United States Patent [19]

Yamaguchi et al.

[11] E **Patent Number: Re. 33,745**

[45] **Reissued Date of Patent: Nov. 19, 1991**

[54] **THIADIAZABICYCLONONANE DERIVATIVES, PROCESSES FOR THEIR PRODUCTION AND HERBICIDAL COMPOSITIONS**

[75] Inventors: **Mikio Yamaguchi**, Shimada; **Yukihiro Watase**, Shizuoka; **Takeshi Kambe**, Tokyo; **Susumu Katou**, Shizuoka, all of Japan

[73] Assignees: **Kumiai Chemical Industry Co., Ltd.**; **Ihara Chemical Industry Co., Ltd.**, both of Tokyo, Japan

[21] Appl. No.: **394,351**

[22] Filed: **Aug. 15, 1989**

Related U.S. Patent Documents

Reissue of:

[64] Patent No.: **4,816,063**
Issued: **Mar. 28, 1989**
Appl. No.: **910,978**
Filed: **Sep. 24, 1986**

[30] **Foreign Application Priority Data**

Mar. 25, 1986 [JP] Japan 61-66567

[51] **Int. Cl.**[5] **C07D 513/04**; A01N 43/82

[52] **U.S. Cl.** **71/90**; 71/87; 544/224; 544/232; 544/235; 544/238

[58] **Field of Search** 71/90, 87; 544/224, 544/232, 235, 238

[56] **References Cited**

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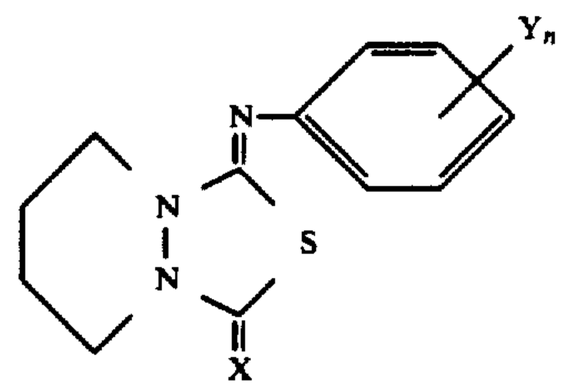
Primary Examiner—Donald G. Daus
Attorney, Agent, or Firm—Oblon, Spivak, McClelland, Maier & Neustadt

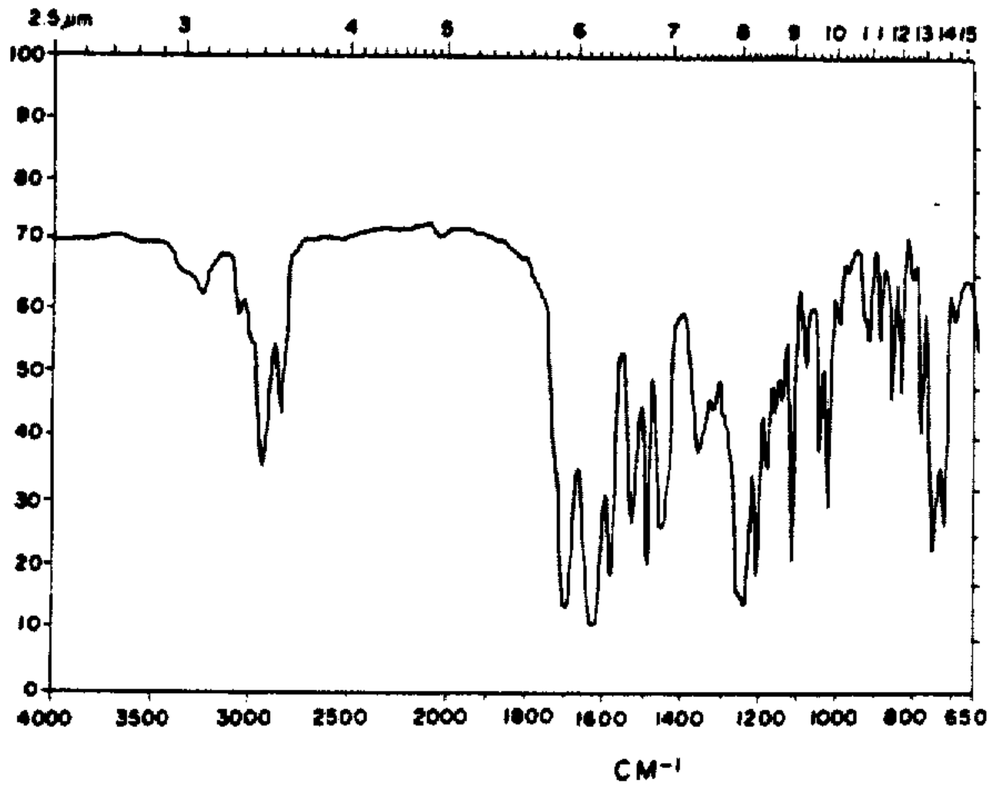
[57] **ABSTRACT**

As a herbicide 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) compound having the formula:

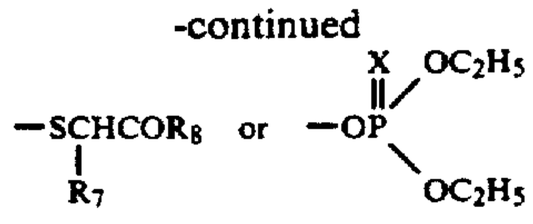


(I)

wherein Y represents halogen, hydroxyl, alkyl, alkoxy which may be substituted by halogen, alkenyloxy, alkynyloxy, phenoxy, cycloalkyloxy, alkoxycarbonylalkyloxy alkoxycarbonylalkenyloxy, alkythiocarbonylalkyloxy, alkynyloxycarbonylalkyloxy, benzyloxycarbonylalkyloxy, trifluoromethyl, benzyloxy, alkenyl, cyanoalkyl, alkylcarbomoyloxy, benzyl, alkoxyalkyl, alkynyloxyalkyl, cycloalkylmethyloxy, alkoxyalkyloxy, phenethyloxy, cycloalkyloxycarbonylalkyloxy, pyrrolidinocarbonyl, phenylcarbonyl,



-continued



n is an integer of from 0 to 3; and X is oxygen or sulfur.

15 Claims, 25 Drawing Sheets

FIGURE 1

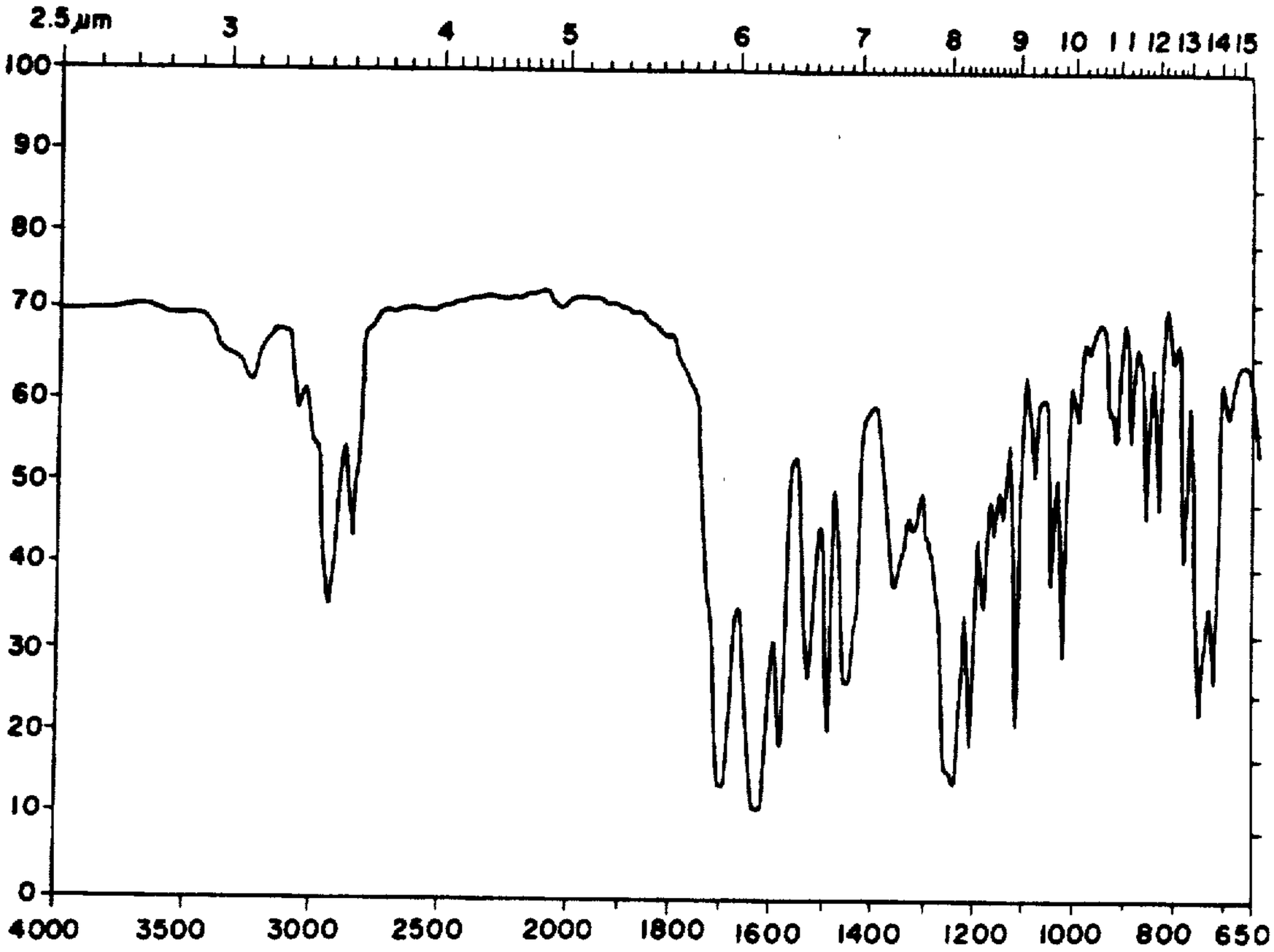


FIGURE 2

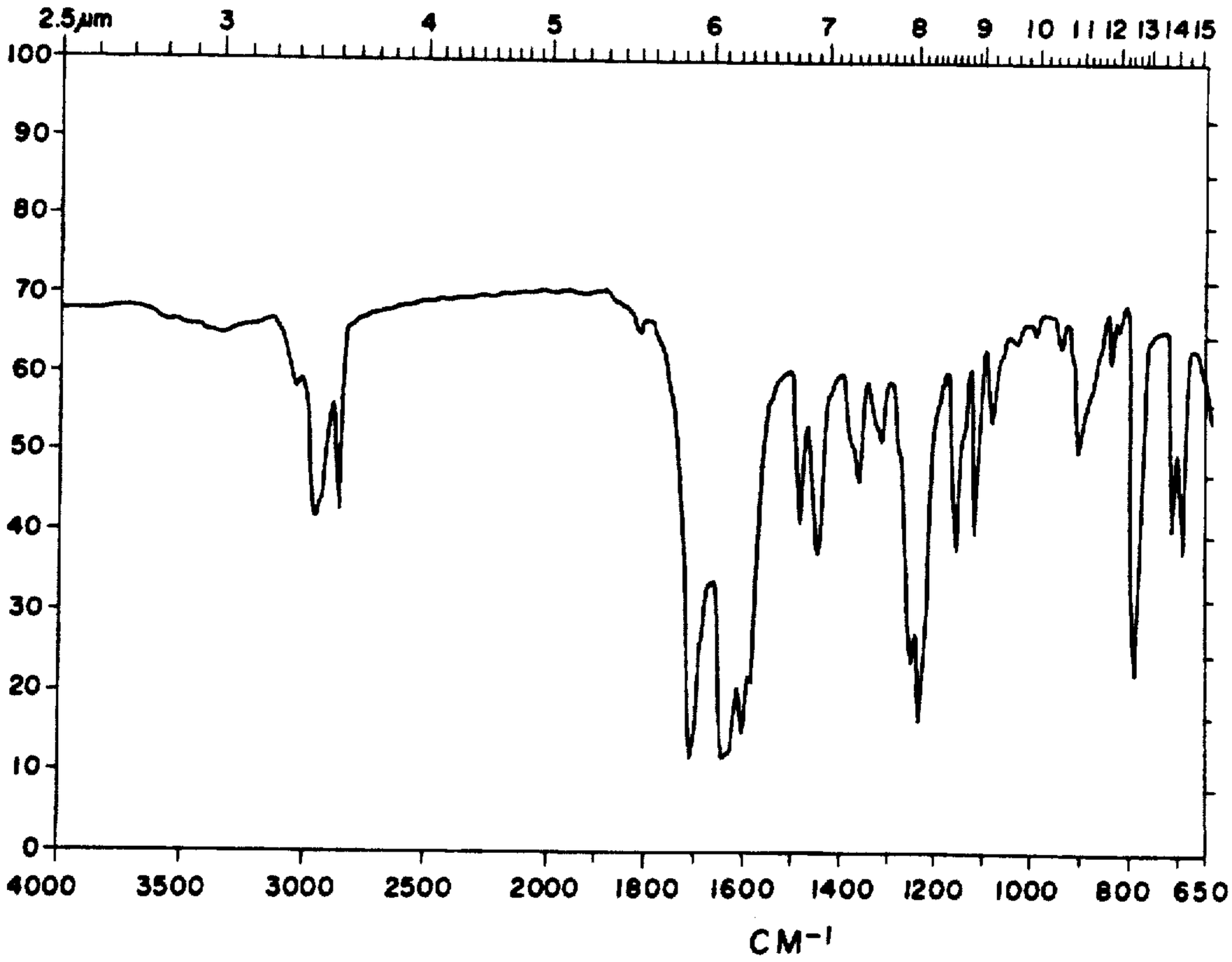


FIGURE 3

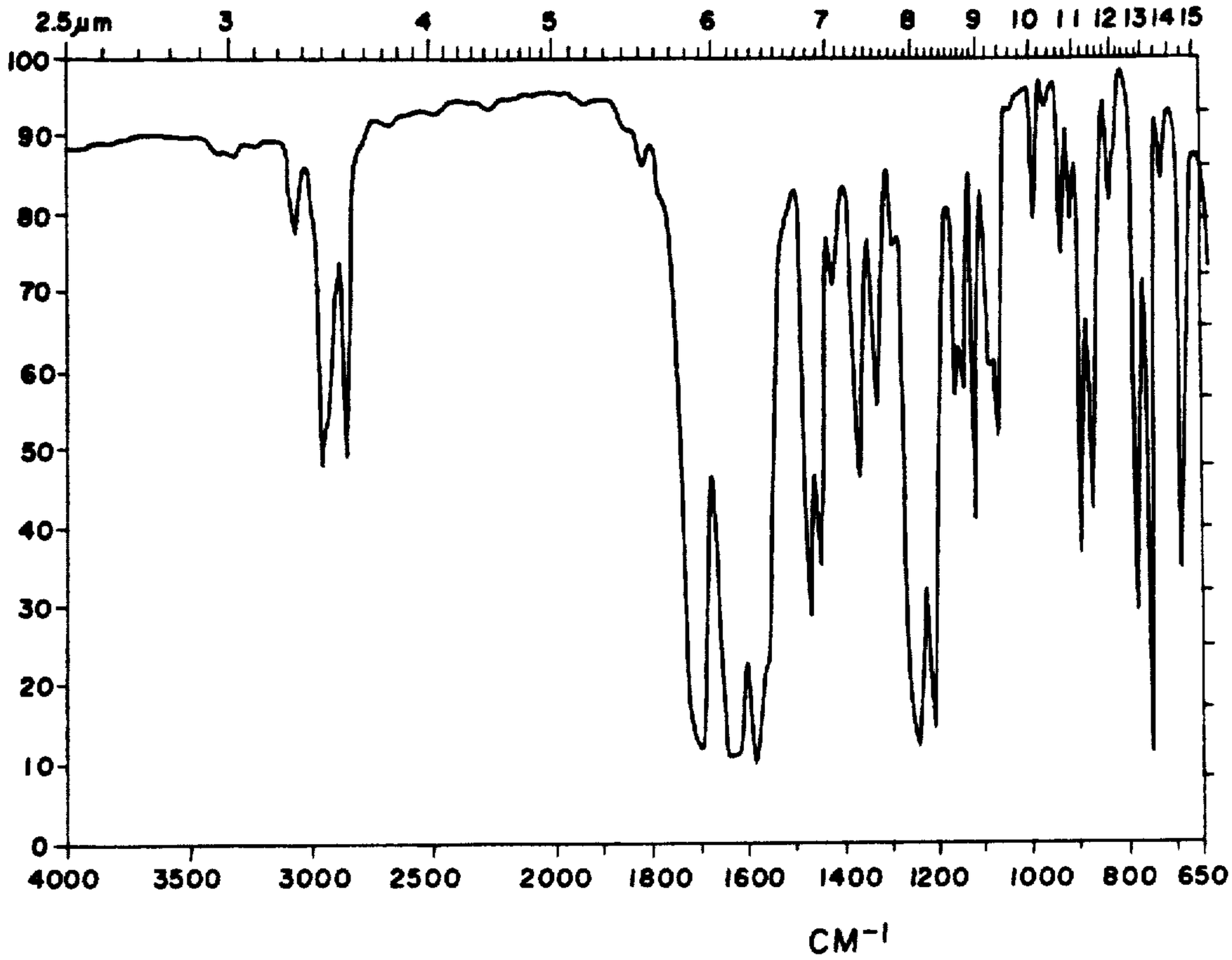


FIGURE 4

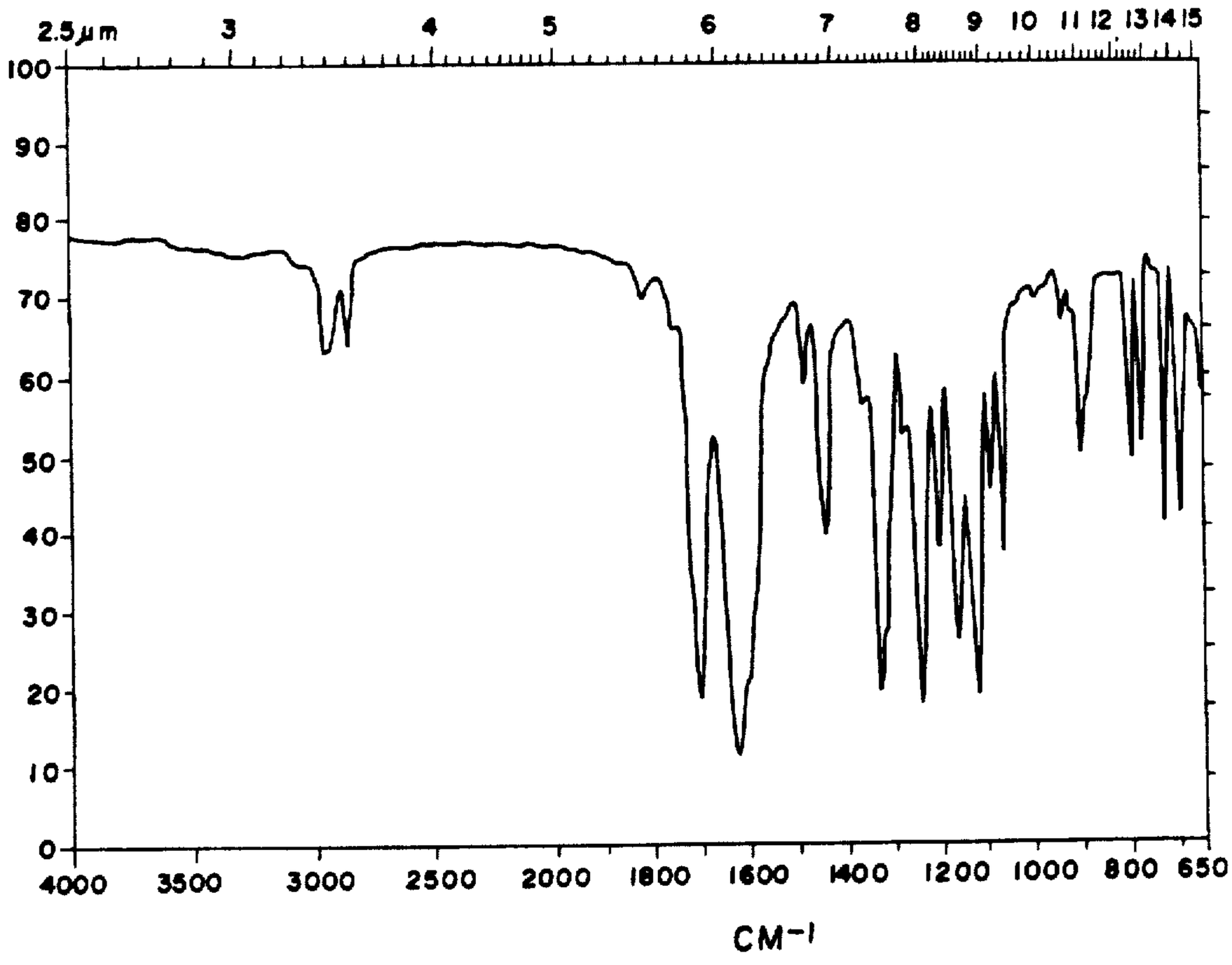


FIGURE 5

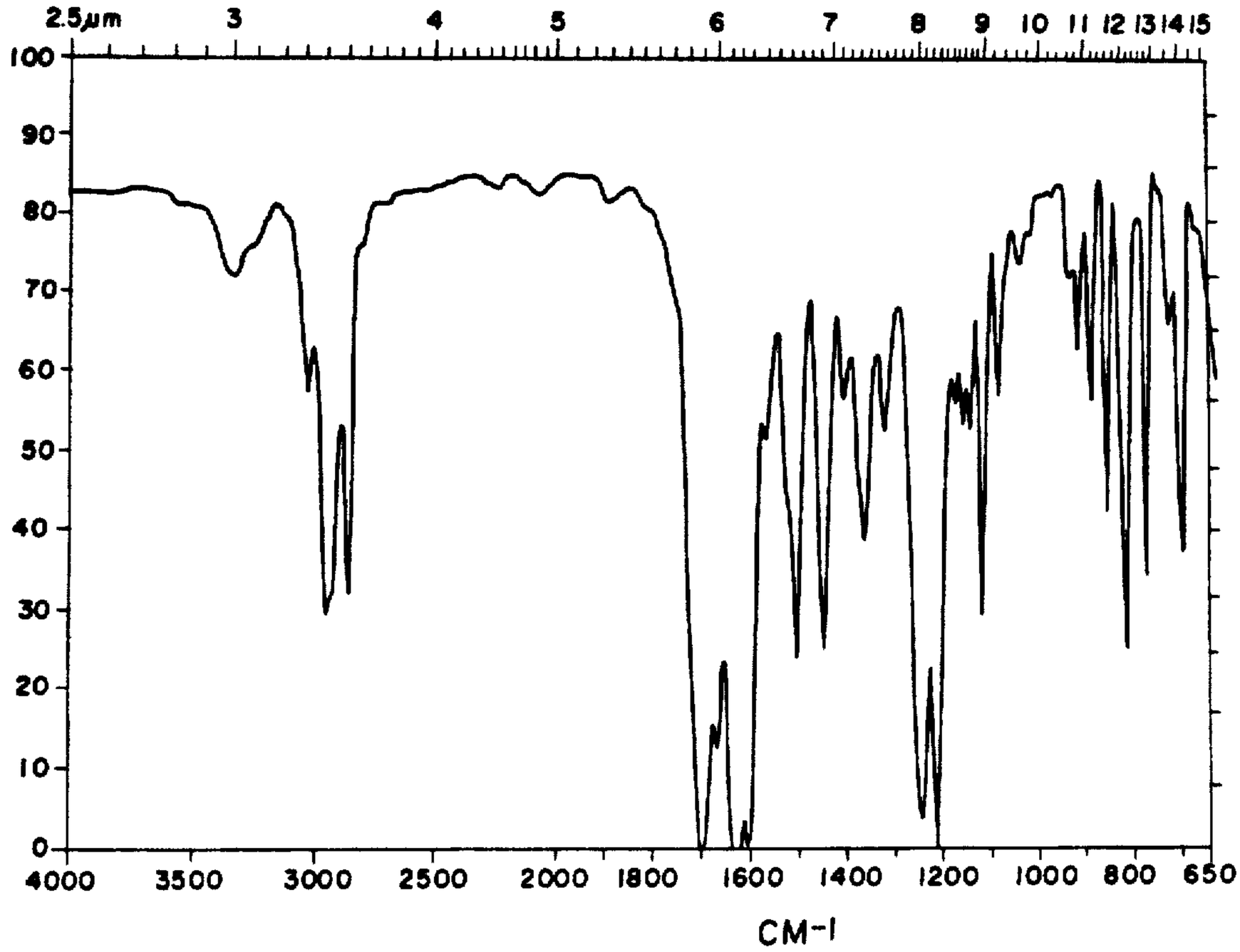


FIGURE 6

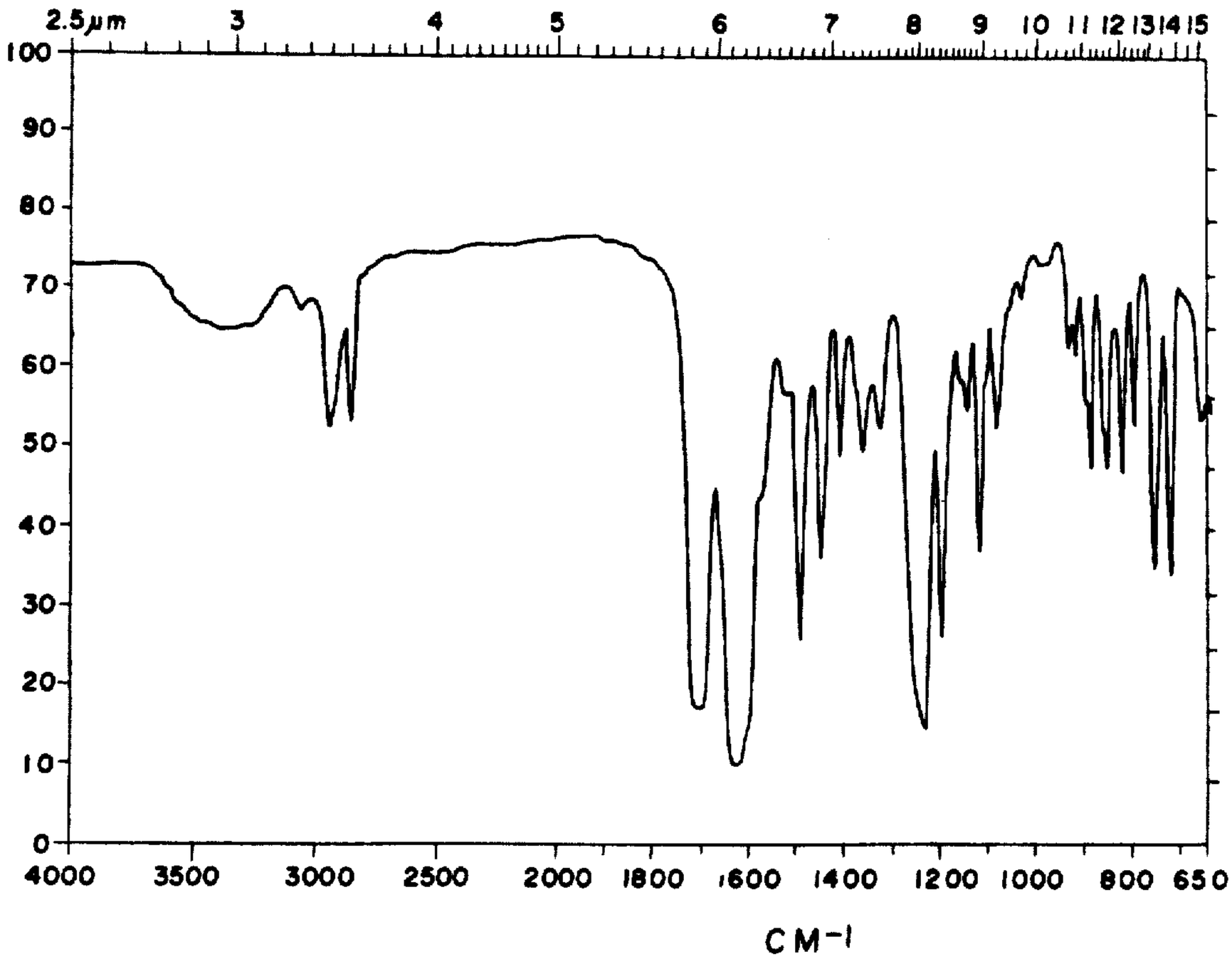


FIGURE 7

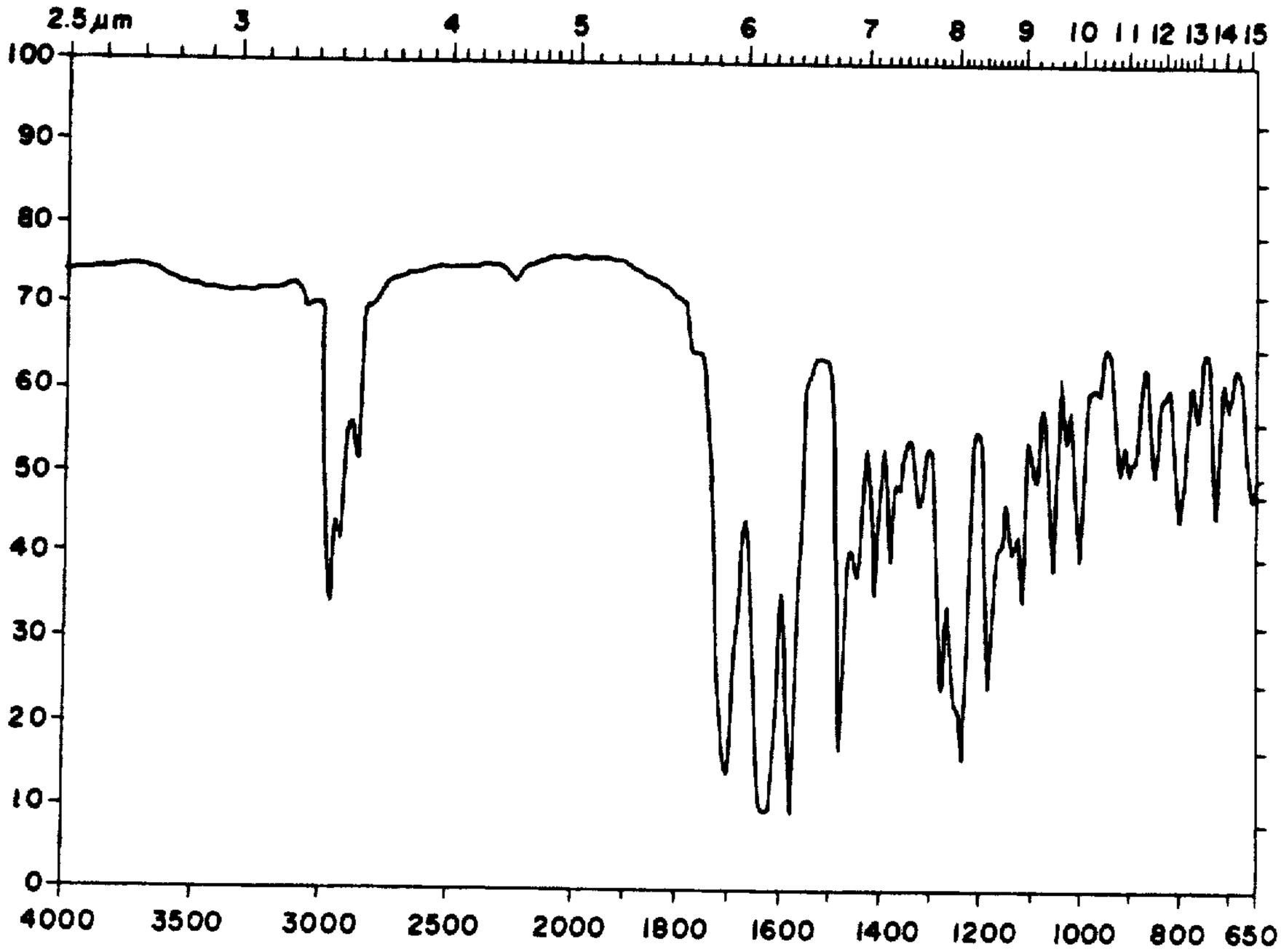


FIGURE 8

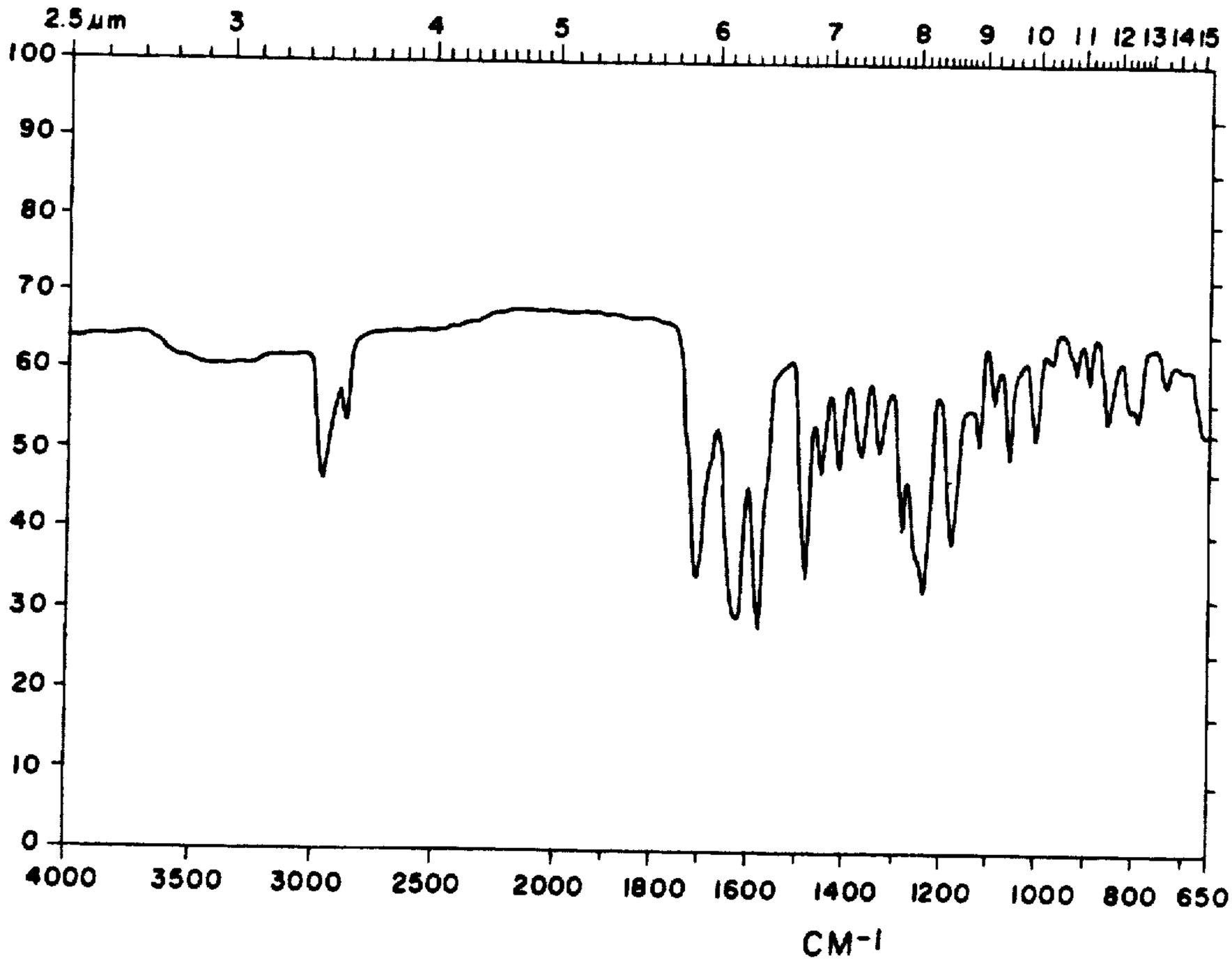


FIGURE 9

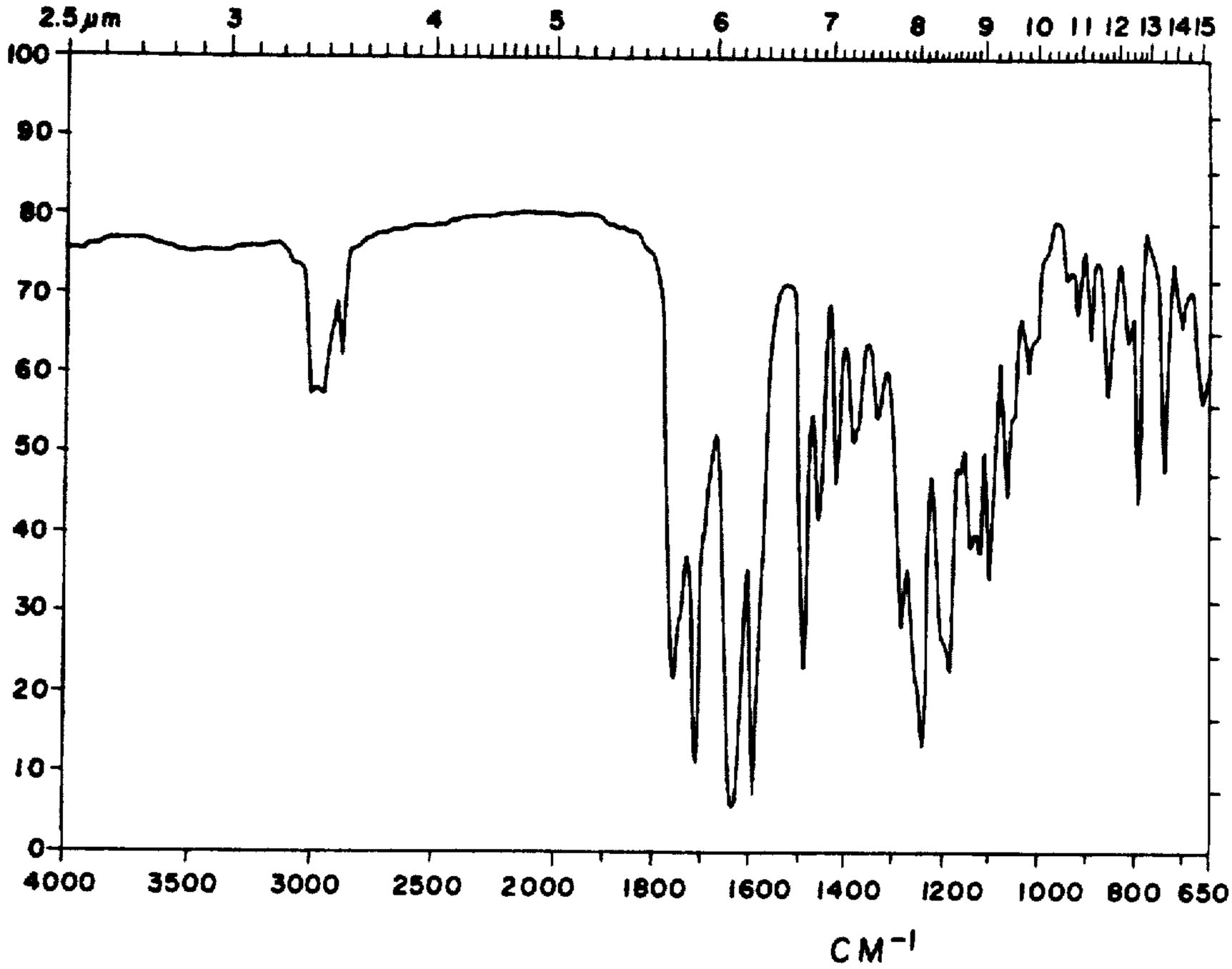


FIGURE 10

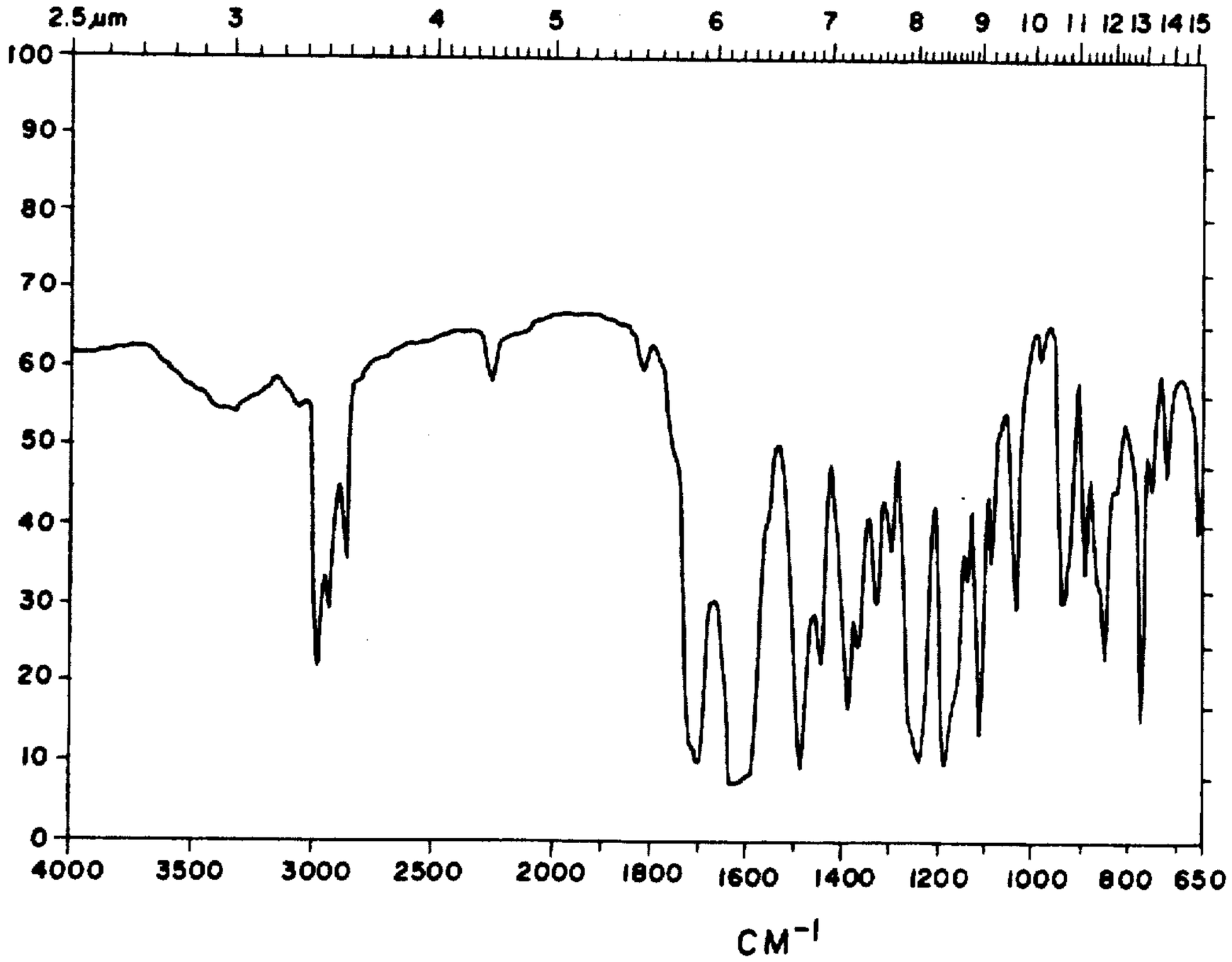


FIGURE 11

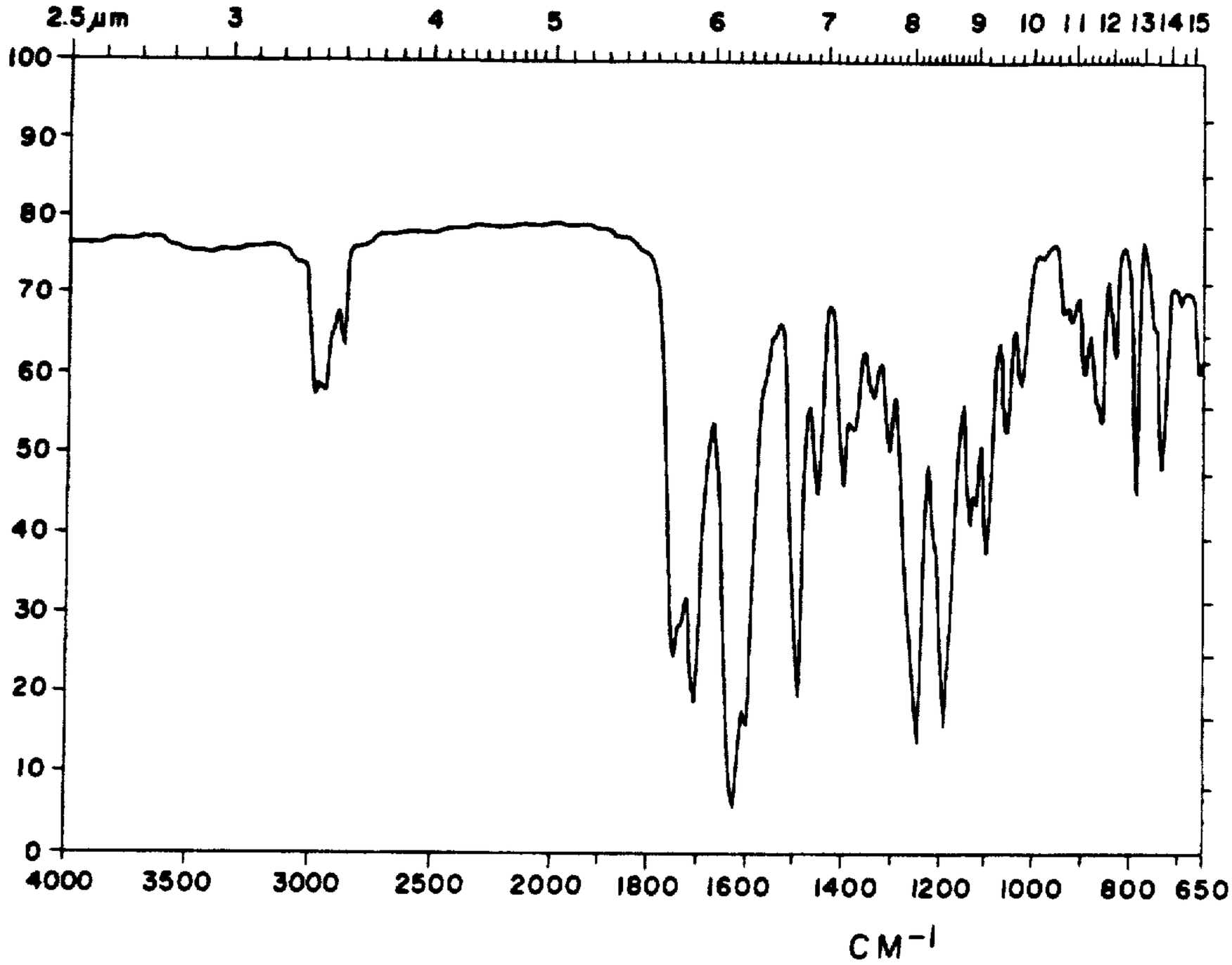


FIGURE 12

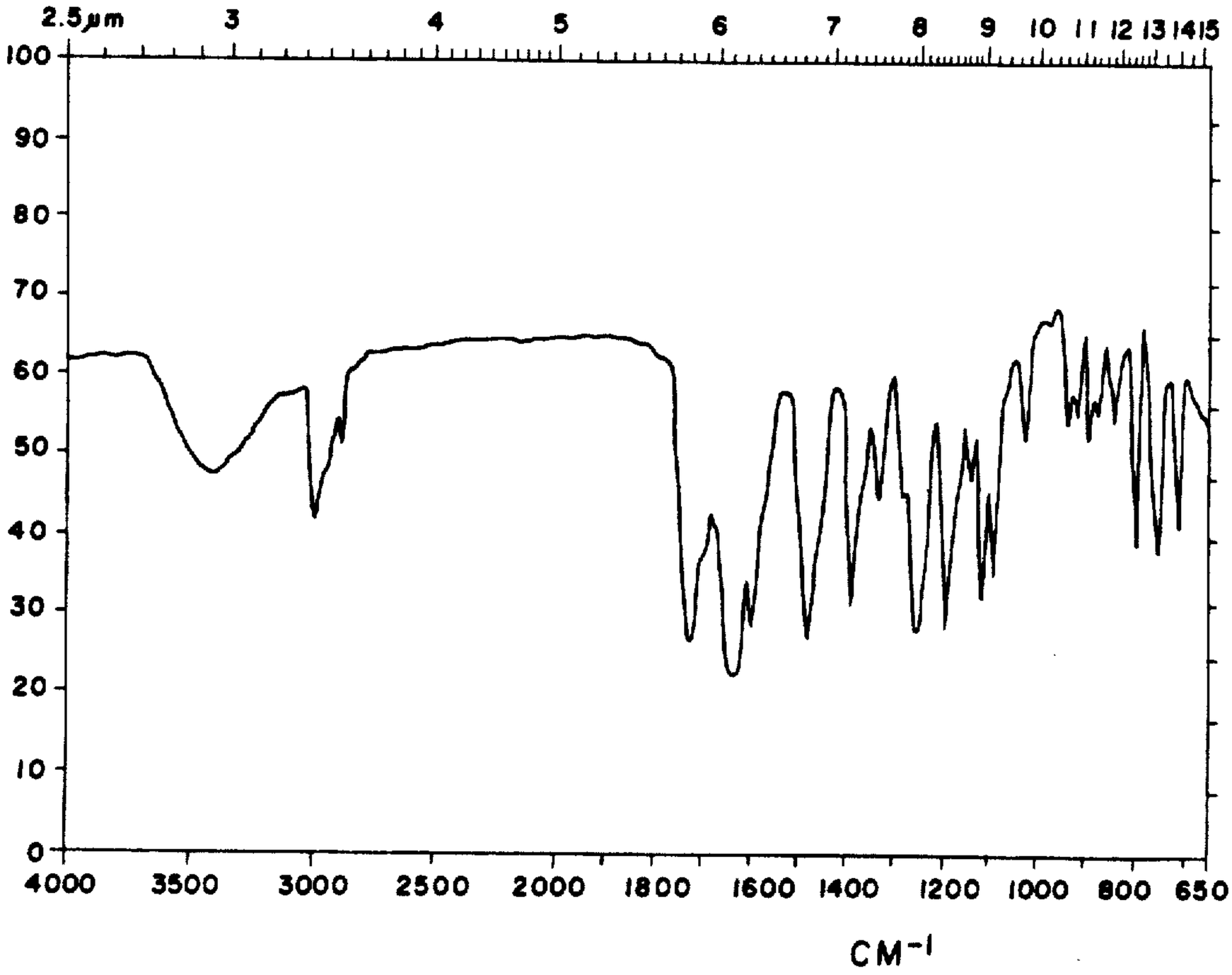


FIGURE 13

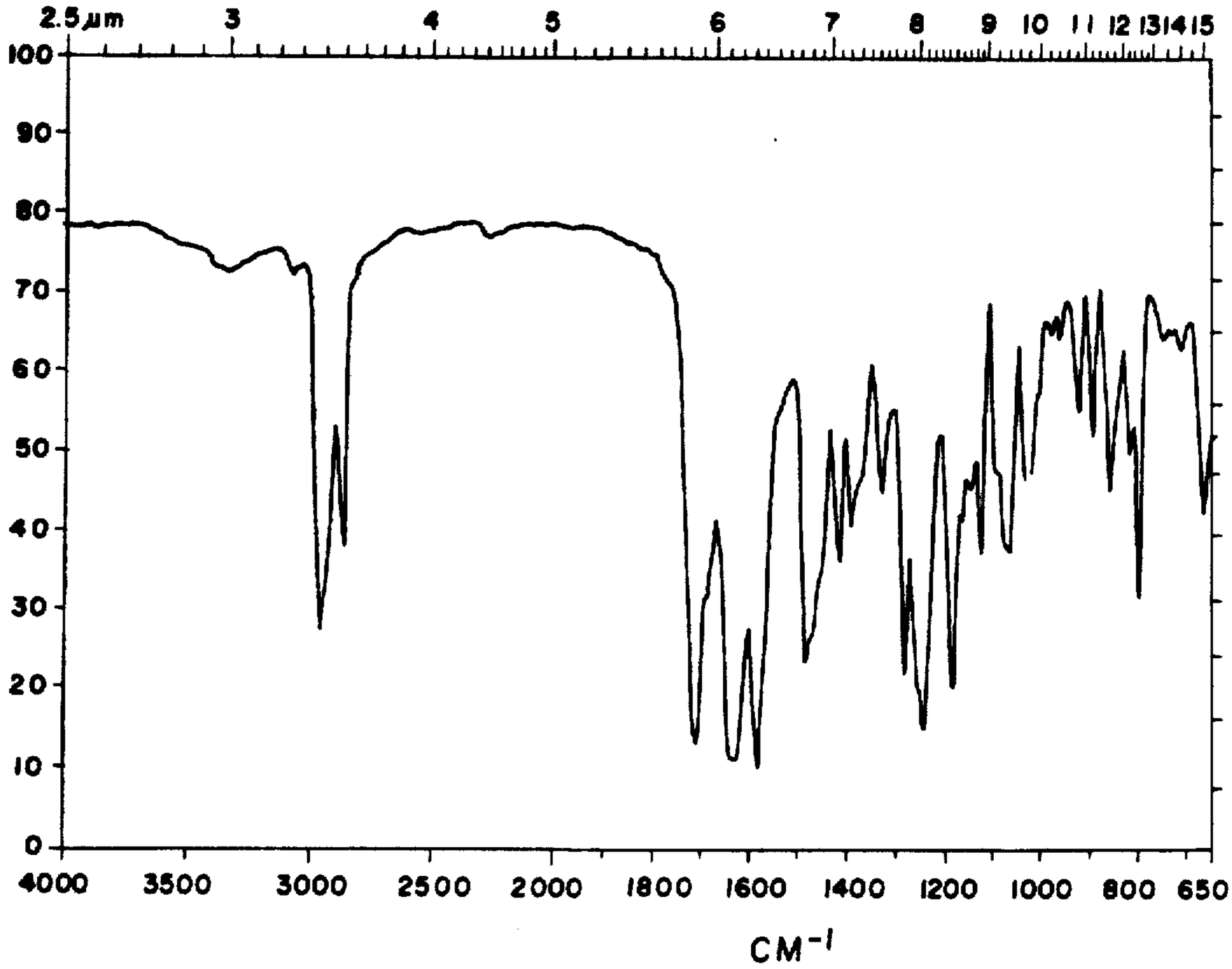


FIGURE 14

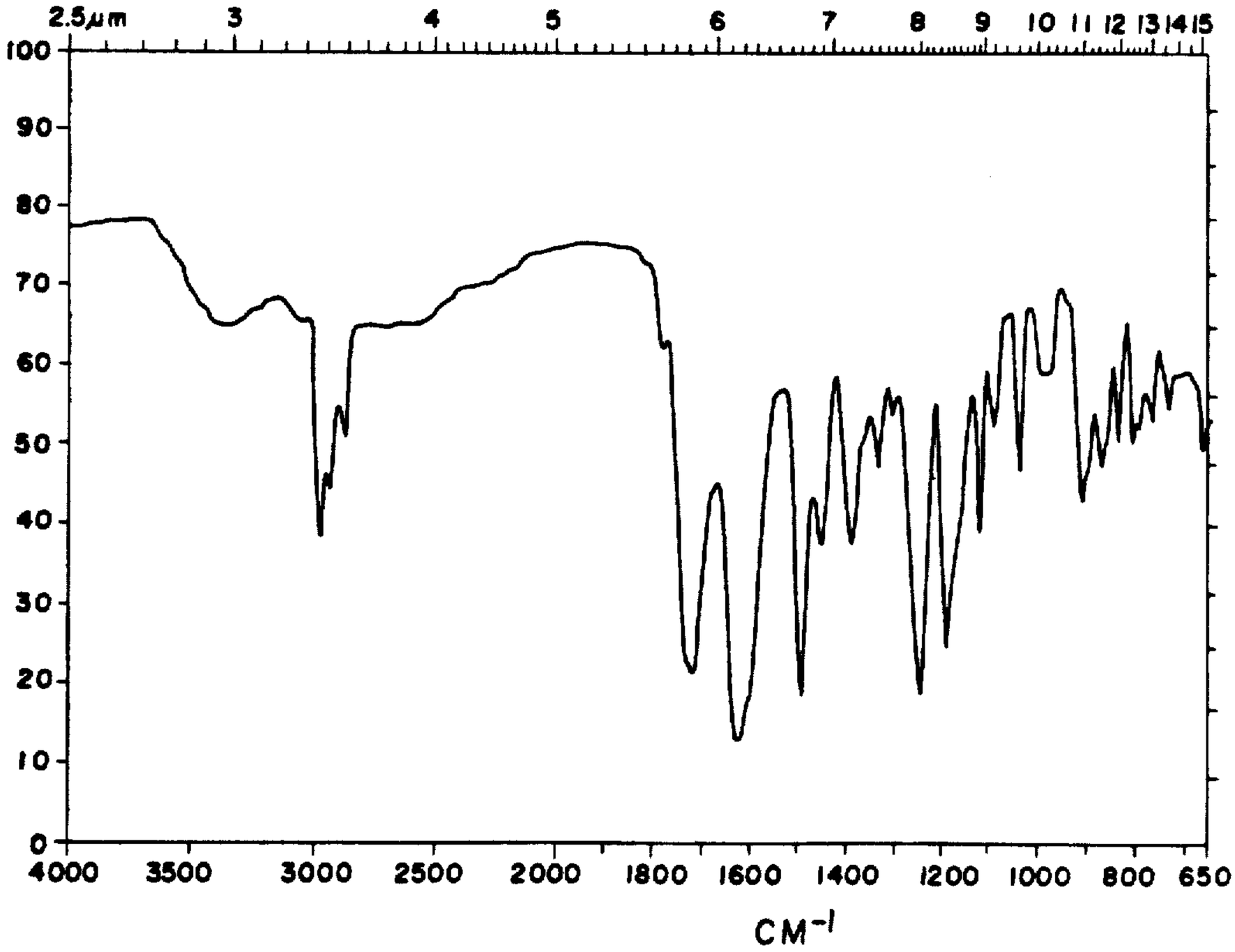


FIGURE 15

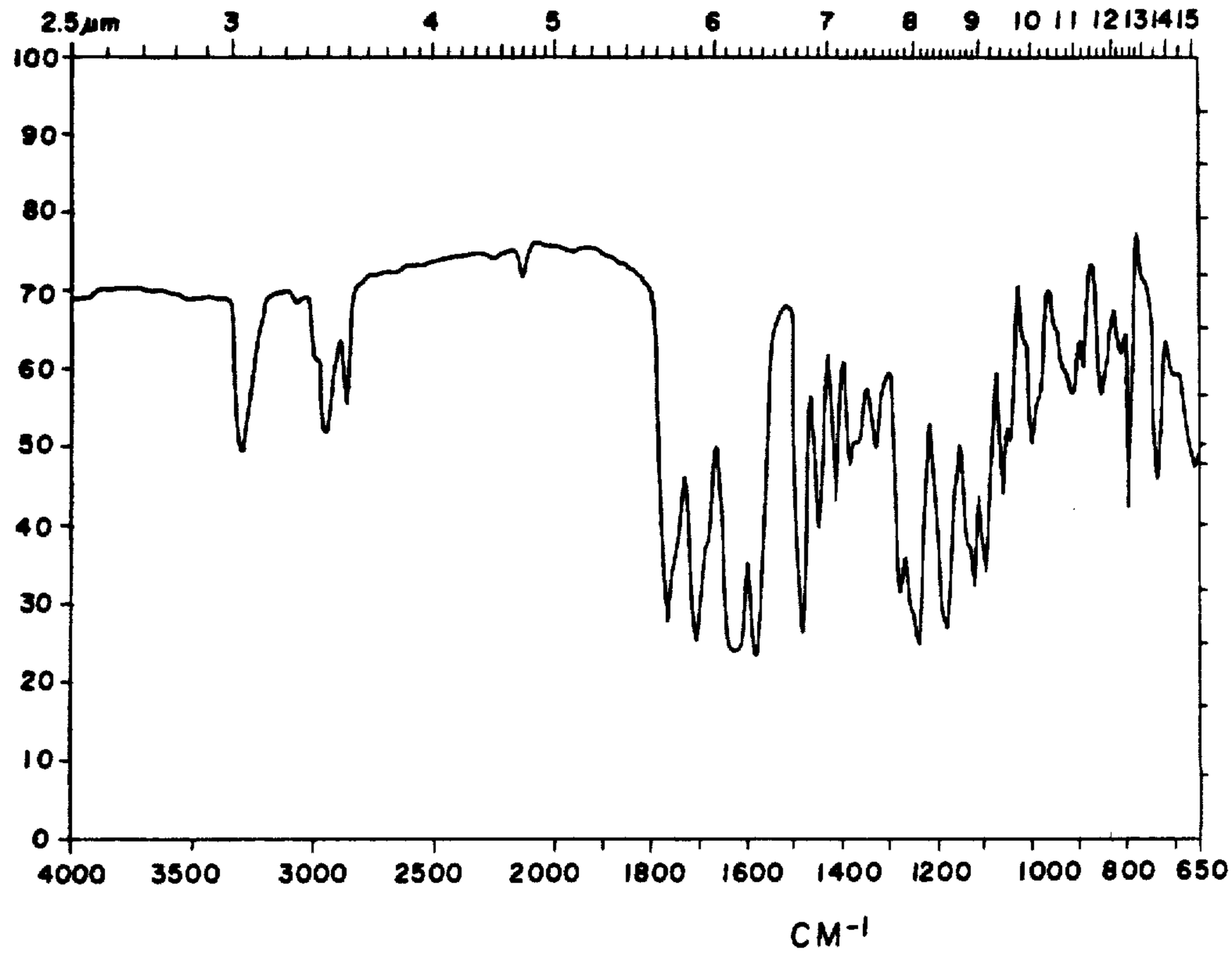


FIGURE 16

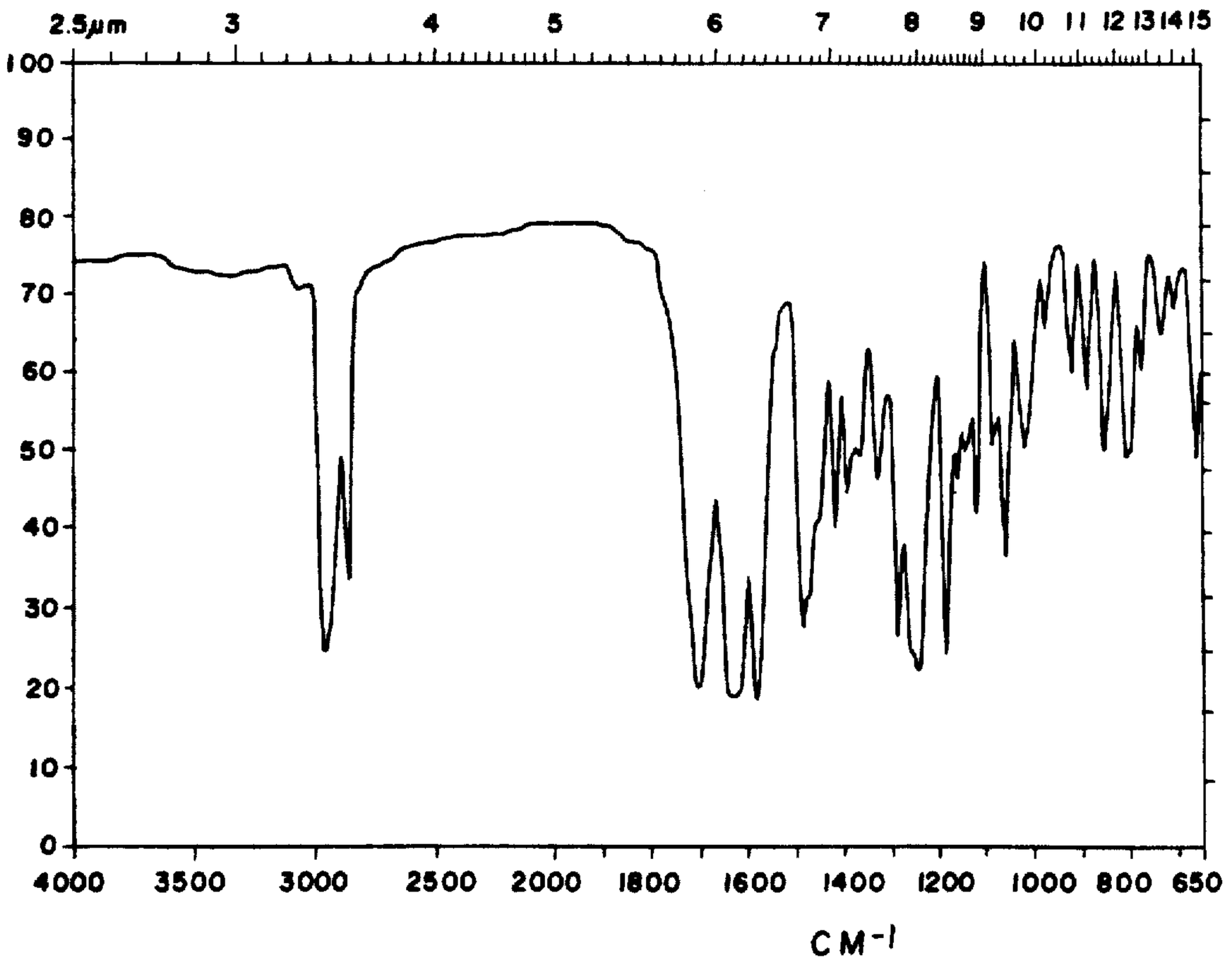


FIGURE 17

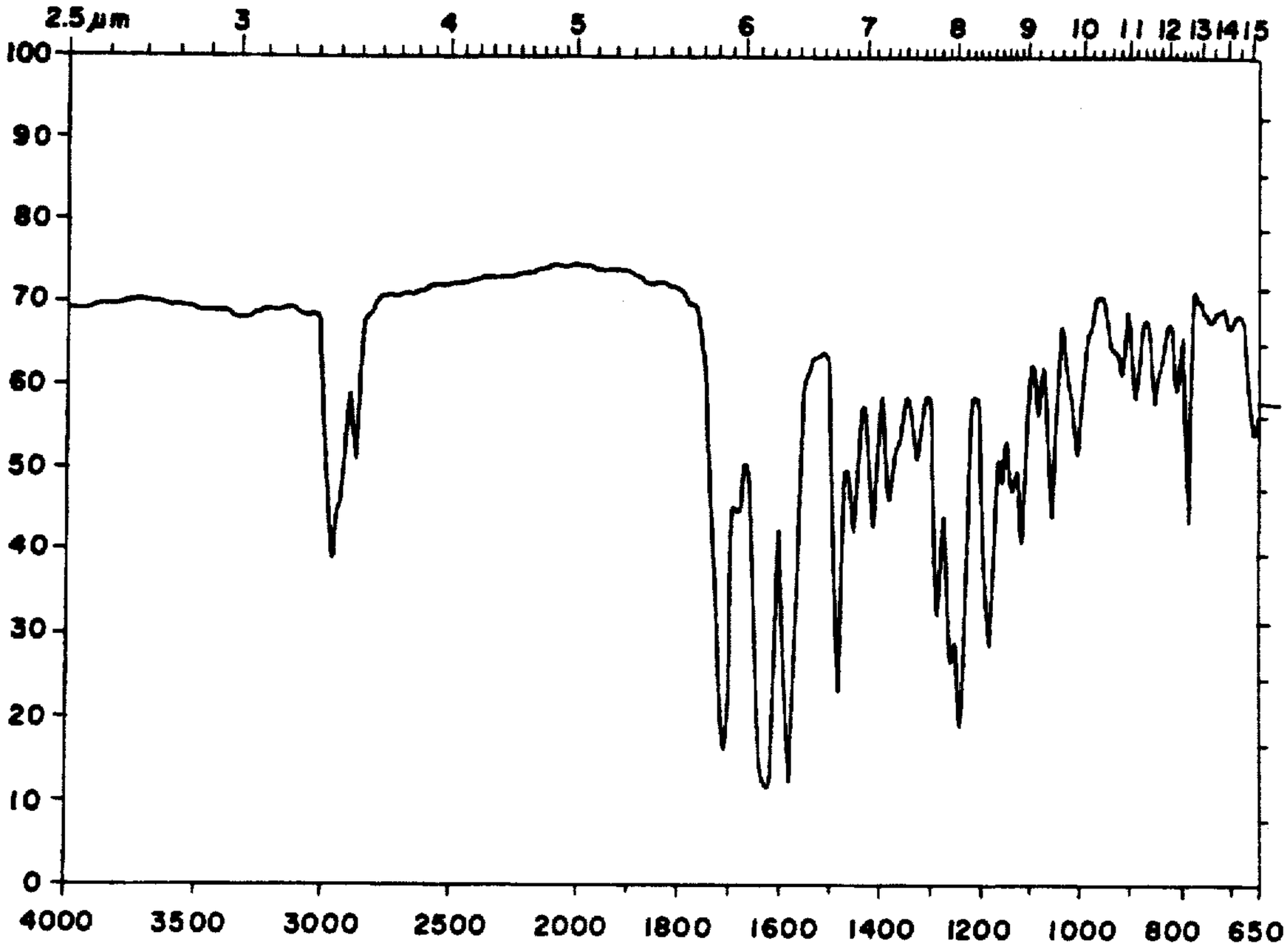
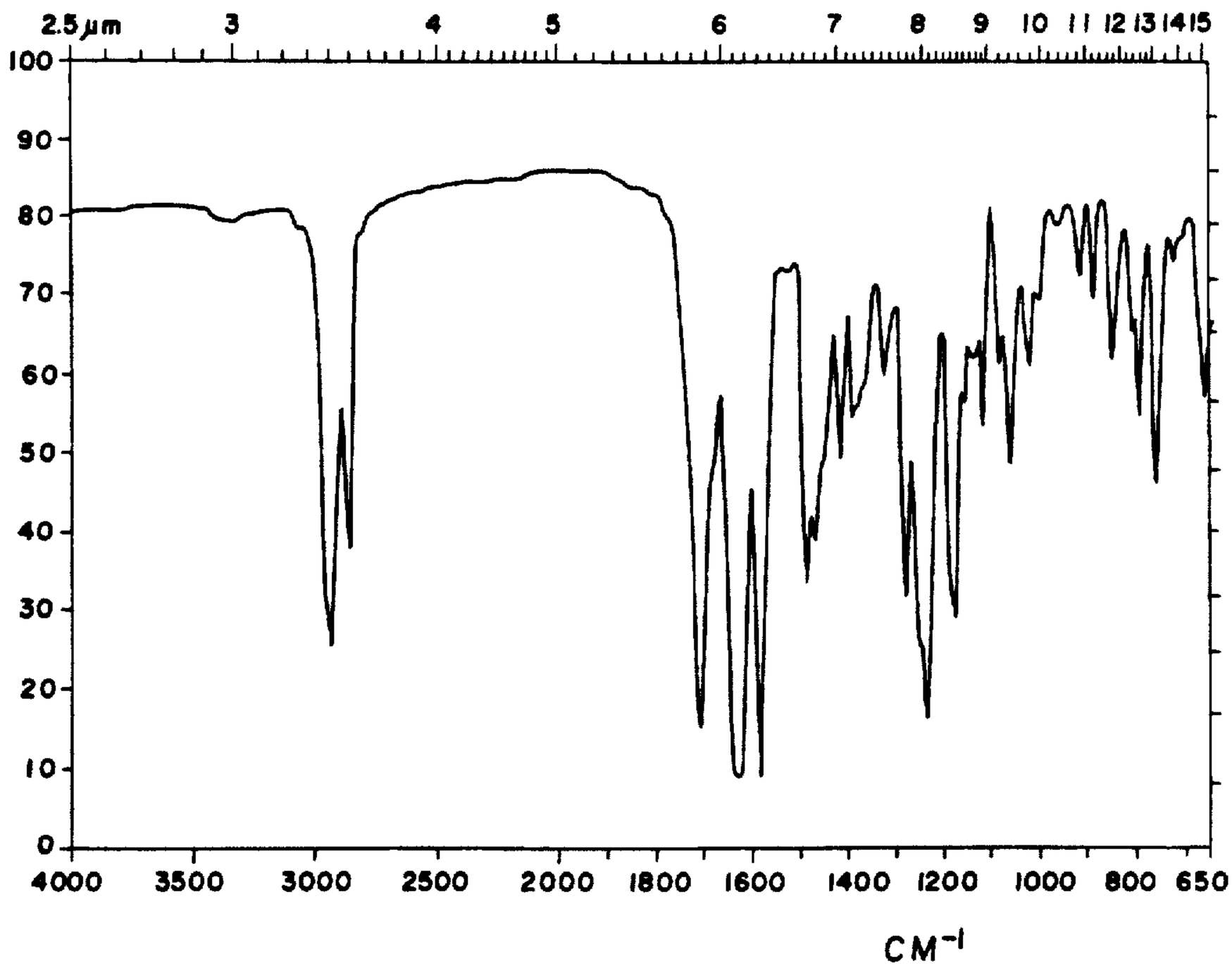


FIGURE 18



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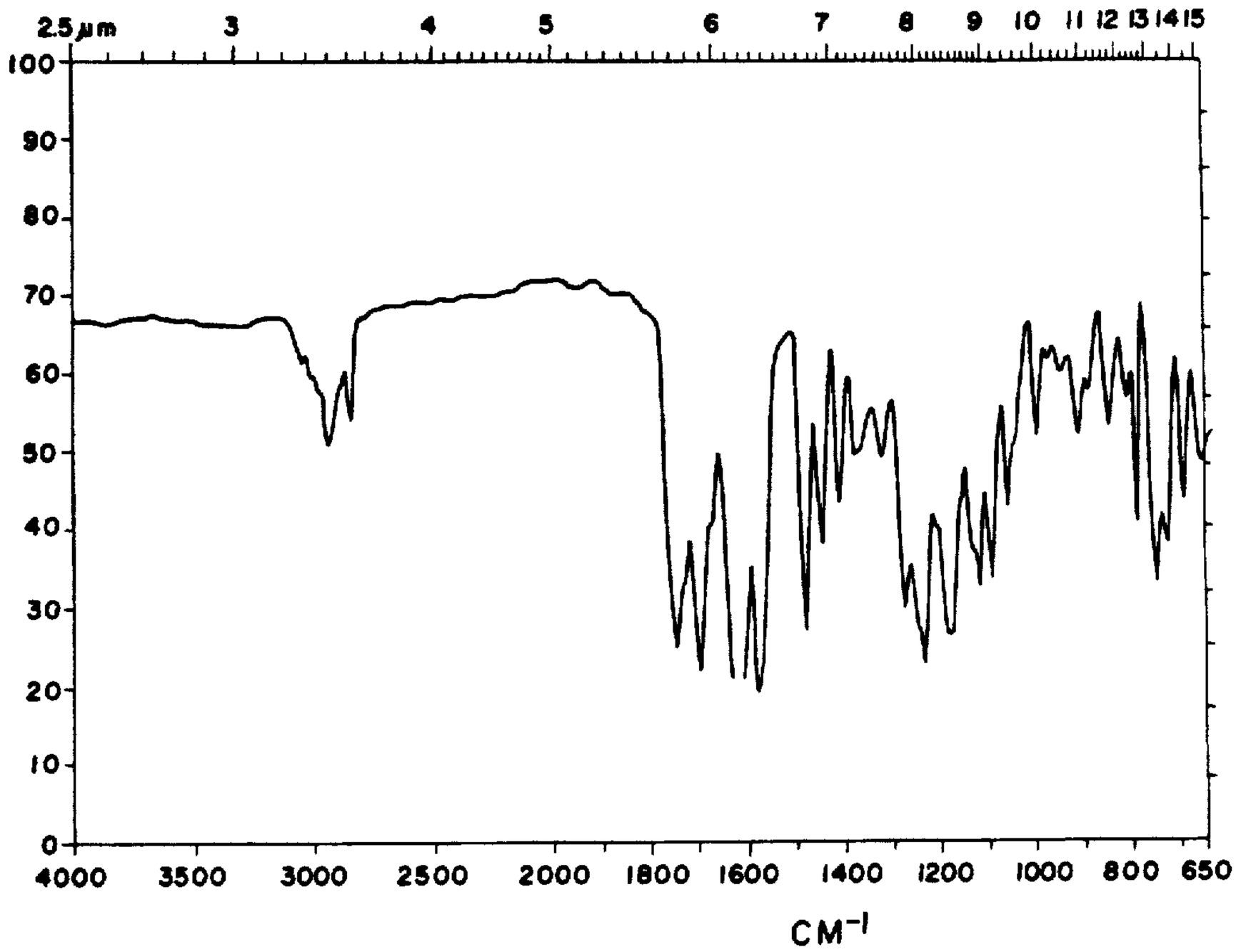
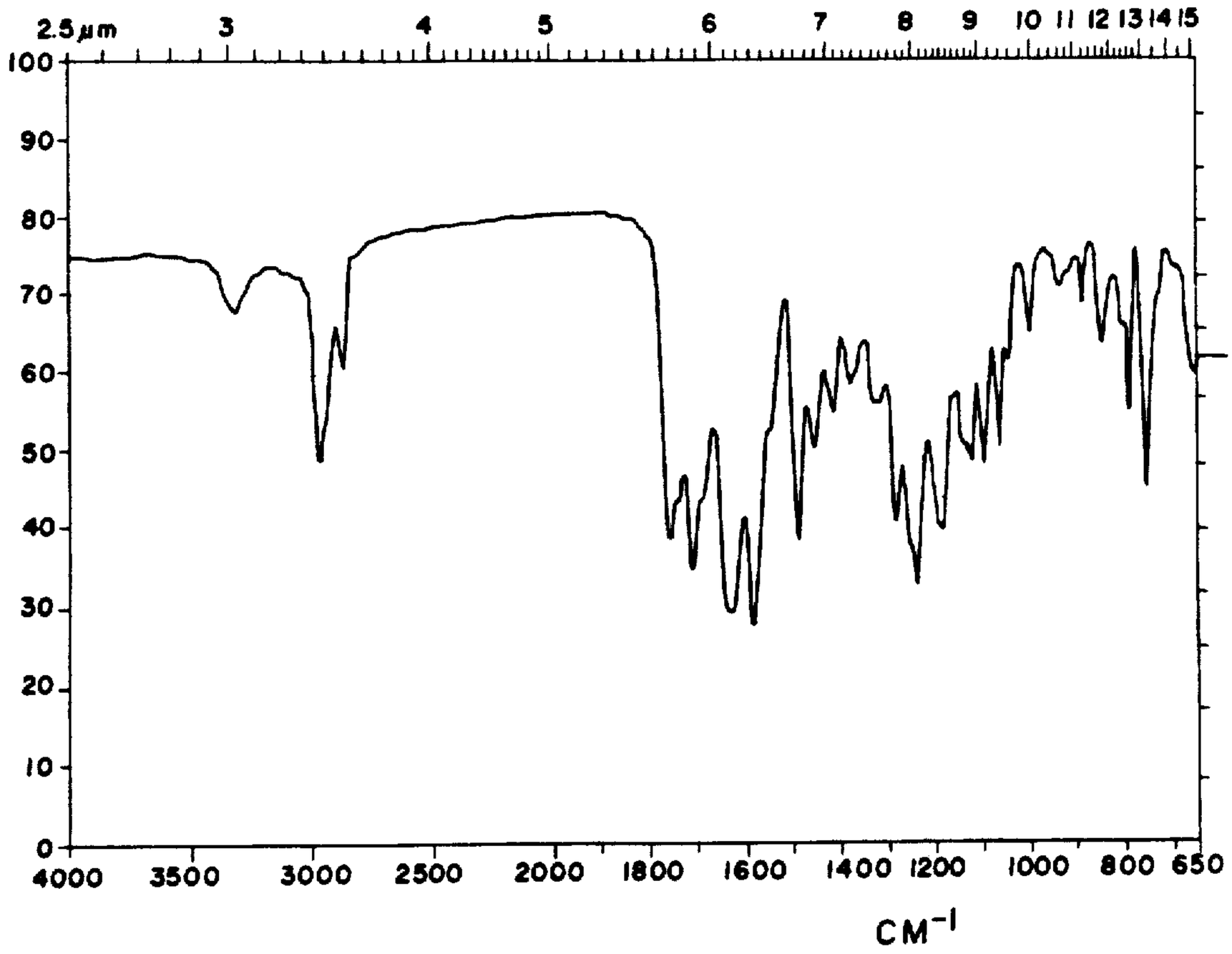
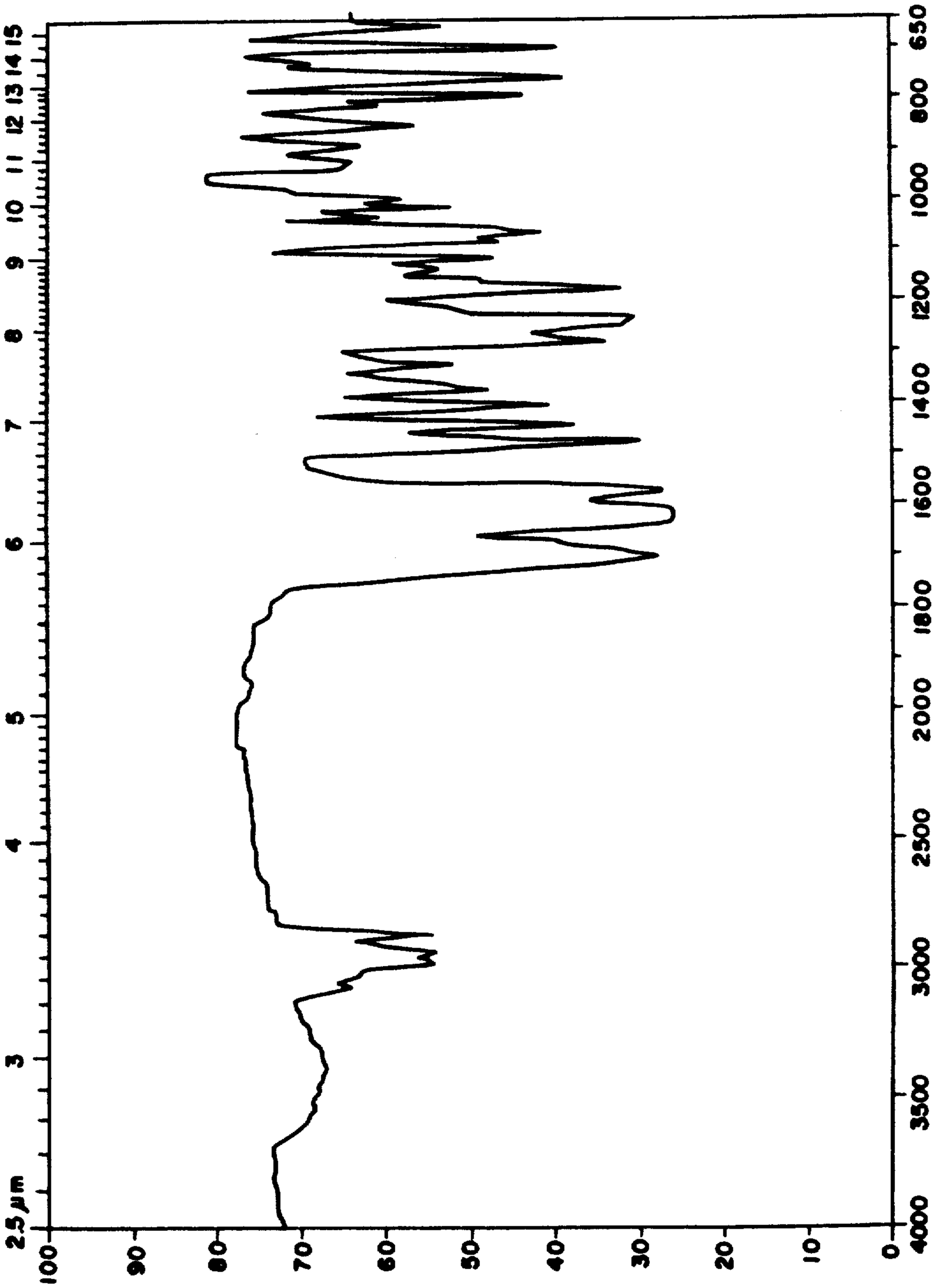


FIGURE 20




2.5 μm
3
4
5
6
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8
9
10
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13
14
15
100
90
80
70
60
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30
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4000
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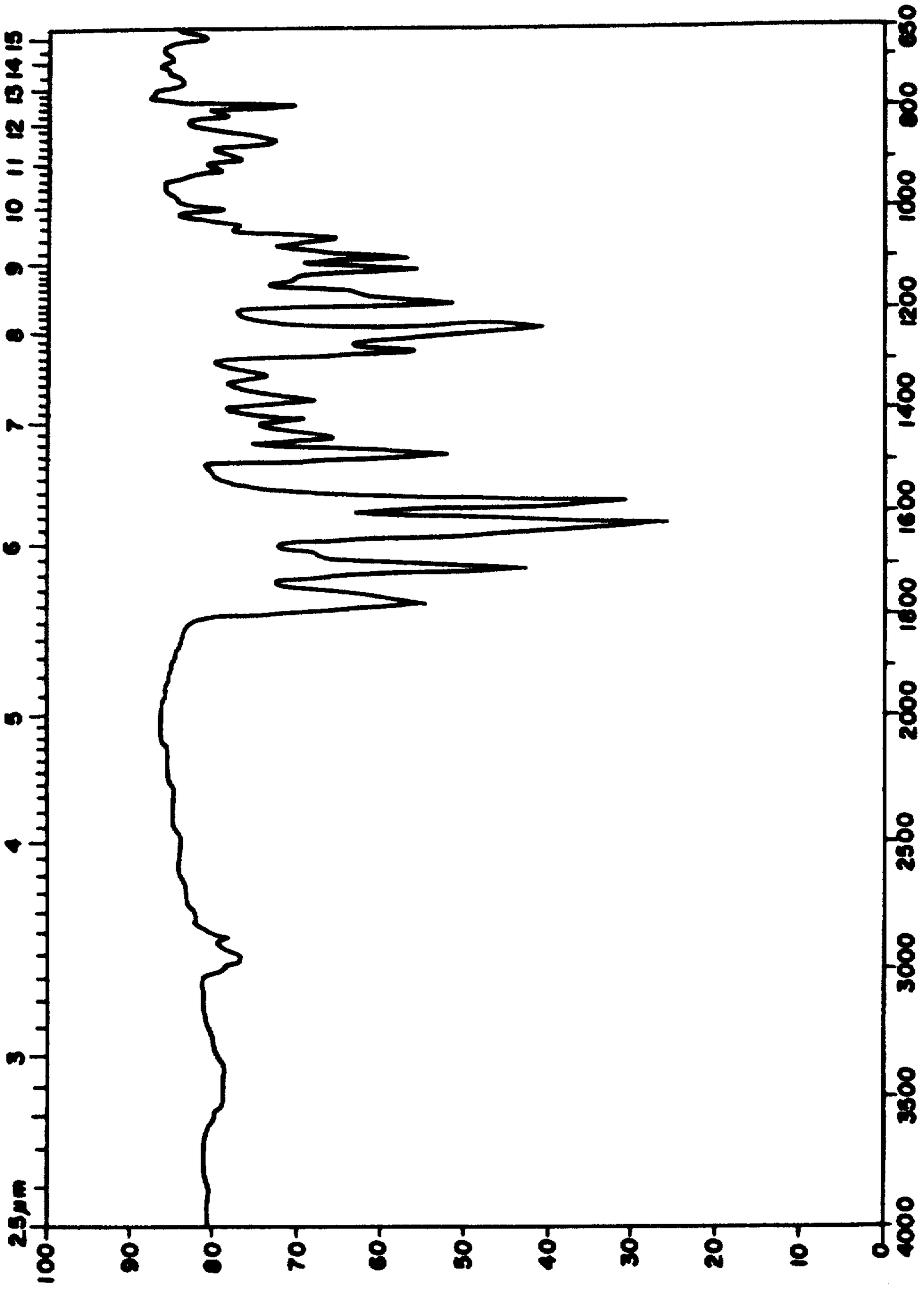
2500
2000
1800
1600
1400
1200
1000
800
650
CM-1

FIGURE 21


2.5μm
3
4
5
6
7
8
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10
11
12
13
14
15
100
90
80
70
60
50
40
30
20
10
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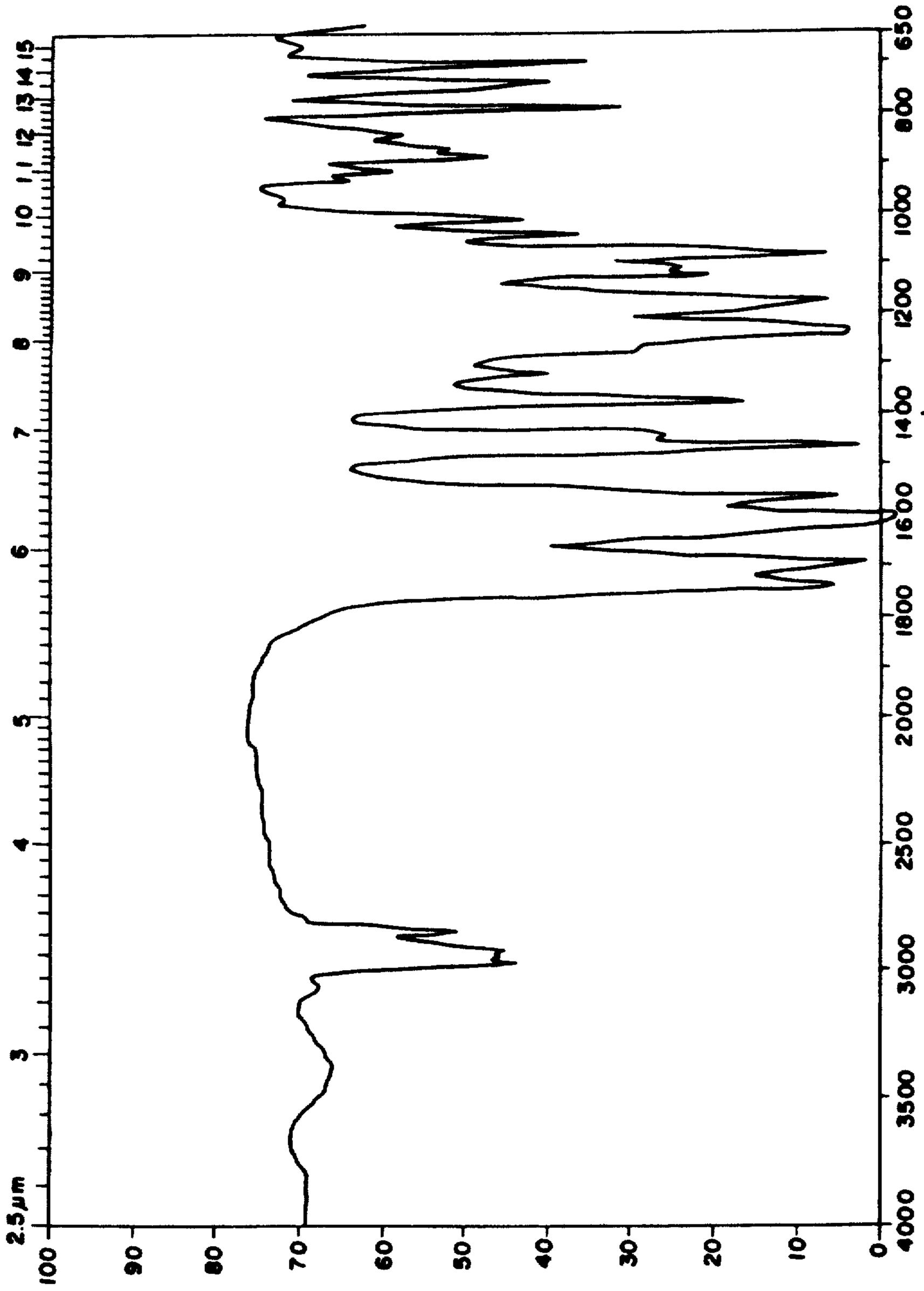
4000
3800
3600
3400
3200
3000
2800
2600
2400
2200
2000
1800
1600
1400
1200
1000
800
600
CM-1

FIGURE 22


2.5μm
3
4
5
6
7
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9
10
11
12
13
14
15
100
90
80
70
60
50
40
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20
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0
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2500
2000
1800
1600
1400
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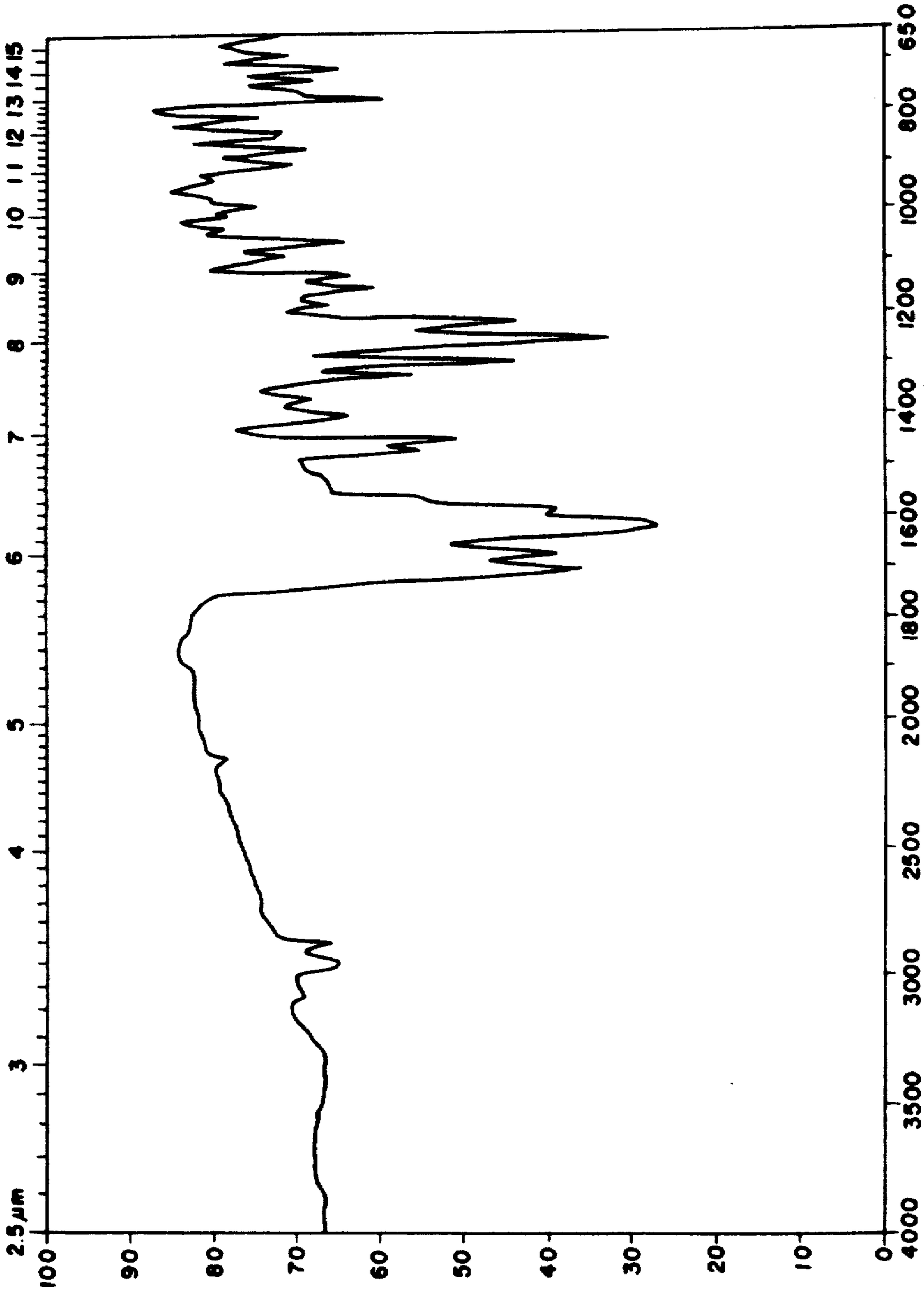
1000
800
650
CM⁻¹

FIGURE 23


2.5 μm
3
4
5
6
7
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9
10
11 12 13 14 15
100
90
80
70
60
50
40
30
20
10
0
4000
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3000
2500
2000
1800
1600
1400
1200
1000
800
650
CM-1

FIGURE 24

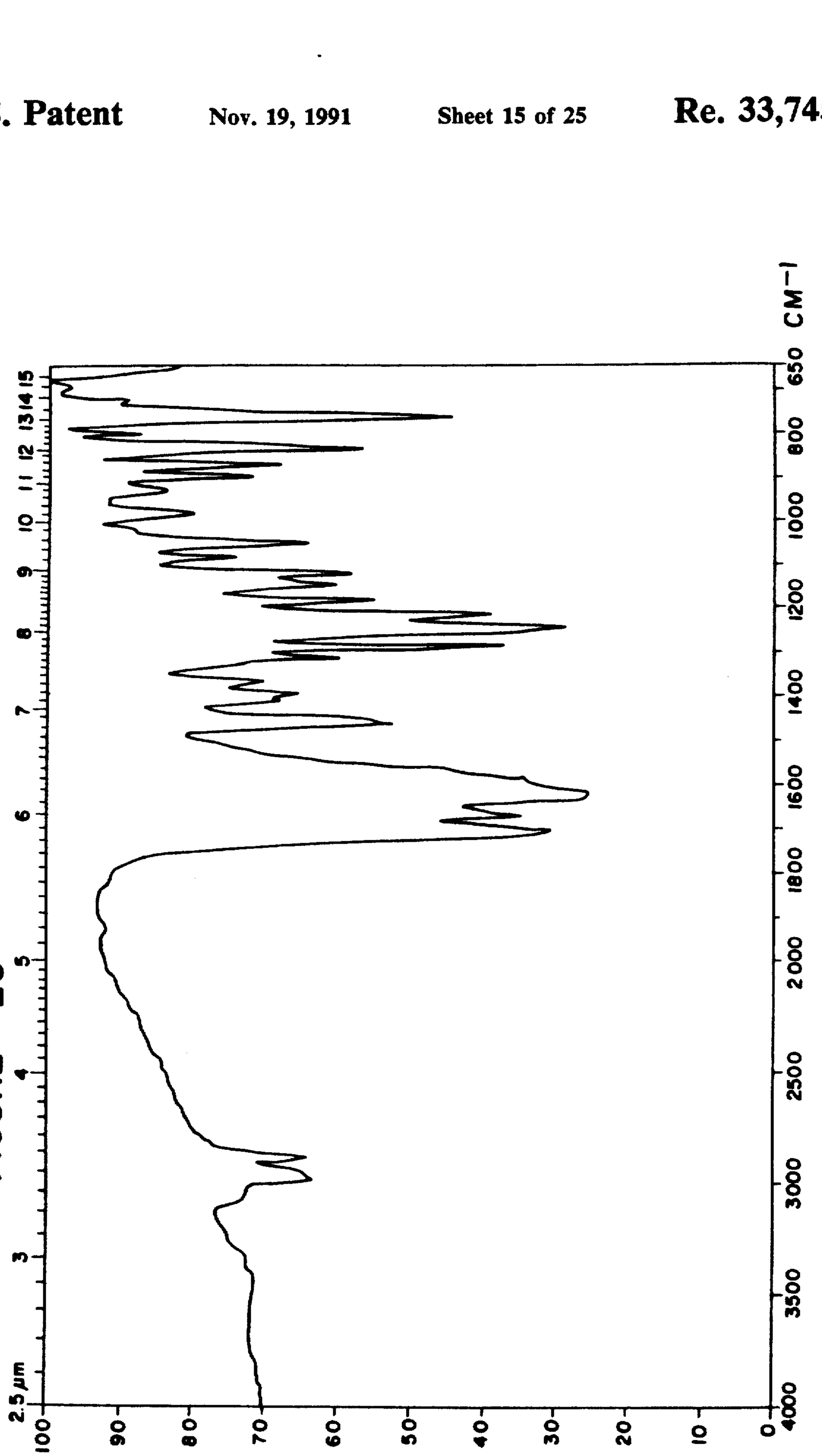

2.5 μm
3
4
5
6
7
8
9
10
11
12
13
14
15
100
90
80
70
60
50
40
30
20
10
0
4000
3500
3000
2500
2000
1800
1600
1400
1200
1000
800
650
CM⁻¹

FIGURE 25

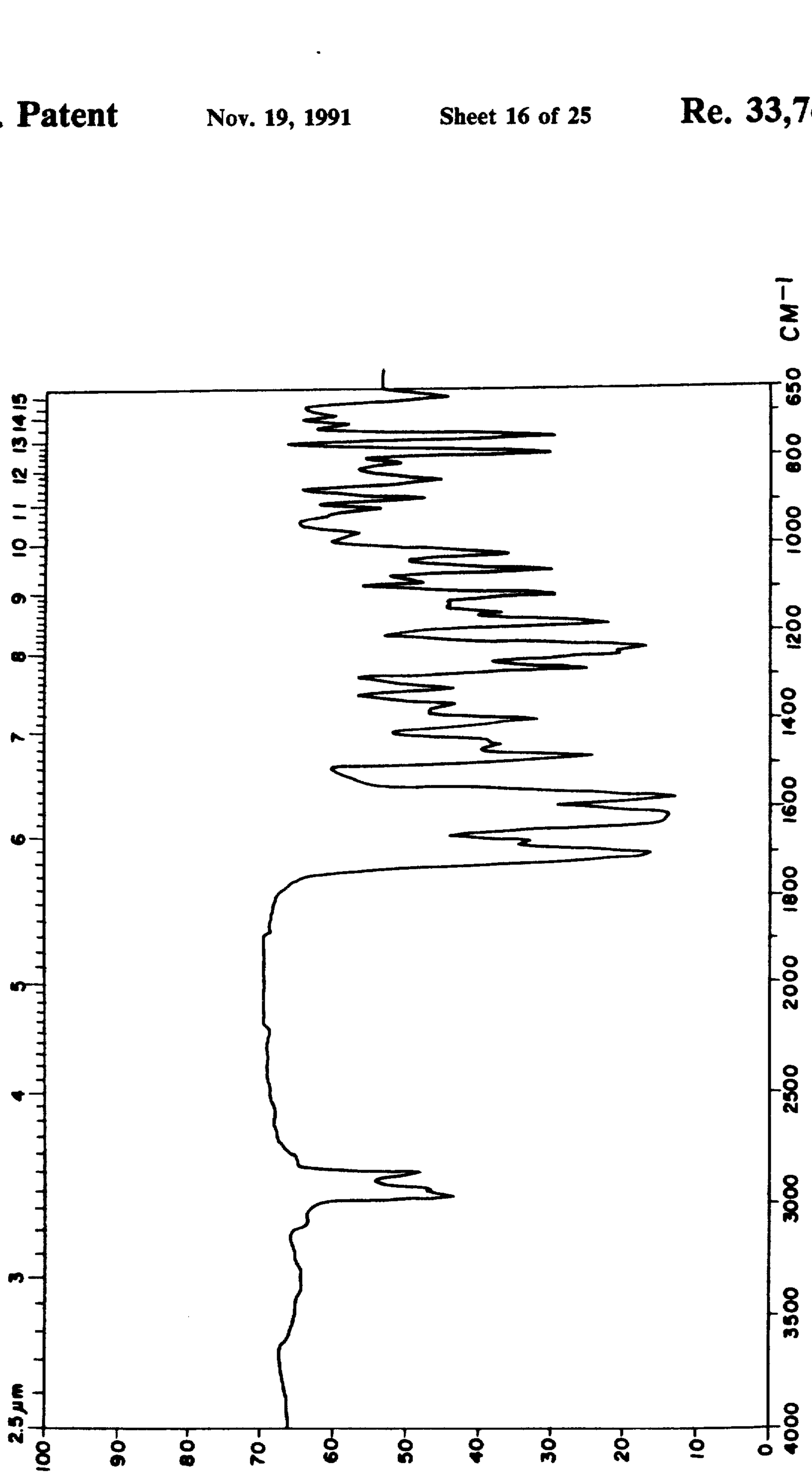
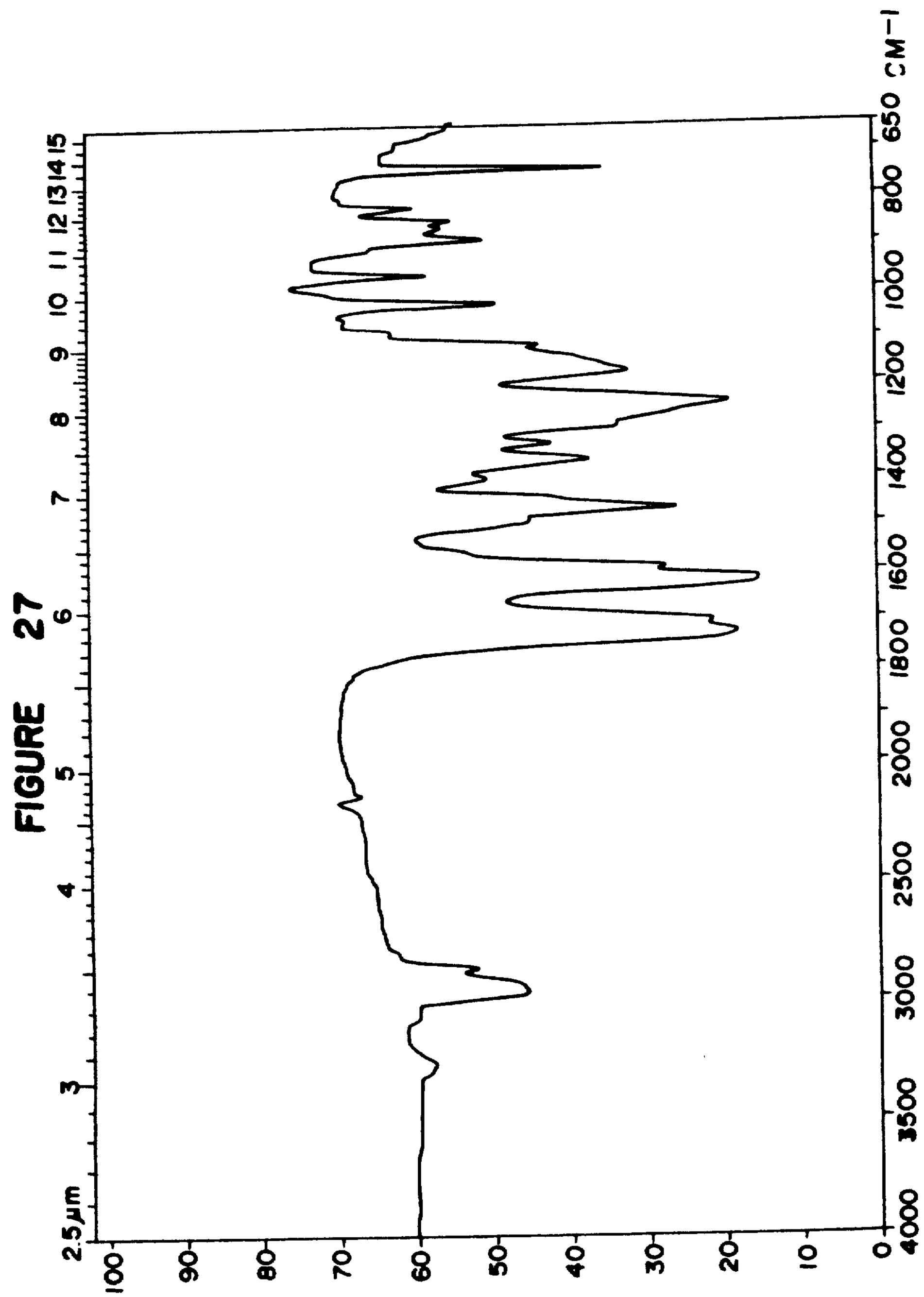

CM⁻¹
650
800
1000
1200
1400
1600
1800
2000
2500
3000
3500
4000
0
10
20
30
40
50
60
70
80
90
100
2.5 μm
3
4
5
6
7
8
9
10
11
12
13
14
15

FIGURE 26


27
FIGURE
2.5μm
3
4
5
6
7
8
9
10
11
12
13
14
15
100
90
80
70
60
50
40
30
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2500
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1800
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1200
1000
800
650
CM⁻¹

FIGURE 28

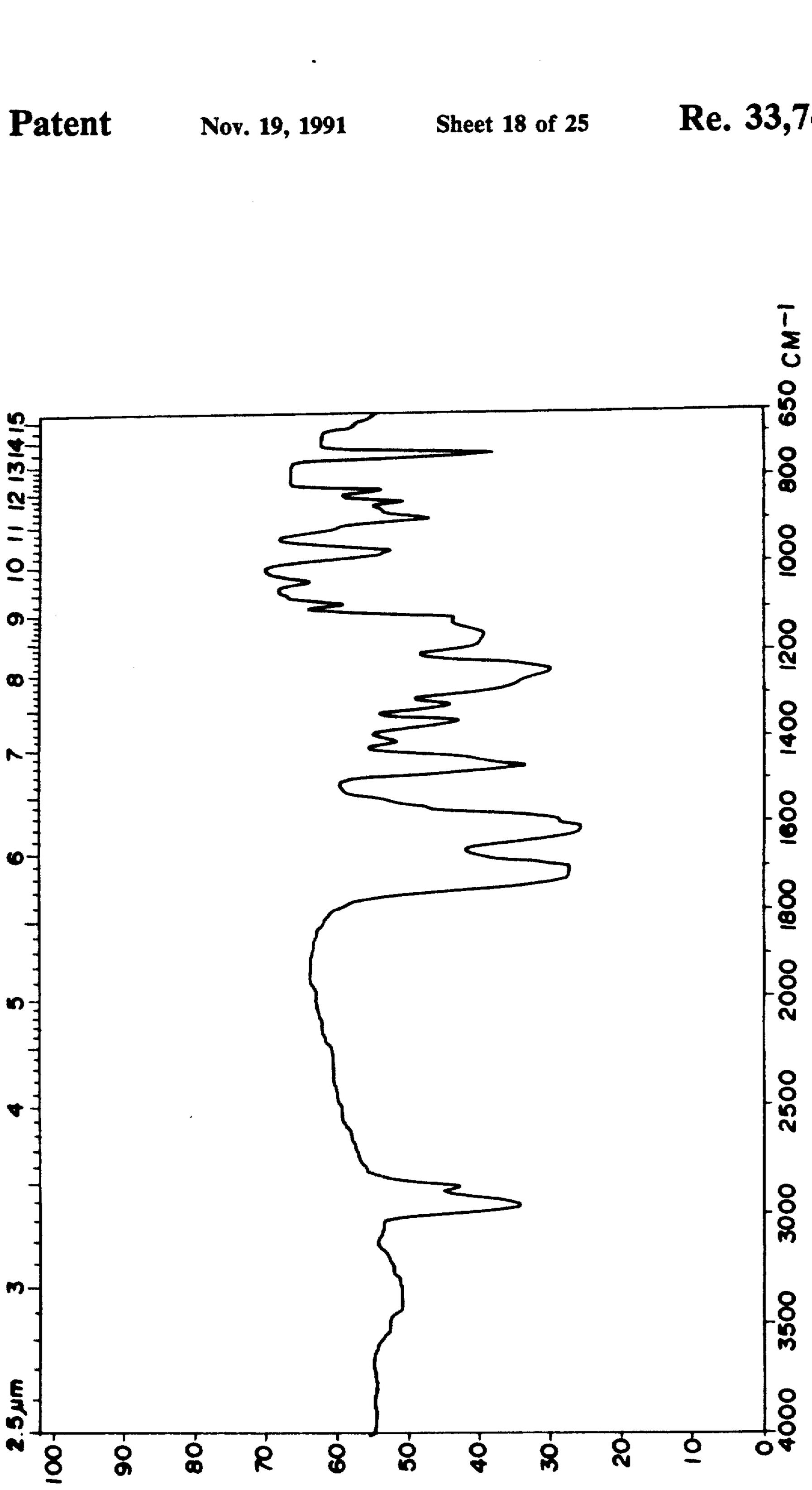
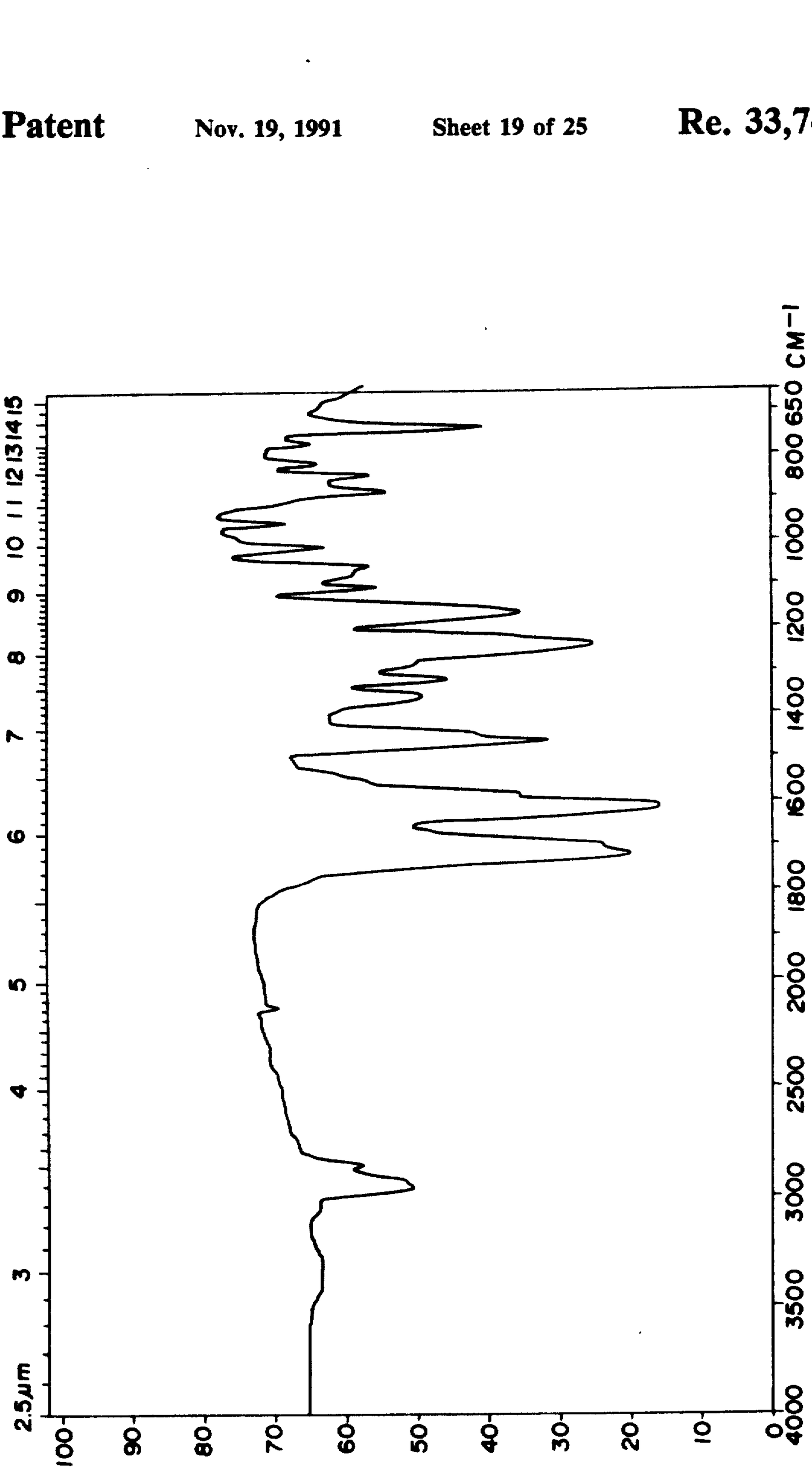

CM⁻¹
15 14 13 12 11 10 9 8 7 6 5 4 3
2.5μm
650 800 1000 1200 1400 1600 1800 2000 2500 3000 3500 4000
100 90 80 70 60 50 40 30 20 10 0

FIGURE 29


CM-1
2.5μm
3
4
5
6
7
8
9
10
11
12
13
14
15
100
90
80
70
60
50
40
30
20
10
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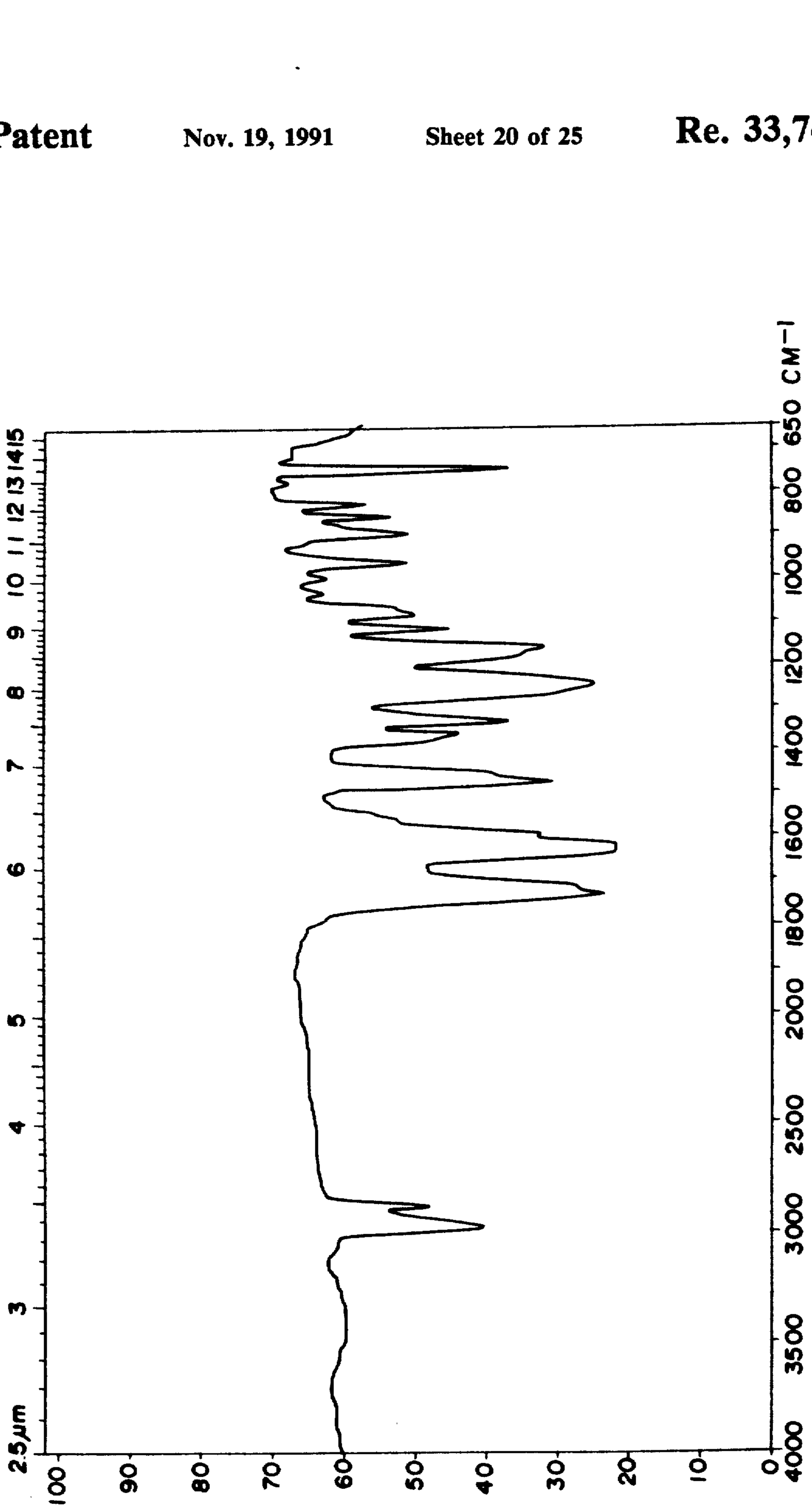
4000
3500
3000
2500
2000
1800
1600
1400
1200
1000
800
650


2.5μm
3
4
5
6
7
8
9
10
11
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15
30
100
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60
50
40
30
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4000
3500
3000
2500
2000
1800
1600
1400
1200
1000
800
650
CM⁻¹

FIGURE

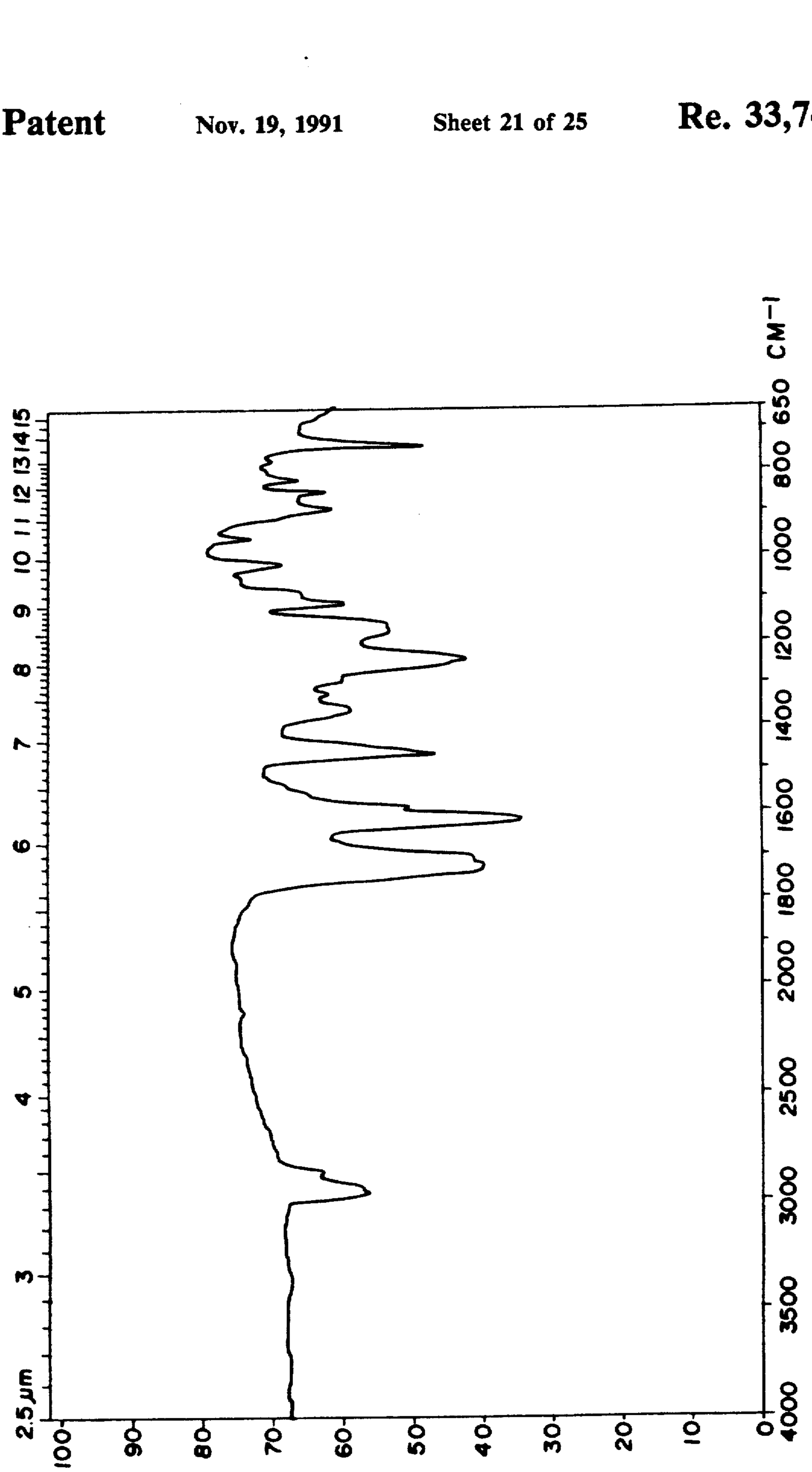
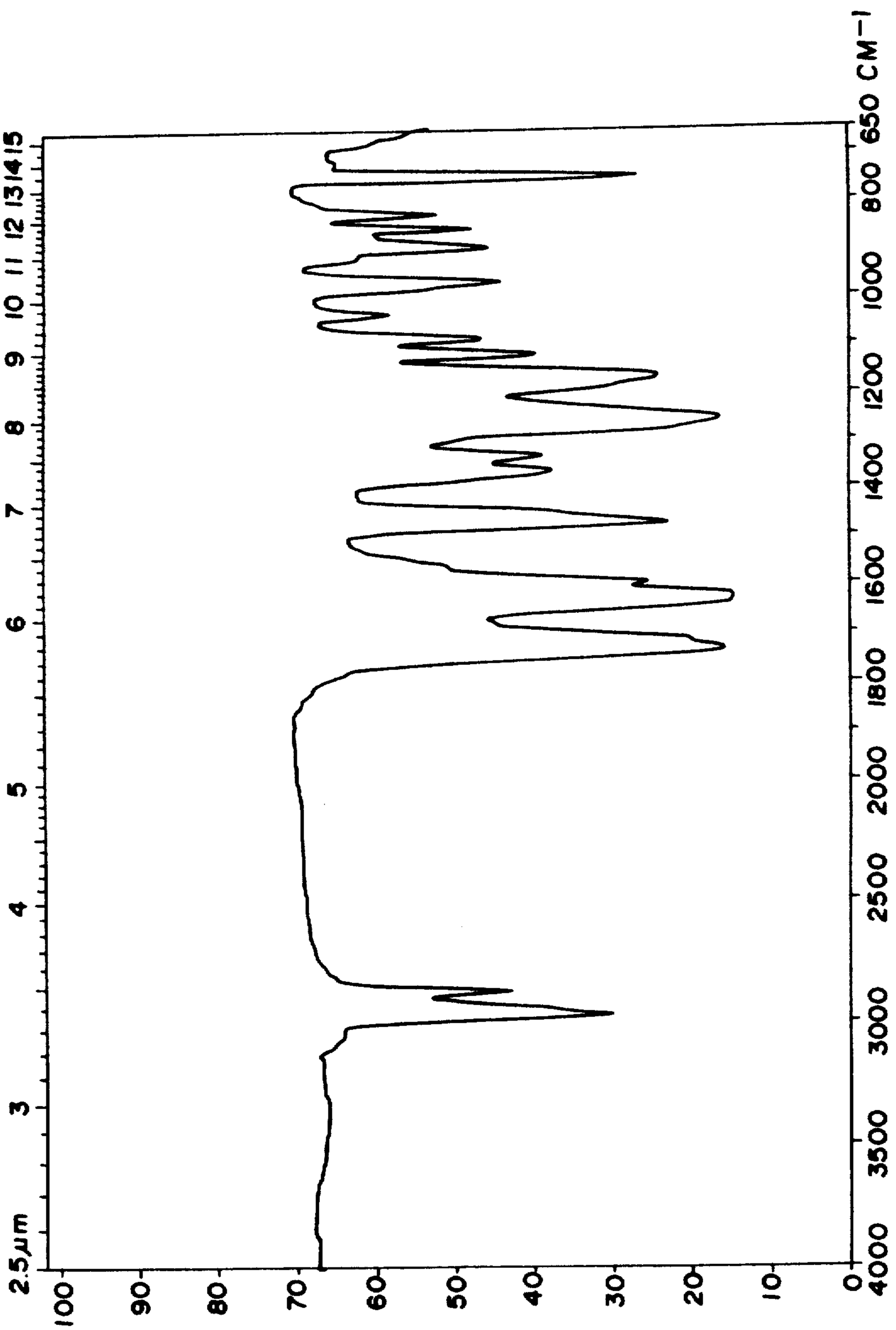

WAVENUMBER, CM⁻¹
4000 3500 3000 2500 2000 1800 1600 1400 1200 1000 800 650
WAVELENGTH, μm
2.5 3 4 5 6 7 8 9 10 11 12 13 14 15
TRANSMITTANCE, %
100 90 80 70 60 50 40 30 20 10 0

FIGURE 31


32
FIGURE
2.5μm
3
4
5
6
7
8
9
10
11
12
13
14
15
100
90
80
70
60
50
40
30
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2500
2000
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1600
1400
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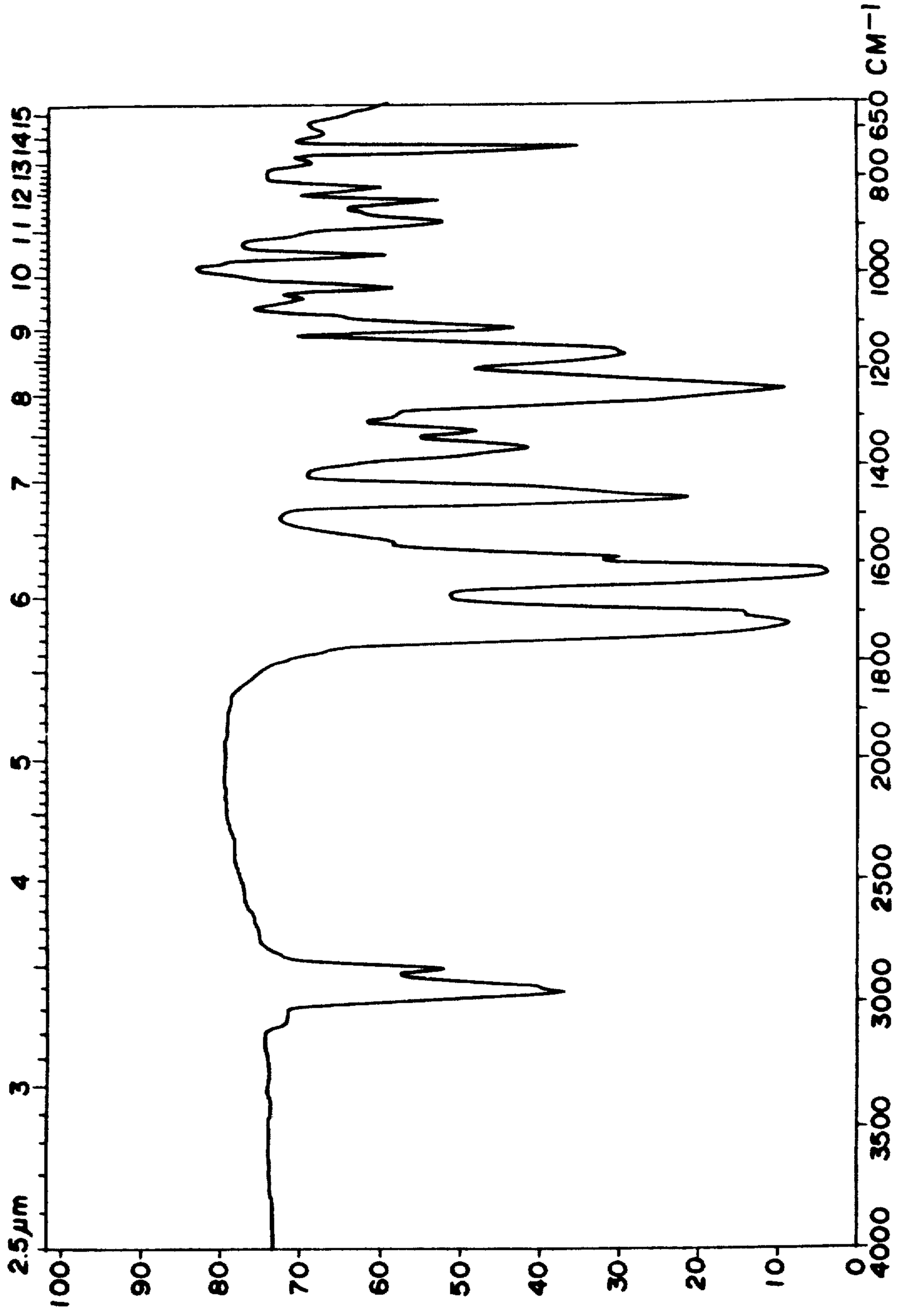
1000
800
650
CM⁻¹


CM⁻¹
2.5μm
3
4
5
6
7
8
9
10
11
12
13
14
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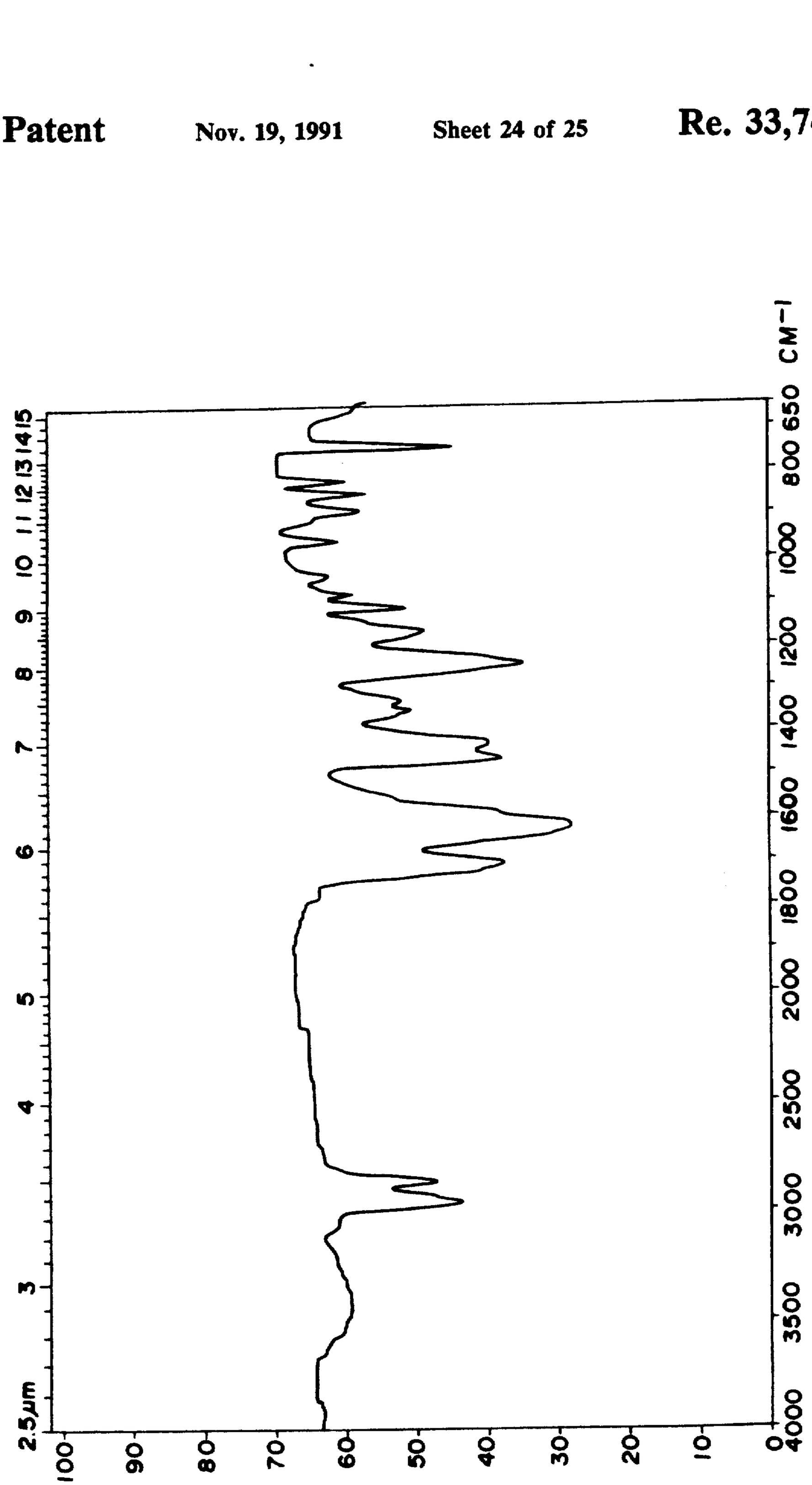
1000
800
650

FIGURE 33


CM⁻¹
2.5μm
3
4
5
6
7
8
9
10
11
12
13
14
15
4000
3500
3000
2500
2000
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1600
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1200
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800
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0
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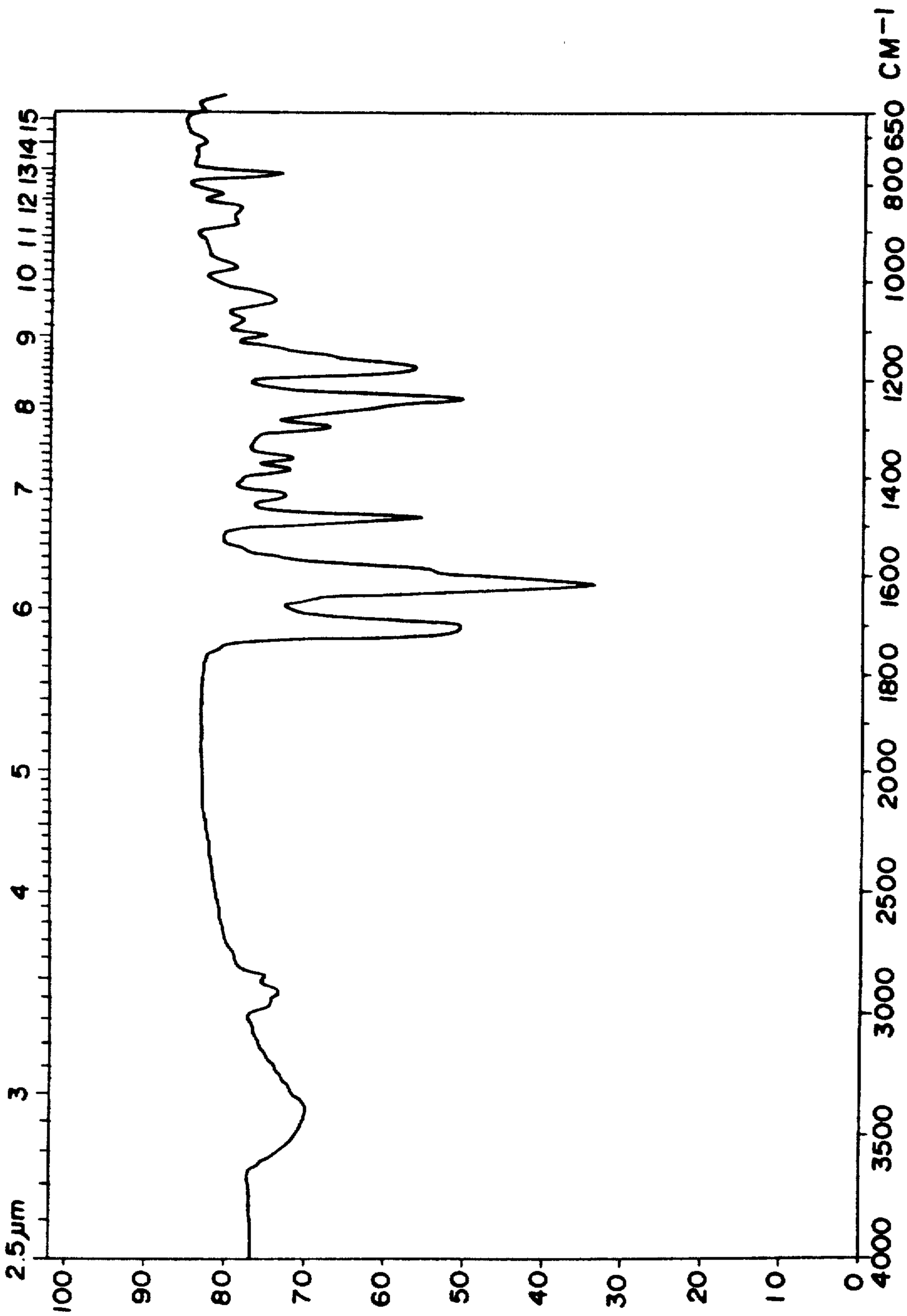
40
50
60
70
80
90
100

FIGURE 34


CM⁻¹
2.5μm
3
4
5
6
7
8
9
10
11
12
13
14
15
100
90
80
70
60
50
40
30
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10
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1800
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1000
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650
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3000
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2000
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FIGURE 35

THIADIAZABICYCLONONANE DERIVATIVES, PROCESSES FOR THEIR PRODUCTION AND HERBICIDAL COMPOSITIONS

Matter enclosed in heavy brackets [ ] appears in the original patent but forms no part of this reissue specification; matter printed in italics indicates the additions made by reissue.

The present invention relates to novel 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) derivatives, processes for their production and herbicidal compositions containing them.

In recent years, a number of herbicides have been developed and actually used, and they have contributed to the reduction of the agricultural work load and to the improvement of the productivity. As a herbicide having a hetero ring, Ronstar [i.e. 5-t-butyl-3-(2,4-dichloro-5-isopropoxyphenyl)-1,3,4-oxadiazoline-2-one] is widely used. However, Ronstar has drawbacks that it is likely to bring about phytotoxicity, and it is not effective against perennial weeds, particularly against Sagitaria pygmaea. Accordingly, a development of a herbicide having improved effectiveness and safety has been desired.

Under the circumstances, the present inventors have conducted extensive researches with an aim to develop a herbicide which satisfies the following conditions, and have finally accomplished the present invention.

(1) It is effective at a low dose.

(2) It is effective against paddy field weeds and (or) against upland field weeds.

(3) It is also effective against perennial weeds.

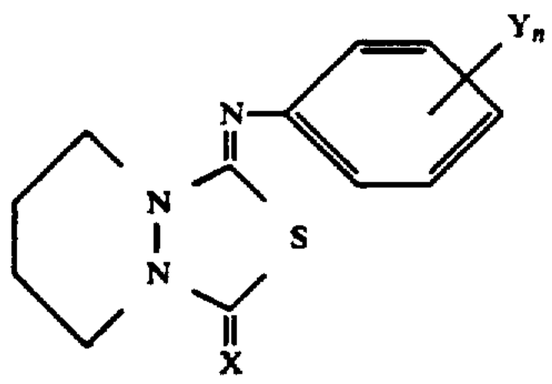
(4) It is effective in a wide range covering the germination stage to the growing stage.

(5) It has excellent residual effects and can be expected to provide stabilized effects.

(6) It exhibits excellent herbicidal effects.

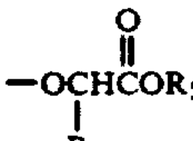
(7) It is highly safe to crop plants.

Thus, the present invention provides a 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) derivative having the formula:

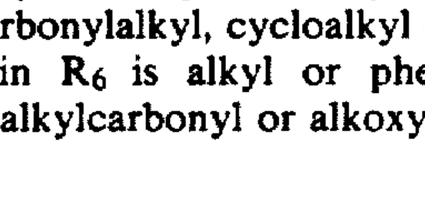


(I)

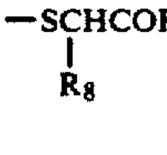
wherein Y which may be the same or different, represents halogen, hydroxy, alkyl, alkoxy which may be substituted by halogen, alkenyloxy which may be substituted by halogen, alkynyloxy, phenoxy, cycloalkyloxy, alkoxycarbonylalkyloxy, alkoxycarbonylalkenyloxy, alkylthiocarbonylalkyloxy, alkynyloxycarbonylalkyloxy, benzyloxycarbonylalkyloxy, trifluoromethyl, benzyloxy which may be substituted by chlorine or alkyl, alkenyl, cyanoalkyl, alkylcarbamoyloxy, benzyl which may be substituted by one or two alkyl, alkoxyalkyl, alkynyloxyalkyl, cycloalkylmethyloxy which may be substituted by halogen, alkoxyalkyloxy, phenethyloxy, cycloalkyloxycarbonylalkyloxy, pyrrolidinocarbonyl, phenylcarbonyl which may be substituted by alkyl,



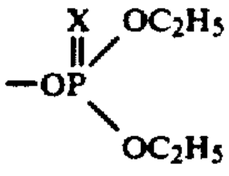
[wherein R_1 is hydrogen, alkyl, phenyl, cycloalkyl, alkoxyalkyl, alkoxycarbonylalkyl or



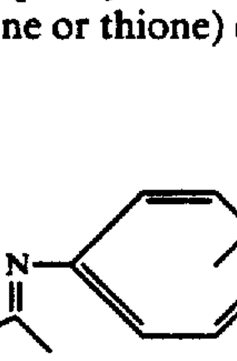
(wherein R_2 is hydrogen or alkoxy), X is oxygen or sulfur],



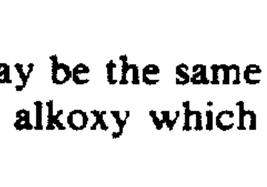
(wherein R_3 is alkyl, alkenyl or alkynyl, and m is 0 or 2),



[wherein R_4 is hydrogen or alkyl, and R_5 is hydrogen, alkyl, alkoxyalkyl, tetrahydrofurfuryl, alkoxyalkyloxyalkyl, alkoxycarbonylalkyl, cycloalkyl or —N=C(CH-3)—R_6 (wherein R_6 is alkyl or phenyl)], —NHR_7 (wherein R_7 is alkylcarbonyl or alkoxycarbonylalkyl),



(wherein R_8 is hydrogen or alkyl, and R_9 is alkoxy, cycloalkyloxy or 1-pyrrolidinyl), or



(wherein X is as defined above); n is an integer of from 0 to 3; and X is oxygen or sulfur.

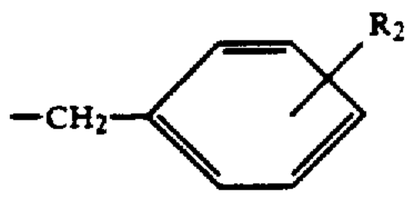
The present invention also provides a process for producing a 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) derivative having the formula:

(IV)

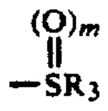
wherein Z which may be the same or different, represents halogen, alkyl, alkoxy which may be substituted

by halogen, alkenyloxy which may be substituted by halogen, alkynyloxy, phenoxy, cycloalkyloxy, alkoxycarbonylalkyloxy, alkylthiocarbonylalkyloxy, alkynyloxycarbonylalkyloxy, benzyloxycarbonylalkyloxy, trifluoromethyl, benzyloxy which may be substituted by chlorine or alkyl, alkenyl, cyanoalkyl, alkylcarbamoyloxy, benzyl which may be substituted by one or two alkyl, alkoxyalkyl, alkynyloxyalkyl, cycloalkylmethyloxy, alkoxyalkyloxy, phenethyloxy, cycloalkyloxycarbonylalkyloxy, pyrrolidinocarbonyl, phenylcarbonyl which may be substituted by alkyl,

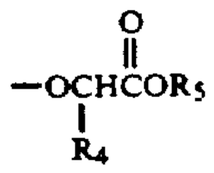
[wherein R_1 is hydrogen, alkyl, phenyl, cycloalkyl, alkoxyalkyl, alkoxycarbonylalkyl or



(wherein R_2 is hydrogen or alkoxy), X is oxygen or sulfur],

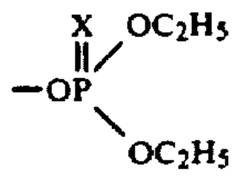


(wherein R_3 is alkyl, alkenyl or alkynyl, and m is 0 or 2),

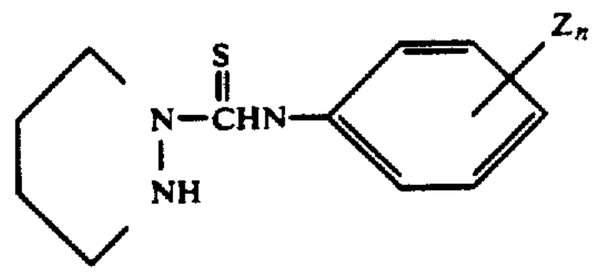


[wherein R_4 is hydrogen or alkyl, and R_5 is hydrogen, alkyl, alkoxyalkyl, tetrahydrofurfuryl, alkoxyalkyloxyalkyl, alkoxycarbonylalkyl, cycloalkyl or —N=C(CH_3)—R_6 (wherein R_6 is alkyl or phenyl)], —NHR_7 (wherein R_7 is alkylcarbonyl),

(wherein R_8 is hydrogen or alkyl, and R_9 is alkoxy, cycloalkyloxy or 1-pyrrolidinyl), or



(wherein X is as defined above); n is an integer of from 0 to 3; and X is oxygen or sulfur, which comprises reacting a compound of the formula:



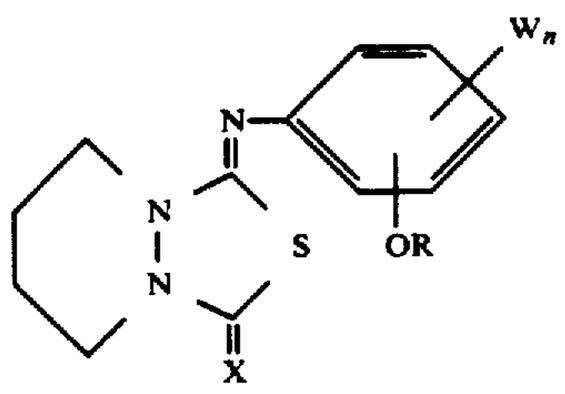
(II)

wherein Z and n are as defined above, with a compound of the formula:

$CXCl_2$

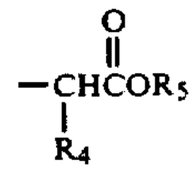
wherein X is as defined above.

Further, the present invention provides a process for producing a 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) derivative having the formula:

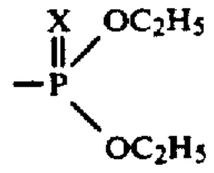


(V)

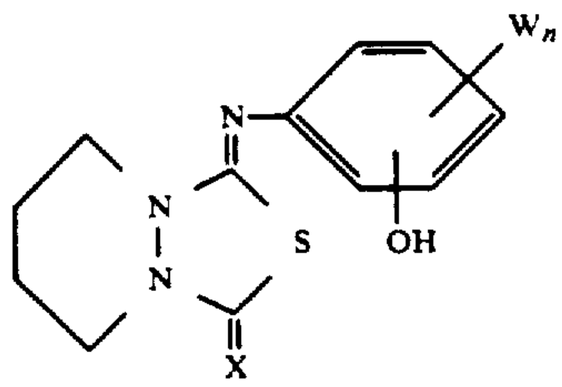
wherein W which may be the same or different, represents halogen; R is alkyl which may be substituted by halogen, alkenyl which may be substituted by halogen, alkynyl, alkoxycarbonylalkyl, alkoxycarbonylalkenyloxy, alkylthiocarbonylalkyl, alkynyloxycarbonylalkyl, benzyloxycarbonylalkyl, benzyl which may be substituted by chlorine or alkyl, alkylcarbamoyl, cycloalkylmethyl which may be substituted by halogen, phenethyl,



[wherein R_4 is hydrogen or alkyl, and R_5 is alkyl, alkoxyalkyl, tetrahydrofurfuryl, alkoxyalkyloxyalkyl, alkoxycarbonylalkyl, cycloalkyl or —N=C(CH_3)—R_6 (wherein R_6 is alkyl or phenyl)], or



(wherein X is oxygen or sulfur); n is an integer of from 0 to 3; and X is oxygen or sulfur, which comprises reacting a compound of the formula:



(VI)

wherein W, X and n are as defined above, with a compound of the formula RT wherein R is as defined above, and T is halogen.

Furthermore, the present invention provides a herbicidal composition comprising a herbicidally effective amount of a compound of the formula I as defined above and a carrier.

Now, the present invention will be described in detail with reference to the preferred embodiments.

In the accompanying drawings:

FIG. **1** is the infrared absorption spectrum of Compound No. 3.

FIG. **2** is the infrared absorption spectrum of Compound No. 6.

FIG. **3** is the infrared absorption spectrum of Compound No. 7.

FIG. **4** is the infrared absorption spectrum of Compound No. 8.

FIG. **5** is the infrared absorption spectrum of Compound No. 9.

FIG. **6** is the infrared absorption spectrum of Compound No. 16.

FIG. **7** is the infrared absorption spectrum of Compound No. 24.

FIG. **8** is the infrared absorption spectrum of Compound No. 25.

FIG. **9** is the infrared absorption spectrum of Compound No. 28.

FIG. **10** is the infrared absorption spectrum of Compound No. 33.

FIG. **11** is the infrared absorption spectrum of Compound No. 36.

FIG. **12** is the infrared absorption spectrum of Compound No. 39.

FIG. **13** is the infrared absorption spectrum of Compound No. 48.

FIG. **14** is the infrared absorption spectrum of Compound No. 49.

FIG. **15** is the infrared absorption spectrum of Compound No. 50.

FIG. **16** is the infrared absorption spectrum of Compound No. 51.

FIG. **17** is the infrared absorption spectrum of Compound No. 52.

FIG. **18** is the infrared absorption spectrum of Compound No. 55.

FIG. **19** is the infrared absorption spectrum of Compound No. 56.

FIG. **20** is the infrared absorption spectrum of Compound No. 59.

FIG. **21** is the infrared absorption spectrum of Compound No. 75.

FIG. **22** is the infrared absorption spectrum of Compound No. 84.

FIG. **23** is the infrared absorption spectrum of Compound No. 86.

FIG. **24** is the infrared absorption spectrum of Compound No. 96.

FIG. **25** is the infrared absorption spectrum of Compound No. 102.

FIG. **26** is the infrared absorption spectrum of Compound No. 144.

FIG. **27** is the infrared absorption spectrum of Compound No. 147.

FIG. **28** is the infrared absorption spectrum of Compound No. 148.

FIG. **29** is the infrared absorption spectrum of Compound No. 149.

FIG. **30** is the infrared absorption spectrum of Compound No. 150.

FIG. **31** is the infrared absorption spectrum of Compound No. 151.

FIG. **32** is the infrared absorption spectrum of Compound No. 152.

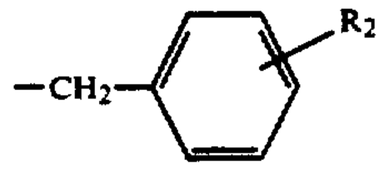
FIG. **33** is the infrared absorption spectrum of Compound No. 153.

FIG. **34** is the infrared absorption spectrum of Compound No. 155.

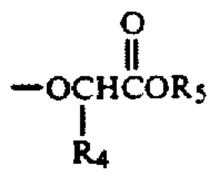
FIG. **35** is the infrared absorption spectrum of Compound No. 157.

In the formula I, Y is preferably halogen, alkoxy which may be substituted by halogen, alkenyloxy which may be substituted by halogen, alkynyloxy, phenoxy, benzyloxy which may be substituted by chlorine or alkyl,

[wherein R_1 is hydrogen, alkyl, phenyl, cycloalkyl, alkoxyalkyl, alkoxycarbonylalkyl or

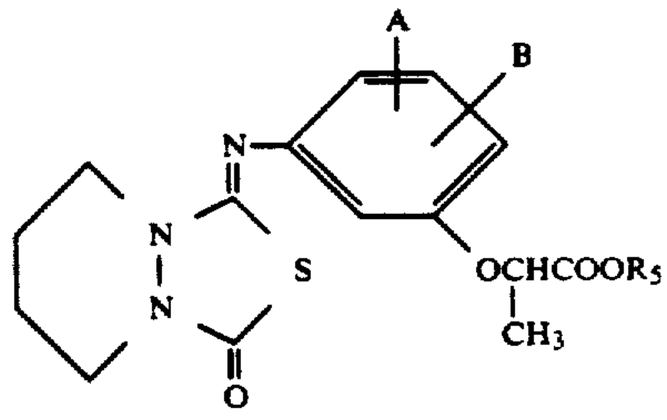


(wherein R_2 is hydrogen or alkoxy), X is oxygen or sulfur],

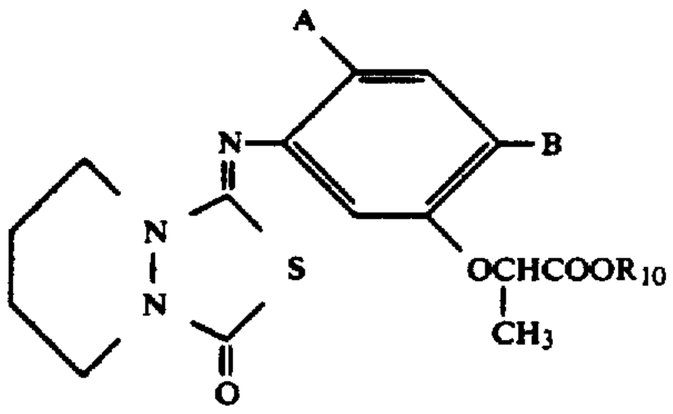


[wherein R_4 is hydrogen or alkyl, and R_5 is hydrogen, alkyl, alkynyl, benzyl, alkoxyalkyl, tetrahydrofurfuryl, alkoxyalkyloxyalkyl, alkoxycarbonylalkyl, cycloalkyl or $-N=C(CH_3)R_6$ (wherein R_6 is alkyl or phenyl)].

A compound having the formula:

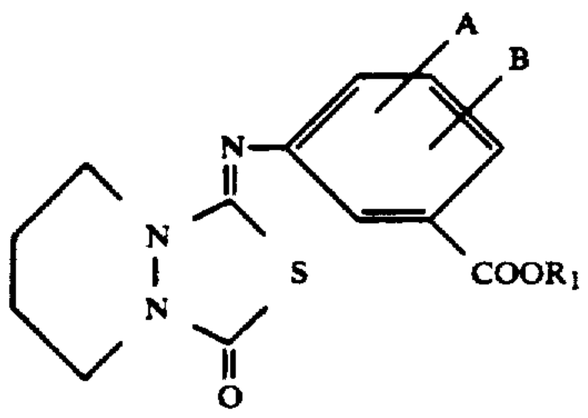


wherein A is hydrogen or halogen, B is halogen, and R_5 is hydrogen, alkyl, alkynyl, benzyl, alkoxyalkyl, tetrahydrofurfuryl, alkoxyalkyloxyalkyl, alkoxycarbonylalkyl, cycloalkyl, or $-N=C(CH_3)R_6$ (wherein R_6 is alkyl or phenyl), is effective particularly as a herbicide for a soybean field. Particularly preferred in this respect is a compound of the formula:

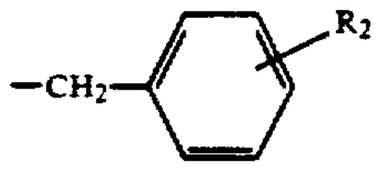


wherein A is hydrogen or halogen, B is halogen, and R_{10} is hydrogen, alkyl, alkynyl, alkoxyalkyl, alkoxyalkyloxyalkyl, or tetrahydrofurfuryl.

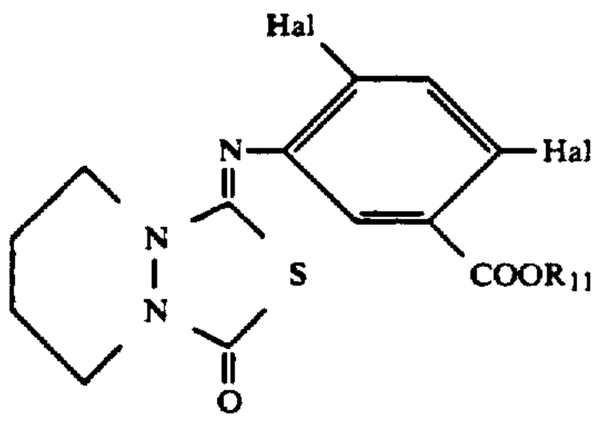
As a herbicide for a non-agricultural field, a compound of the formula:



wherein A is hydrogen or halogen, B is halogen, and R_1 is hydrogen, alkyl, phenyl, cycloalkyl, alkoxyalkyl, alkoxycarbonylalkyl or



(wherein R_2 is hydrogen or alkoxy), is particularly useful. Particularly preferred in this respect is a compound having the formula:



wherein Hal is halogen, and R_{11} is hydrogen or alkyl.

Typical examples of the compound of the formula I are presented in Table 1.

TABLE 1

Compound No.	X	Y_n
1	O	H
2	O	2-CH_3
3	O	2-OCH_3
4	O	2-F
5	O	2-Cl
6	O	3-CH_3
7	O	3-Cl
8	O	3-CF_3
9	O	4-CH_3
10	O	4-OCH_3

TABLE 1-continued

Compound No.	X	Y_n
11	O	4-F
12	O	4-Cl
13	O	4-Br
14	O	4-I
15	O	4-OCH_2—C_6H_4—Cl (para)
16	O	2-F, 4-Cl
17	O	2-F, 4-Br
18	O	3-OCH_3, 4-Cl
19	O	3-OC_2H_5, 4-Cl
20	O	3-OC_3H_7-n, 4-Cl
21	O	3-OC_3H_7-i, 4-Cl
22	O	3-OCH_2CH=CH_2, 4-Cl
23	O	3-OCH_2C≡CH, 4-Cl
24	O	3-OC_4H_9-s, 4-Cl
25	O	3-Cyclopentoxy, 4-Cl
26	O	3-O—C_6H_5, 4-Cl
27	O	3-OCH_2—C_6H_5, 4-Cl
28	O	3-$OCH(CH_3)CO_2$—C_2H_5, 4-Cl
29	O	3-$OCH_2CO_2C_2H_5$, 4-Cl
30	O	3,4-Cl_2
31	O	3-OCH_3, 4-Br
32	O	3-OCH_2C≡CH, 4-Br
33	O	2-F, 4-Cl, 5-OC_3H_7-i
34	O	2-F, 4-Cl, 5-OCH_2CH=CH_2
35	O	2-F, 4-Cl, 5-OCH_2C≡CH
36	O	2-F, 4-Cl, 5-$OCH(CH_3)CO_2$—C_2H_5
37	O	2-F, 4-Cl, 5-OCH_2—C_6H_5
38	O	2-F, 4-Cl, 5-O—C_6H_5
39	O	2,4-Cl_2, 5-OC_3H_7-i
40	O	2,4-Cl_2, 5-O—C_6H_5
41	S	4-Cl
42	S	3-OC_3H_7-i, 4-Cl
43	S	2-F, 4-Cl, 5-OC_3H_7-i
44	S	2-F, 4-Cl, 5-O—C_6H_5
45	S	2,4-Cl_2, 5-OC_3H_7-i
46	O	3-OH, 4-Cl
47	O	2-F, 4-Cl, 5-$OCH_2CO_2C_2H_5$
48	O	3-OC_4H_9-n, 4-Cl
49	O	2-F, 4-Cl, 5-OC_4H_9-s
50	O	3-$OCH(CH_3)CO_2$—CH_2C≡CH, 4-Cl
51	O	3-OC_5H_{11}-n, 4-Cl
52	O	3-OC_5H_{11}-s, 4-Cl
53	O	3-OC_4H_9-i, 4-Cl
54	O	4-CF_3
55	O	3-OC_8H_{17}-n, 4-Cl
56	O	3-$OCH(CH_3)CO_2$—CH_2—C_6H_5, 4-Cl
57	O	3-OCH_2CH=CH—CH_3, 4-Cl
58	O	3-OC_3H_7-i, 4-Br
59	O	3-$OCH(CH_3)CO_2$—C_4H_9-n, 4-Cl
60	O	3-OC_2H_4CH=CH_2, 4-Cl
61	O	3-$OCH_2C(CH_3)$=CH_2, 4-Cl
62	O	3-OCH_2—C_6H_4—Cl (para), 4-Cl
63	O	3-OC_4H_9-i, 4-Br
64	O	3-OCH_2CH=CH_2, 4-Br
65	O	3-OCH_2—C_6H_4—Cl (ortho), 4-Cl
66	O	3-OCH_2—C_6H_4—CH_3 (para), 4-Cl
67	O	2-F, 4-Cl, 5-CH_2OCH_3
68	O	3-OCH_2CH=$C(CH_3)_2$, 4-Cl
69	O	3-$OCH(CH_3)COS$—C_2H_5, 4-Cl
70	O	3-$OCH(CH_3)CO_2$—C_4H_9-i, 4-Cl
71	O	3-$OCH(CH_3)CO_2C_2H_4OCH_3$, 4-Cl
72	O	3-$NHCH(CH_3)CO_2C_2H_5$, 4-Cl
73	O	3-OC_2H_4Cl, 4-Cl
74	O	3-OCH_2—C_6H_4—Cl (metha), 4-Cl
75	O	3-$OCH(CH_3)$—C_6H_5, 4-Cl
76	O	3-$CH_2OC_2H_5$, 4-Cl
77	O	3-Cyclohexyloxy, 4-Cl
78	O	3-Cyclohexyimethyloxy, 4-Cl
79	O	3-OC_2H_4C≡CH, 4-Cl
80	O	3-(1-Cyclohexyloxycarbonylethoxy), 4-Cl
81	O	3-$OCH(CH_3)CH$=CH_2, 4-Cl
82	O	2-F, 4-Cl, 5-OCH_2—C_6H_4—Cl (para)
83	O	2-Br, 4-Cl, 5-$OCH_2CO_2C_2H_5$
84	O	3-$OCH(CH_3)CO_2N$=$C(CH_3)_2$, 4-Cl
85	O	3-$OPO(OC_2H_5)_2$, 4-Cl
86	O	2,4-Cl_2, 5-$OCH(CH_3)CO_2C_2H_5$
87	O	3-CH_2OCH_2C≡CH, 4-Cl
88	O	3-$OCONHC_2H_5$, 4-Cl
89	O	3-$NHCOC_2H_5$, 4-Cl

TABLE 1-continued

Compound No.	X	Y_n
90	O	3-OCONHCH$_3$, 4-Cl
91	O	3,5-Cl$_2$
92	O	3-OCH(CH$_3$)CO$_2$—N=C(CH$_3$)C$_6$H$_5$, 4-Cl
93	O	3-CH$_2$—C$_6$H$_5$, 4-Cl
94	O	3-OCH$_2$CH=CHCl, 4-Cl
95	O	3-CO$_2$C$_2$H$_5$, 4-Cl
96	O	3-COC$_6$H$_5$, 4-Cl
97	O	3-CO$_2$—C$_3$H$_7$-i, 4-Cl
98	O	3-CO$_2$—C$_4$H$_9$-n, 4-Cl
99	O	3-C$_3$H$_7$-n, 4-Cl
100	O	3-CH$_2$—C$_6$H$_4$—CH$_3$ (para), 4-Cl
101	O	3-CH$_2$—C$_6$H$_3$—(CH$_3$)$_2$(2,5), 4-Cl
102	O	3-CO—C$_6$H$_4$—CH$_3$ (para), 4-Cl
103	O	3-CH=CHCH$_3$, 4-Cl
104	O	3-SC$_2$H$_5$, 4-Cl
105	O	3-SO$_2$C$_2$H$_5$, 4-Cl
106	O	3-Pyrrolidinocarbonyl, 4-Cl
107	O	3-CO$_2$—CH(CH$_3$)CO$_2$C$_2$H$_5$, 4-Cl
108	O	3-CO$_2$—C$_2$H$_4$OCH$_3$, 4-Cl
109	O	3-SCH$_2$CH=CH$_2$, 4-Cl
110	O	3-SCH$_2$C≡CH, 4-Cl
111	O	3-SO$_2$CH$_2$CH=CH$_2$, 4-Cl
112	O	2-F, 4-Cl, 5-CO$_2$C$_2$H$_5$
113	O	3-Cyclopentoxycarbonyl, 4-Cl
114	O	3-COSC$_2$H$_5$, 4-Cl
115	O	3-CH$_2$CN, 4-Cl
116	O	3-CO$_2$—C$_6$H$_5$, 4-Cl
117	O	3-CO$_2$—CH$_2$—C$_6$H$_5$, 4-Cl
118	O	3-OCH$_2$CH=CCl$_2$, 4-Cl
119	O	2-F, 4-Cl, 5-CO$_2$—CH$_2$C$_6$H$_4$—OCH$_3$ (para)
120	O	2-F, 4-Cl, 5-CO$_2$H
121	O	2-F, 4-Cl, 5-CO$_2$—C$_3$H$_7$-i
122	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$CH$_3$
123	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_3$H$_7$-i
124	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_4$H$_9$-i
125	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—CH$_2$C≡CH
126	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—N=C=(CH$_3$)$_2$
127	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—N=C.(CH$_3$)—C$_6$H$_5$
128	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_2$H$_4$OCH$_3$
129	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_2$H$_4$—OC$_2$H$_5$
130	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_2$H$_4$—OC$_3$H$_7$-i
131	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_2$H$_4$—OC$_4$H$_9$-n
132	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_2$H$_4$—OC$_2$H$_4$OCH$_3$
133	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—CH.(CH$_3$)CH$_2$OCH$_3$
134	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—C$_3$H$_6$—OC$_2$H$_5$
135	O	2-F, 4-Cl, 5-(1-Tetrahydrofurfuryloxy-carbonylethoxy)
136	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—CH$_2$—CO$_2$C$_2$H$_5$
137	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—CH.(CH$_3$)CO$_2$CH$_3$
138	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$—CH.(CH$_3$)CO$_2$C$_2$H$_5$
139	O	3-OPS(OC$_2$H$_5$)$_2$, 4-Cl
140	O	3-OC$_2$H$_4$C$_6$H$_5$, 4-Cl
141	O	3-OC$_2$H$_4$OCH$_3$, 4-Cl
142	O	3-CH$_3$, 4-Cl
143	O	3-OCH$_2$CCl=CH$_2$, 4-Cl
144	O	3-(2,2-Dichlorocyclopropylmethoxy), 4-Cl
145	O	3-SCHCO$_2$C$_2$H$_5$, 4-Cl (CH$_3$)
146	O	3-SCHCO$_2$—(H), 4-Cl (CH$_3$)
147	O	2-F, 4-Cl, 5-SCH(CH$_3$)CO$_2$C$_2$H$_5$
148	O	2-F, 4-Cl, 5-SCH$_2$CO$_2$—(H)
149	O	2-F, 4-Cl, 5-SCH(CH$_3$)CO$_2$C$_2$H$_5$
150	O	2-F, 4-Cl, 5-SCH(CH$_3$)CO$_2$—(H)
151	O	2-F, 4-Cl, 5-SCH(C$_2$H$_5$)CO$_2$C$_2$H$_5$
152	O	2-F, 4-Cl, 5-SCH(C$_2$H$_5$)CO$_2$—(H)
153	O	2-F, 4-Cl, 5-SCH(C$_3$H$_7$)CO$_2$C$_2$H$_5$
154	O	2-F, 4-Cl, 5-SCH(CH$_3$)CON(C$_4$H$_8$)
155	O	2-F, 4-Cl, 5-SCH(C$_2$H$_5$)CON(C$_4$H$_8$)
156	O	2-F, 4-Cl, 5-OCH(CH$_3$)CO$_2$H
157	O	2-F, 4-Cl, 5-OCH(CH$_3$)CH=CHCO$_2$C$_2$H$_5$
158	O	2-F, 4-Cl, 5-CO$_2$CH$_3$
159	O	2-F, 4-Cl, 5-CO$_2$C$_3$H$_7$-n
160	O	2-F, 4-Cl, 5-CO$_2$C$_4$H$_9$-n

The melting points and refractive indexes of the Compound Nos. 1 to 157 are shown in Table 2.

TABLE 2

Compound No.	Melting point (°C.)	Refractive index n_D^{20}
1	78–82	
2	74–78	
3		1.6124
4	83–85	
5	73–75	
6		1.6208
7		1.6368
8		1.5641
9		1.6256
10	116–119	
11	69–72	
12	82–85	
13	69–70	
14	80–82	
15	150–154	
16		1.6121
17	98–100	
18	125–128	
19	143–145	
20	102–105	
21	66–69	
22	119–122	
23	130–132	
24		1.5794
25		1.6145
26	83–85	
27	114–116	
28		1.5840
29	94–96	
30	116–118	
31	118–121	
32	132–135	
33		Not measurable
34	106–109	
35	132–134	
36		1.5666
37	99–101	

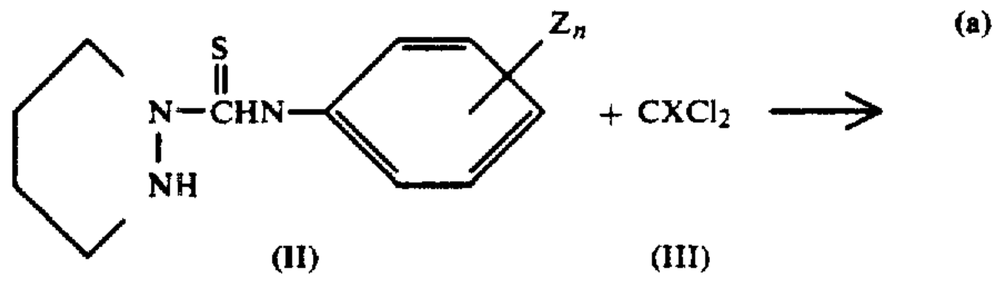
TABLE 2-continued

Compound No.	Melting point (°C.)	Refractive index n_D^{20}
38	116–119	
39		1.6050
40	111–114	
41	96–97	
42	98–101	
43	77–80	
44	138–141	
45	71–73	
46	130–132	
47	93–96	
48		1.6050
49		Not measurable
50		1.5915
51		1.5951
52		1.5878
53	72–75	
54	103–107	
55		1.5673
56		1.5961
57	83–86	
58	84–88	
59		1.5732
60	77–78	
61	89–95	
62	94–96	
63	92–95	
64	116–119	
65	100–103	
66	162–164	
67	84–87	
68	78–81	
69		1.5962
70		1.5661
71		1.5710
72		1.5755
73	110–114	
74	120–122	
75		Not measurable
76	73–76	
77		1.6043
78	97–103	
79	85–88	
80		1.5771
81		1.5970
82	129–132	
83	116–118	
84		Not measurable
85		1.5759
86		Not measurable
87		1.6946
88	143–146	
89	117–120	
90	167–170	
91	111–114	
92	44–46	
93	154–156	
94	87–90	
95		1.6043
96		Not measurable
97		1.5843
98		1.5775
99		1.5678
100		1.6255
101	127–129	
102		Not measurable
103		1.6320
104	120–122	
105		1.6213
106	119–121	
107		1.5881
108		1.5936
109	66–68	
110		1.6641
111		1.6225
112		1.6002
113		1.5979
114		1.6321
115	120–122	
116		1.6297
117		1.6024

TABLE 2-continued

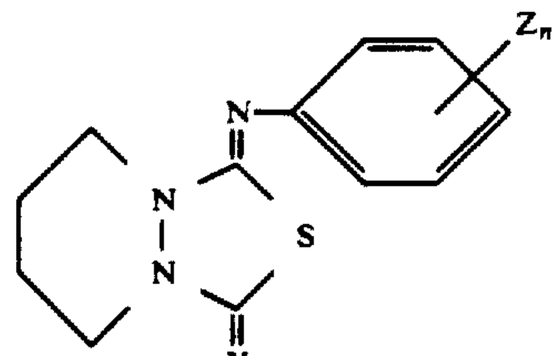
Compound No.	Melting point (°C.)	Refractive index n_D^{20}
118		1.6058
119		1.6052
120	223–226	
121		1.5734
122	106–109	
123		1.5549
124	69–72	
125		1.5735
126	116–119	
127	52–55	
128		1.5749
129		1.5678
130		1.5590
131		1.5575
132		1.5672
133		1.5580
134		1.5691
135		1.5751
136		1.5690
137		1.5647
138		1.5538
139		1.5995
140	115–118	
141	68–71	
142	82–84	
143	77–79	
144		Not measurable
145		1.5969
146		1.5972
147		1.5990
148		Not measurable
149		1.5925
150		1.602
151		1.5738
152		1.5963
153		1.5761
154	136–138	
155		Not measurable
156	120–122	
157		Not measurable
158	79–81	
159	57–59	
160		1.5791

The compound of the formula I may be prepared by the following processes.



(a)

(II) (III)



X

(IV)

wherein X, Z and n are as defined above. This process can be conducted by reacting the compound of the formula II with the compound of the formula III in the presence of a base.

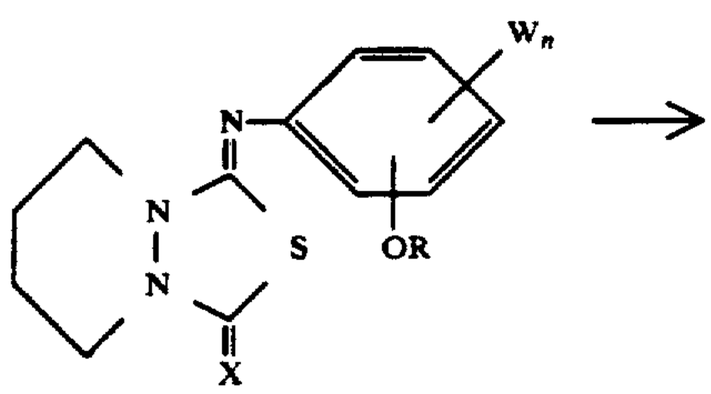
As the base, there may be mentioned an aliphatic tertiary amine such as triethylamine or trimethylamine; an aromatic tertiary amine such as pyridine, picoline or

quinoline; or an inorganic base such as sodium hydroxide, potassium hydroxide, potassium carbonate or sodium carbonate.

The above reaction is preferably conducted in a solvent. As such a solvent, there may be mentioned a chlorine-containing hydrocarbon such as dichloromethane, chloroform or carbon tetrachloride; an ether such as diethyl ether, tetrahydrofuran or dioxane; a hydrocarbon such as n-hexane, benzene or toluene; an aliphatic ketone such as acetone or methyl ethyl ketone; dimethylsulfoxide; or N,N-dimethylformamide.

The above-mentioned condensation reaction can be completed in from 1 to 7 hours at a temperature within a range of from −20° C. to the boiling point of the solvent.

(b)



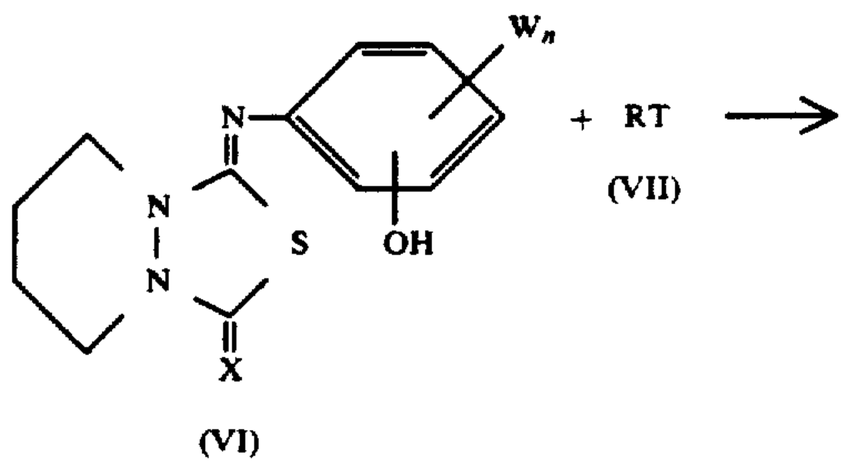
(V)

(VI)

wherein X, W, R and n are as defined above.

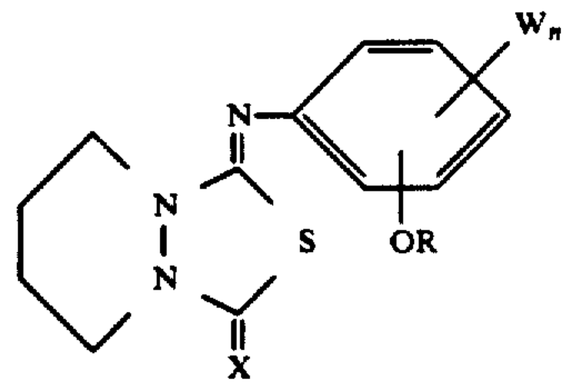
This process can be conducted by reacting the compound of the formula V with an acid such as hydrobromic acid or aluminum chloride in the presence of a solvent, for instance, a fluorine-containing hydrocarbon such as chloroform or carbon tetrachloride, or a hydrocarbon such as benzene or toluene. This reaction can be completed in from 1 to 7 hours at a temperature within a range of from 0° C. to the boiling point of the solvent.

(c)



(VI)

-continued



(V)

wherein W, X, R, T and n are as defined above.

This process can be conducted by reacting the compound of the formula VI with the compound of the formula VII in the presence of the same base and solvent as used in process (a) at a temperature within a range of from −20° C. to the boiling point of the solvent for from 1 to 7 hours.

Now, the present invention will be described in further detail with reference to Examples. However, it should be understood that the present invention is by no means restricted by these specific Examples.

Firstly, Examples for the preparation of the compounds of the present invention will be described.

Preparation Example 1

9-(4-Chlorophenylimino)-8-thia-1,6-diazabicyclo[4.3.0-]nonane-7-one (Compound No. 12)

In a reaction flask, 3.2 g (13 mmol) of 1,2-tetramethylene-1-(4-chlorophenylthiocarbamoyl)-hydrazine, 2.2 g (28 mmol) of pyridine ad 20 ml of dichloromethane, were charged, and a dichloromethane solution containing 1.5 g (15 mmol) of phosgene was dropwise added while cooling the mixture with ice water. After the dropwise addition, the mixture was stirred at room temperature for 3 hours to complete the reaction. The reaction solution was washed with water, and dried over anhydrous sodium sulfate, and then the solvent was distilled off to obtain a crude product. This crude product was recrystallized from isopropyl ether to obtain 3.2 g (yield: 74%) of white crystals. Melting point: 82°–85° C.

Preparation Example 2

9-(4-Chlorophenylimino)-8-thia-1,6-diazabicyclo[4.3.0-]nonane-7-thione (Compound No. 41)

Into a reaction flask, 2.4 g (9 mmol) of 1,2-tetramethylene-1-(4-chlorophenylthiocarbamoyl)-hydrazine, 2.0 g (25 mmol) of pyridine and 20 ml of dichloromethane, were charged, and 1.3 g (11 mmol) of thiophosgene was dropwise added while cooling the mixture with ice water. After the dropwise addition, the mixture was stirred at room temperature for 3 hours to complete the reaction. After the completion of the reaction, the reaction solution was washed with water, and dried over anhydrous sodium sulfate, and the solvent was distilled off to obtain a crude product. This crude product was recrystallized from isopropyl ether to obtain 2.1 g (yield: 75%) of brown crystals. Melting point: 96°–97° C.

Preparation Example 3

9-(4-Chloro-3-hydroxyphenylimino)8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one (Compound No. 46)

Into a reaction flask equipped with a Dimroth condenser, 1.7 g (5 mmol) of 9-(4-chloro-3-isopropoxyphenylimino)-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one, 1.3 g (10 mmol) of aluminum chloride and 50 ml of chloroform, were charged, and refluxed under heating for 2 hours to complete the reaction.

The reaction solution was poured into ice water, and the organic layer was washed with water and dried over anhydrous sodium sulfate, and the solvent was distilled off to obtain a crude product.

The crude product was purified by column chromatography to obtain 1.2 g (yield: 80%) of colorless crystals. Melting points: 130°–132° C.

Preparation Example 4

9-(4-Chloro-2-fluoro-3-propargyloxyphenylimino)-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one (Compound No. 35)

Into a reaction flask equipped with a Dimroth condenser, 2.2 g (7 mmol) of 9-(4-chloro-2-fluoro-3-hydroxyphenylimino)-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one, 1.1 g (9 mmol) of propargyl bromide, 1.3 g (9 mmol) of potassium carbonate and 20 ml of acetonitrile, were charged, and refluxed under heating for 3 hours to complete the reaction.

The reaction solution was filtered, concentrated, extracted with ethyl acetate, and then washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off to obtain a crude product, which was then recrystallized from isopropyl ether to obtain 1.6 g (yield: 64%) of white crystals. Melting point: 132°–134° C.

Preparation Example 5

9-(4-Chloro-2-fluoro-5-ethoxycarbonylphenylimino)-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one (Compound No. 112)

Into a reaction flask, 3.8 g (11 mmol) of 1,2-tetramethylene-1-(4-chloro-2-fluoro-5-ethoxycarbonylphenylthiocarbamoyl)hydrazine, 2.1 g (26 mmol) of pyridine and 20 ml of dichloromethane, were charged, and a dichloromethane solution containaing 1.3 g (13 mmol) of phosgene, was dropwise added while cooling the mixture with ice water. After the dropwise addition, the mixture was stirred at room temperature for 1 hour to complete the reaction. The reaction solution was washed with water and dried over anhydrous sodium sulfate, and then the solvent was distilled off to obtain a crude product. This crude product was purified by column chromatography to obtain 2.8 g (yield: 68%) of colorless sticky substance. Refractive index: n_D^{20} 1.6002.

Preparation Example 6

9-{4-Chloro-2-fluoro-5-(1-methoxyethoxycarbonylethoxy)phenylimino}-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one (Compound No. 128)

Into a reaction flask equipped with a Dimroth condenser, 1.9 g (6 mmol) of 9-(4-chloro-2-fluoro-3-hydroxyphenylimino)-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one, 1.3 g (6 mmol) of methoxyethyl 2-bromopropionate, 1.0 g (6 mmol) of potassium carbonate and 20 ml of acetonitrile, were charged, and refluxed under heating for 40 minutes to complete the reaction. The reaction solution was filtered, concentrated, extracted with ethyl ether, and then washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off to obtian a crude product, which was purified by column chromatography to obtain 1.2 g (yield: 46.1%) of slightly brown liquid. Refractive index: n_D^{20} 1.5749

Preparation Example 7

9-{4-Chloro-3-[1-(ethoxycarbonyl)ethylamino]-phenylimino}-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one (Compound No. 72)

Into a reaction flask equipped with a Dimroth condenser, 2.2 g (7.4 mmol) of 9-(4-chloro-3-aminophenylimino)-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-one, 15 ml of ethyl 2-bromopropionate and 2.0 g (24 mmol) of sodium hydrogen carbonate, were charged, and refluxed under heating for 4 hours to complete the reaction. The reaction solution was filtered, concentrated and extracted with ethyl acetate, and then washed with water and dried over anhydrous sodium sulfate. The solvent was distilled off to obtain a crude product, which was purified by column chromatography to obtain 1.5 g (yield: 52%) of colorless oily substance. Refractive index: n_D^{20} 1.5755

The 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) derivative of the formula I is useful as an active ingredient for a herbicide. When the compound of the formula I of the present invention is used as a herbicide for a paddy rice field, an upland field, an orchard or a non-agricultural field, the active ingredient can be used in a suitable formulation depending upon the particular purpose. Usually, the active ingredient is diluted with an inert liquid or solid carrier, and used in the form of a formulation such as a dust, a wettable powder, an emulsifiable concentrate, a granule, etc., if necessary by adding a surfactant and other additives. Further, the compound of the present invention may be used in combination with an insecticide, a nematocide, a fungicide, other herbicides, a plant growth controlling agent, a fertilizer, etc., as the case requires.

Now, the formulations will be described in detail with reference to typical Formulation Examples. In the following Formulation Examples, "parts" means "parts by weight".

Formulation Example 1

Wettable power

10.0 parts of Compound No. 43, 0.5 part of Emulgen (trademark of Kao Soap Co., Ltd.) 810, 0.5 part of Demol trademark of Kao Soap Co., Ltd.) N, 20.0 parts of Kunilite (trademark of Kunimine Kogyo K.K.) 201, and 69.0 parts of Zeeklite (trademark of Zeeklite Co., Ltd.) CA, were mixed and pulverized to obtain a wettable powder containing 10% of an active ingredient.

Formulation Example 2

Wettable powder

10.0 parts of Compound No. 33, 0.5 part of Emulgen 810, 0.5 part of Demol N, 20.0 parts of Kunilite 201, 5.0 parts of Carplex 80 and 64.0 parts of Zeeklite CA, were mixed and pulverized to obtain a wettable powder containing 10% of the active ingredient.

Formulation Example 3

Emulsifiable concentrate

To 30 parts of Compound No. 19, 60 parts of a mixture of xylene and isophorone in equal amounts and 10 parts of surfactant Sorpol (trademark of Toho Kagaku Kogyo K.K.) 800A, were added, and the mixture was thoroughly mixed to obtain 100 parts of an emulsifiable concentrate.

Formulation Example 4

Granules

10 parts of water was added to 10 parts of Compound No. 21, 80 parts of a filler obtained by mixing talc and bentonite in a ratio of 1:3, 5 parts of white carbon and 5 parts of surfactant Sorpol N 800A, and the mixture was thoroughly kneaded to obtain a paste, which was extruded from sieve openings having a diameter of 0.7 mm and dried, and then cut into a length of from 0.5 to 1 mm, to obtain 100 parts of granules.

The compounds of the formula of the present invention exhibit excellent herbicidal effects at a very low dose in a wide range from the germination stage to the growing stage of annual weeds such as barnyardgrass (Echinochloa crus-galli), umbrella-plant (Cyperus difformis L.), monochoria (Monochoria vaginalis Presl), spike-flowered rotala (Rotalal indica Koehne), false pimpernel (Lindernia procumbens Philcox) and Dopatrium junceum Hamilt, and perennial weeds such as bulrush (Scirpus juncoides Roxb.), slender spikerush (Eleocharis acicularis Roem. et Schult.), water plantain (Alisma canaliculatum A. Br. et Bouche), Sagittaria (Sagittaria pygmaea Miq.) and cyperus sp. (Cyperus serotinus Rottb.) which grow in paddy fields. At the same time, they have high selectivity for paddy field rice. Further, they exhibit high herbicidal effects, by soil treatment or by foliage treatment, against various weeds in the upland fields, for example, broad leaf weeds such as smart weed (Polygonum nodosum L.), pigweed (Amaranthus retroflexus), lambsquaters (Chenopodium album), common chickweed (Stellaria media), speed well (Veronica persica), wild mustard (Brassica kaber var. pinnatifida) and cocklebur (Xanthium strumarium), cyperaceous weeds such as rice flatsedge (Cyperus iria L.), and gramineous weeds such as barnyardgrass, large crabgrass (Digitaria sanguinalis) and green foxtail (Setaria viridis). At the same time, they have a feature that they are highly safe to crop plants such as upland rice, wheat, soybean and corn.

The dose of the compound of the present invention is usually within a range of from 10 g to 15 kg/ha. More specifically, the dose is usually from 30 g to 5 kg/ha for upland fields, from 10 g to 1 kg/ha for paddy rice fields, and from 200 g to 5 kg/ha for non-agricultural fields.

Further, the compounds of the present invention have excellent residual effects, and show stabilized effects for a long period of time also in paddy fields. They are also useful for orchard, grassland, lawn and non-agricultural fields.

Now, the herbicidal effects of the herbicides of the present invention will be described with reference to Text Examples.

Text Example 1

Herbicidal test by soil treatment of paddy field

Into a 100 cm^2 porcelain pot, paddy field soil was filled and puddled. Then, seeds of barnyardgrass, umbrella plant, monochoria and bulrush were sown, and water was introduced to a depth of 3 cm.

Next day, the wettable powder prepared in accordance with Formulation Example 1, was diluted with water and dropwise applied to the surface of the water. The amount of the active ingredient applied, was 400 g/10a. Then, the pot was left in a green house. Twenty one days after the application, the herbicidal effects were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 4.

TABLE 3

Index	Herbicidal effects and phytotoxicity
5	Withered
4.5	Herbicidal effect (or phytotoxicity) in a range of 90 to 99%
4	Herbicidal effect (or phytotoxicity) in a range of 80 to 89%
3.5	Herbicidal effect (or phytotoxicity) in a range of 70 to 79%
3	Herbicidal effect (or phytotoxicity) in a range of 60 to 69%
2.5	Herbicidal effect (or phytotoxicity) in a range of 50 to 59%
2	Herbicidal effect (or phytotoxicity) in a range of 40 to 49%
1.5	Herbicidal effect (or phytotoxicity) in a range of 30 to 39%
1	Herbicidal effect (or phytotoxicity) in a range of 20 to 29%
0.5	Herbicidal effect (or phytotoxicity) in a range of 1 to 19%
0	No herbicidal effect (or no phytotoxicity)

TABLE 4

Compound No.	Herbicidal effects			
	Barnyard-grass	Umbrella plant	Monochoria	Bulrush
1	4	5	5	4
2	2.5	2	5	3.5
3	4	5	5	2.5
4	4	4	5	4
6	4	5	5	3.5
7	5	5	5	4
8	3	4	5	5
9	5	5	5	5
10	5	5	5	5
11	5	5	5	5
12	5	5	5	5
13	5	5	5	5
14	5	5	5	5
15	4	4	5	5
16	5	5	5	5
17	5	5	5	5
18	5	5	5	5
19	5	5	5	5
20	5	5	5	5
21	5	5	5	5
22	5	5	5	5
23	5	5	5	5
24	5	5	5	5
25	5	5	5	5
26	5	5	5	5
27	5	5	5	5
28	5	5	5	5
29	5	5	5	5
30	5	5	5	5
31	5	5	5	5
32	5	5	5	5
33	5	5	5	5
34	5	5	5	5
35	5	5	5	5
36	5	5	5	5
37	5	5	5	5
38	5	5	5	5
39	5	5	5	5
40	5	5	5	5
41	5	5	5	5

TABLE 4-continued

Compound No.	Herbicidal effects: Barnyard-grass	Umbrella plant	Monochoria	Bulrush
42	5	5	5	5
43	5	5	5	5
44	5	5	5	5
45	5	5	5	5
46	5	5	5	5
47	5	5	5	5
48	5	5	5	5
49	5	5	5	5
50	5	5	5	5
51	5	5	5	5
52	5	5	5	5
53	5	5	5	5
54	5	5	5	5
55	5	5	5	5

In pot A, three germinated tubers of each of flat sedge and sagittaria, were embedded in the surface layer of the soil, and two seedlings of two rice plants of 2.2 leaf stage, were transplanted in a depth of 2 cm.

In pot B, seeds of barnyardgrass, hardstem bulrush, narrow leaf water plantain, monochoria and umbrella plant were sown in the surface layer of the soil.

The day after the seeding and transplantation, a prescribed amount of a wettable powder of each compound formulated in accordance with Formulation Example 1, was diluted with water and dropwise applied by a pipette.

Thirty days after application, the herbicidal effect and phytotoxicity were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 5.

TABLE 5

Compound No.	Dose of active ingredient (g/10 a)	(Part 1) Barnyard-grass	Umbrella plant	Monochoria	Bulrush	(Part 2) Water plantain	Sagittaria	Cyperus sp	Transplanted paddy field rice
13	50	5	5	5	4.5	5	5	5	0.5
	25	4.5	5	5	4	5	5	5	0
18	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	5	5	0
19	12.5	5	5	5	5	5	5	5	0
	6.3	5	5	5	5	5	4.5	5	0
20	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
21	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
22	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
23	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
24	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	4	5	0
26	25	5	5	5	5	5	4	5	1
	12.5	5	5	5	5	5	2	5	0
30	50	5	5	5	5	5	5	5	0.5
	25	5	5	5	5	5	5	5	0
31	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	5	5	0
32	6.3	5	5	5	5	5	5	5	1
	3.2	5	5	5	5	5	5	5	0.5
33	6.3	5	5	5	5	5	5	5	0.5
	3.2	5	5	5	5	5	5	5	0
34	6.3	5	5	5	5	5	5	5	1
	3.2	5	5	5	5	5	5	5	0.5
35	6.3	5	5	5	5	5	5	5	3.5
	3.2	5	5	5	5	5	5	5	3
36	12.5	5	5	5	5	5	5	4	2
	6.3	5	5	5	4	5	5	2	1
37	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
38	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
39	25	5	5	5	5	5	5	5	1
	12.5	5	5	5	3.5	5	5	2	0
42	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
43	12.5	5	5	5	5	5	5	5	0
	6.3	5	5	5	5	5	5	5	0
Ronstar	50	5	5	5	5	5	4.5	5	2
	25	5	5	5	5	5	2.5	5	2
	12.5	5	5	5	5	5	1.5	3	1

Test Example 2

Low dose test in soil treatment of irrigated paddy field

Into a 1/5,000a Wagner pot, paddy field soil was filled and puddled, and water was introduced to a depth of 3 cm.

Text Example 3

The herbicidal test in soil treatment of upland field

Into a 120 cm^2 plastic pot, upland field soil was filled, and seeds of barnyardgrass, large crabgrass, smart weed, pigweed, lambsquaters and rice flatsedge were sown and covered with soil.

A wettable powder of each test compound formulated in accordance with Formulation Example 1, was diluted with water in an amount of 100 liter/10a and uniformly applied to the surface of soil by means of a small size spray at a dose of 400 g/10a of the active ingredient. After the application, the pot was left for 21 days in a green house, and then the herbicidal effects were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 6.

TABLE 6

Compound No.	Herbicidal effects					
	Barnyard-grass	Large crabgrass	Smart weed	Pig-weed	Lambs-quaters	Rice flatsedge
4	4	5	5	5	5	5
11	5	5	5	5	5	5
12	5	5	5	5	5	5
16	5	5	5	5	5	5
17	5	5	5	5	5	5
20	5	5	5	5	5	5
22	4	5	5	5	5	5
23	4	5	5	5	5	5
24	5	5	5	5	5	5
28	5	5	4	5	5	5
31	5	5	5	5	5	5
33	5	5	5	5	5	5
34	5	5	5	5	5	5
35	5	5	5	5	5	5
36	5	5	5	5	5	5

Text Example 4

The herbicidal test in foliage treatment in upland field

Into a 120 cm^2 plastic pot, upland field soil was filled, and seeds of barnyardgrass, large crabgrass, smart weed, pigweed, lambsquater and rice flatsedge, were sown, and grown in a green house until barnyardgrass grew to the 3 leaf stage. When barnyardgrass reached the 3 leaf stage, a wettable powder of each test compound formulated in accordance with Formulation Example 1 was diluted with water in an amount of 100 liter/10 a and applied to the foliage of the plants from above by a small size spray at a dose of 400 g/10 a of the active ingredient. After the application, the pot was left for 21 days in a green house, and then the herbicidal effects were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 7.

TABLE 7

Compound No.	Herbicidal effects					
	Barnyard-grass	Large crabgrass	Smart weed	Pig-weed	Lambs-quaters	Rice flatsedge
7	5	5	5	5	5	5
11	5	5	5	5	5	5
12	5	5	5	5	5	5
13	5	5	5	5	5	5
14	5	5	5	5	5	5
16	5	5	5	5	5	5
17	5	5	5	5	5	5
20	5	5	5	5	5	5
21	4	5	5	5	5	5
22	5	5	5	5	5	5
23	5	5	5	5	5	5
24	5	5	5	5	5	5
28	5	5	5	5	5	5
31	4	5	5	5	5	5
32	4	5	5	5	5	5
33	5	5	5	5	5	5
34	5	5	5	5	5	5
35	5	5	5	5	5	5
36	5	5	5	5	5	5
37	5	5	5	5	5	5
38	4	5	5	5	5	5

TABLE 7-continued

Compound No.	Herbicidal effects					
	Barnyard-grass	Large crabgrass	Smart weed	Pig-weed	Lambs-quaters	Rice flatsedge
39	5	5	5	5	5	5
41	4	5	5	5	5	5
42	4	5	5	5	5	5
43	5	5	5	5	5	5
44	4	5	5	5	5	5

Text Example 5

Herbicidal test in the soil treatment of paddy rice field

Into a 10 cm^2 porcelain pot, paddy field soil was filled and puddled, and seeds of barnyardgrass, umbrella plant, monochoria and bulrush were sown. Then, water was introduced to the depth of 3 cm.

Next day, a wettable powder prepared in accordance with Formulation Example 1, was diluted with water and dropwise applied to the surface of water. The amount of the active ingredient applied was 400 g/10a. The pot was left in a green house, and twenty one days after the application, the herbicidal activities were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 8.

TABLE 8

Compound No.	Herbicidal effects			
	Barnyard-grass	Umbrella plant	Monochoria	Bulrush
56	5	5	5	5
57	5	5	5	5
58	5	5	5	5
59	5	5	5	5
60	5	5	5	5
61	5	5	5	5
62	5	5	5	5
63	5	5	5	5
64	5	5	5	5
65	5	5	5	5
66	5	5	5	5
67	5	5	5	5
68	5	5	5	5
69	5	5	5	5
70	5	5	5	5
71	5	5	5	5
72	5	5	5	5
73	5	5	5	5
74	5	5	5	5
75	5	5	5	5
76	5	5	5	5
77	5	5	5	5
78	5	5	5	5
79	5	5	5	5
80	5	5	5	5
81	5	5	5	5
82	5	5	5	5
84	5	5	5	5
85	5	5	5	5
86	5	5	5	5
87	5	5	5	5
88	5	5	5	5
89	5	5	5	5
90	5	5	5	5
92	5	5	5	5
93	5	5	5	5
94	5	5	5	5
95	5	5	5	5
96	5	5	5	5
97	5	5	5	5
98	5	5	5	5
99	5	5	5	5
100	5	5	5	5
102	5	5	5	5
103	5	5	5	5
104	5	5	5	5

TABLE 8-continued

Compound No.	Herbicidal effects			
	Barnyard-grass	Umbrella plant	Monochoria	Bulrush
105	5	5	5	5
106	5	5	5	5
107	5	5	5	5
108	5	5	5	5
109	5	5	5	5
110	5	5	5	5
111	5	5	5	5
112	5	5	5	5
113	5	5	5	5
114	5	5	5	5
115	5	5	5	5
122	5	5	5	5
123	5	5	5	5
124	5	5	5	5
125	5	5	5	5
126	5	5	5	5
127	5	5	5	5
128	5	5	5	5
129	5	5	5	5
130	5	5	5	5
131	5	5	5	5
132	5	5	5	5
133	5	5	5	5
136	5	5	5	5
137	5	5	5	5
138	5	5	5	5
139	5	5	5	5
140	5	5	5	5
141	5	5	5	5
142	5	5	5	5
143	5	5	5	5

TABLE 8-continued

Compound No.	Herbicidal effects			
	Barnyard-grass	Umbrella plant	Monochoria	Bulrush
144	5	5	5	5

Text Example 6

Low dose test in the soil treatment of irrigated paddy rice field

Into a 1/5,000a Wagner pot, paddy field soil was filled and puddled, and then water was introduced to a depth of 3 cm.

In pot A, three germinated tubers of each of water nutgrass and sagittaria, were embedded in the surface layer of the soil, and two seedlings of two paddy field rice plants of 2.2 leaf stage, were transplanted in a depth of 2 cm.

In pot B, seeds of barnyardgrass, bulrush, water plantain, monochoria and umbrella plant, were sown in the surface layer of the soil.

The day after the seeding and transplantation, a prescribed amount of a wettable powder of each compound formulated in accordance with Formulation Example 1, was diluted with water and dropwise applied by a pippet.

Thirty days after the application, the herbicidal effects were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 9.

TABLE 9

Compound No.	Dose of active ingredient (g/10 a)	Barnyard-grass	Umbrella plant	Monochoria	Bulrush	Water plantain	Sagittaria	Flat sedge	Transplanted paddy field rice
57	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
58	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
59	25	5	5	5	5	5	5	5	1
60	12.5	5	5	5	5	5	5	5	1
	6.3	5	5	5	5	5	5	5	0
61	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	5	5	0
63	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
64	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
67	6.3	5	5	5	5	5	5	5	1
68	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	4	5	0
69	50	5	5	5	5	5	5	4	0
	25	5	5	5	5	5	5	4	0
72	50	5	5	5	5	5	5	5	1
73	50	5	5	5	5	5	5	3	0
76	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
77	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
79	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	5	5	0
81	50	5	5	5	5	5	5	5	0.5
	25	5	5	5	5	5	5	5	0.5
94	25	5	5	5	5	5	5	5	1
	12.5	5	5	5	5	5	4	4	1
95	50	5	5	5	5	5	5	5	2
	25	5	5	5	5	5	5	5	1
98	50	5	5	5	5	5	5	5	0
103	50	5	5	5	5	5	5	4	1
122	25	5	5	5	5	5	5	5	0
	12.5	5	5	5	5	5	5	5	0
123	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	5	5	0
124	50	5	5	5	5	5	5	5	1

TABLE 9-continued

Compound No.	Dose of active ingredient (g/10 a)	Barnyard-grass	Umbrella plant	Monochoria	Bulrush	Water plantain	Sagittaria	Flat sedge	Transplanted paddy field rice
	25	5	5	5	5	5	5	5	0
141	6.3	5	4	5	5	5	5	5	1
143	50	5	5	5	5	5	5	5	0
	25	5	5	5	5	5	5	5	0
144	50	5	5	5	5	5	5	5	2
	25	5	5	5	5	5	5	5	1
Ronstar	50	5	5	5	5	5	5	5	2
	25	5	5	5	5	5	2	5	2
	12.5	5	5	5	5	5	1	2	1

Test Example 7

Herbicidal test in the soil treatment of upland field

To a 120 cm^2 plastic pot, upland field soil was filled and seeds of barnyardgrass, large crabgrass, smart weed, pigweed, lambsquater and rice flatsedge were sown and covered with soil. A wettable powder of each compound formulated in accordance with Formulation Example 1, was diluted with water in an amount of 100 liter/10a and uniformly applied to the surface of the soil by a small size spray at a dose of 400 g/10a of the active ingredient. After the treatment, the pot was left in a green house for 21 days, and then the herbicidal effects were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 10.

TABLE 10

Compound No.	Herbicidal effects: Barnyard-grass	Large crabgrass	Smart weed	Pig-weed	Lambs-quaters	Rice flatsedge
58	4	5	5	5	5	5
59	4	5	5	5	5	5
63	3	5	5	5	5	5
64	3	5	5	5	5	5
67	5	5	5	5	5	5
71	3	5	5	5	5	5
76	3	5	5	5	5	5
81	4	5	5	5	5	5
95	5	5	4	5	5	5
103	3	5	5	5	5	5
107	3	5	5	5	5	5
108	3	5	5	5	5	5
112	5	5	5	5	5	5
113	3	5	5	5	5	5
114	4	5	5	5	5	5
123	3	5	5	5	5	5
128	5	5	5	5	5	5
129	5	5	4	5	5	5
130	5	5	5	5	5	5
133	5	5	5	5	5	5
141	5	5	5	5	5	5
142	4	5	5	5	5	5

Text Example 8

Herbicidal test in the foliage treatment of upland field

Into a 120 cm^2 plastic pot, upland field soil was filled, and seeds of barnyardgrass, large crabgrass, smart weed, pigweed, lambsquater and rice flatsedge, were sown, and grown in a green house until barnyardgrass grew to the 3 leaf stage. When barnyardgrass reached the 3 leaf stage, a wettable powder of each test compound prepared in accordance with Formulation Example 1 was diluted with water in an amount of 100 liter/10a and applied to the foliage of the plants from above by a small size spray at a dose of 400 g/10a of the active ingredient. After the application, the pot was left in a green house for 21 days, and then the herbicidal effects were evaluated in accordance with the standards identified in Table 3. The results are shown in Table 11.

TABLE 11

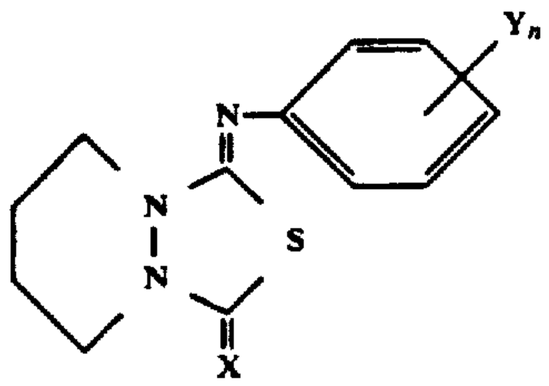
Compound No.	Herbicidal effects: Barnyard-grass	Large crabgrass	Smart weed	Pig-weed	Lambs-quarters	Rice flat-sedge
57	4	5	5	5	5	5
58	5	5	5	5	5	5
59	5	5	5	5	5	5
60	4	5	5	5	5	5
61	5	5	5	5	5	5
63	5	5	5	5	5	5
64	5	5	5	5	5	5
67	5	5	5	5	5	5
68	5	4	5	5	5	5
69	5	5	5	5	5	5
70	5	5	5	5	5	5
71	5	5	5	5	5	5
72	5	5	5	5	5	5
73	4	4	5	5	5	5
76	4	5	5	5	5	5
77	4	4	5	5	5	5
80	4	4	5	5	5	5
81	5	5	5	5	5	5
84	5	5	5	5	5	5
85	5	5	5	5	5	5
86	5	5	5	5	5	5
87	5	5	5	5	5	5
88	3	3	5	5	5	5
89	3	4	5	5	5	5
92	5	4	5	5	5	5
93	5	5	5	5	5	5
94	4	5	5	5	5	5
95	5	5	5	5	5	5
97	5	5	5	5	5	5
98	5	5	5	5	5	5
99	4	5	5	5	5	5
102	4	4	5	5	5	5
103	5	5	5	5	5	5
104	4	5	5	5	5	5
105	4	5	5	5	5	5
106	4	4	5	5	5	5
107	5	5	5	5	5	5
108	5	5	5	5	5	5
109	5	5	5	5	5	5
110	5	5	5	5	5	5
111	4	5	5	5	5	5
112	5	5	5	5	5	5
113	5	5	5	5	5	5
114	5	5	5	5	5	5
123	5	5	5	5	5	5
123	5	5	5	5	5	5
125	5	5	5	5	5	5
126	5	5	5	5	5	5
127	5	5	5	5	5	5
128	5	5	5	5	5	5
129	5	5	5	5	5	5
130	5	5	5	5	5	5
132	5	5	5	5	5	5
133	5	5	5	5	5	5
136	5	4	5	5	5	5
139	4	5	5	5	5	5
141	5	5	5	5	5	5

TABLE 11-continued

Compound No.	Herbicidal effects					
	Barnyard-grass	Large crabgrass	Smart weed	Pig-weed	Lambs-quarters	Rice flat-sedge
142	5	5	5	5	5	5
144	4	5	5	5	5	5

We claim:

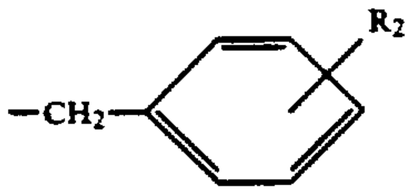
1. A 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) compound having the formula:



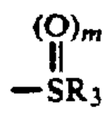
(I)

wherein Y which may be the same or different, represents chlorine, bromine, fluorine, hydroxyl, lower alkyl, lower alkoxy which may be substituted by chlorine, bromine or fluorine, lower alkenyloxy which may be substituted by chlorine, bromine or fluorine, lower alkynyloxy, phenoxy, lower cycloalkyloxy, lower alkoxycarbonyl-lower alkenyloxy, lower alkylthiocarbonyl-lower alkyloxy, lower alkynyloxycarbonyl-lower alkyloxy, benzyloxycarbonyl-lower alkyloxy, trifluoromethyl, benzyloxy which may be substituted by chlorine or lower alkyl, lower alkenyl, cyano-lower alkyl, lower alkylcarbamoyloxy, benzyl which may be substituted by one or two lower alkyl, lower alkoxy-lower alkyl, lower alkynyloxy-lower alkyl, lower cycloalkylmethyloxy which may be substituted by chlorine, bromine or fluorine, lower alkoxy-lower alkyloxy, phenethyloxy, lower cycloalkyloxycarbonyl-lower alkyloxy, pyrrolidinocarbonyl, phenylcarbonyl which may be substituted by lower alkyl,

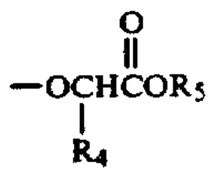
wherein R_1 is hydrogen, lower alkyl, phenyl, lower cycloalkyl, lower alkoxy-lower alkyl, lower alkoxycarbonyl-lower alkyl or



(wherein R_2 is hydrogen or lower alkoxy), X is oxygen or sulfur,

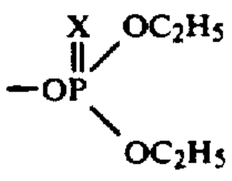


(wherein R_3 is lower alkyl, lower alkenyl or lower alkynyl, and m is 0 or 2),



wherein R_4 is hydrogen or lower alkyl, and R_5 is hydrogen, lower alkyl, lower alkoxy-lower alkyl, tetrahydrofurfuryl, lower alkoxy-lower alkyloxy-lower alkyl, lower alkoxycarbonyl-lower alkyl or $N{=}C(CH_3){-}R_6$ (wherein R_6 is lower alkyl or phenyl), $-NHR_7$ (wherein R_7 is lower alkylcarbonyl or lower alkoxycarbonyl-lower alkyl),

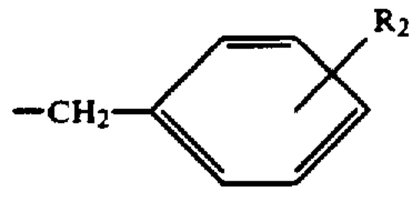
(wherein R_8 is hydrogen or lower alkyl, and R_9 is lower alkoxy, lower cycloalkyloxy or pyrrolidinyl), or



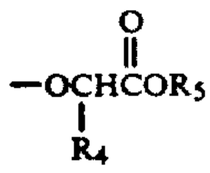
(wherein X is as defined above); n is an integer or from 0 to 3; and X is oxygen or sulfur.

2. The compound according to claim 1, wherein Y is chlorine, bromine or fluorine, lower alkoxy which may be substituted by chlorine, bromine or fluorine, lower alkenyloxy which may be substituted by chlorine, bromine or fluorine, lower alkynyloxy, phenoxy, benxyloxy which may be substituted by chlorine or lower alkyl,

wherein R_1 is hydrogen, lower alkyl, phenyl, lower cycloalkyl, lower alkoxy-lower alkyl, lower alkoxycarbonyl-lower alkyl or

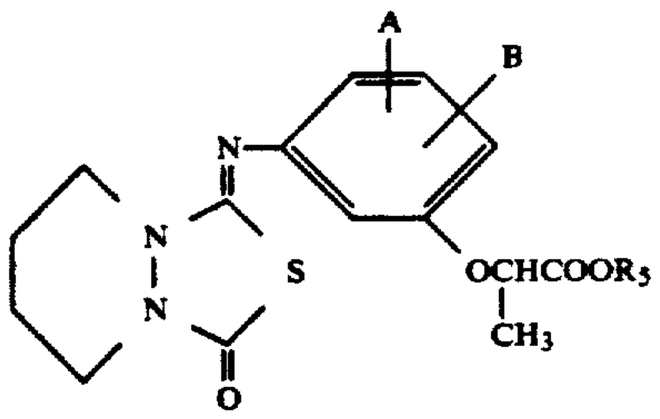


(wherein R_2 is hydrogen or lower alkoxy), X is oxygen or sulfur,



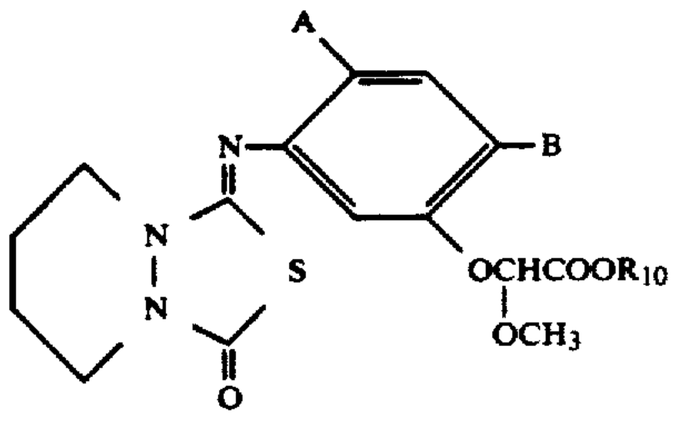
wherein R_4 is hydrogen or lower alkyl, and R_5 is hydrogen, lower alkyl, lower alkynyl, benzyl, lower alkoxy-lower alkyl, tetrahydrofurfuryl, lower alkoxy-lower alkyloxy-lower alkyl, lower alkoxycarbonyl-lower alkyl, lower cycloalkyl, or $-N{=}C(CH_3)R_6$ (wherein R_6 is lower alkyl or phenyl).

3. The compound according to claim 1, which has the formula:



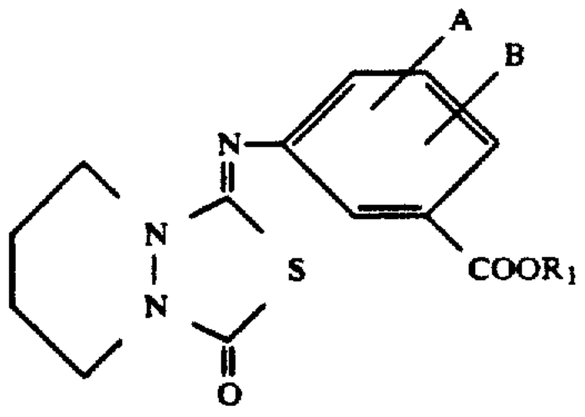
wherein A is hydrogen, chlorine, bromine or fluorine, B is chlorine, bromine or fluorine, and R_5 is hydrogen, lower alkyl, lower alkynyl, benzyl, lower alkoxy-lower alkyl, tetrahydrofurfuryl, lower alkoxy-lower alkyloxy-lower alkyl, lower alkoxycarbonyl-lower alkyl, lower cycloalkyl, or $-N=C(CH_3)R_6$ (wherein R_6 is lower alkyl or phenyl).

4. The compound according to claim **1**, which has the formula:

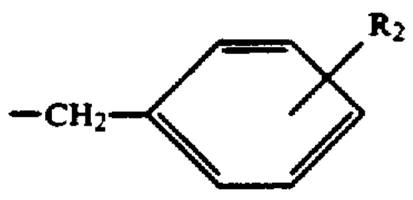


wherein A is hydrogen, chlorine, bromine or fluorine, B is chlorine, bromine or fluorine, and R_{10} is hydrogen, lower alkyl, lower alkynyl, lower alkoxy-lower alkyl, lower alkoxy-lower alkyloxy-lower alkyl, or tetrahydrofurfuryl.

5. The compound according to claim **1**, which has the formula:

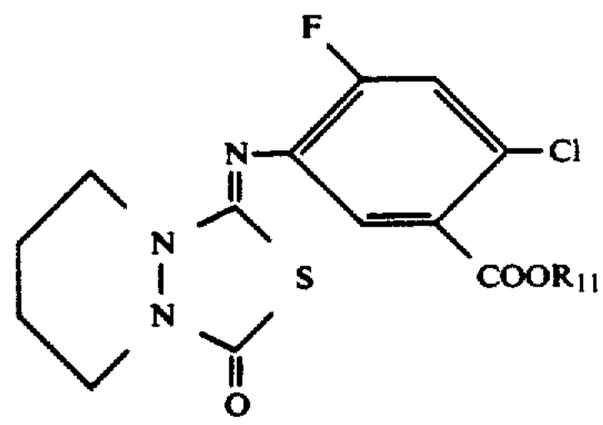


wherein A is hydrogen, chlorine, bromine or fluorine, B is chlorine, bromine or fluorine, and R_1 is hydrogen, lower alkyl, phenyl, lower cycloalkyl, lower alkoxy-lower alkyl, lower alkoxycarbonyl-lower alkyl or



(wherein R_2 is hydrogen or lower alkoxy).

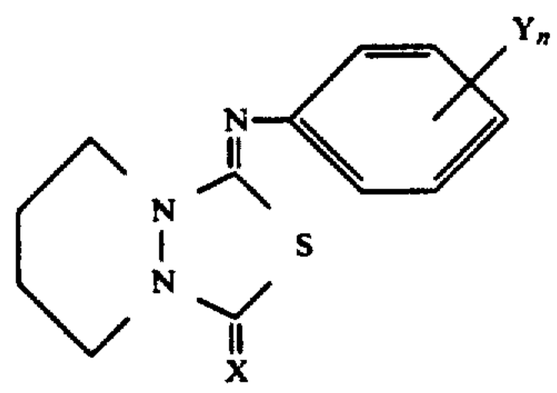
6. The compound according to claim **1**, which has the formula:



wherein [Hal is chlorine, bromine or fluorine, and] R_{11} is hydrogen or lower alkyl.

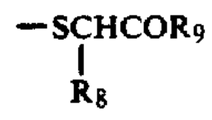
7. A herbicidal composition comprising a herbicidally effective amount of a compound of the formula I as defined in claim **1** and a carrier.

8. A 9-phenylimino-8-thia-1,6-diazabicyclo[4.3.0]nonane-7-(one or thione) compound having the formula:



(I)

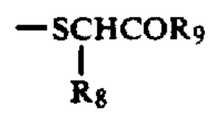
wherein Y is at least one halogen and a group represented by the formula:



wherein R_8 is hydrogen or lower alkyl, and R_9 is lower alkoxy, lower cycloalkyloxy or pyrrolidinyl; X is oxygen or sulfur, and n is an integer from 2 to 3.

9. The compound according to claim **8**, wherein R_9 is lower alkoxy.

10. The compound according to claim **8**, wherein n is 3 having two chlorine or fluorine radicals and one radical of the formula



wherein R_8 is hydrogen or lower alkyl, and R_9 is lower alkoxy, lower cycloalkoxy or pyrrolidinyl.

11. The compound according to claim **9**, wherein n is an integer from 2 to 3.

12. The compound according to claim **11**, wherein n is 3 having two chlorine or fluorine radicals and one radical of the formula:

wherein R_8 is hydrogen or lower alkyl, and R_9 is lower alkoxy, lower cycloalkoxy or pyrrolidinyl.

13. The compound according to claim 6, wherein R_{11} is ethyl.

14. The compound according to claim 6, wherein R_{11} is hydrogen.

15. The compound according to claim 6, wherein R_{11} is isopropyl.

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