IS BROKEN
 713  NBA=NB+78
      NNBA=NBA
      GO TO 200
 101  L(ML,NB)=3
 802  IF(L(ML,NB+79)-2)715,716,717
```

```
 716   L(ML,NB+79)=5
       GO TO 718
 717   GOG=1.
 715   L(ML,NB+79)=3
C MOLECULE IS BROKEN
 718   NBA=NB+79
       NNBA=NBA
       GO TO 200
 41    IF(1-L(ML,NB+78))720,720,721
 721   IF(1-L(ML,NB+77))722,722,21
 720   L(ML,NB)=3
 724   IF(L(ML,NB+78)-2)725,726,727
 726   L(ML,NB+78)=5
       GO TO 728
 727   GOG=1.
 725   L(ML,NB+78)=3
C MOLECULE IS BROKEN
 728   NBA=NB+78
       NNBA=NBA
       GO TO 200
 722   IF(L(ML,NB+77)-2)730,731,732
 731   L(ML,NB+77)=5
       GO TO 733
 732   GOG=1.
 730   L(ML,NB+77)=3
C MOLECULE IS BROKEN
 733   NBA=NB+77
       NNBA=NBA
 200   GAG=GOG
       CALL MVAP(0,1,0)
 400   IF(IND)12,12,402
 12    NBA=NNBA
       GAG=0
       CALL MLNK(0,1,0)
 402   NBA=NNBA
C CHECK TO RIGHT NEXT
       GAG=GOG
       CALL MVAP(1,0,67)
 404   IF(IND)405,405,24
 405   GAG=0
       CALL MLNK(1,0,67)
       GO TO 24
 21    NBA=NB+78
```

```
      GAG=0
      CALL MLNK(0,1,0)
      GAG=0
      NBA=NB+78
      CALL MLNK(1,0,67)
      GO TO 24
 4    IF(NB-156)480,480,481
 480  NA=NB/2
 740  IF(2*NA-NB)741,756,741
 741  IF(1-L(ML,NB-78))742,742,743
 743  IF(1-L(ML,NB-77))744,744,22
 742  L(ML,NB)=3
 745  IF(L(ML,NB-78)-2)746,747,748
 747  L(ML,NB+78)=5
      GO TO 750
 748  GOG=1.
 746  L(ML,NB-78)=3
C MOLECULE IS BROKEN
 750  NBA=NB-78
      NNBA=NBA
      GO TO 201
 744  L(ML,NB)=3
 751  IF(L(ML,NB-77)-2)752,753,754
 753  L(ML,NB-77)=5
      GO TO 755
 754  GOG=1.
 752  L(ML,NB-77)=3
C MOLECULE IS BROKEN
 755  NBA=NB-77
      NNBA=NBA
      GO TO 201
 756  IF(1-L(ML,NB-78))757,757,758
 758  IF(1-L(ML,NB-79))759,759,22
 757  L(ML,NB)=3
 760  IF(L(ML,NB-78)-2)761,762,763
 762  L(ML,NB-78)=5
      GO TO 764
 763  GOG=1.
 761  L(ML,NB-78)=3
C MOLECULE IS BROKEN
 764  NBA=NB-78
      NNBA=NBA
      GO TO 201
```

```
 759   L(ML,NB)=3
 765   IF(L(ML,NB-79)-2)766,767,768
 767   L(ML,NB-79)=5
       GO TO 769
 768   GOG=1.
 766   L(ML,NB-79)=3
C MOLECULE IS BROKEN
 769   NBA=NB-79
       NNBA=NBA
 201   GAG=GOG
       CALL MVAP(0,1,0)
 770   IF(IND)771,771,772
 771   NBA=NNBA
       GAG=0
       CALL MLNK(0,1,0)
 772   NBA=NNBA
C CHECK TO RIGHT NEXT
       GAG=GOG
       CALL MVAP(1,0,67)
 773   IF(IND)774,774,24
 774   GAG=0
       CALL MLNK(1,0,67)
       GO TO 24
 22    NBA=NB-78
       GAG=0
       CALL MLNK(0,1,0)
       GAG=0
       NBA=NB+78
       CALL MLNK(1,0,67)
       GO TO 24
C BROKEN BOND IS A CROSSLINK
 481   DO 485 K=1,40
 482   IF(NB-K-157)483,484,484
 484   NBK=NB-K
 486   IF(L(ML,NBK)-1)487,485,485
 485   CONTINUE
C ALL CROSSLINKS TO THE LEFT CHECKED
 487   K=2*K
       DO 488 KK=1,K
       NX=NB-156-KK+NB-157
 489   IF(L(ML,NX)-1)488,490,490
 488   CONTINUE
       NB1=0
```

```
      GO TO 495
 490  NB1=NX
C CHECKED ADJACENT BONDS TO THE LEFT-NB LESS 78
C MUST ALSO CHECK ASSUMED NB FOR GAG WHEN ORIGINAL NB IS CROSSLINK
C LC=1 IF NB IS 3,4,OR,5 ANDNB+LB-LA IS 3,4,OR,5
 495  DO 496 KK=1,K
      NX=NB-156-KK+NB-157-78
 497  IF(L(ML,NX)-1)496,498,498
 496  CONTINUE
      NB2=0
      GO TO 500
C CHECKED OPPOSITE BONDS TO THE LEFT-NB MORE THAN 78
 498  NB2=NX
 500  NBA1=NB
 502  IF(NB1)503,503,504
 504  NB=NB1
      GAG=0
 801  IF(L(ML,NB1)-2)640,640,641
 640  LC=0
      GO TO 642
 641  LC=1
 642  CALL MLNK(1,0,67)
 503  IF(NB2)800,800,506
 506  NB=NB2
      GAG=0
 803  IF(L(ML,NB2)-2)644,644,645
 644  LC=0
      GO TO 646
 645  LC=1
 646  CALL MLNK(1,0,67)
 800  NB=NBA1
      GO TO 24
 483  DO 705 K=1,40
 702  IF(NB+K-196)703,703,704
 703  NX=NB+K
 706  IF(L(ML,NX)-1)710,705,705
 705  CONTINUE
C ALL CROSSLINKS TO THE RIGHT CHECKED
 704  K=K-1
 710  K=2*K
      DO 550 KK=1,K
      NX=NB-157+KK+NB-157
 511  IF(L(ML,NX)-1)550,513,513
```

```
 550  CONTINUE
      NB3=0
      GO TO 530
 513  NB3=NX
C CHECKED ADJACENT BONDS TO THE RIGHT-NB LESS 66
 530  DO 551 KK=1,K
      NX=NB-79+KK+NB-157
 531  IF(L(ML,NX)-1)551,533,533
 551  CONTINUE
      NB4=0
      GO TO 540
C CHECKED OPPOSITE BOND TO THE RIGHT-NB MORE THAN 66
 533  NB4=NX
 540  NBA1=NB
 541  IF(NB3)542,542,543
 543  NB=NB3
      GAG=0
 805  IF(L(ML,NB3)-2)780,780,781
 780  LC=0
      GO TO 782
 781  LC=1
 782  CALL MLNK(0,1,0)
 542  IF(NB4)544,544,545
 545  NB=NB4
 806  IF(L(ML,NB4)-2)790,790,791
 790  LC=0
      GO TO 792
 791  LC=1
 792  CALL MLNK(0,1,0)
 544  NB=NBA1
      GO TO 24
 11   NDUP=NDUP+1
 24   CONTINUE
      XI=IX
      DUPN=NDUP
      TIME=LOGF(19600./(19600.-600.*XI+DUPN-BVOL))
      EE=IEE
      VOL=14.*(E-EE)
      PCVOL=100.*VOL/(1400.*158.)
      DIMENSION A(70),T(70)
      A(IX)=VOL
      T(IX)=TIME
```

```
600   IF(IX-1)601,601,602
602   IY=IX-1
      DWT=(A(IX)-A(IY))/(T(IX)-T(IY))
      GO TO 603
601   DWT=A(IX)/T(IX)
603   WRITE OUTPUT TAPE 3,30,PCVOL,TIME,DWT,NDUP,VOL,BVOL,E,EE,XI
30    FORMAT(1X,3F16.6,I10,5F12.1)
25    CONTINUE
      CALL EXIT
      END
      SUBROUTINE MVAP(LAD,LBD,LCD)
      DIMENSION L(100,200)
      FREQUENCY 30(1,1,5),33(1,0,1),39(1,1,5),42(1,0,1),44(5,1,1),46(0,1
     1,5),901(0,1,1),902(1,1,5),410(1,0,1),801(3,1,1),52(1,0,1),54(1,1,0
     2),920(0,1,5),721(0,1,1),725(1,1,5),420(1,0,1),802(5,1,1),59(1,0,1)
     3,60(1,1,0),363(2,1,1)
      COMMON L,NB,NBA,E,BVOL,IEE,IND,LA,LB,LC,ML,GOG,GAG
      LA=LAD
      LB=LBD
C CHECK IF BROKEN MOLECULE RESULTS IN VAPORIZATION
      DO 29 K=1,7
30    IF((LB-LA)*NB+(LA-LB)*K*(1-2*LA)+79*LA+78*(NB/79)*(LA-LB))35,
     135,32
32    NBK=NB+(LA-LB)*K
33    IF(L(ML,NBK)-2)29,29,35
29    CONTINUE
C CHECKED FOR BROKEN MOLECULE ON ADJACENT SIDE
      GO TO 45
35    LN=K
      DO 38 K=1,7
39    IF((LB-LA)*NBA+(LA-LB)*K*(1-2*LA)+79*LA+78*(NBA/79)*(LA-LB))
     143,43,41
41    NBK=NBA+(LA-LB)*K
42    IF(L(ML,NBK)-2)38,38,43
38    CONTINUE
      GO TO 45
C CHECKED FOR BROKEN MOLECULE ON OPPOSITE SIDE
43    LN1=K
44    IF(LN+LN1-10)46,46,45
45    IND=0
      RETURN
46    IF(LN-1)946,901,946
901   IF(LC)946,946,902
```

```
 902  IF((LB-LA)*NB+(LA-LB)*(1-2*LA)+79*LA+78*(NB/79)*(LA-LB))920,
     1920,905
 905  NLN=NB+LB-LA
 410  IF(L(ML,NLN)-2)946,946,920
 946  DO 50 K=1,LN
C MOLECULE HAS VAPORIZED
      NLN=NB+(LA-LB)*K+LB-LA
 801  IF(L(ML,NLN)-1)51,52,52
 51   BVOL=BVOL+1.
 52   IF(L(ML,NLN)-2)54,50,54
 54   IF(L(ML,NLN)-5)800,50,800
 800  E=E+1.
 50   L(ML,NLN)=4
C VAPORIZED ADJACENT BONDS
 920  IF(LN1-1)756,721,756
 721  IF(GAG)756,756,725
 725  IF((LB-LA)*NBA+(LA-LB)*(1-2*LA)+79*LA+78*(NBA/79)*(LA-LB))
     1780,780,758
 758  NLN=NBA+LB-LA
 420  IF(L(ML,NLN)-2)756,756,780
 756  DO 57 K=1,LN1
      NLN=NBA+(LA-LB)*K+LB-LA
 802  IF(L(ML,NLN)-1)58,59,59
 58   BVOL=BVOL+1.
 59   IF(L(ML,NLN)-2)60,80,60
 60   IF(L(ML,NLN)-5)811,80,811
 811  E=E+1.
 80   L(ML,NLN)=4
C VAPORIZED OPPOSITE BONDS
 57   CONTINUE
      KA=LN1/2
      KB=NB/2
      KAA=LN1-2*KA
      KBB=NB-2*KB
      KC=K/2+KAA*KBB*LB+KAA*(1-KBB)*LA
      DO 400 K=1,KC
      NYY=NB+LB+156-NB/2-(NB/79)*39+(LA-LB)*K
 363  IF(L(ML,NYY)-1)63,400,400
 63   BVOL=BVOL+1.
 400  L(ML,NYY)=4
C VAPORIZED CROSSLINKS
 780  IND=1
```

```
      RETURN
      END
      SUBROUTINE MLNK(LAD,LBD,LCD)
      DIMENSION L(100,200)
      FREQUENCY 100(1,1,5),107(0,1,5),600(0,1,1),810(5,1,1),610(1,0,1),
     1811(0,1,1),105(0,1,5),815(5,1,1),133(1,0,1),156(5,1,1),620(1,0,1),
     2161(5,1,1),163(1,0,1),630(1,0,1),152(0,1,1)
      COMMON L,NB,NBA,E,BVOL,IEE,IND,LA,LB,LC,ML,GOG,GAG
      LA=LAD
      LB=LBD
C CHECK IF BROKEN CROSSLINK GIVES BROKEN MOLECULE
      DO 104 K=1,6
      NY=NB+LB+156-NB/2-(NB/79)*39+(LA-LB)*K
 100  IF(197*LA+(LB-LA)*NY-156*LB)105,105,106
 106  IF(L(ML,NY)-1)107,104,104
 104  CONTINUE
      K=6
      GO TO 109
 107  IF(K-1)108,108,109
 108  KA=NB/2
      KB=NB-2*KA
 600  IF(LB*(1-KB)+LA*KB)40,41,40
 41   RETURN
   40 NY=NB+LA-LB
      IF(L(ML,NY)-1)240,241,241
  240 RETURN
  241 L(ML,NB)=2
      E=E+1.
      RETURN
 109  KA=NB/2
      KB=NB-2*KA
      K=2*K+LB*(1-KB)+LA*KB
      DO 110 KK=1,K
      NBA=NB+78+(LA-LB)*KK-(NB/79)*156
 810  IF(L(ML,NBA)-1)110,150,150
 110  CONTINUE
C CHECKED IF OPPOSITE BOND IS BROKEN TO GIVE BROKEN MOLECULE
      GO TO 151
 150  L(ML,NB)=3
 610  IF(L(ML,NBA)-2)717,718,747
 718  L(ML,NBA)=5
      GO TO 719
 747  GAG=1.
```

```
 717   L(ML,NBA)=3
 719   CALL MVAP(LA,LB,LC)
C MOLECULE IS BROKEN
       NBAA=NBA
 811   IF(IND)151,151,152
 105   IF(K-1)130,130,131
 130   RETURN
 131   KA=NB/2
       KB=NB-2*KA
       K=K-1
       K=2*K+LB*(1-KB)+LA*KB
       DO 120 KK=1,K
       NLN=NB+(LA-LB)*KK+(LB-LA)
 815   IF(L(ML,NLN)-1)132,133,133
 132   BVOL=BVOL+1.
 133   IF(L(ML,NLN)-2)134,135,120
 135   IEE=IEE+1
 134   E=E+1.
       L(ML,NLN)=2
 120   CONTINUE
       RETURN
C VAPORIZED ADJACENT BONDS
 151   DO 155 KK=1,K
       NBA=NB+(LA-LB)*KK
 156   IF(L(ML,NBA)-1)155,158,158
 155   CONTINUE
 620   IF(L(ML,NB)-3)650,152,650
C CHECKED IF ADJACENT BOND IS BROKEN TO GIVE BROKEN MOLECULE
 158   DO 168K=1,KK
       NBA=NB+(LA-LB)*K+LB-LA
 161   IF(L(ML,NBA)-1)162,163,163
 162   BVOL=BVOL+1.
 163   IF(L(ML,NBA)-2)164,165,166
 165   IEE=IEE+1
 164   E=E+1.
       L(ML,NBA)=4
C VAPORIZED ADJACENT BONDS
       GO TO 168
 166   E=E+1.
 168   CONTINUE
 630   IF(L(ML,NB)-3)650,152,650
C IF MOLECULE IS BROKEN,CHECK BOTH DIRECTIONS
```

```
 152  IF(LA)200,200,201
 200  LA=1
      LB=0
      LCC=67
      GO TO 210
 201  LA=0
      LB=1
      LCC=0
 210  NBA=NBAA
      CALL MVAP(LA,LB,LCC)
 650  RETURN
      END
)1 (X-  -   (G7                                                                  070SQU00
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13. ABSTRACT

The random degradation of four and six-membered ring ladder polymers were investigated by means of a digital computer and the results compared to a single chain polymer undergoing degradation under identical conditions. The percent vaporization versus time and the rate of weight loss versus time was plotted and significant differences were obtained. The results indicate that ladder polymers should have increased stability over single chain polymers undergoing random degradation.

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