Figure 1

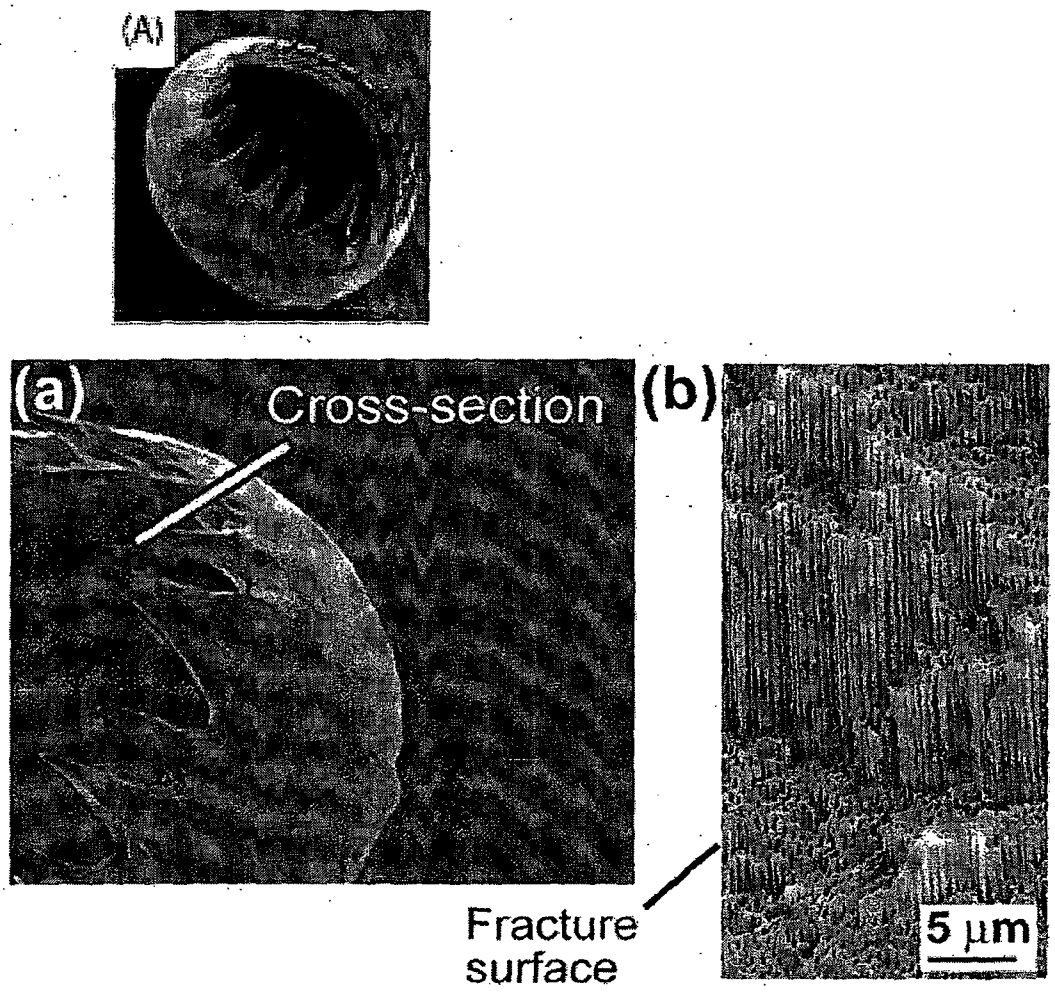


Figure 2

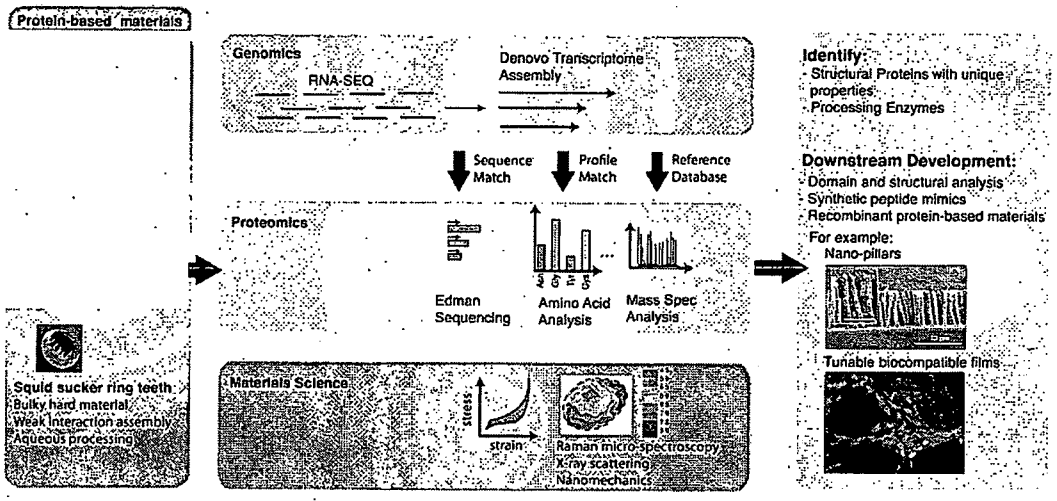


Figure 3

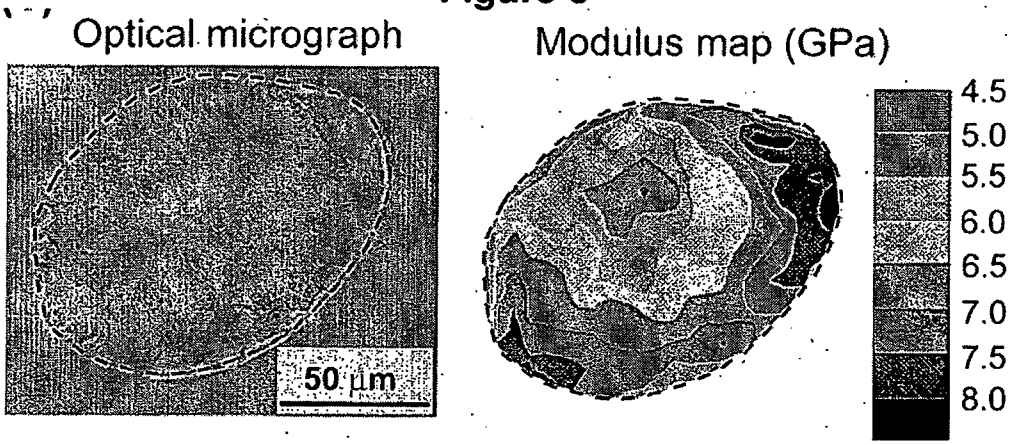


Figure 4

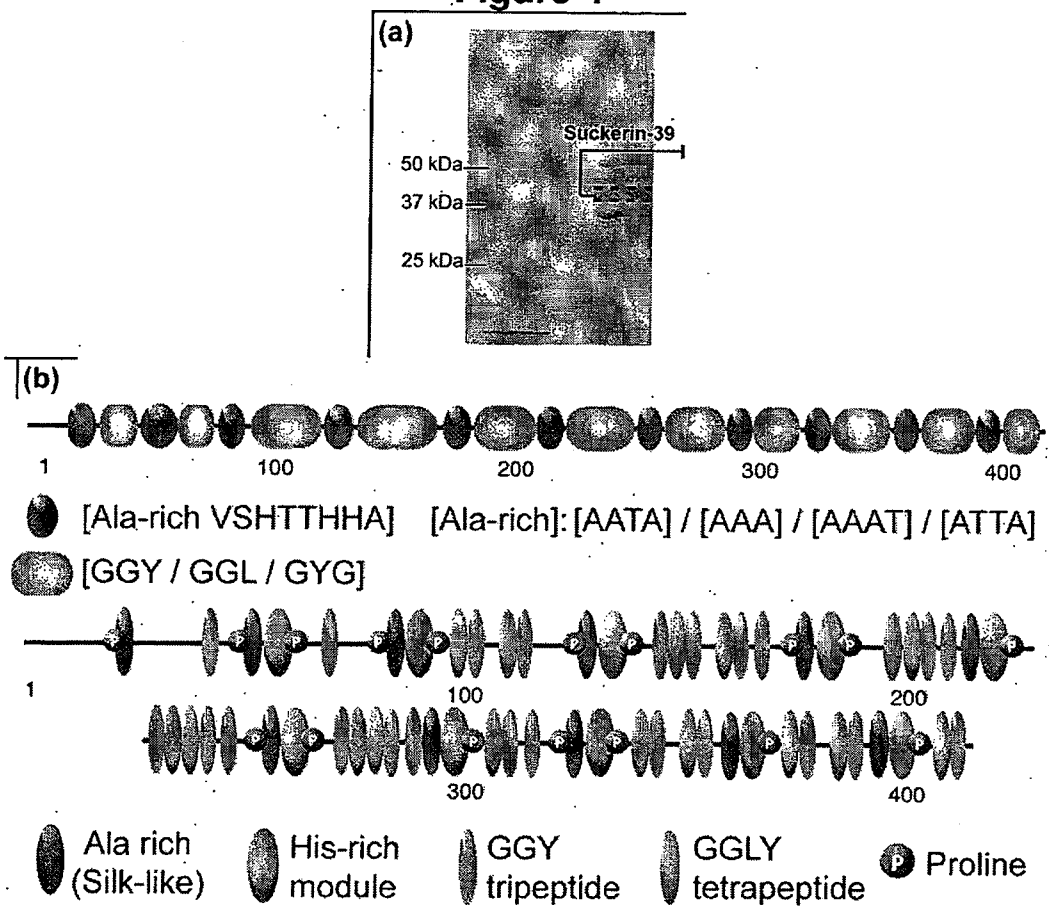

(a)
Suckerin-39
50 kDa
37 kDa
25 kDa
(b)
1
100
200
300
400
[Ala-rich VSHTTHHA]
[Ala-rich]: [AATA] / [AAA] / [AAAT] / [ATTA]
[GGY / GGL / GYG]
1
100
200
300
400
Ala rich (Silk-like)
His-rich module
GGY tripeptide
GGLY tetrapeptide
Proline

Figure 5 A

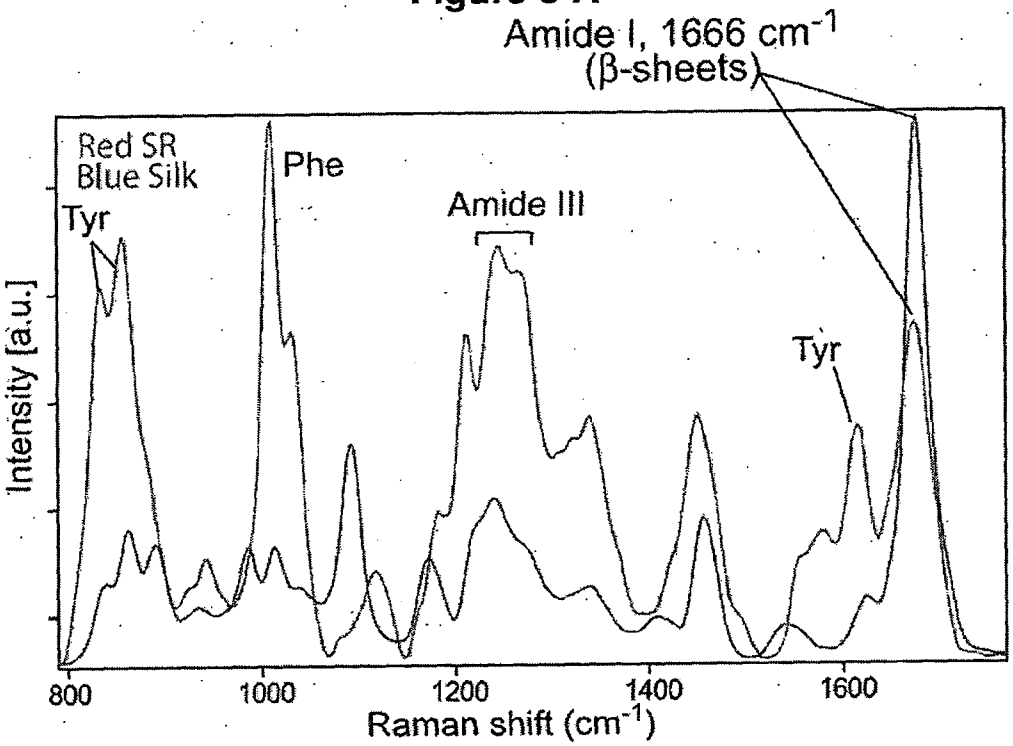


Figure 5B

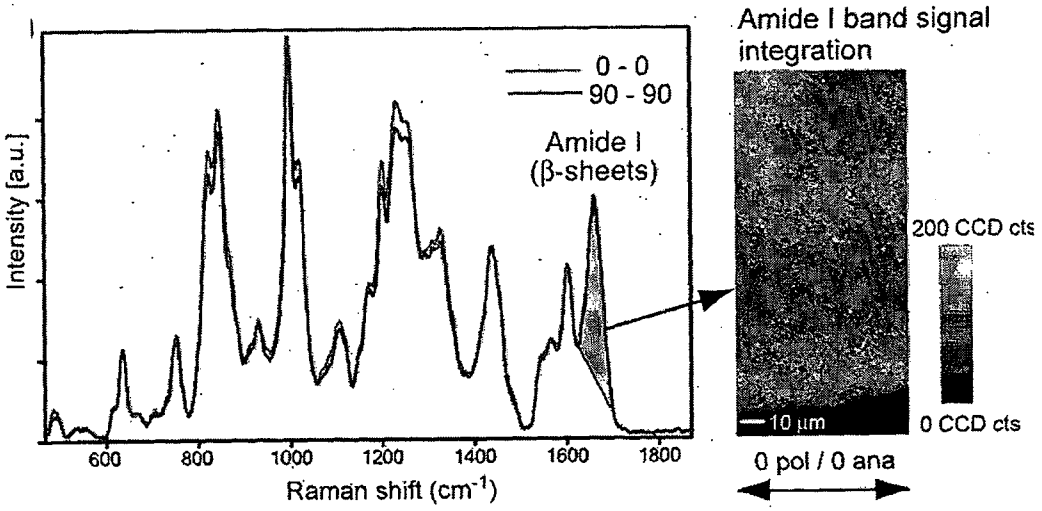
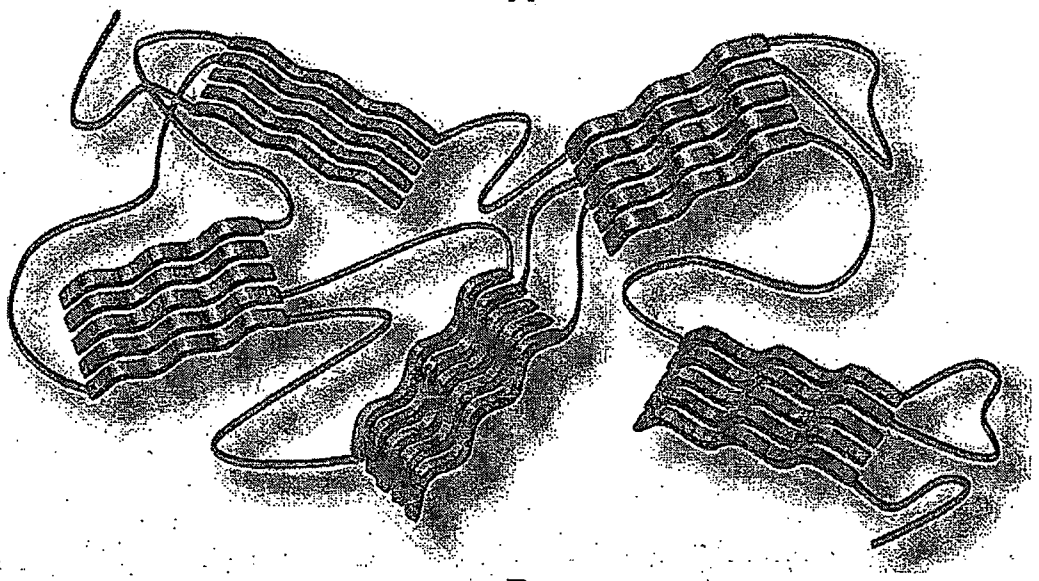


Figure 6

A



B

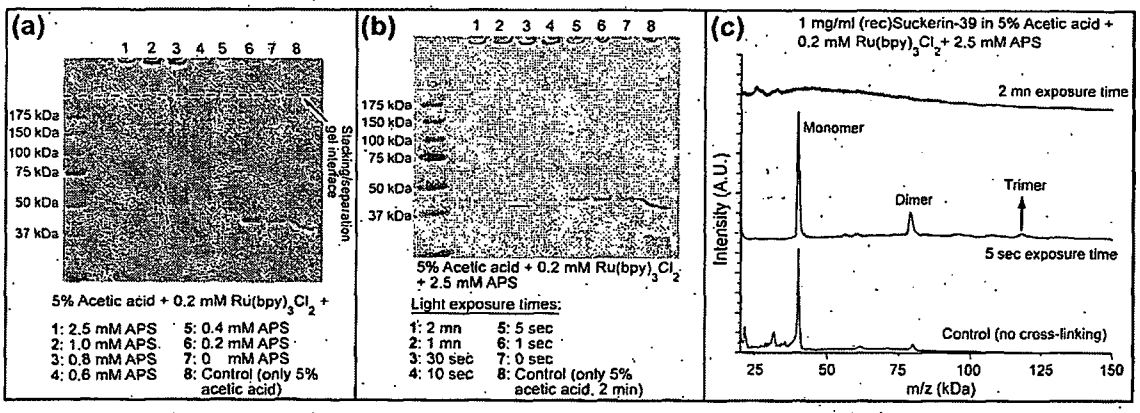

(a)
1 2 3 4 5 6 7 8
175 kDa
150 kDa
100 kDa
75 kDa
50 kDa
37 kDa
Stacking/separation gel interface
5% Acetic acid + 0.2 mM Ru(bpy)3Cl2 +
1: 2.5 mM APS
2: 1.0 mM APS
3: 0.8 mM APS
4: 0.6 mM APS
5: 0.4 mM APS
6: 0.2 mM APS
7: 0 mM APS
8: Control (only 5% acetic acid)
(b)
1 2 3 4 5 6 7 8
175 kDa
150 kDa
100 kDa
75 kDa
50 kDa
37 kDa
5% Acetic acid + 0.2 mM Ru(bpy)3Cl2 + 2.5 mM APS
Light exposure times:
1: 2 mn
2: 1 mn
3: 30 sec
4: 10 sec
5: 5 sec
6: 1 sec
7: 0 sec
8: Control (only 5% acetic acid, 2 min)
(c)
1 mg/ml (rec)Suckerin-39 in 5% Acetic acid + 0.2 mM Ru(bpy)3Cl2 + 2.5 mM APS
2 mn exposure time
Monomer
Dimer
Trimer
5 sec exposure time
Control (no cross-linking)
Intensity (A.U.)
25
50
75
100
125
150
m/z (kDa)

Figure 6C

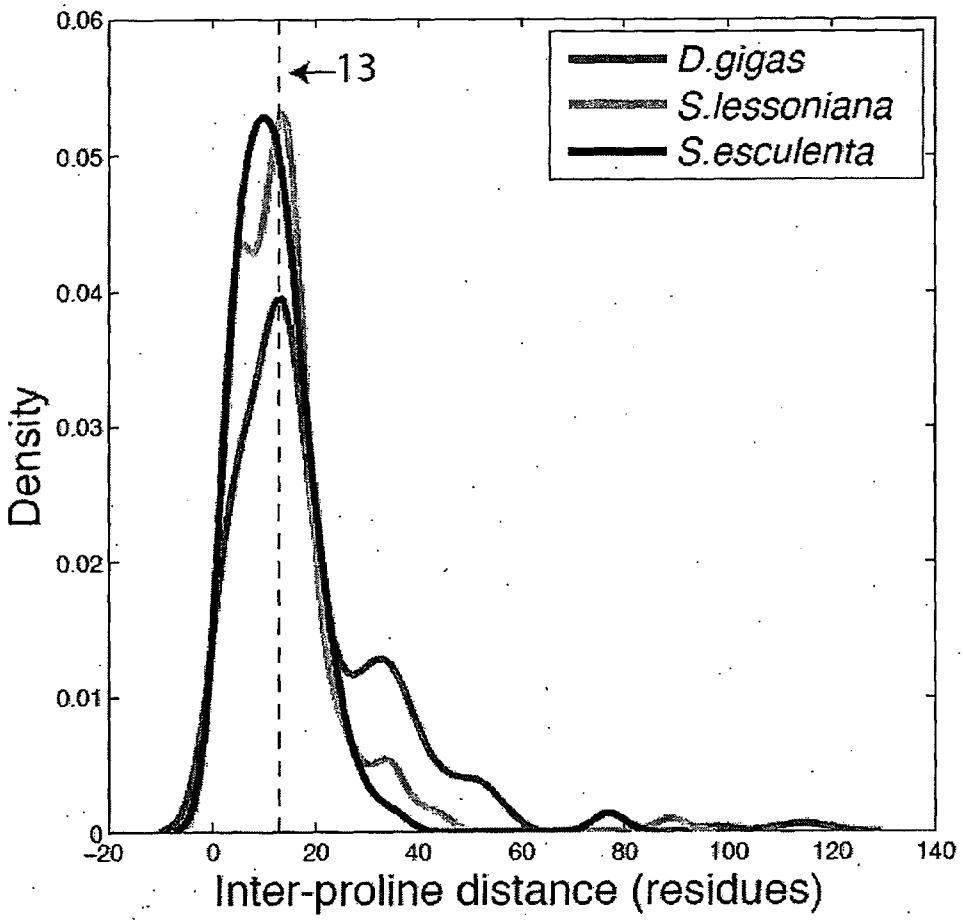

0.06
0.05
0.04
0.03
0.02
0.01
0
Density
←13
D.gigas
S.lessoniana
S.esculenta
−20
0
20
40
60
80
100
120
140
Inter-proline distance (residues)

Figure 7

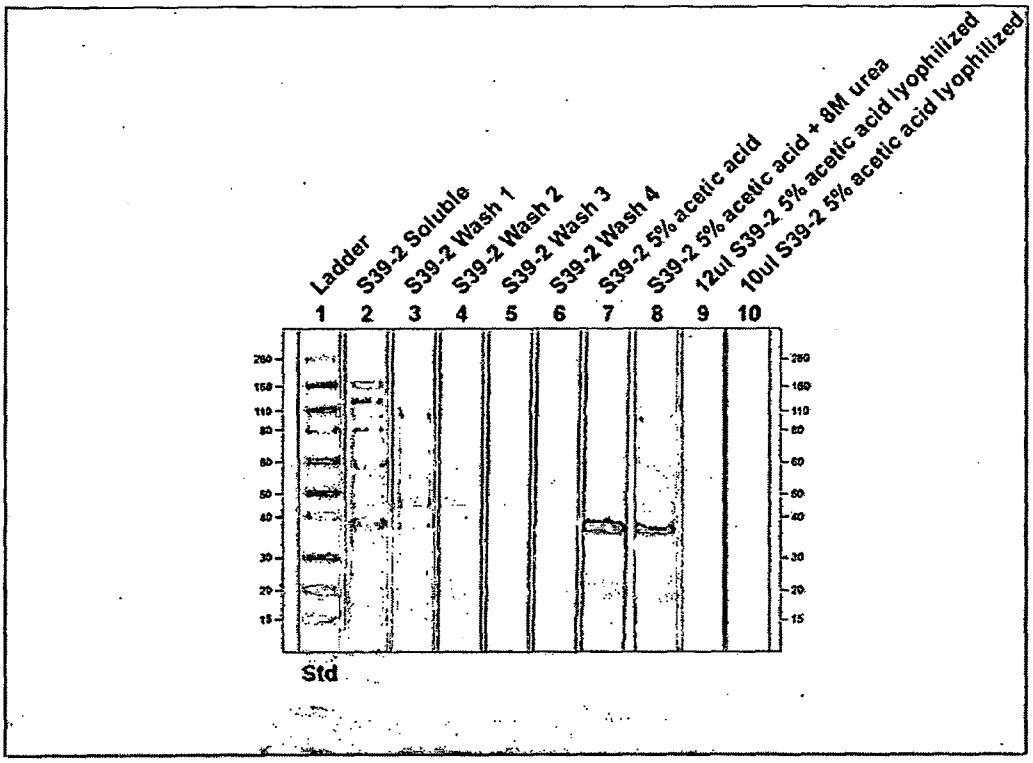


Figure 8

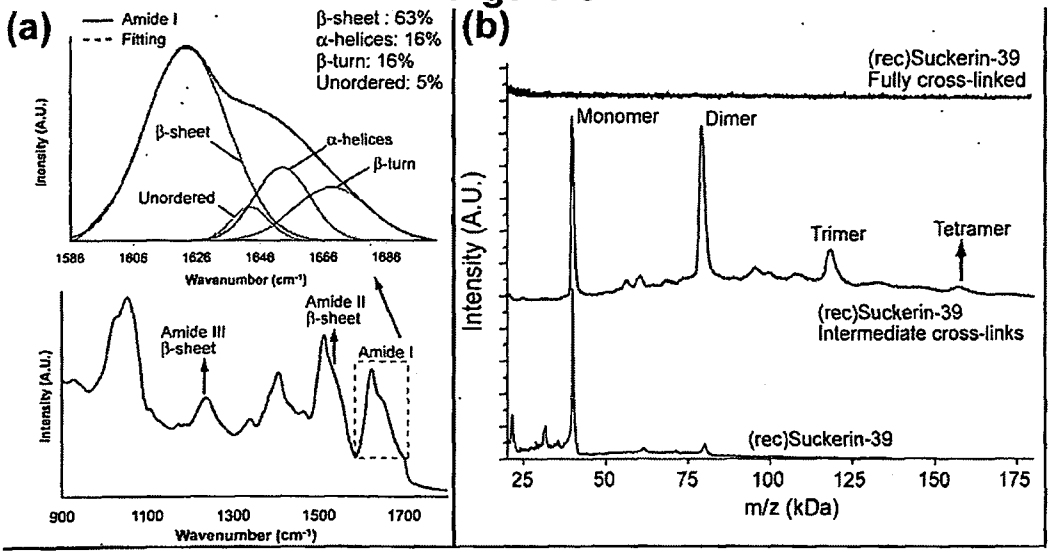
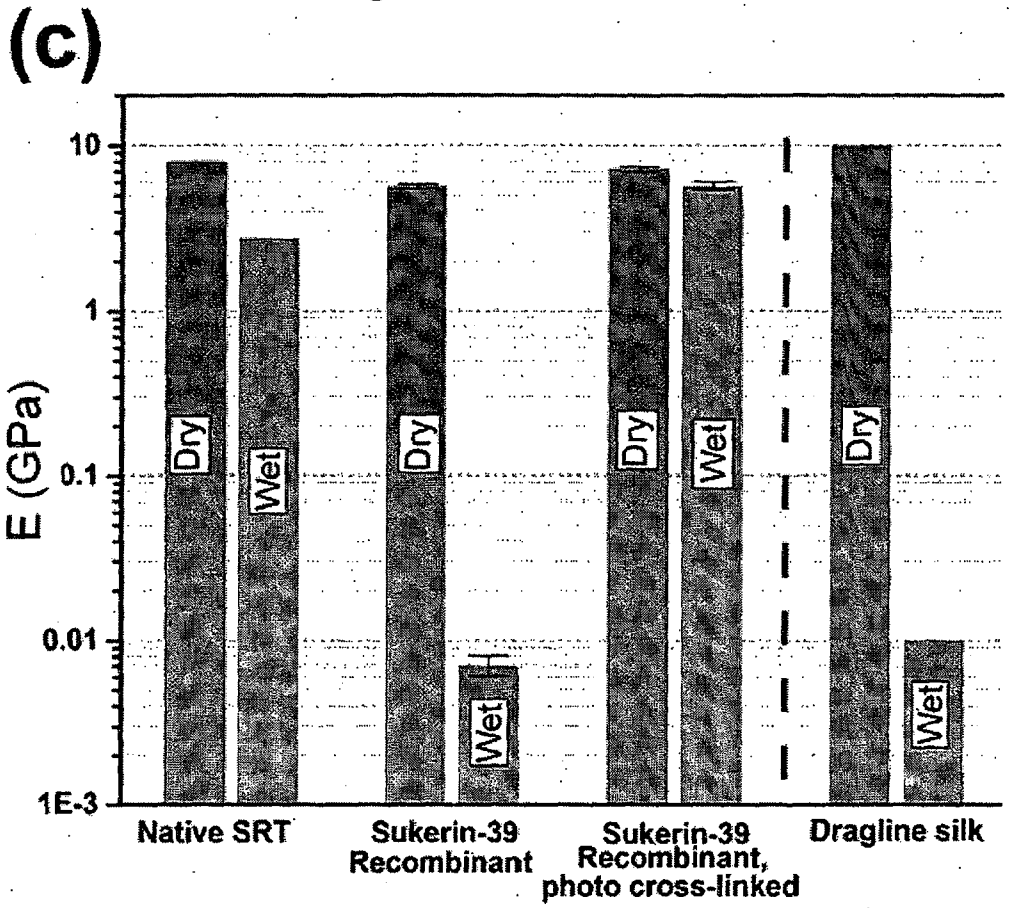


Figure 8 continued



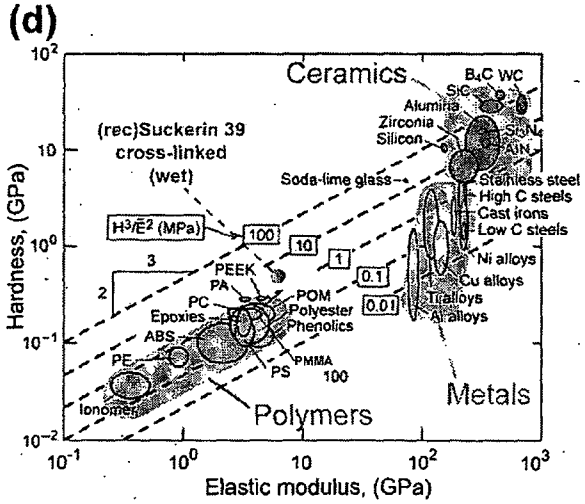
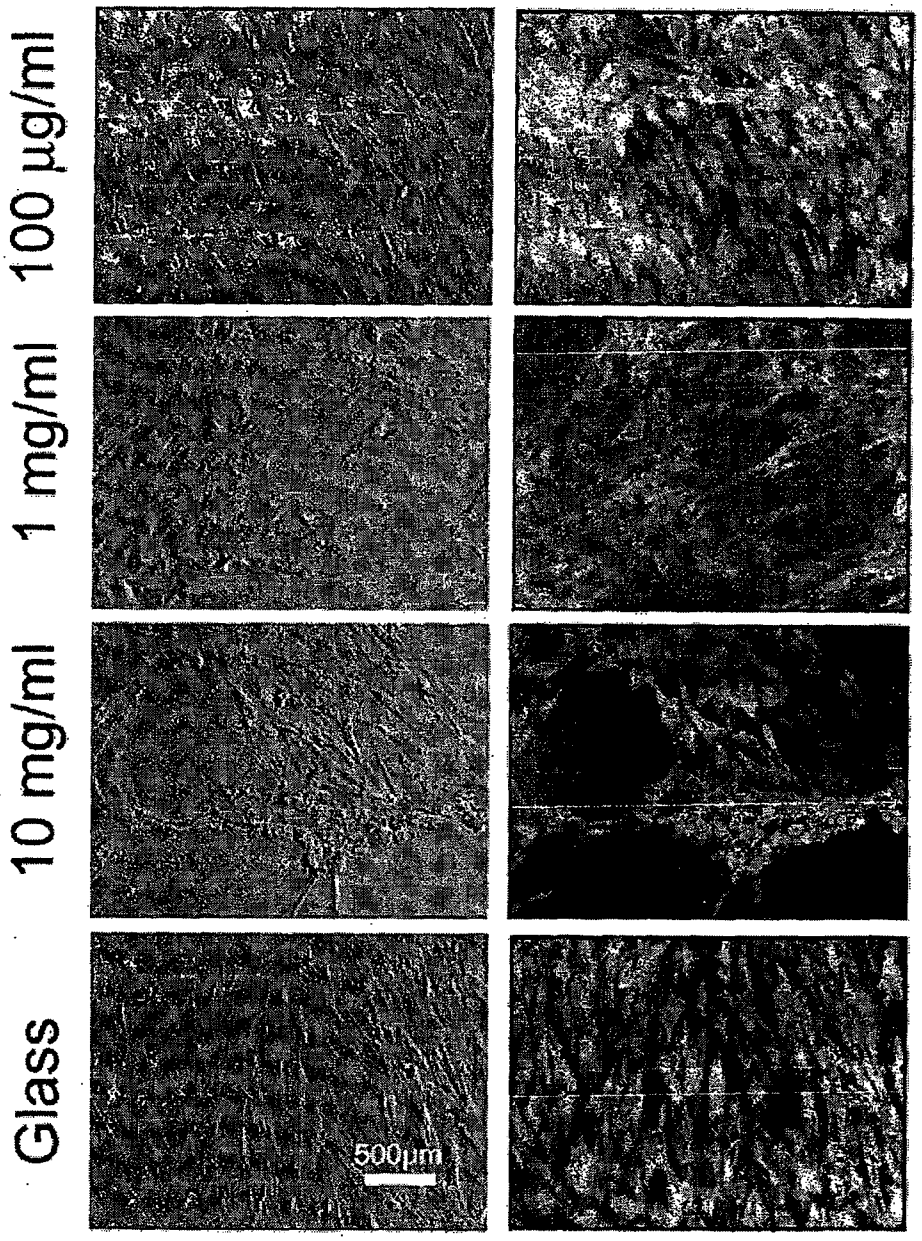


Figure 8 continued



(e)

Figure 9

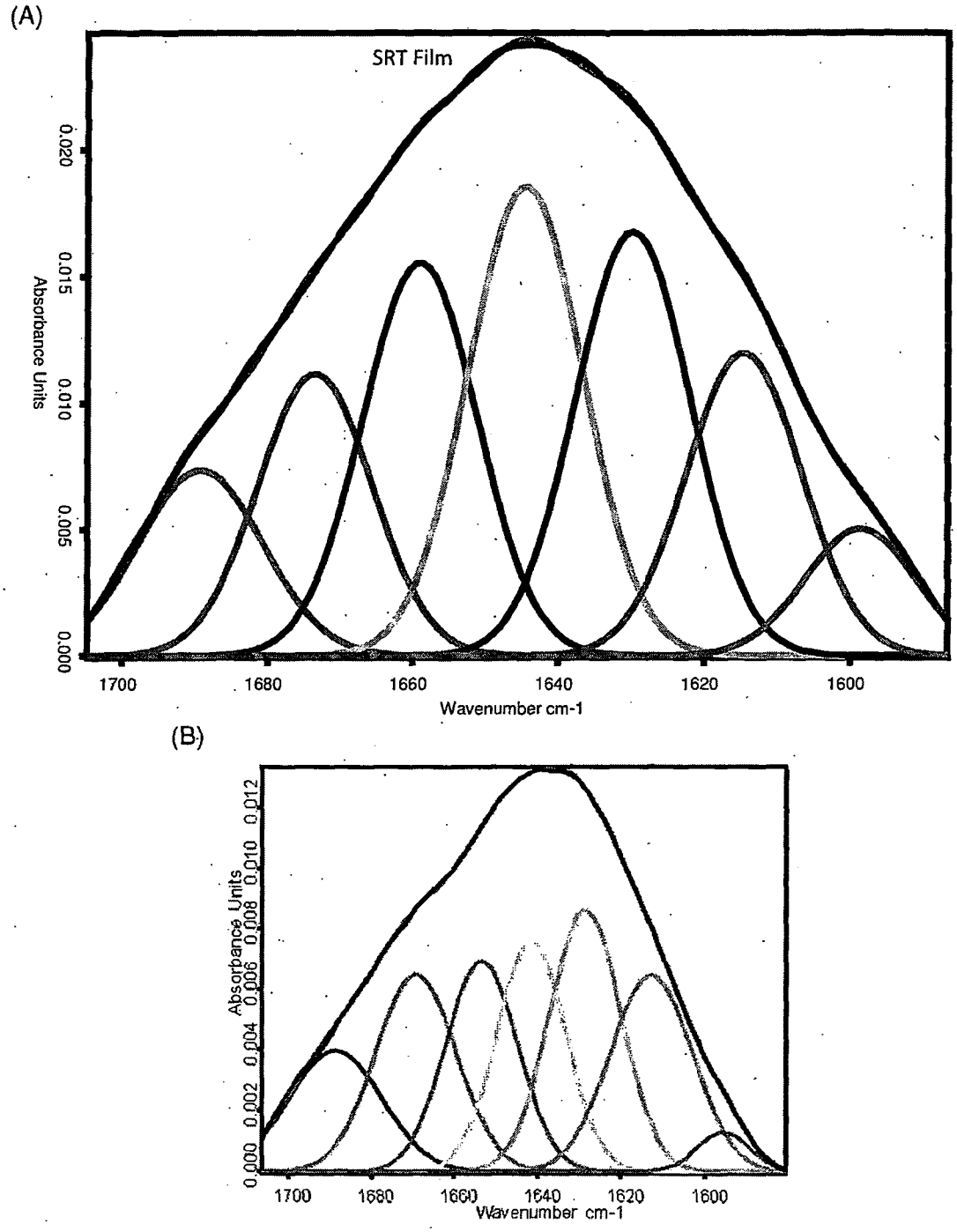

(A)
SRT Film
Absorbance Units
0.000
0.005
0.010
0.015
0.020
1700
1680
1660
1640
1620
1600
Wavenumber cm-1
(B)
Absorbance Units
0.000
0.002
0.004
0.006
0.008
0.010
0.012
1700
1680
1660
1640
1620
1600
Wavenumber cm-1

Figure 10 A

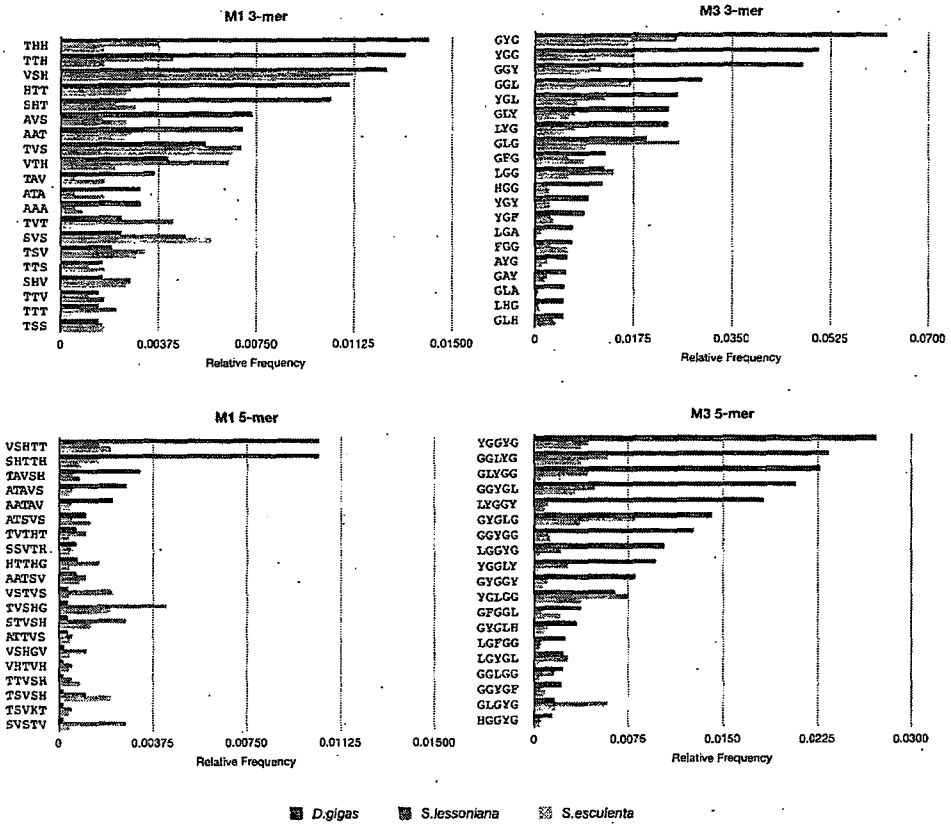

M1 3-mer
THH
TTH
VSH
HTT
SHT
AVS
AAT
TVS
VTH
TAV
ATA
AAA
TVT
SVS
TSV
TTS
SHV
TTV
TTT
TSS
0
0.00375
0.00750
0.01125
0.01500
Relative Frequency
M3 3-mer
GYG
YGG
GGY
GGL
YGL
GLY
LYG
GLG
GFG
LGG
HGG
YGY
YGF
LGA
FGG
AYG
GAY
GLA
LHG
GLH
0
0.0175
0.0350
0.0525
0.0700
Relative Frequency
M1 5-mer
VSHTT
SHTTH
TAVSH
ATAVS
AATAV
ATSVS
TVTHT
SSVTH
HTTHG
AATSV
VSTVS
TVSHG
STVSH
ATTVS
VSHGV
VHTVH
TTVSH
TSVSH
TSVKT
SVSTV
0
0.00375
0.00750
0.01125
0.01500
Relative Frequency
M3 5-mer
YGGYG
GGLYG
GLYGG
GGYGL
LYGGY
GYGLG
GGYGG
LGGYG
YGGLY
GYGGY
YGLGG
GFGGL
GYGLH
LGFGG
LGYGL
GGLGG
GGYGF
GLGYG
HGGYG
0
0.0075
0.0150
0.0225
0.0300
Relative Frequency
D.gigas
S.lessoniana
S.esculenta

Figure 10B

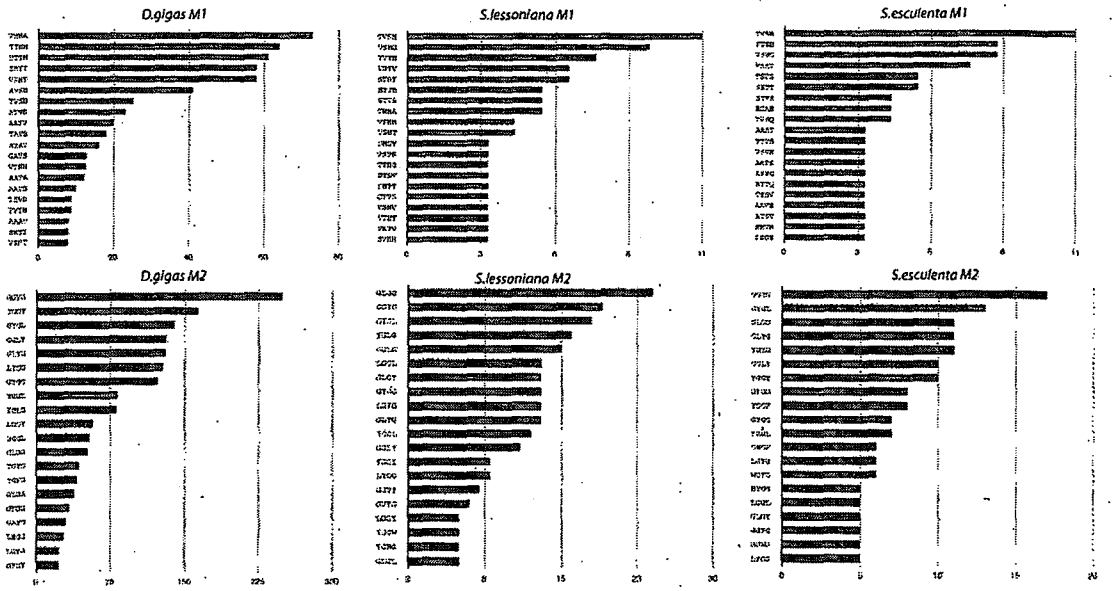


Figure 10 C

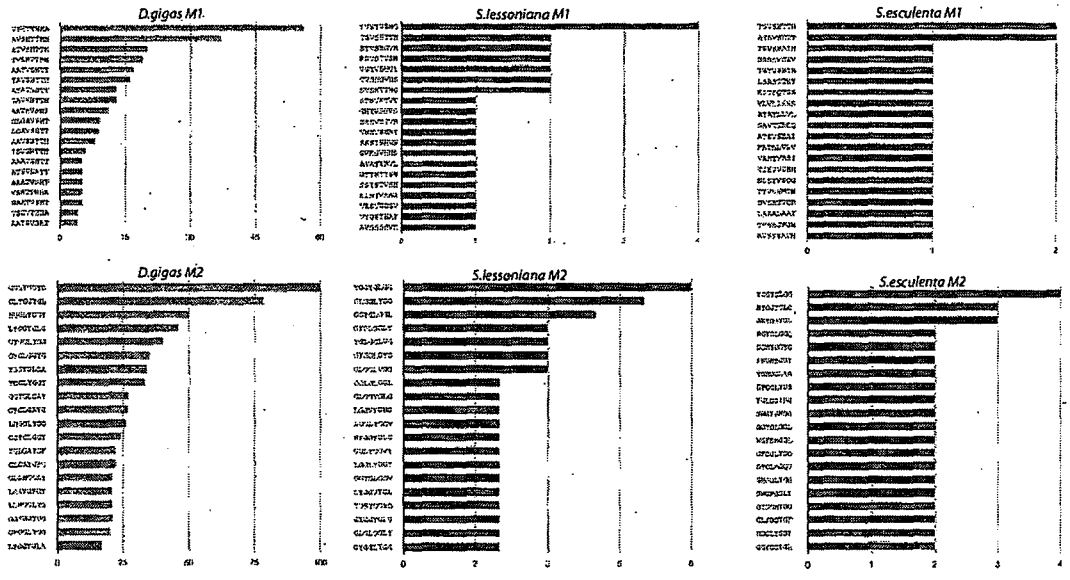


Figure 11 A

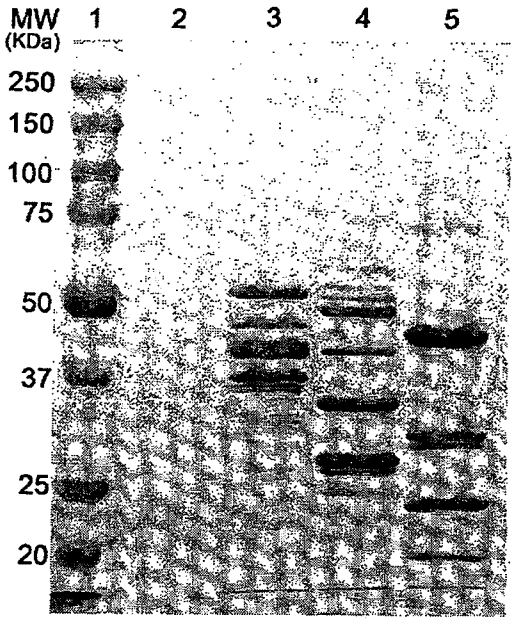


Figure 11 B

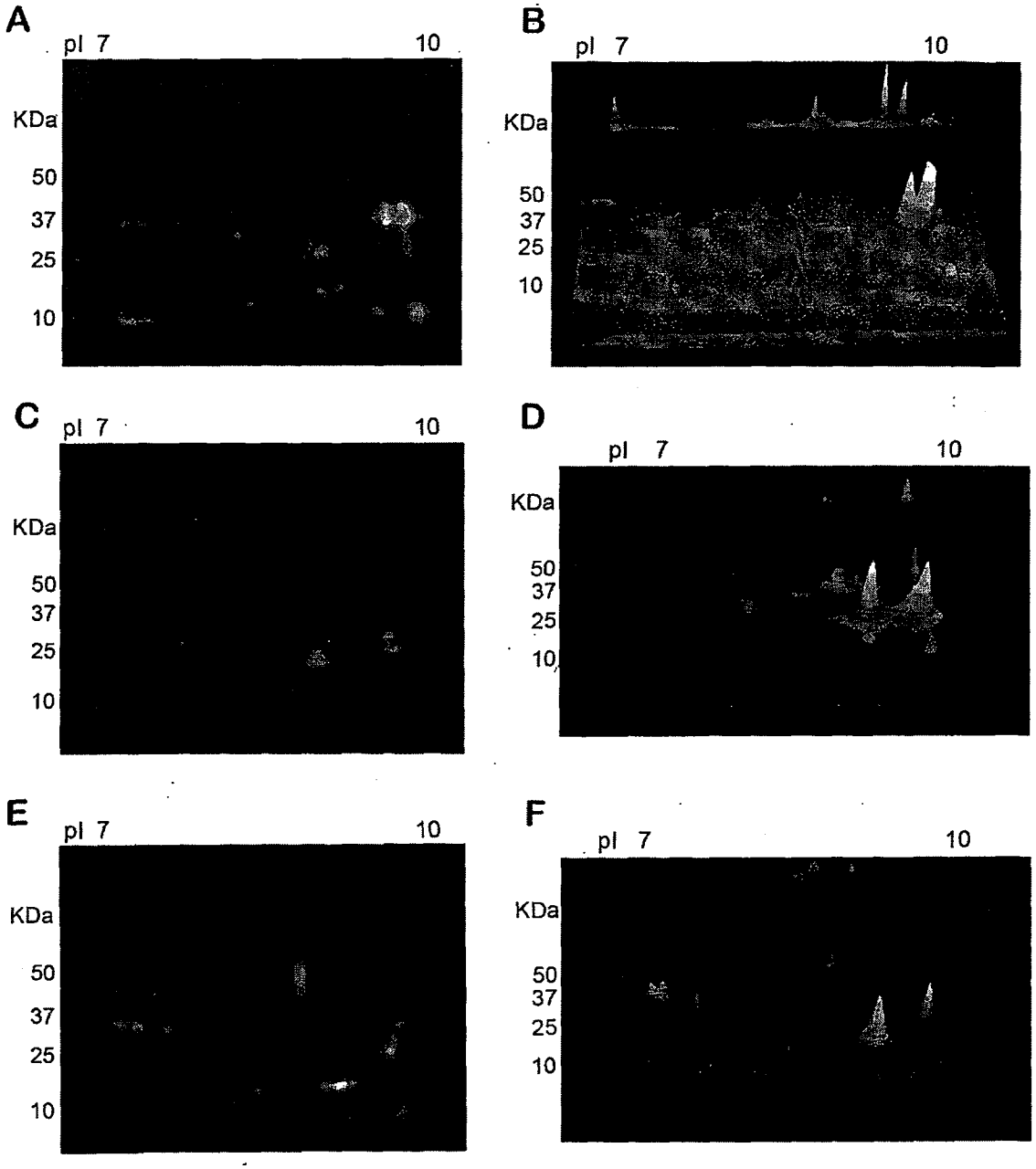

A
pI 7
10
KDa
50
37
25
10
B
pI 7
10
KDa
50
37
25
10
C
pI 7
10
KDa
50
37
25
10
D
pI 7
10
KDa
50
37
25
10
E
pI 7
10
KDa
50
37
25
10
F
pI 7
10
KDa
50
37
25
10

Figure 12

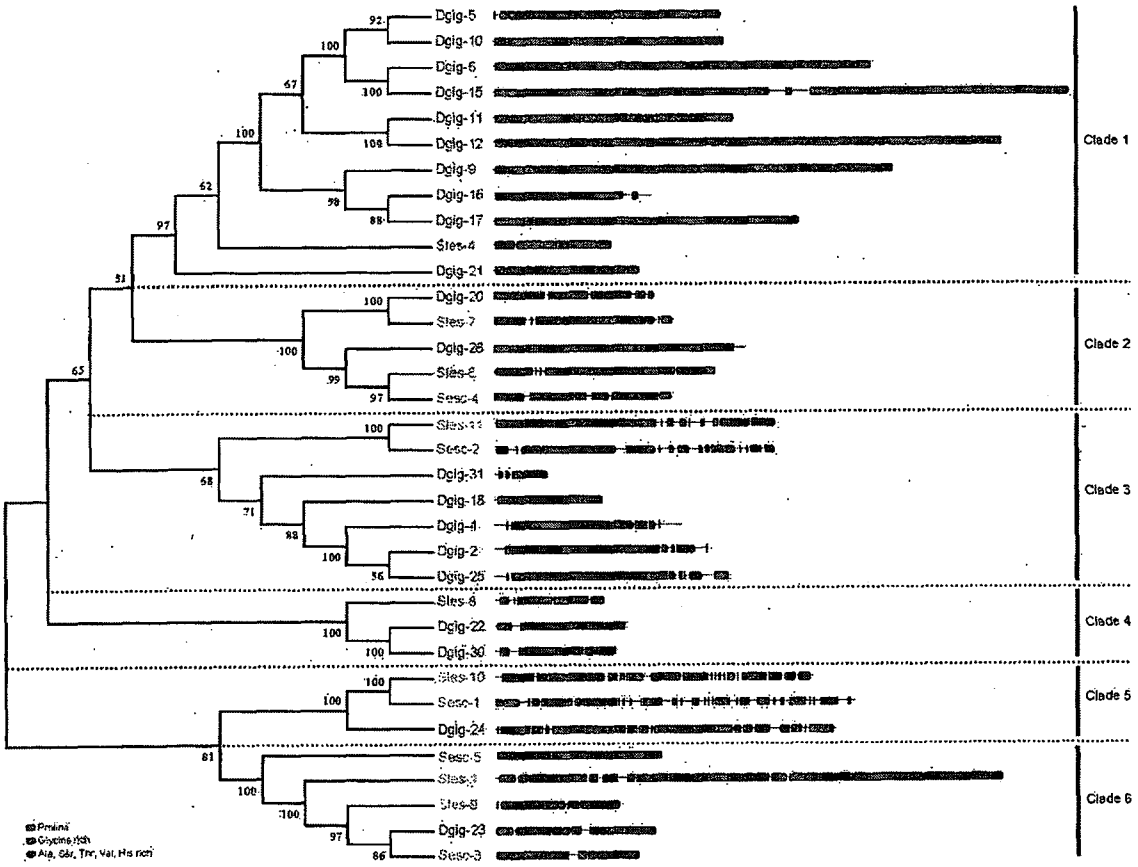
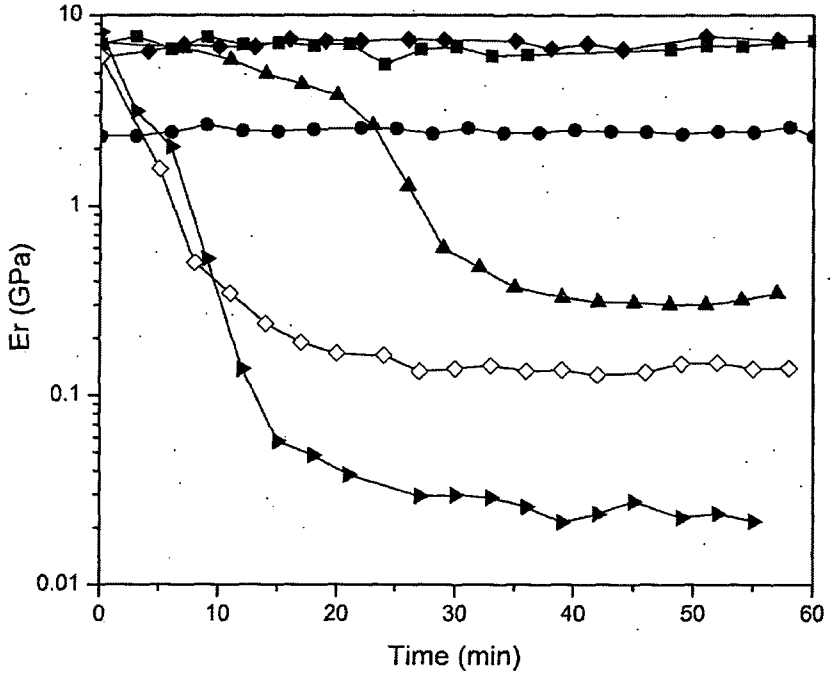
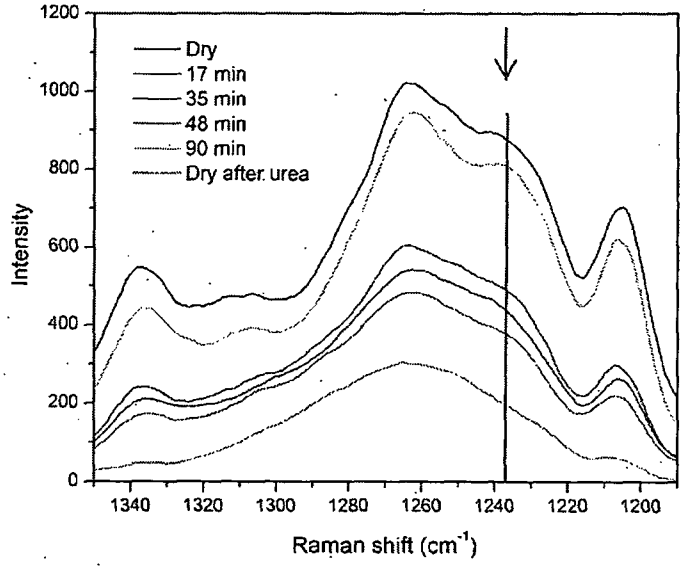

Dgig-5
Dgig-10
Dgig-6
Dgig-15
Dgig-11
Dgig-12
Dgig-9
Dgig-16
Dgig-17
Sles-4
Dgig-21
Dgig-20
Sles-7
Dgig-26
Sles-5
Sesc-4
Sles-11
Sesc-2
Dgig-31
Dgig-18
Dgig-1
Dgig-2
Dgig-25
Sles-8
Dgig-22
Dgig-30
Sles-10
Sesc-1
Dgig-24
Sesc-5
Sles-3
Sles-9
Dgig-23
Sesc-3
Clade 1
Clade 2
Clade 3
Clade 4
Clade 5
Clade 6

Figure 13


10
1
0.1
0.01
Er (GPa)
0
10
20
30
40
50
60
Time (min)


Dry
17 min
35 min
48 min
90 min
Dry after urea
Intensity
1200
1000
800
600
400
200
0
1340
1320
1300
1280
1260
1240
1220
1200
Raman shift (cm-1)