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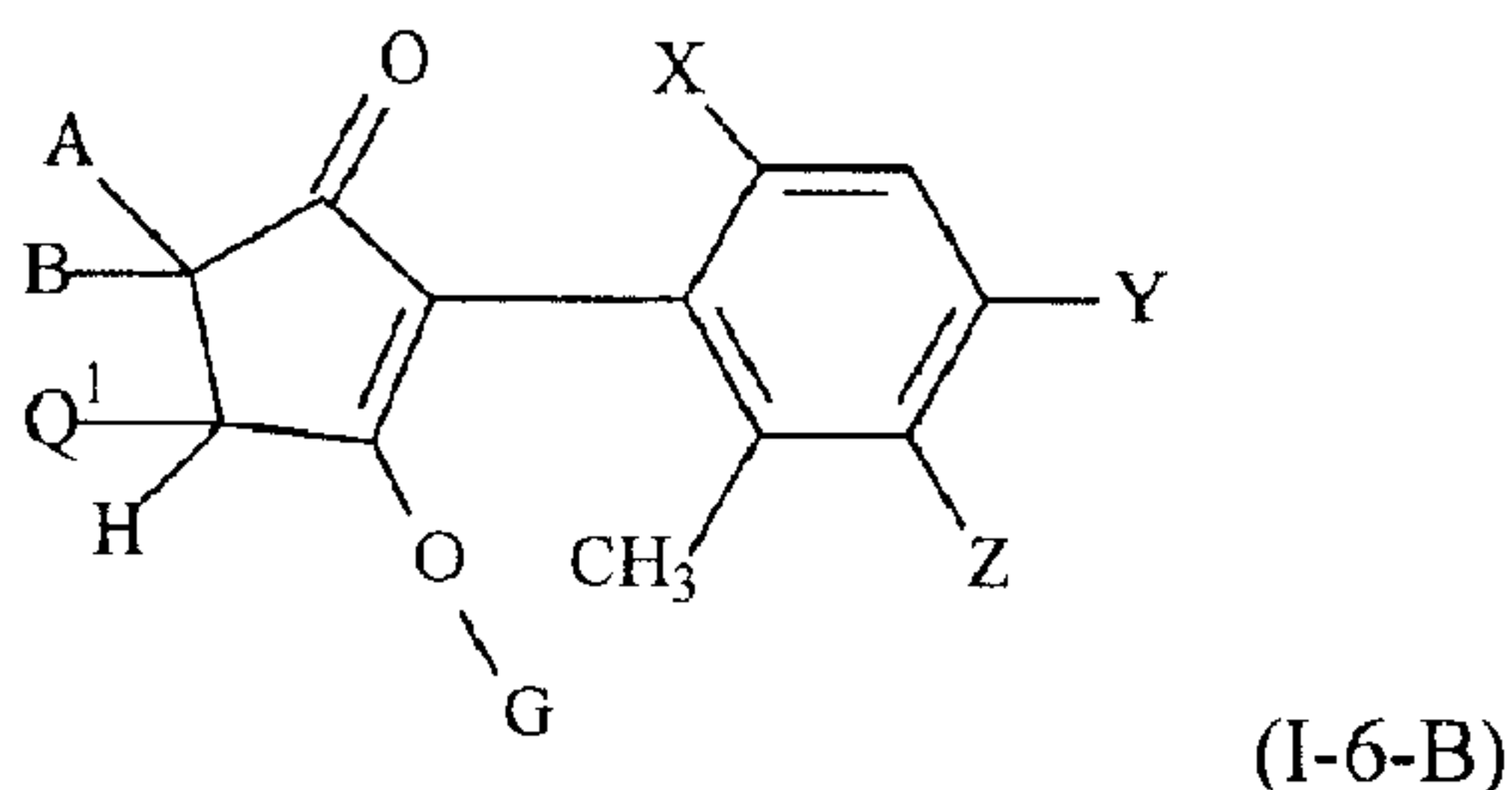
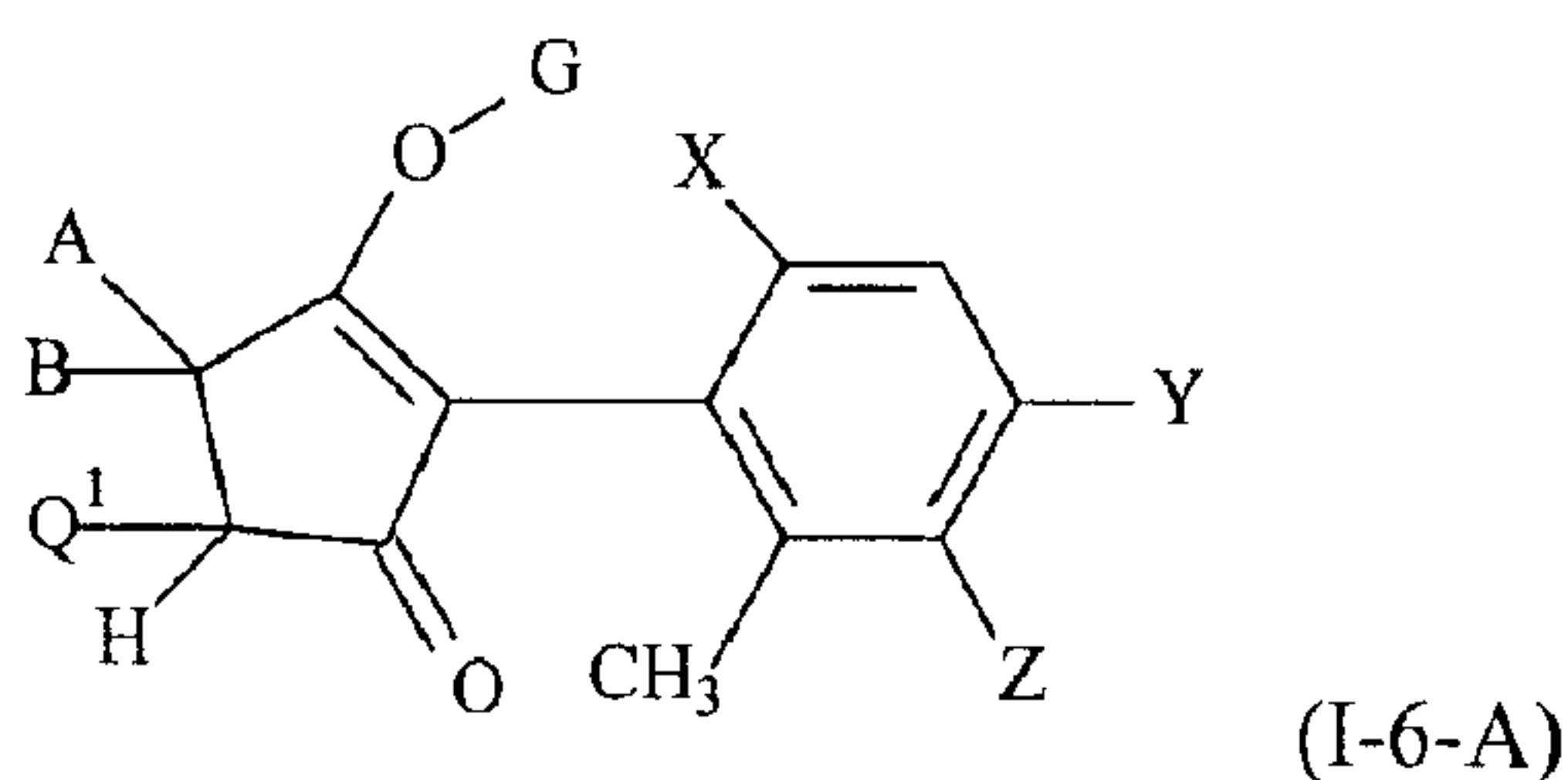
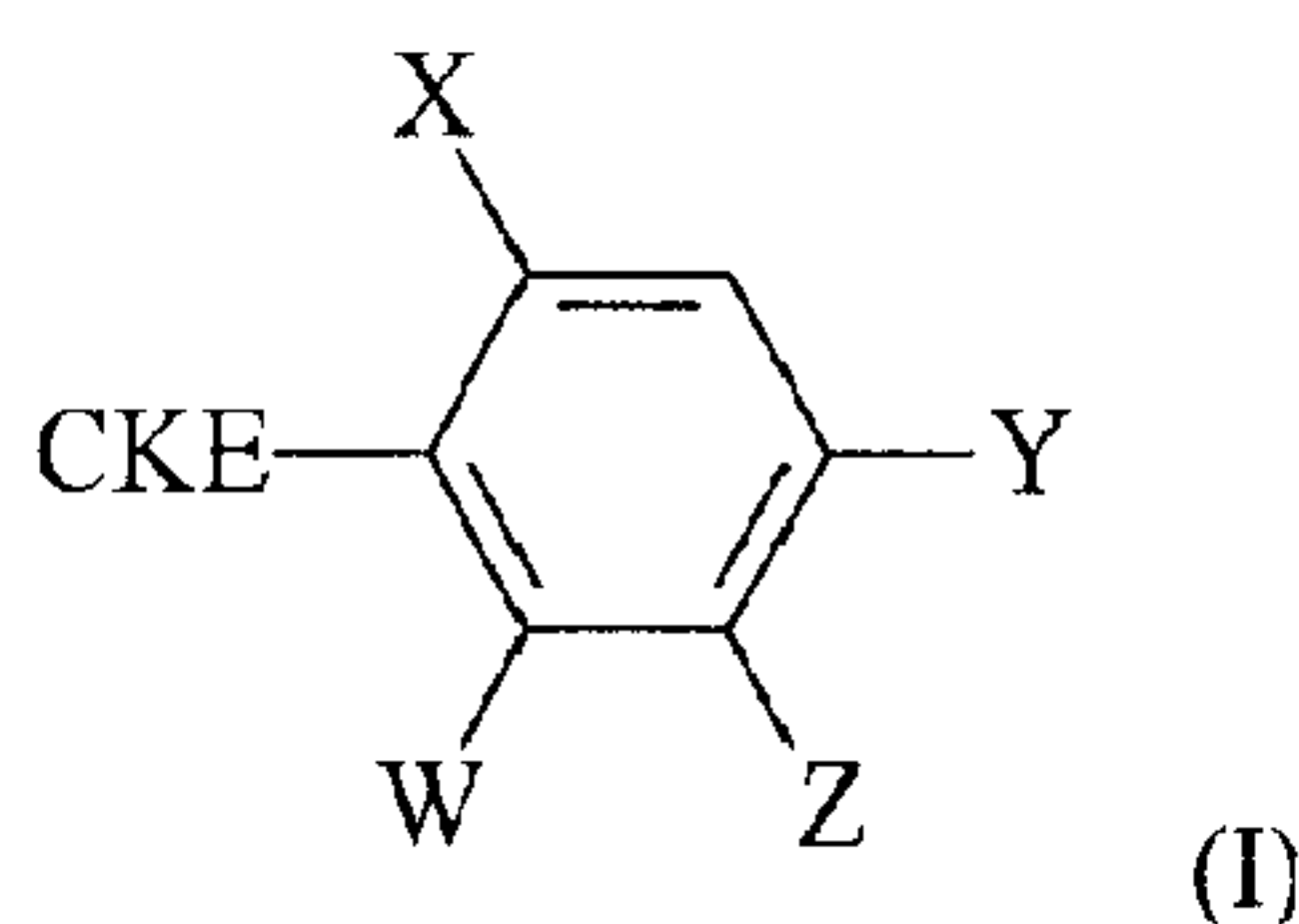
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(54) Titre : CETOENOLS CYCLIQUES SUBSTITUES PAR PHENYLE C₂ UTILISES COMME AGENTS DE LUTTE
CONTRE LES PARASITES ET COMME HERBICIDES

(54) Title: C₂-PHENYL-SUBSTITUTED CYCLIC KETO-ENOLS AS PESTICIDES AND HERBICIDES



(57) Abrégé/Abstract:

The present invention relates to novel C₂-phenyl-substituted cyclic ketoenols of the general formula (I), (I-6-A) or (I-6-B): (see



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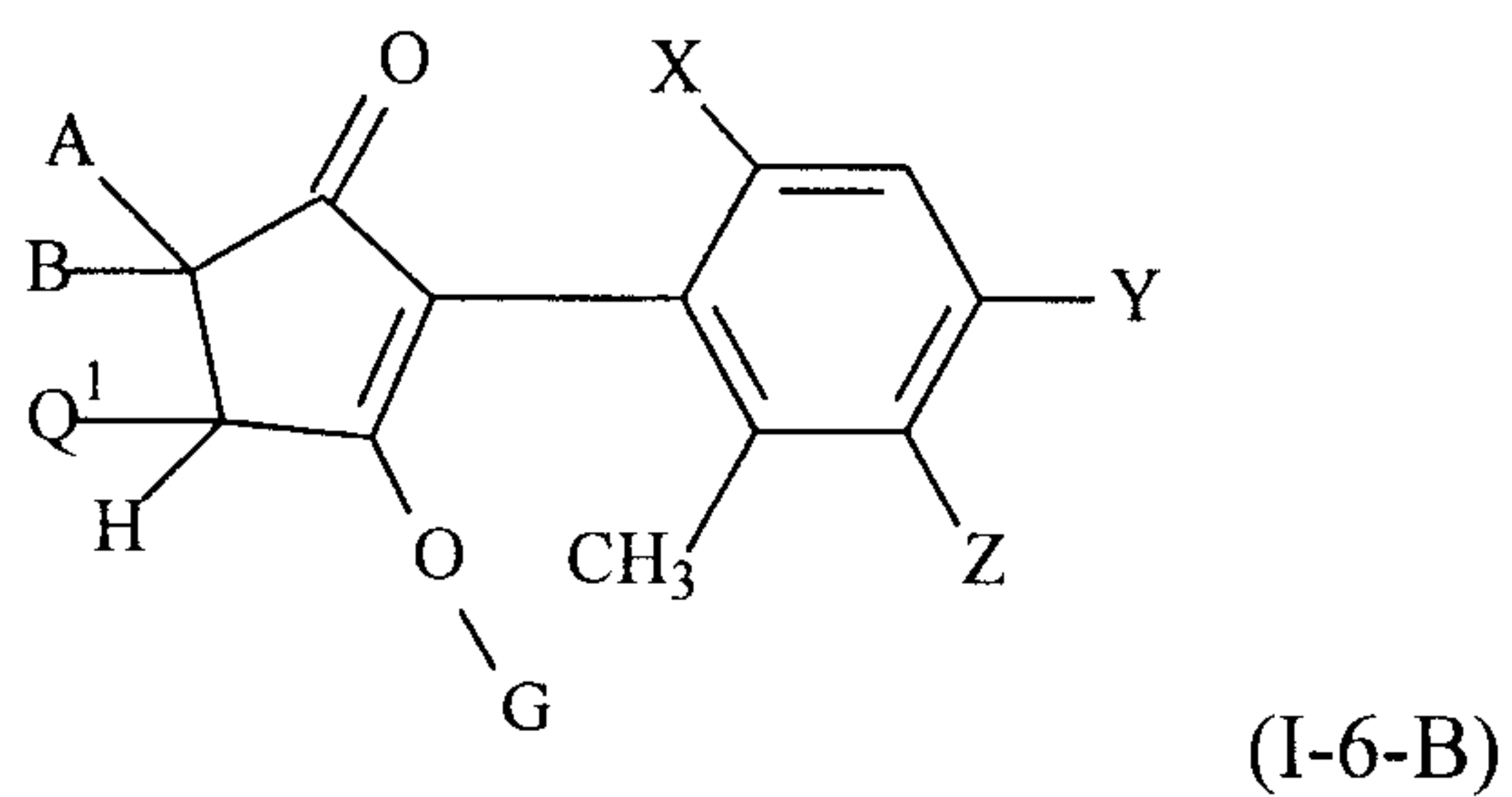
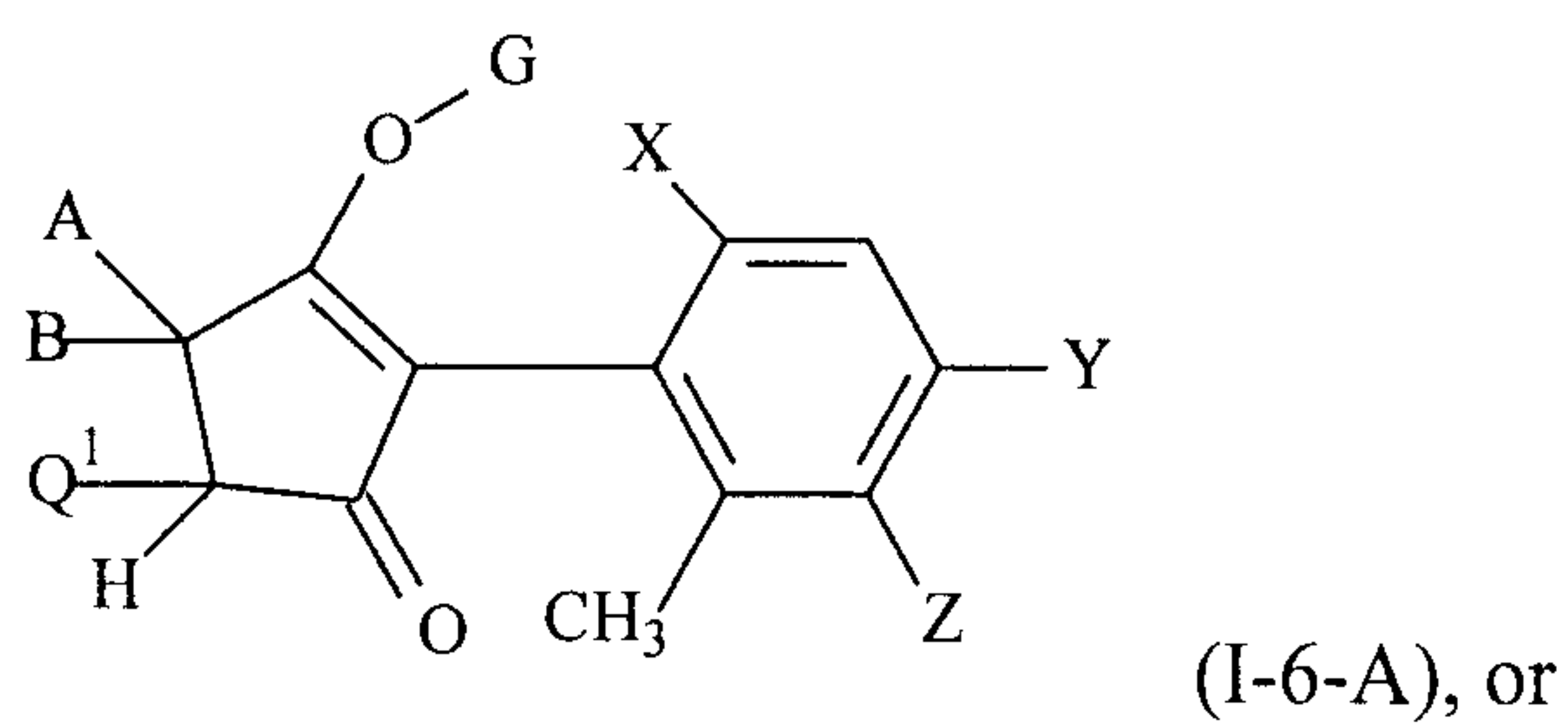
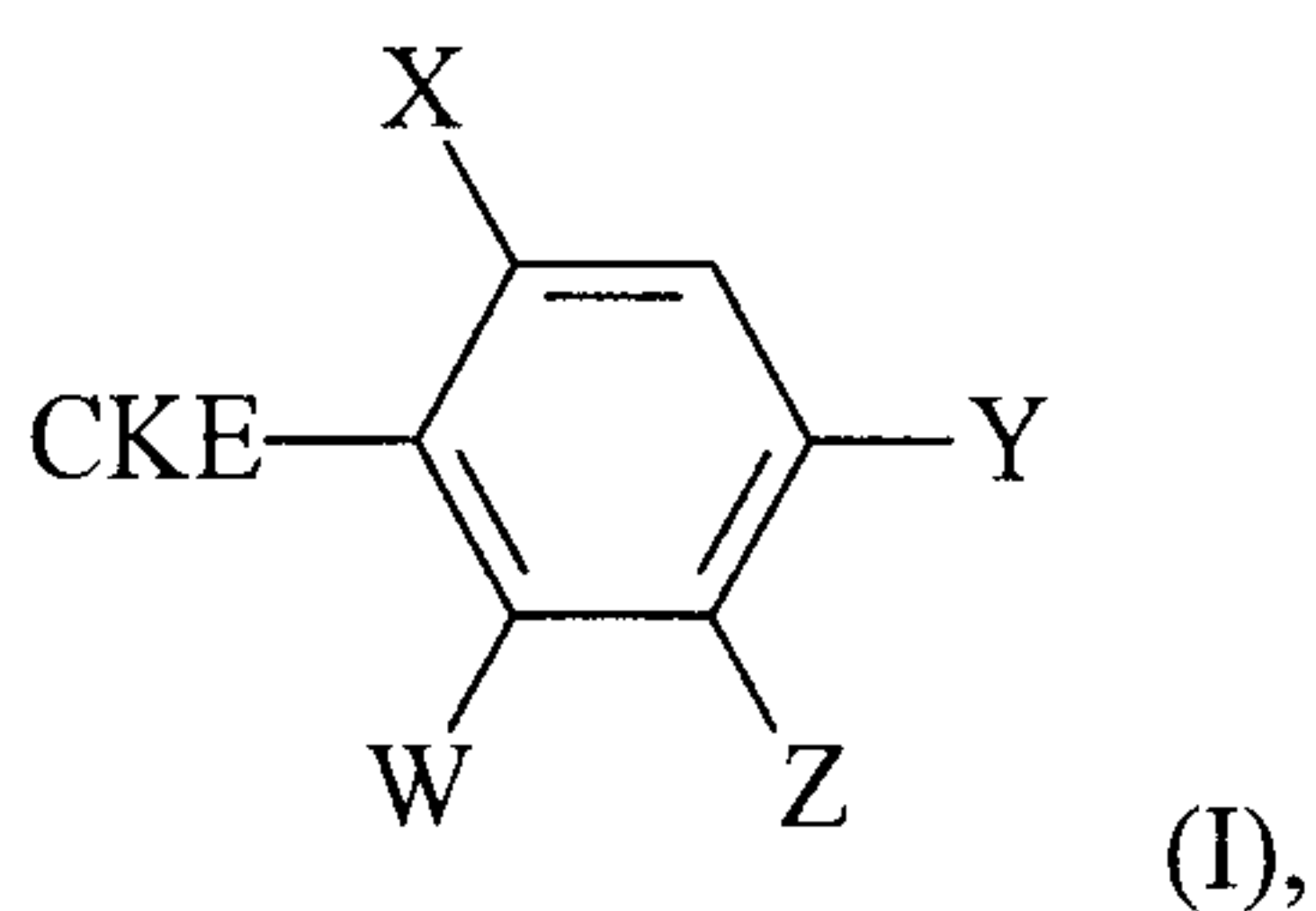
(57) **Abrégé(suite)/Abstract(continued):**

formula I), (see formula I-6-A), or (see formula I-6-B) to processes for their preparation and to their use as pesticides and herbicides. The pesticidal and herbicidal properties of the compounds, particularly at low application rates, and their compatibility with plants are desirable compared to known pesticides and herbicides.

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Abstract

The present invention relates to novel C₂-phenyl-substituted cyclic ketoenols of the general formula (I), (I-6-A) or (I-6-B):



to processes for their preparation and to their use as pesticides and herbicides. The pesticidal and herbicidal properties of the compounds, particularly at low application rates, and their compatibility with plants are desirable compared to known pesticides and herbicides.

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C₂-Phenyl-Substituted Cyclic Keto-Enols as Pesticides and Herbicides

This is a divisional application of Canadian Patent Application No. 2,404,868, filed March 21, 2001. It should be understood that the expression "the present invention" or the like used in this specification encompasses not only the subject matter of this divisional application but that of the parent application also.

- 5 The present invention relates to novel C₂-phenyl-substituted cyclic ketoenols, to a plurality of processes for their preparation and to their use as pesticides and herbicides.

Pharmacological properties of 3-acyl-pyrrolidine-2,4-diones have already been described (S. Suzuki et al. Chem. Pharm. Bull. 15 1120 (1967)). Furthermore, N-phenylpyrrolidine-2,4-diones have been synthesized by R. Schmierer and H. Mildenberger (Liebigs Ann. Chem. 1985, 1095). A biological
10 activity of these compounds has not been described.

EP-A-0 262 399 and GB-A-2 266 888 disclose compounds of a similar structure (3-aryl-pyrrolidine-2,4-diones) of which, however, no herbicidal, insecticidal or acaricidal activity has become known. Unsubstituted bicyclic 3-aryl-pyrrolidine-2,4-dione derivatives (EP-A-355 599 and EP-A-415 211) and substituted monocyclic 3-aryl-pyrrolidine-2,4-dione derivatives (EP-A-377 893 and
15 EP-A-442 077) having herbicidal, insecticidal or acaricidal activity are known.

Also known are polycyclic 3-arylpyrrolidine-2,4-dione derivatives (EP-A-442 073) and 1H-arylpyrrolidine-dione derivatives (EP-A-456 063, EP-A-521 334, EP-A-596 298, EP-A-613 884, EP-A-613 885, WO 94/01 997, WO 95/26 954, WO 95/20 572, EP-A-0 668 267, WO 96/25 395, WO 96/35 664, WO 97/01 535, WO 97/02 243, WO 97/36 868, WO 97/43275, WO 98/05638,
20 WO 98/06721, WO 98/25928, WO 99/16748, WO 99/24437, WO 99/43649, WO 99/48869 and WO 99/55673).

It is known that certain substituted Δ^3 -dihydrofuran-2-one derivatives have herbicidal properties (cf. DE-A-4 014 420). The synthesis of the tetrionic acid derivatives used as starting materials (such as, for example, 3-(2-methyl-phenyl)-4-hydroxy-5-(4-fluorophenyl)- Δ^3 -dihydrofuran-2-one) is
25 likewise described in DE-A-4 014 420.

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Compounds of a similar structure are known from the publication Campbell et al., J. Chem. Soc., Perkin Trans. 1, 1985, (8) 1567-76, without any insecticidal and/or acaricidal activity being mentioned. Furthermore, 3-aryl- Δ^3 -dihydrofuranone derivatives having herbicidal, acaricidal and insecticidal properties are known from
5 EP-A-528 156, EP-A-0 647 637, WO 95/26 345, WO 96/20 196, WO 96/25 395, WO 96/35 664, WO 97/01 535, WO 97/02 243, WO 97/36 868, WO 98/05638, WO 98/25928, WO 99/16748, WO 99/43649, WO 99/48869 and WO 99/55673. 3-Aryl- Δ^3 -dihydrothiophenone derivatives are likewise known (WO 95/26 345, 96/25 395, WO 97/01 535, WO 97/02 243, WO 97/36 868, WO 98/05638, WO 98/25928, WO
10 99/16748, WO 99/43649, WO 99/48869, WO 99/55673).

Certain phenyl-pyrone derivatives which are unsubstituted in the phenyl ring have already become known (cf. A.M. Chirazi, T. Kappe and E. Ziegler, Arch. Pharm. 309, 558 (1976) and K.-H. Boltze and K. Heidenbluth, Chem. Ber. 91, 2849), a
15 possible use of these compounds as pesticides not being mentioned. Phenyl-pyrone derivatives which are substituted in the phenyl ring and have herbicidal, acaricidal and insecticidal properties are described in EP-A-588 137, WO 96/25 395, WO 96/35 664, WO 97/01 535, WO 97/02 243, WO 97/16 436, WO 97/19 941, WO 97/36 868, WO 98/05638, WO 99/43649, WO 99/48869 and WO 99/55673.

20 Certain 5-phenyl-1,3-thiazine derivatives which are unsubstituted in the phenyl ring have already become known (cf. E. Ziegler and E. Steiner, Monatsh. 95, 147 (1964), R. Ketcham, T. Kappe and E. Ziegler, J. Heterocycl. Chem. 10, 223 (1973)), a possible use of these compounds as pesticides not being mentioned. 5-Phenyl-1,3-
25 thiazine derivatives which are substituted in the phenyl ring and have herbicidal, acaricidal and insecticidal activity are described in WO 94/14 785, WO 96/02 539, WO 96/35 664, WO 97/01 535, WO 97/02 243, WO 97/02 243, WO 97/36 868, WO 99/05638, WO 99/43649, WO 99/48869 and WO 99/55673.

30 It is known that certain substituted 2-arylcyclopentanediones have herbicidal and acaricidal properties (cf., for example, US-4 283 348; 4 338 122; 4 436 666;

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4 526 723; 4 551 547; 4 632 698; WO 96/01 798; WO 96/03 366, WO 97/14 667 and
also WO 98/39281, WO 99/43649, WO 99/48869, WO 99/55673). Moreover,
compounds of a similar structure are known; 3-hydroxy-5,5-dimethyl-2-phenylcyclo-
pent-2-en-1-one from the publication Micklefield et al., Tetrahedron, (1992), 7519-
5 26 and the natural product involutine (-)-cis-5-(3,4-dihydroxyphenyl)-3,4-dihydroxy-
2-(4-hydroxyphenyl)-cyclopent-2-en-one from the publication Edwards et al., J.
Chem. Soc. S, (1967), 405-9. An insecticidal or acaricidal activity is not described.
Moreover, 2-(2,4,6-trimethylphenyl)-1,3-indanedione is known from the publication
J. Economic Entomology, 66, (1973), 584 and the Offenlegungsschrift
10 DE-A 2 361 084, with herbicidal and acaricidal activities being mentioned.

It is known that certain substituted 2-arylcyclohexanediones have herbicidal and
acaricidal properties (US-4 175 135, 4 209 432, 4 256 657, 4 256 658, 4 256 659,
4 257 858, 4 283 348, 4 303 669, 4 351 666, 4 409 153, 4 436 666, 4 526 723,
15 4 613 617, 4 659 372, DE-A 2 813 341, and also Wheeler, T.N., J. Org. Chem. 44,
4906 (1979)), WO 99/43649, WO 99/48869, WO 99/55673).

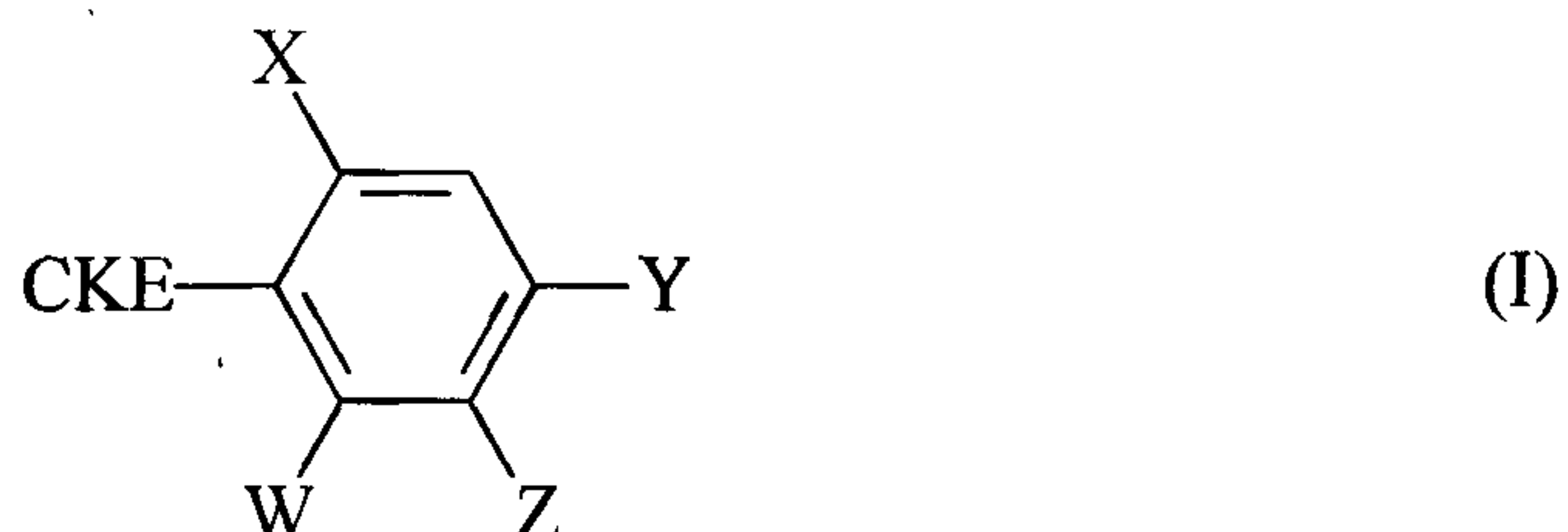
However, the activity and the activity spectrum of these compounds are, in particular
at low application rates and concentrations, not always entirely satisfactory.
20 Furthermore, the compatibility of these compounds with plants is not always
sufficient.

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- 3a -

Summary of Invention

In one compound aspect, the parent invention relates to a compound of the general formula (I):



5 in which:

- (α) W represents H, methyl, ethyl or i-propyl;
 X represents methyl, ethyl, i-propyl or vinyl;
 Y represents H, methyl, ethyl, i-propyl, vinyl or ethinyl;
 Z represents methyl, ethyl, n-propyl or i-butyl; or

- 10 (β) W represents methyl;
 X and Y are as defined under (α);
 Z represents H;

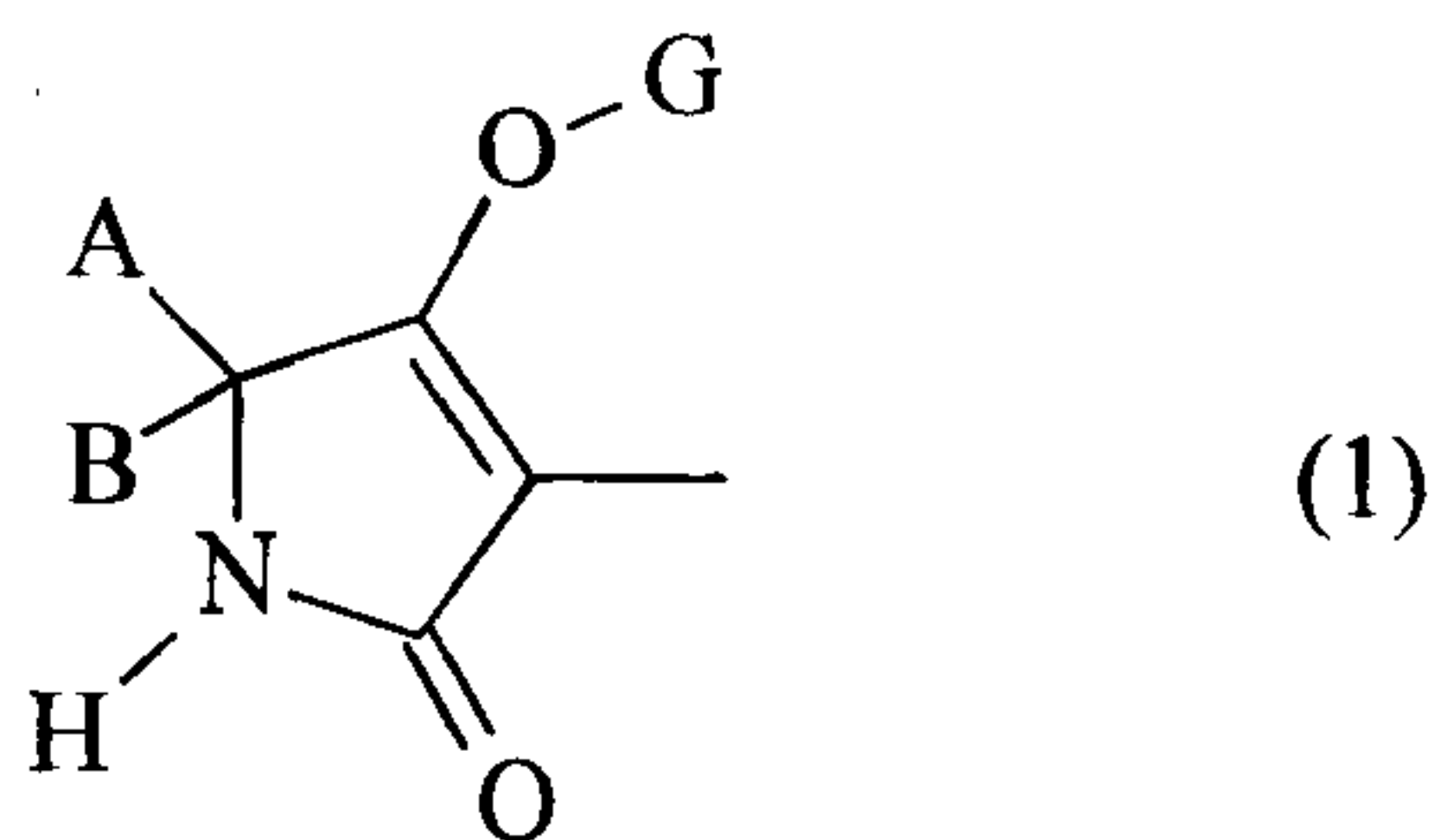
with the provisos that: (i) at least one of the radicals W, X, Y or Z represents a chain having at least two carbon atoms, and (ii) at most only one of the radicals X and Y

- 15 represents vinyl; and

CKE represents the group:

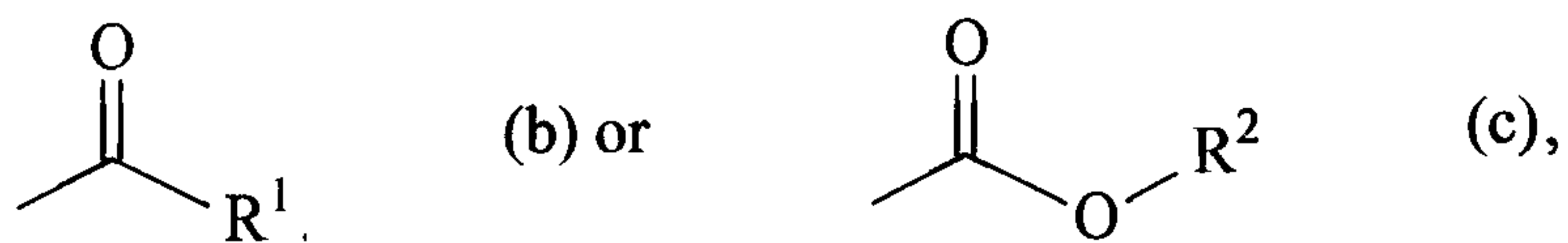
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- 3b -



wherein:

G represents H (a), or represents one of the groups:



5 R^1 represents C_1 - C_6 -alkyl or C_1 - C_2 -alkoxymethyl,

R^2 represents C_1 - C_4 -alkyl,

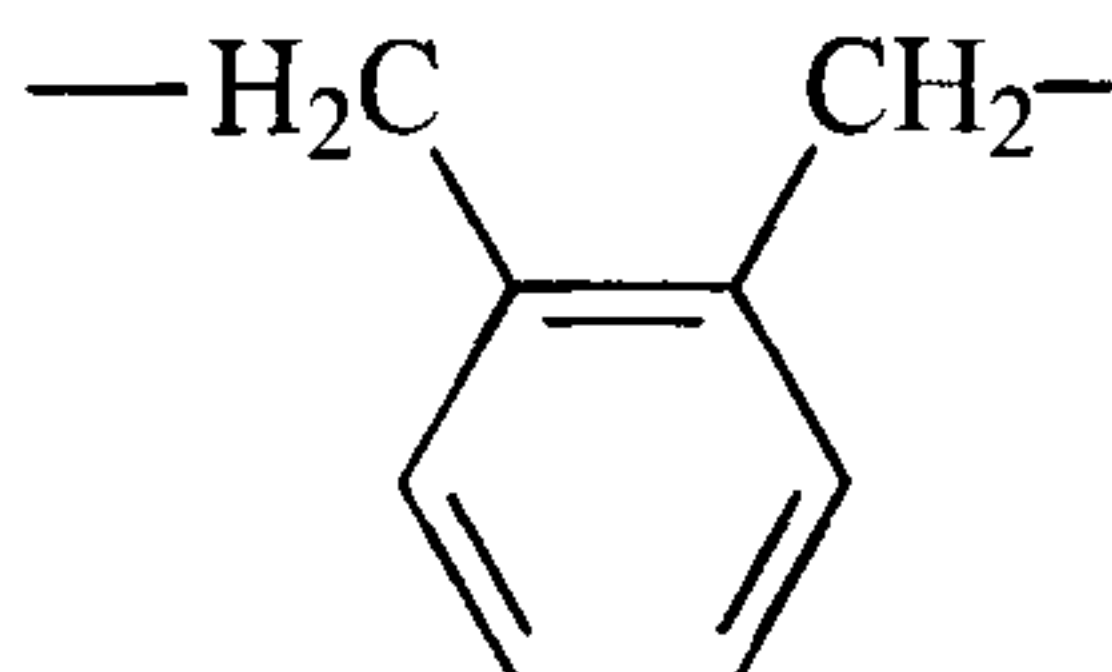
A represents methyl,

B represents methyl,

10 A, B and the carbon atoms to which they are attached represent saturated C_5 - C_6 -cycloalkyl in which optionally one ring member is replaced by O and which is optionally monosubstituted by methyl, ethyl, methoxy or ethoxy,

or

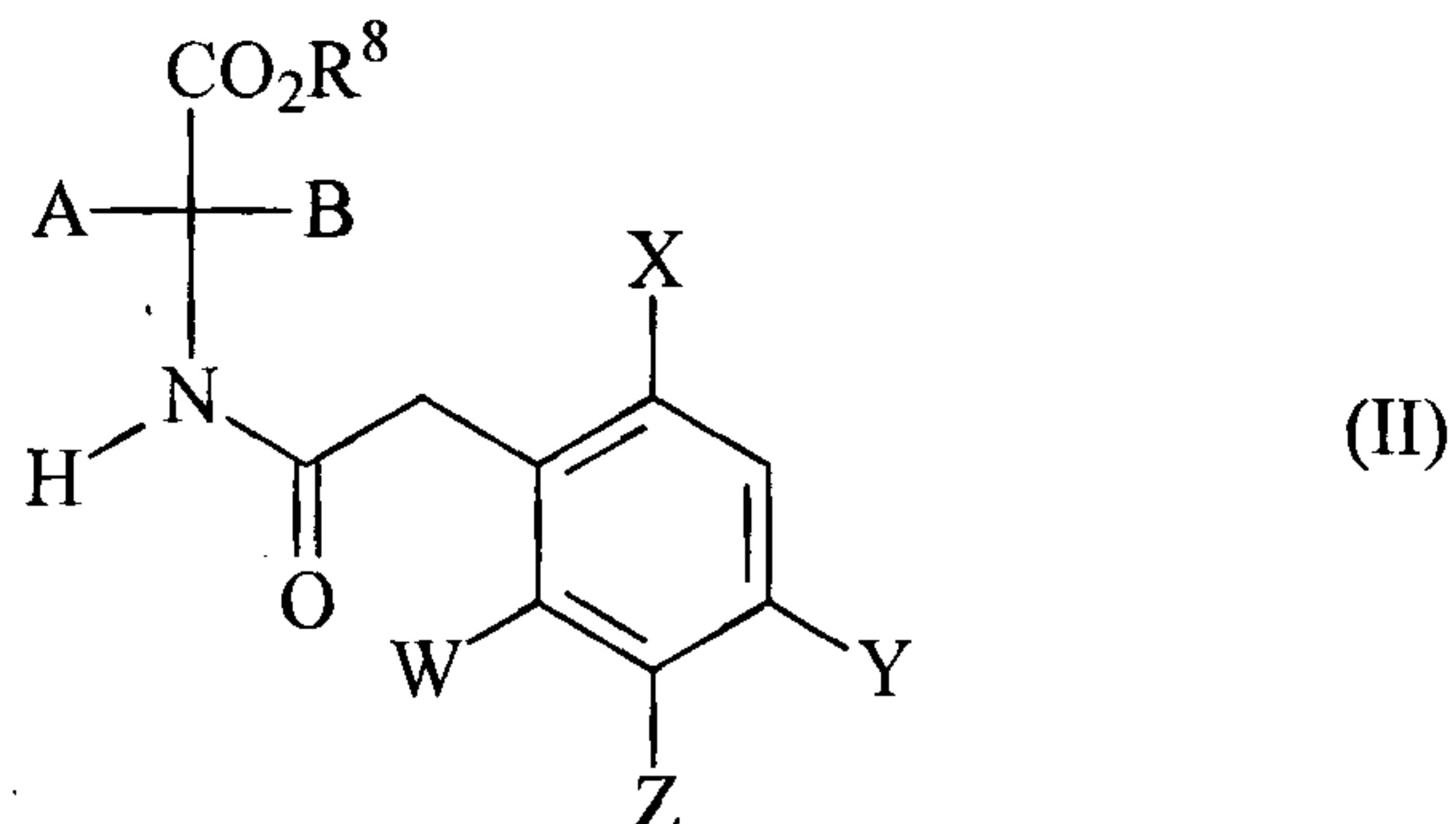
A and B together represent



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In a further compound aspect, the parent invention relates to a compound of the general formula (II):

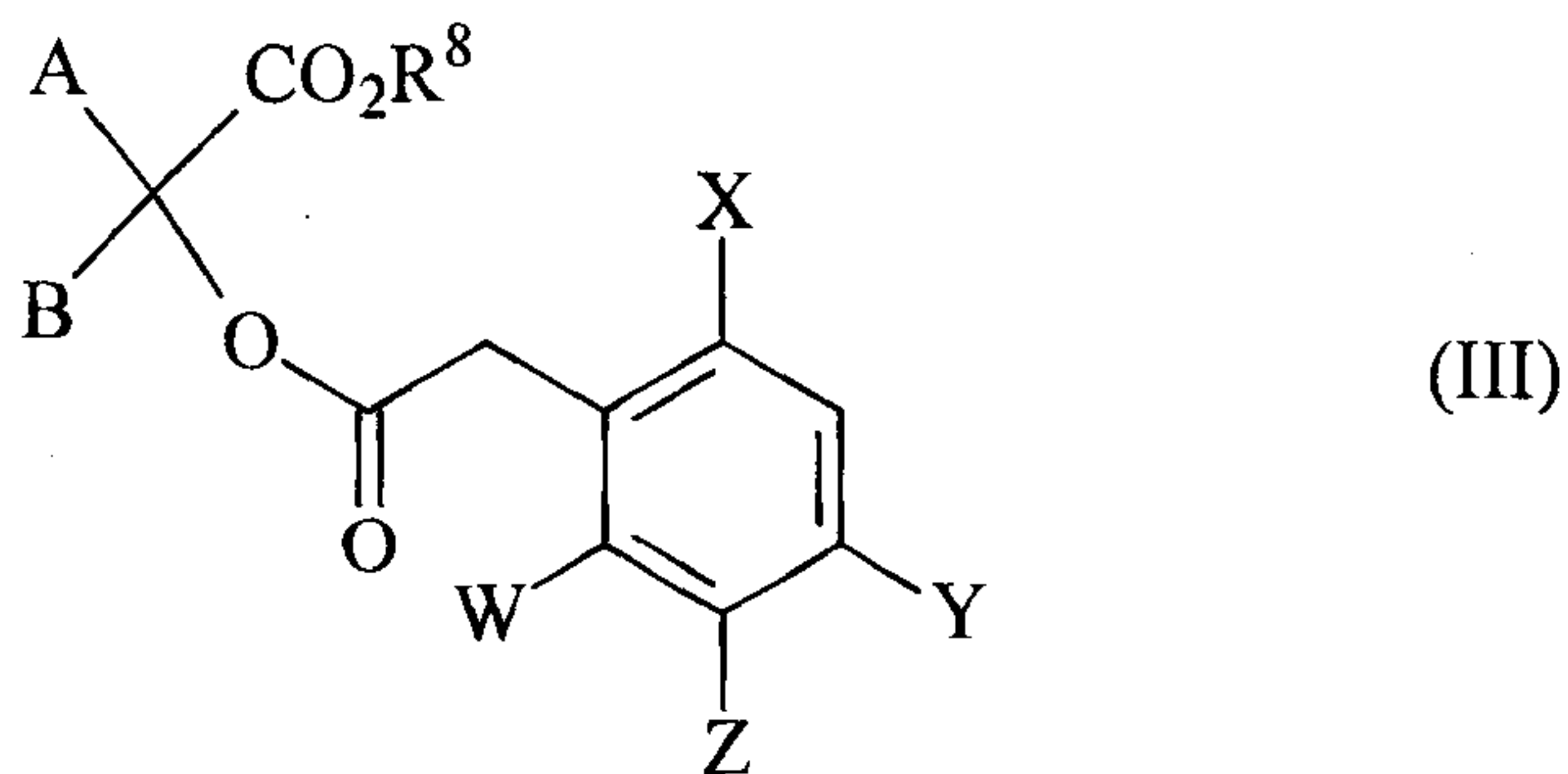


in which:

5 A, B, W, X, Y and Z are each as defined above; and

 R⁸ represents C₁-C₆ alkyl.

In a still further compound aspect, the parent invention relates to a compound of the general formula (III):



10 in which:

 A, B, W, X, Y and Z are each as defined above; and

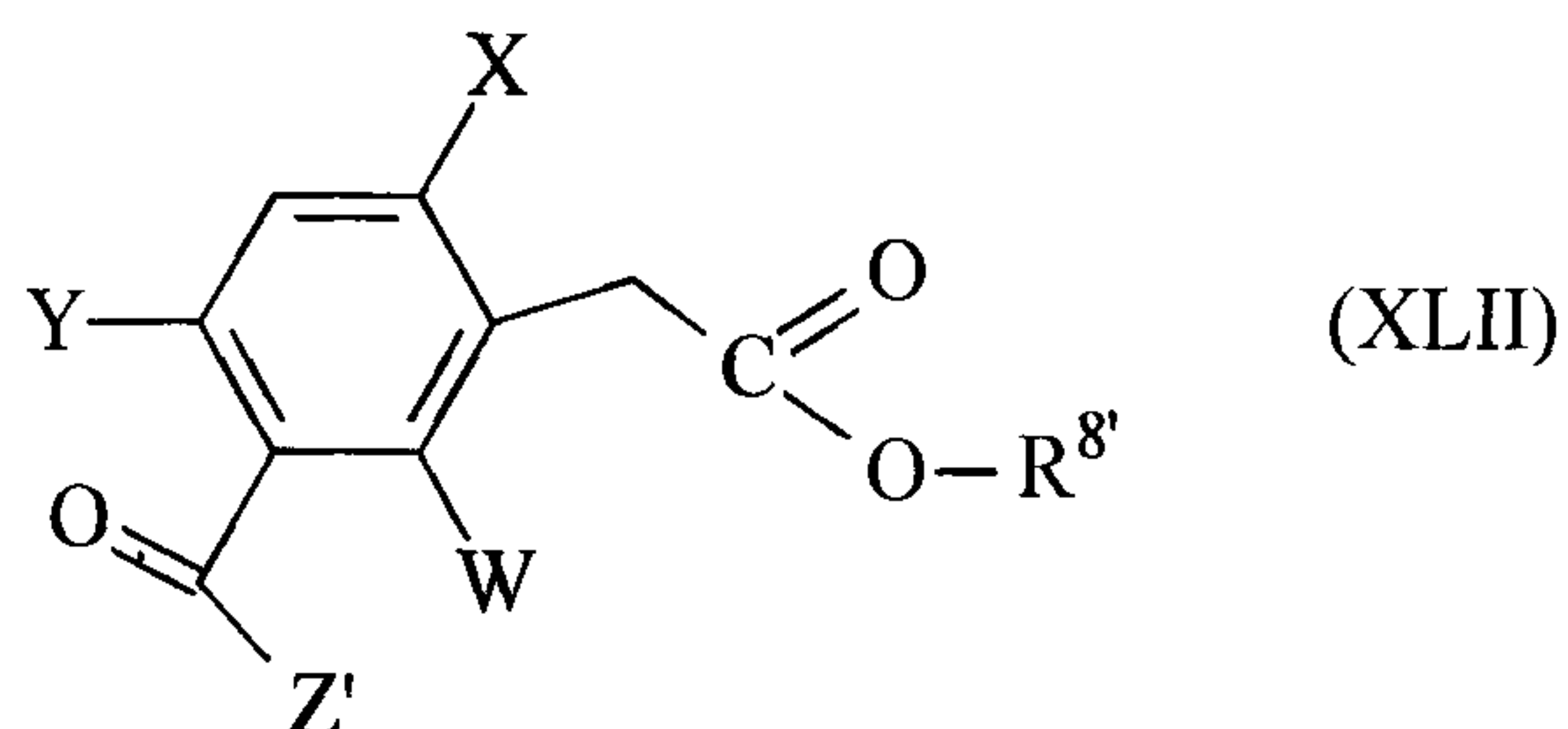
 R⁸ represents C₁-C₆ alkyl.

In a yet further compound aspect the parent invention relates to a compound of the general

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- 3e -

formula (XLII):



in which:

W, X and Y are each as defined above;

5 R^{8'} represents C₁-C₆ alkyl; and

Z' represents C₁-C₆ alkyl.

In a process aspect, the parent invention relates to a process for preparing a compound of the general formula (I) as defined above,

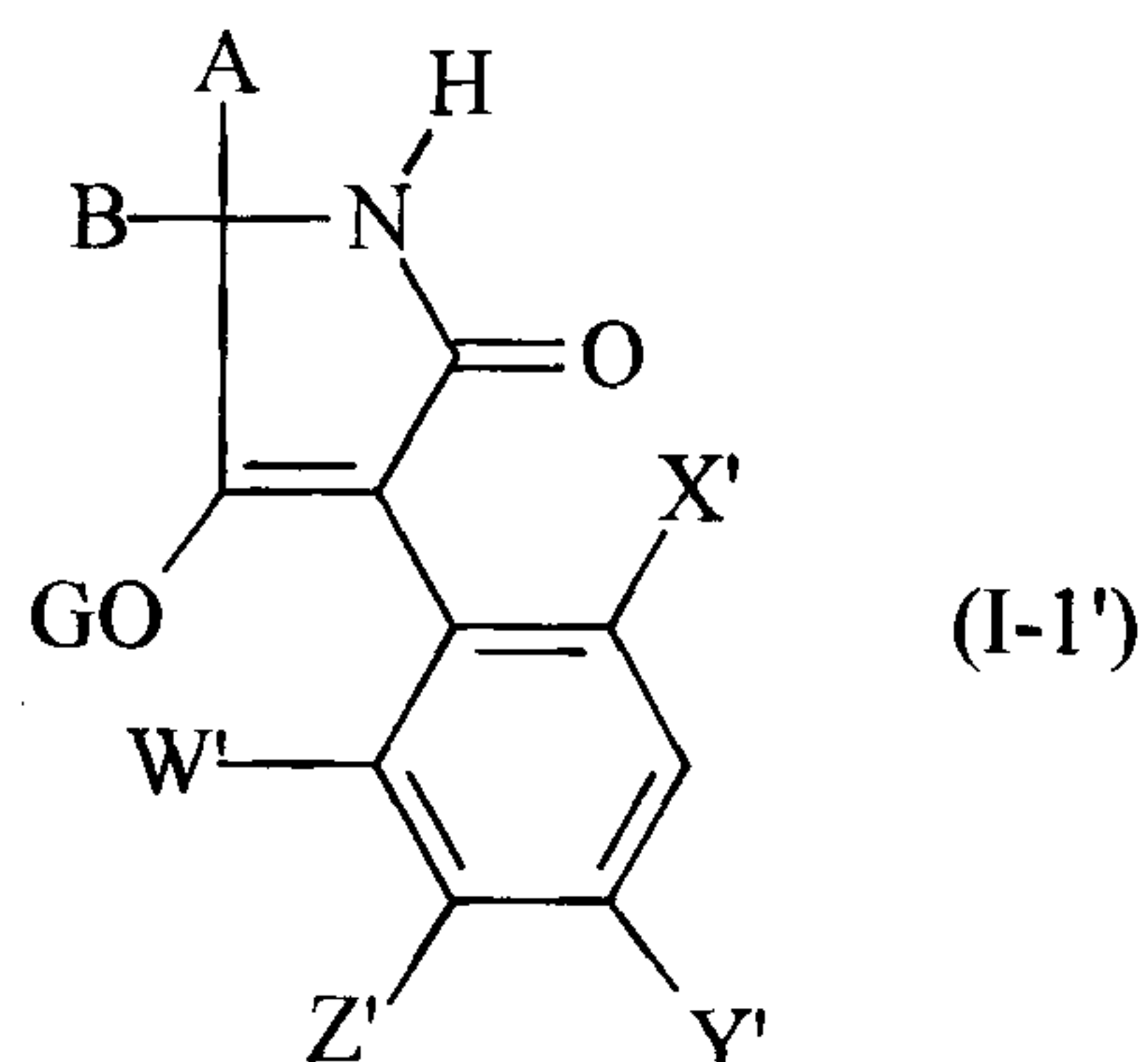
which comprises:

10 (A) the intramolecular condensation of an N-acylamino acid ester of the general formula (II) as defined above, in the presence of a diluent and in the presence of a base; or

(H) for a compound of the general formula (I), wherein one radical X or Y represents vinyl, reacting a compound of the general formula (I-1'):

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- 3f -



in which:

A, B, and G are each as defined above,

W' represents H, methyl, ethyl or i-propyl,

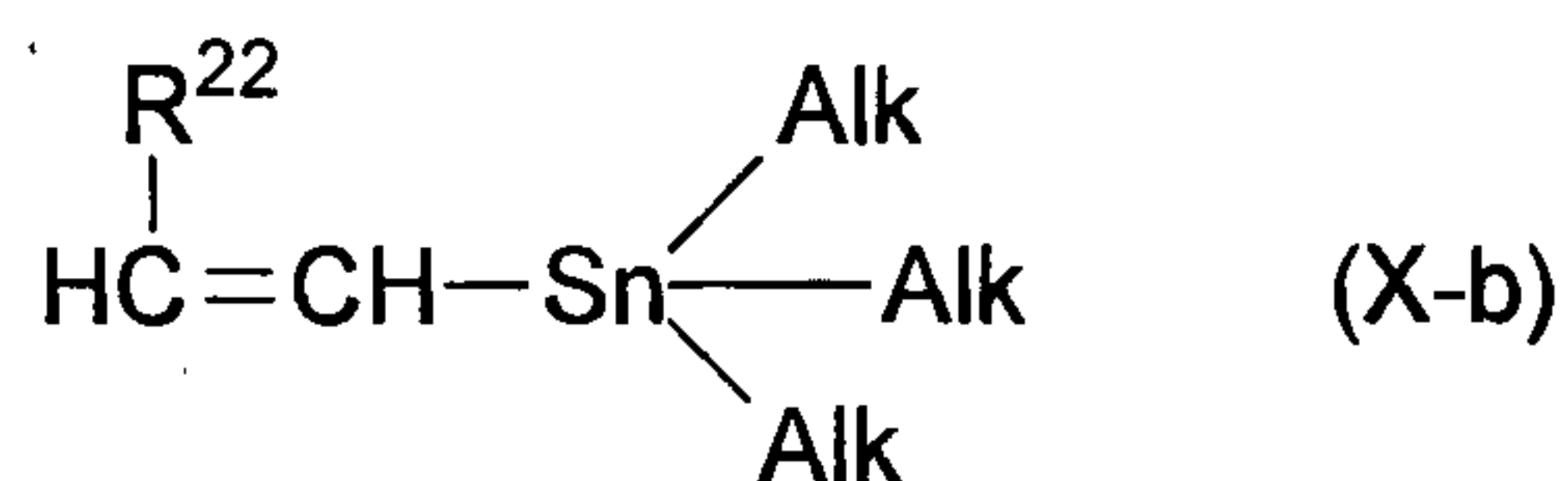
5 X' represents methyl, ethyl, i-propyl, Cl, Br or I,

Y' represents H, methyl, ethyl, i-propyl, Cl, Br or I, and

Z' represents H, methyl, ethyl, n-propyl or i-butyl,

with the provisos that: (i) at least one of the radicals W', X', Y' and Z' represent a chain having at least two carbon atoms, (ii) for preparing a compound of the general
 10 formula (I) wherein X represents vinyl, a compound of the general formula (I-1'), wherein X' represents Cl, Br or I, is reacted with a compound of the general formula (X-b), as defined below, and (iii) for preparing a compound of the general formula (I) wherein Y represents vinyl, a compound of the general formula (I-1'), wherein Y' represents Cl, Br or I, is reacted with a compound of the general formula (X-b), as defined below,

15 with a vinylstannane of the general formula (X-b):



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- 3g -

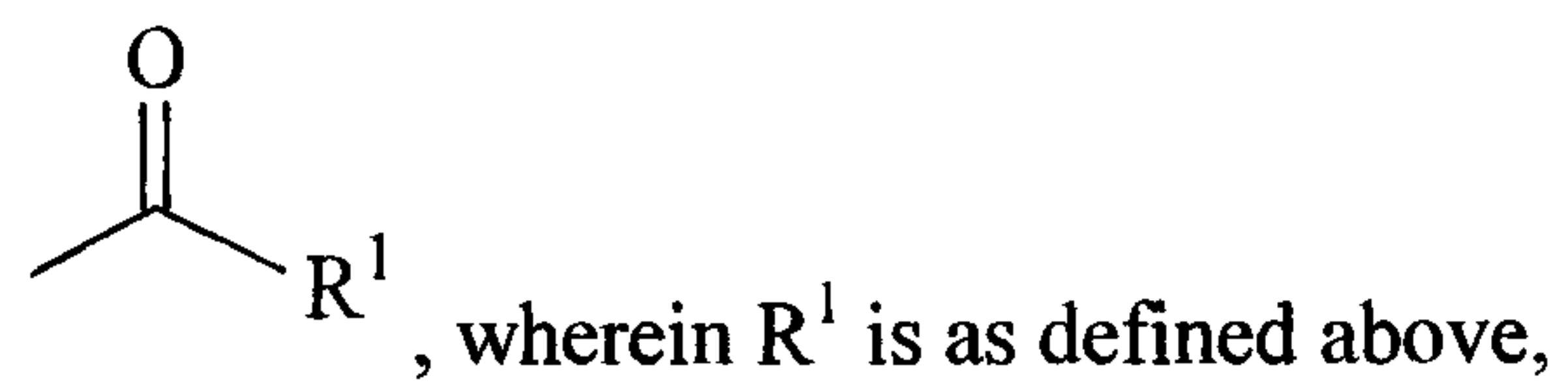
in which:

alk represents C₁-C₄-alkyl, andR²² represents hydrogen,

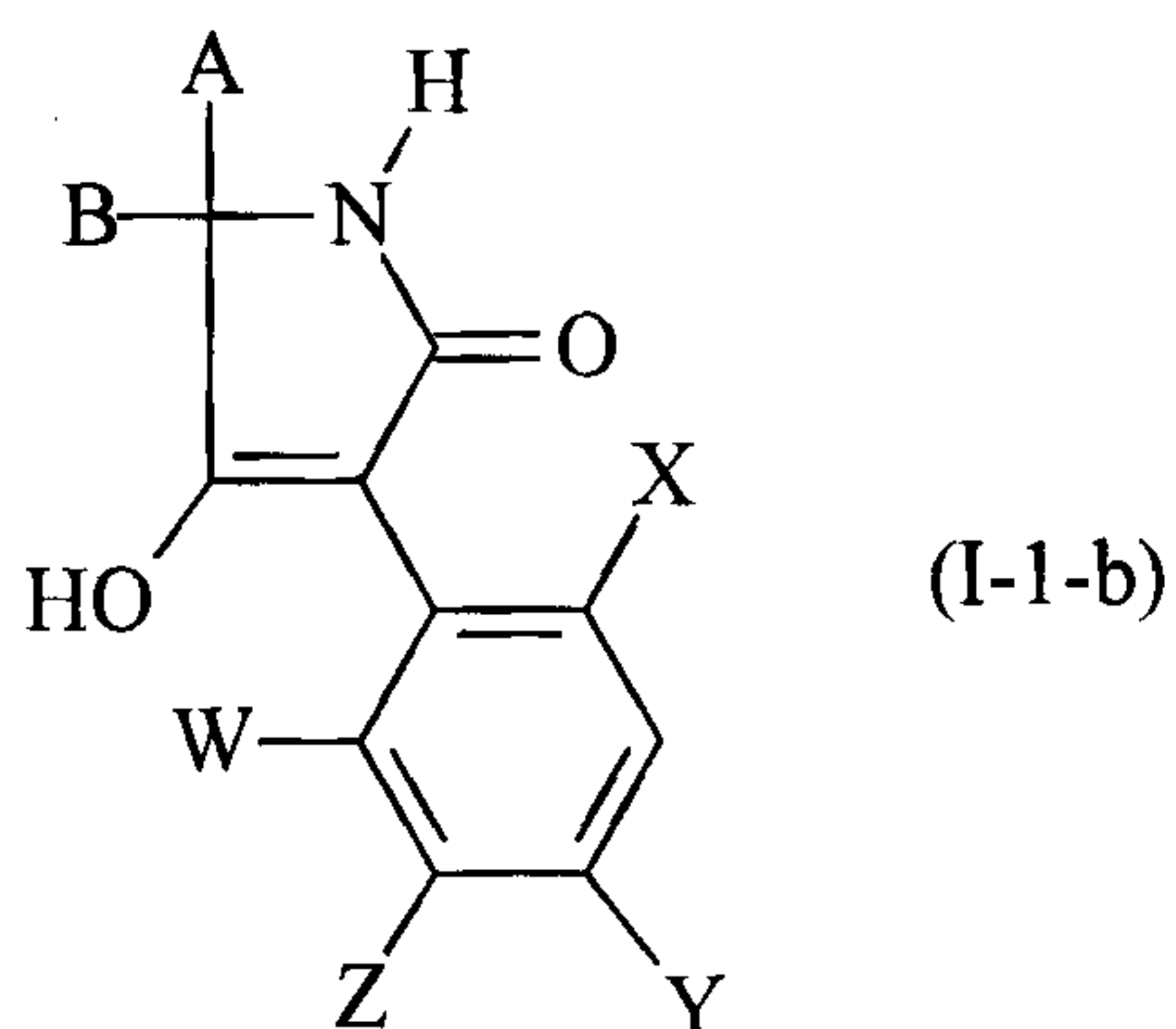
in the presence of a solvent and, optionally, in the presence of a base and a

5 catalyst; or

(I) for a compound of the general formula (I), wherein G represents:

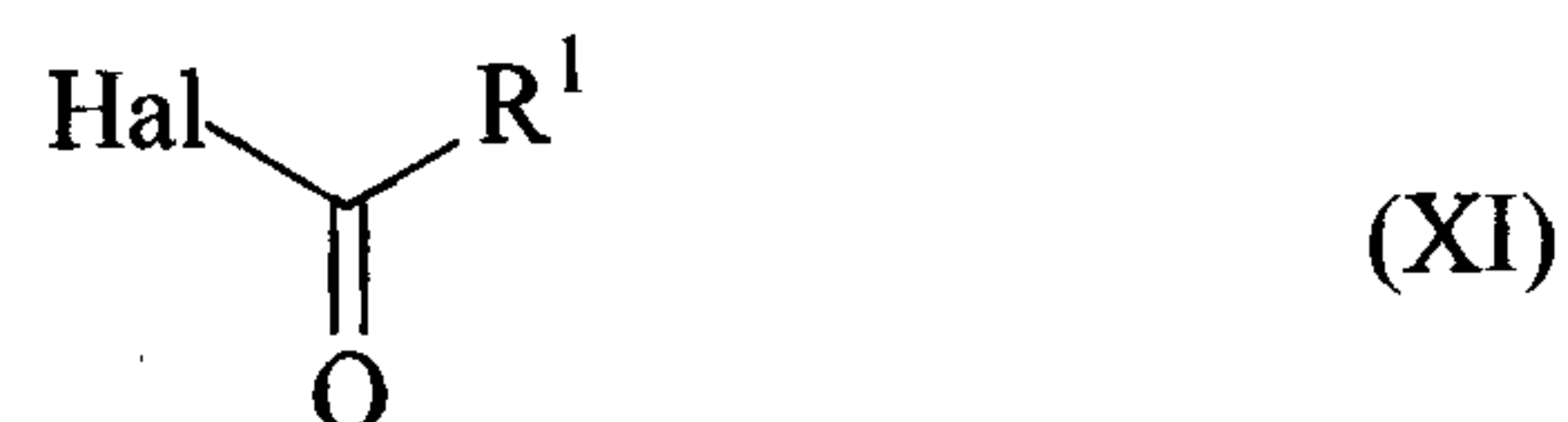


by reacting a component of the general formula (I-1-b):



10 in which A, B, W, X, Y and Z are each as defined above, with:

(α) an acid halide of the general formula (XI):



in which:

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- 3h -

R^1 is as defined above, and

Hal represents a halogen atom,

or

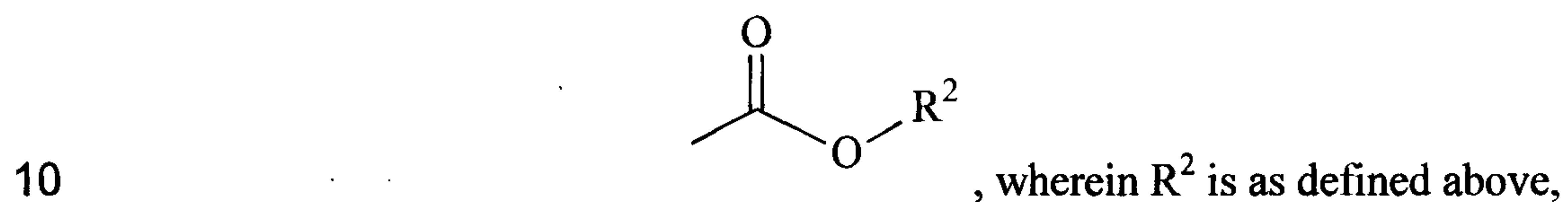
(β) a carboxylic anhydride of the general formula (XII):



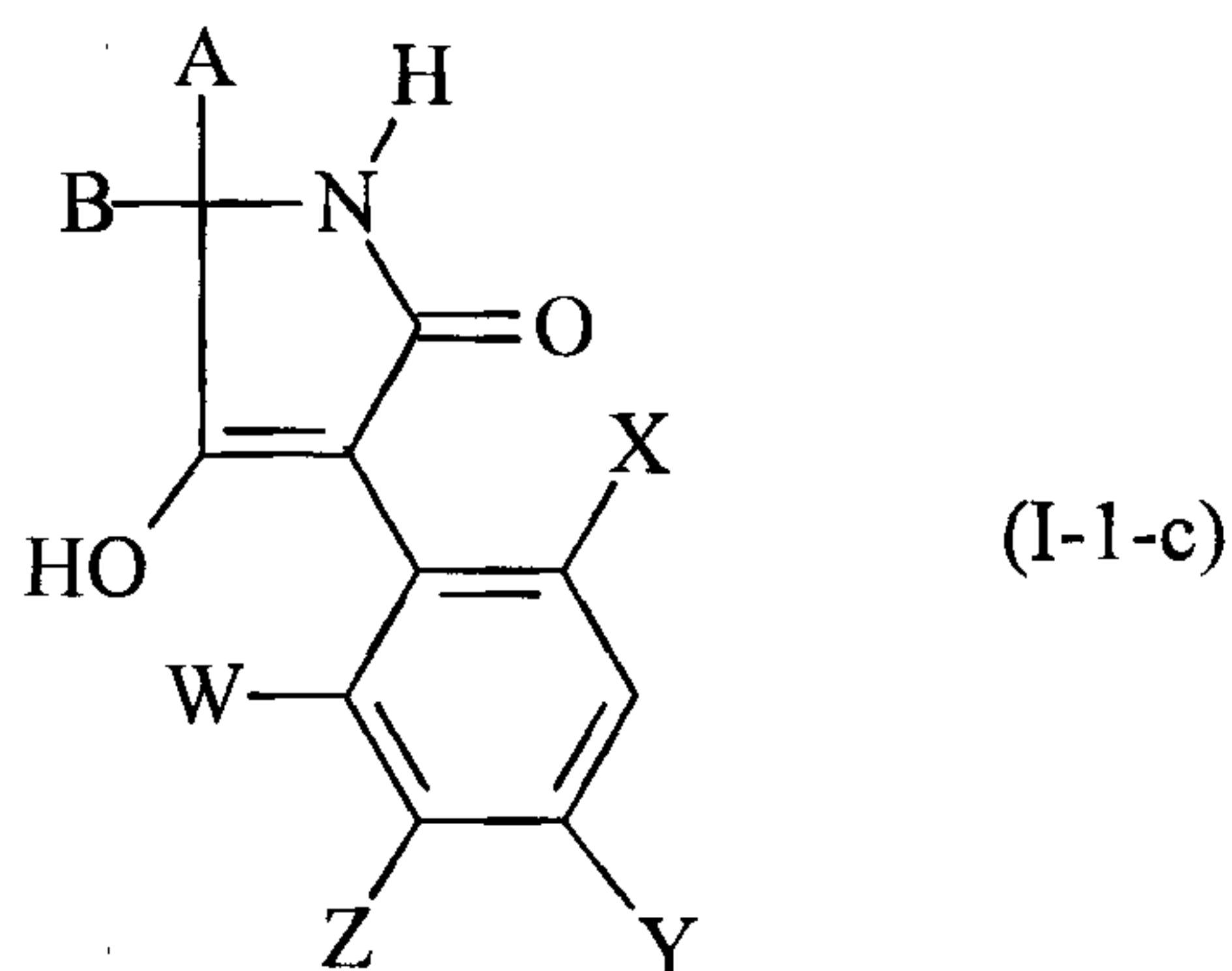
in which R^1 is as defined above,

optionally in the presence of a diluent and, optionally, in the presence of an acid binder; or

(J) for a compound of the general formula (I), wherein G represents



by reacting a compound of the general formula (I-1-c):



in which A, B, W, X, Y and Z are each as defined above, with a chloroformic ester of the general formula (XIII):

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- 3i -



in which R^2 is as defined above,

optionally in the presence of a diluent and, optionally, in the presence of an acid binder.

- 5 The parent invention further relates to a pesticide or a herbicide, comprising at least one compound of the general formula (I) as defined above.

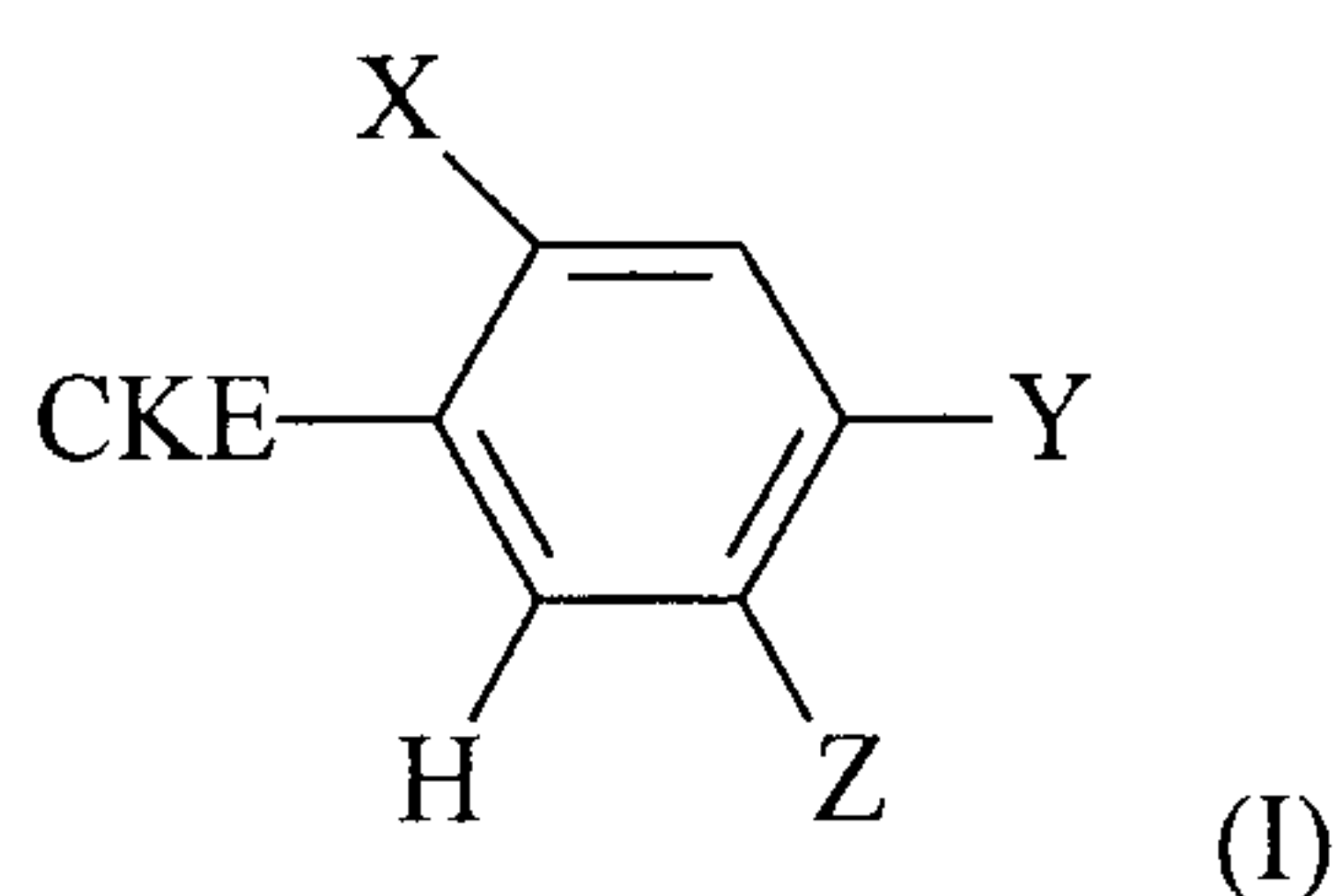
The parent invention further relates to a method for controlling animal pests and undesirable vegetation, wherein at least one compound of the general formula (I) as defined above is allowed to act on the pests or their habitat, or the undesirable vegetation.

- 10 The parent invention further relates to use of a compound of the general formula (I) as defined above for controlling animal pests and undesirable vegetation.

The parent invention further relates to a process for preparing a pesticide or herbicide, comprising mixing at least one compound of the general formula (I) as defined above with at least one of an extender or a surfactant.

- 15 The parent invention further relates to use of a compound of the general formula (I) as defined above for preparing a pesticide or a herbicide.

In one compound aspect, the divisional invention relates to a compound of the general formula (I):



- 20 in which:

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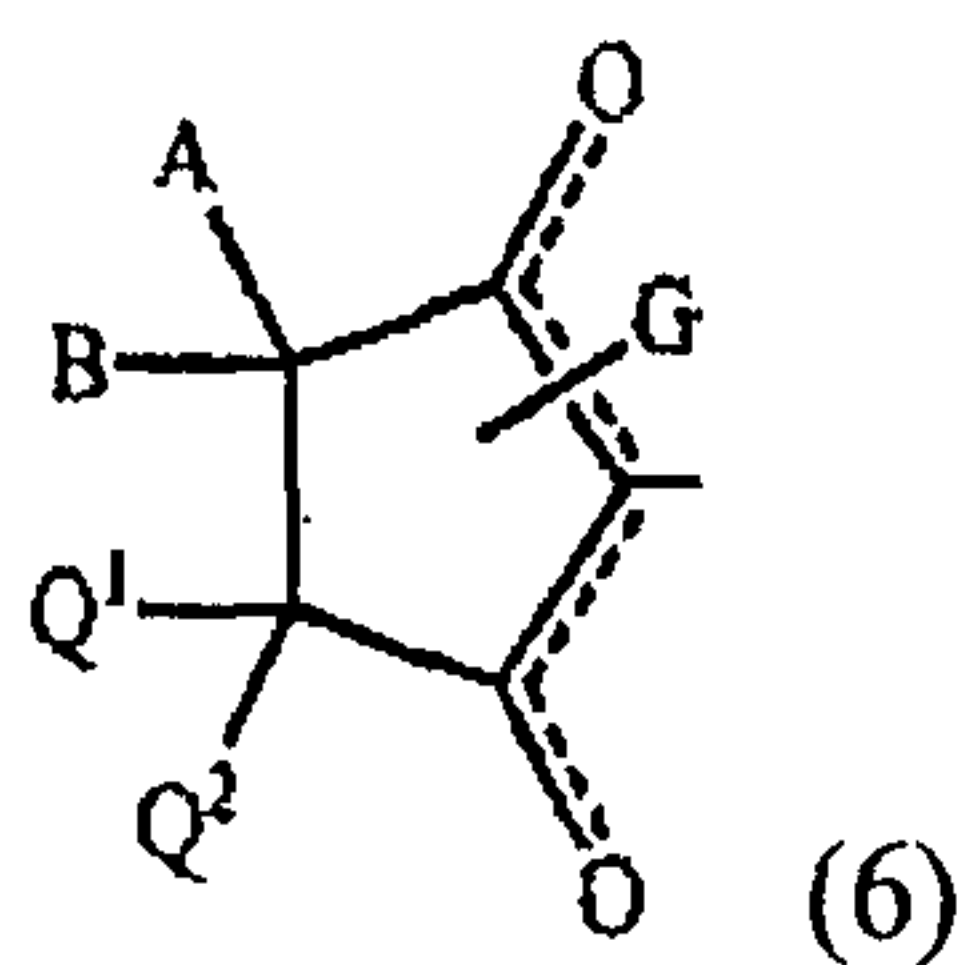
X represents methyl, ethyl, i-propyl or vinyl;

Y represents H, methyl, ethyl, i-propyl, vinyl or ethinyl;

Z represents H, methyl, ethyl, n-propyl or i-butyl,

with the provisos that: (i) at least one of the radicals X, Y or Z represents a chain having at
 5 least two carbon atoms, and (ii) only one of the radicals X or Y represents vinyl; and

CKE represents the group:



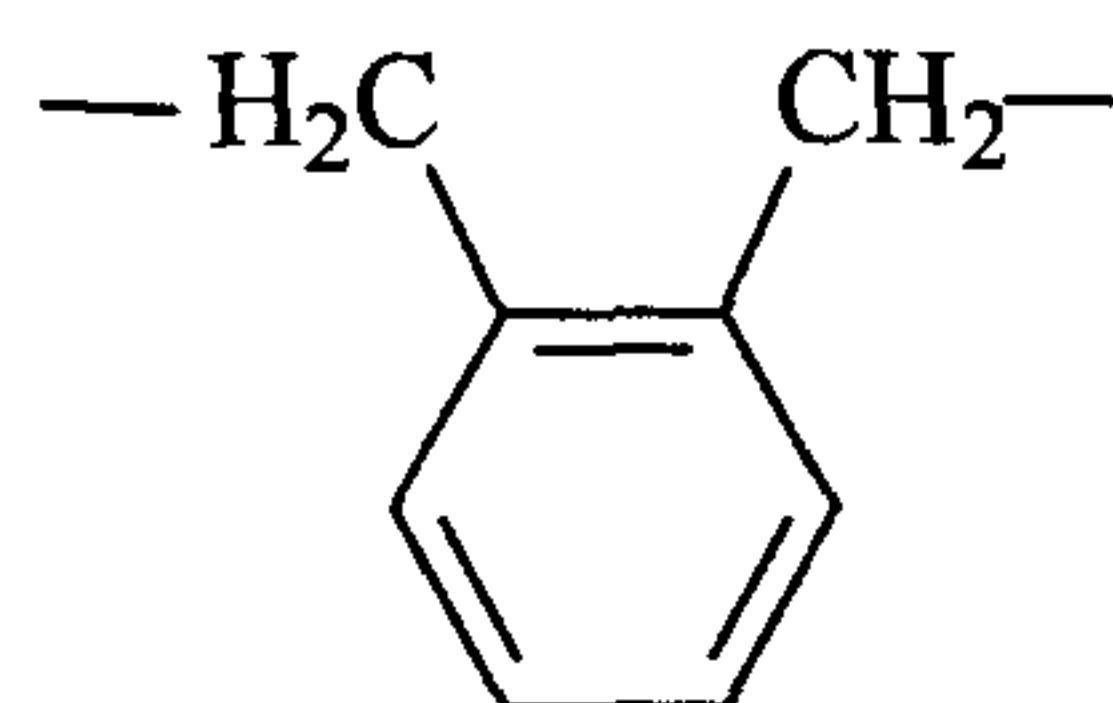
wherein:

A represents methyl,

10 B represents methyl, or

A, B and the carbon atom to which they are attached represent saturated C₅-C₆-cycloalkyl in which optionally one ring member is replaced by O and which is optionally mono substituted by methyl, ethyl, methoxy or ethoxy, or

A and B together represent:



15 , or

when B represents methyl, A and Q¹ together represent C₃-C₄-alkanediyl, or

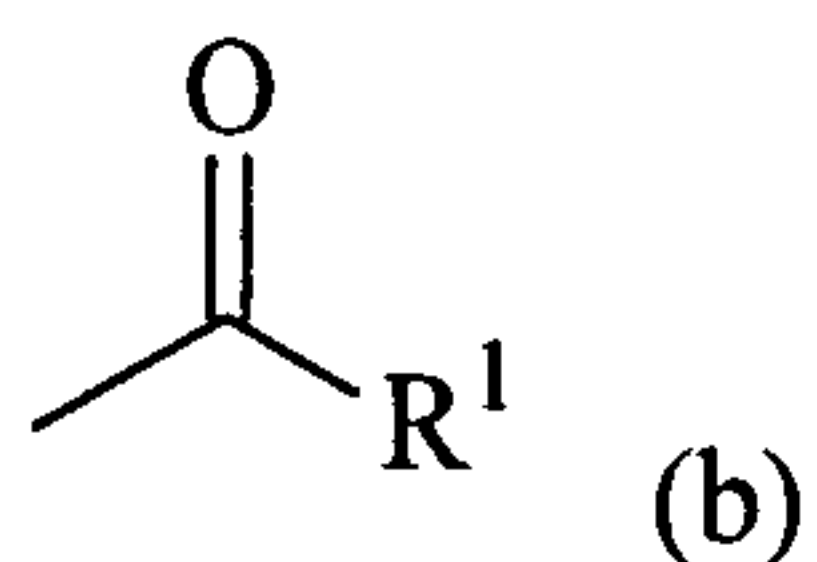
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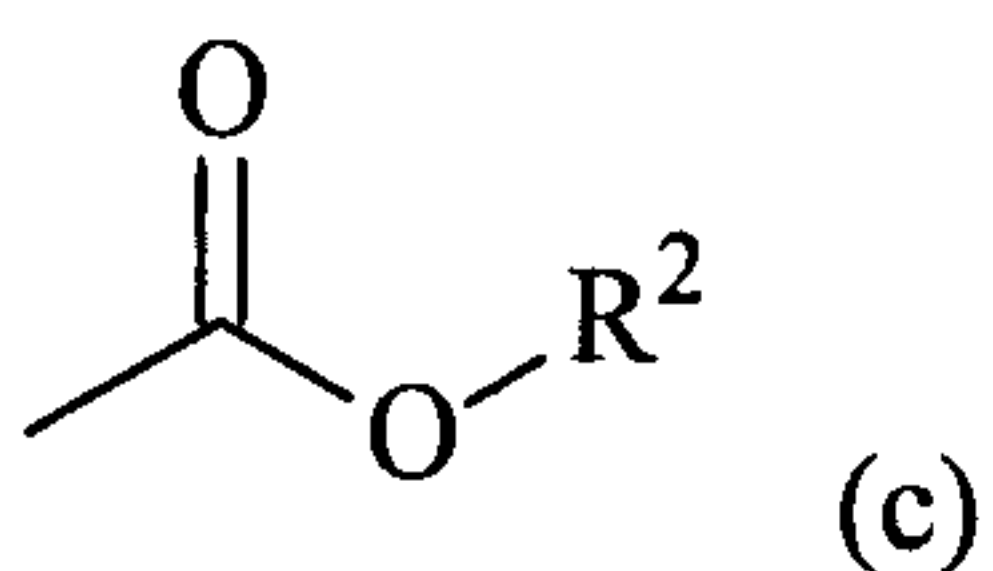
when A represents methyl, B and Q^2 each represent H, or

Q_1 and Q_2 represent H,

G represents a hydrogen atom or one of the groups:



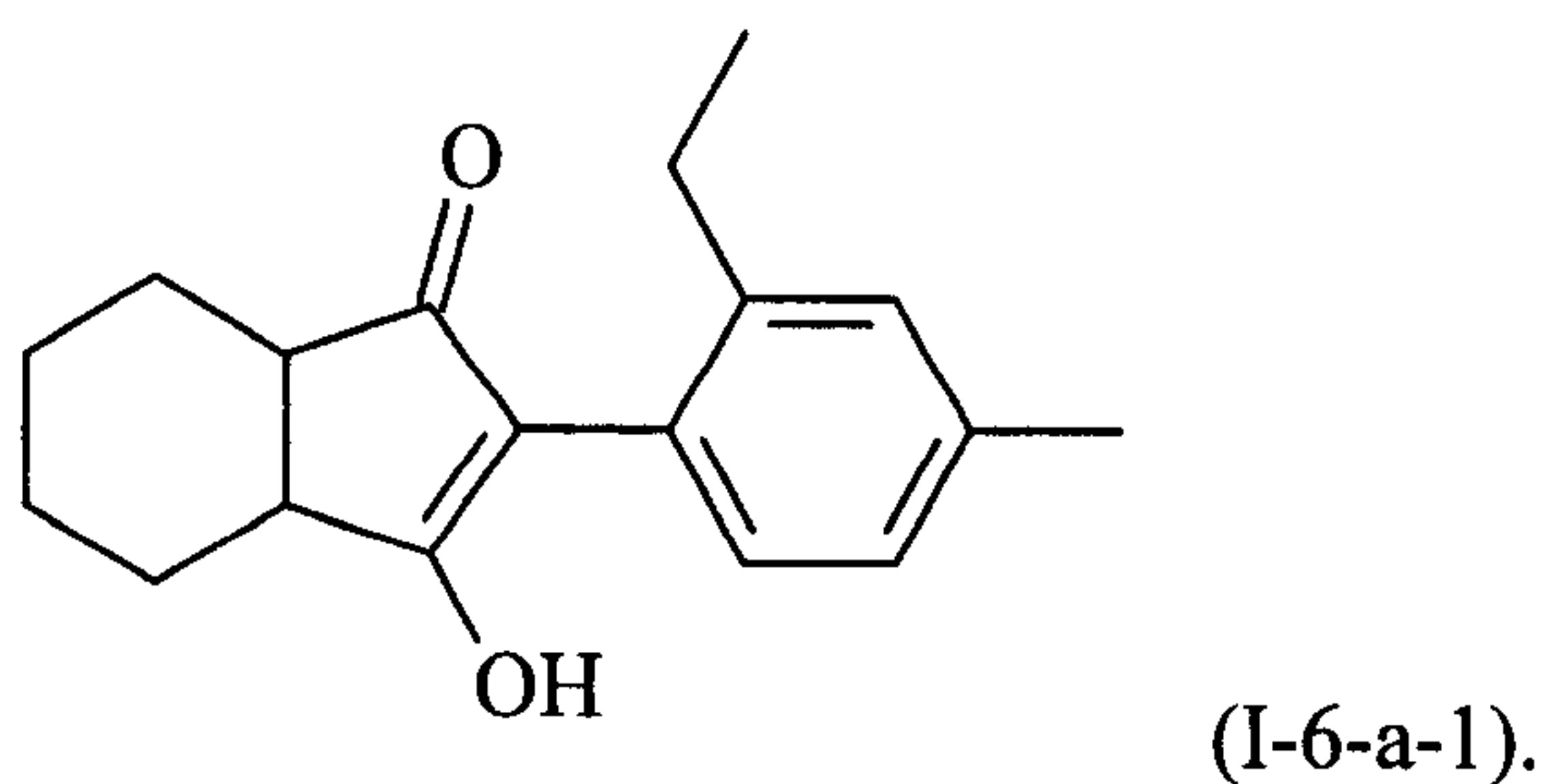
5 or



R^1 represents C_1 - C_6 alkyl or C_1 - C_2 -alkoxymethyl, and

R^2 represents C_1 - C_4 -alkyl.

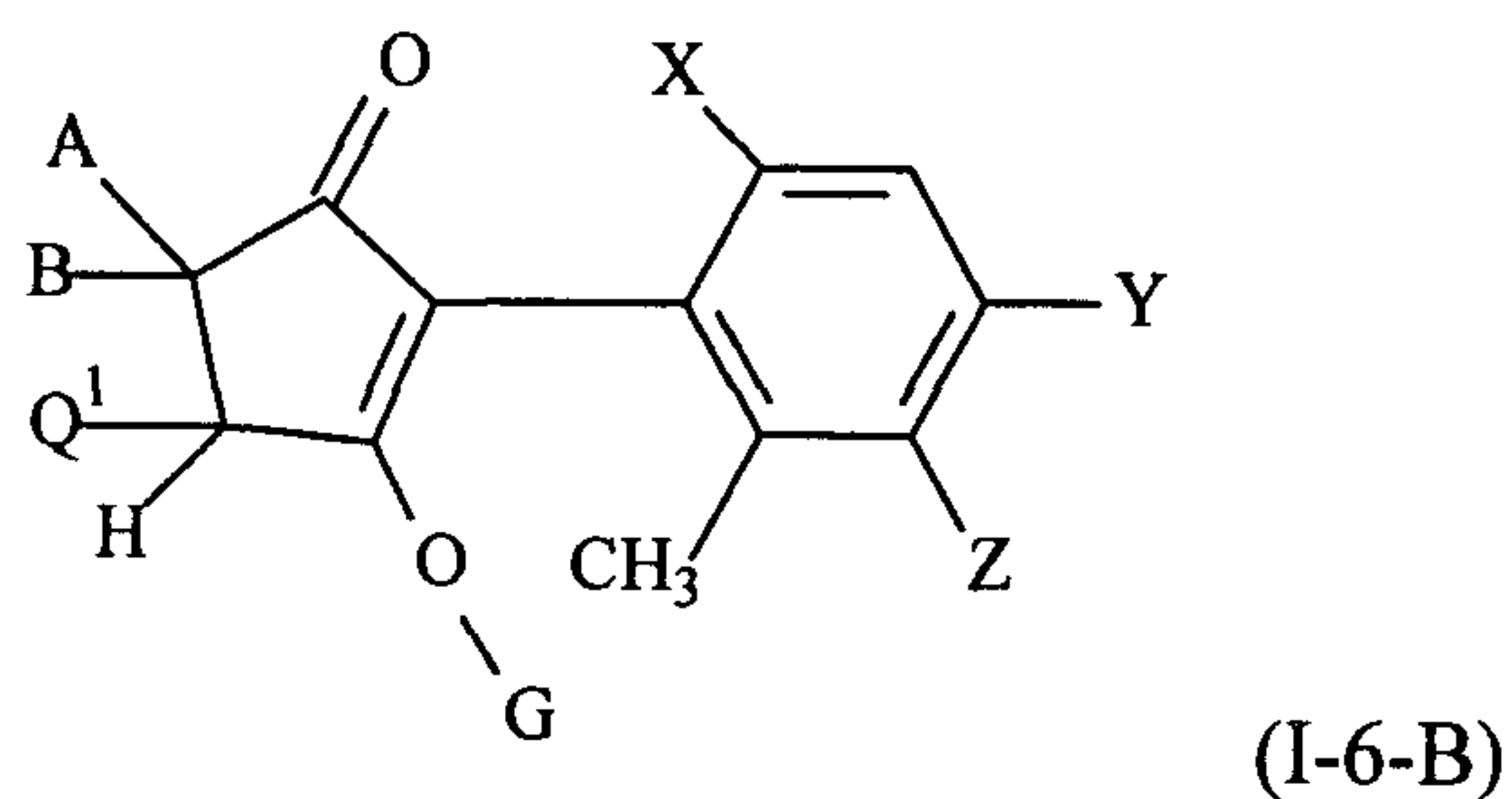
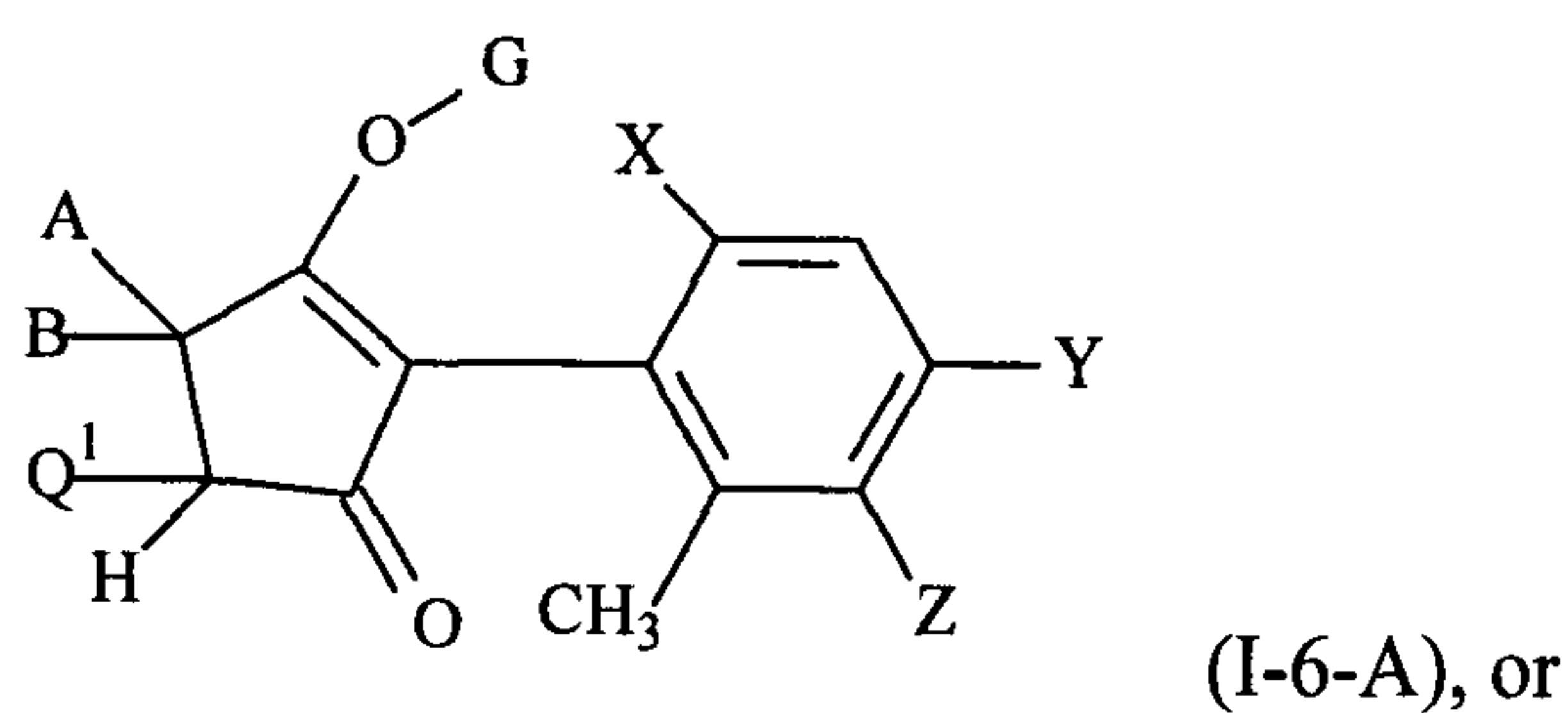
10 In a further compound aspect, the divisional invention relates to a compound of the formula (I-6-a-1):



In a still further compound aspect, the divisional invention relates to a compound of the general formula (I-6-A) or (I-6-B):

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in which:

X represents methyl, ethyl, n-propyl, iso-propyl, vinyl or ethinyl;

5 Y represents H, methyl, ethyl, i-propyl, vinyl or ethinyl;

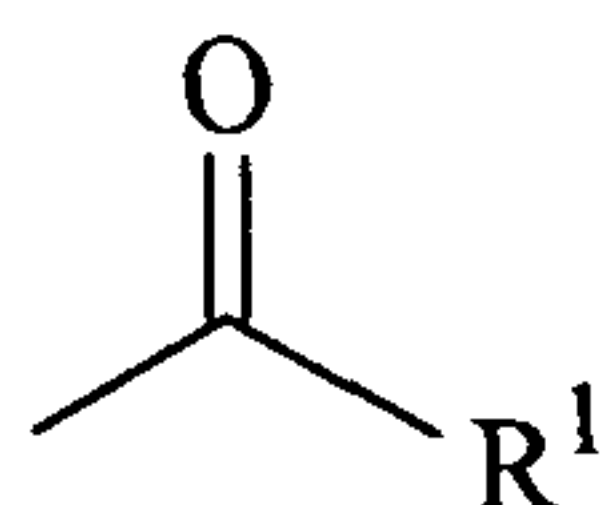
Z represents H, methyl, ethyl, n-propyl, i-butyl, vinyl or ethinyl;

with the provisos that: (i) at least one of the radicals X, Y or Z represents a chain having at least two carbon atoms, (ii) at most only one of the radicals X, Y or Z represents vinyl or ethinyl, and (iii) X is not ethyl when Z is H and Y is methyl;

10 B represents H, methyl or ethyl;

A and Q¹ together represent C₄-alkanediyl; and

G represents a hydrogen atom or the group:



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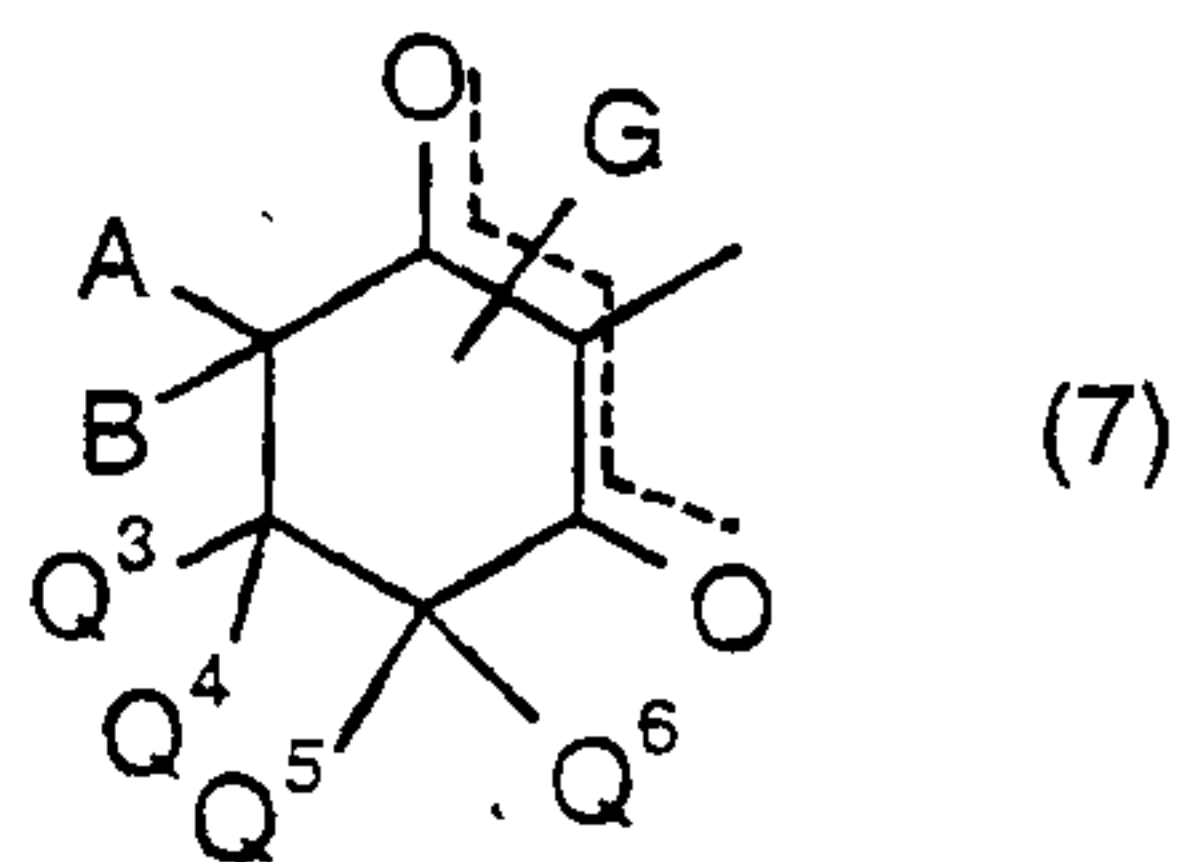
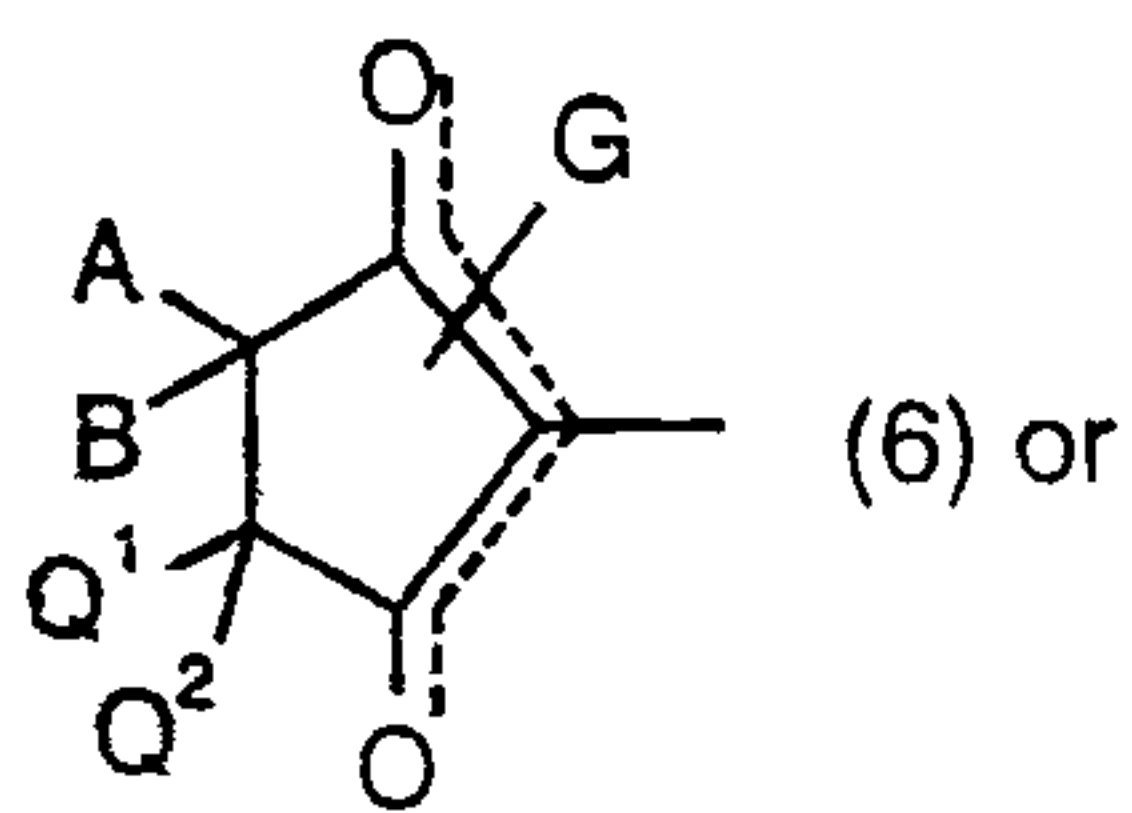
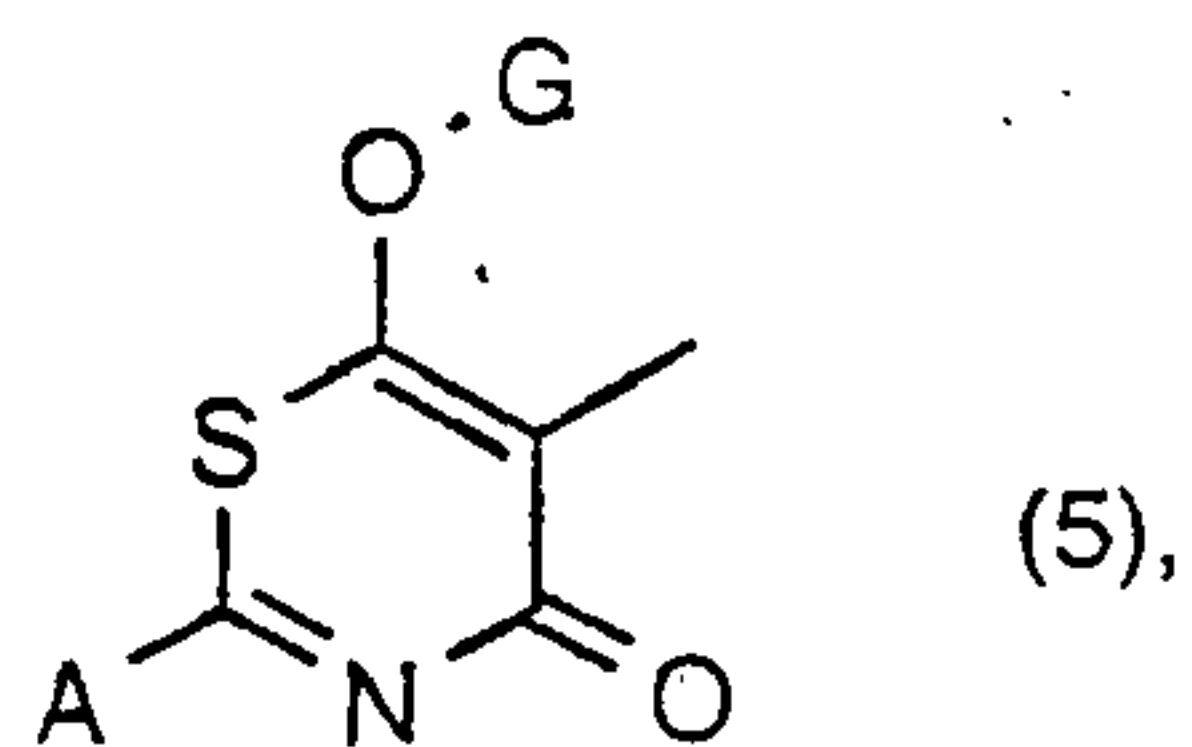
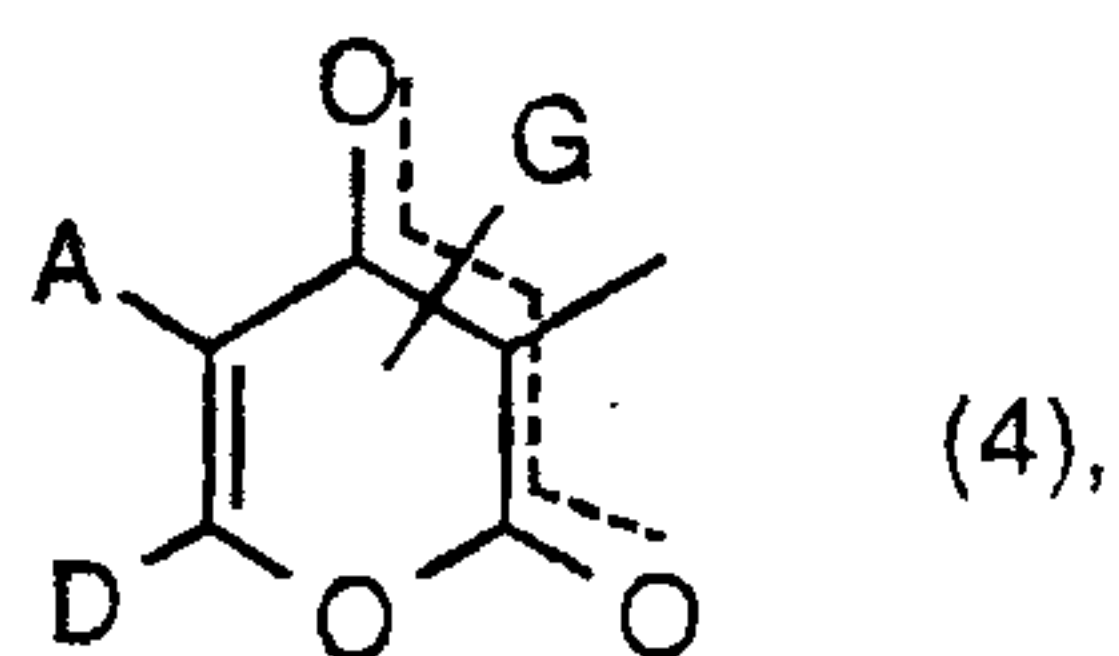
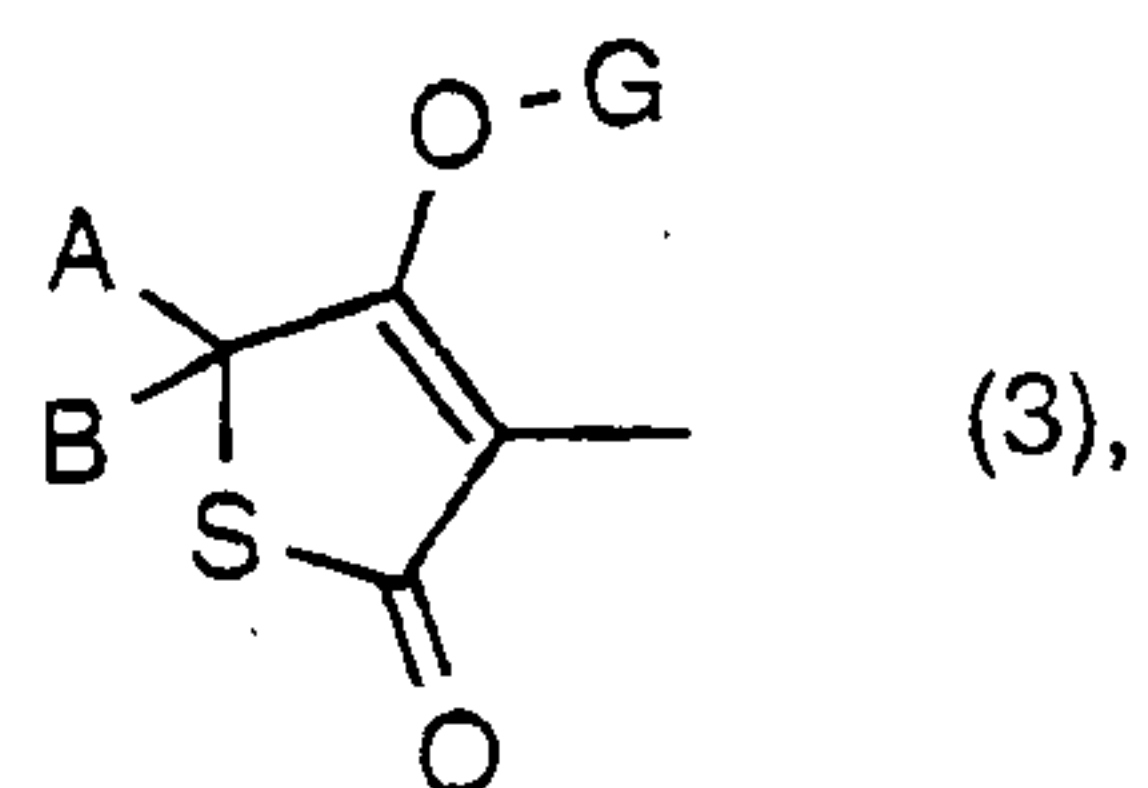
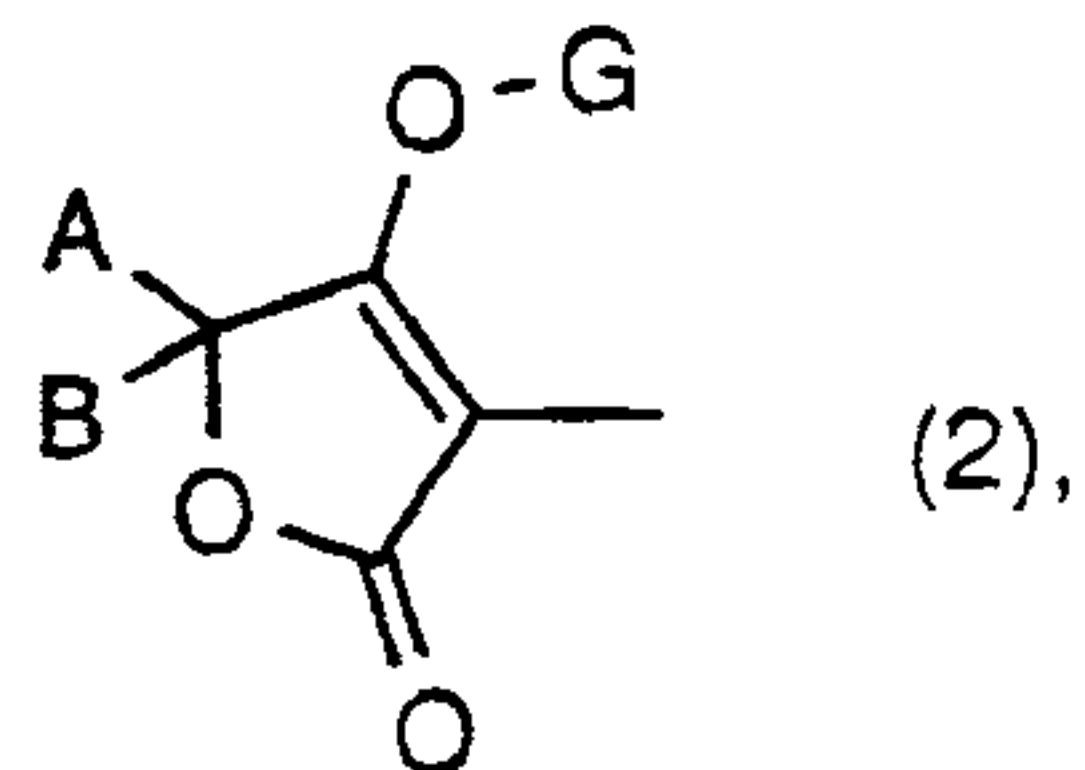
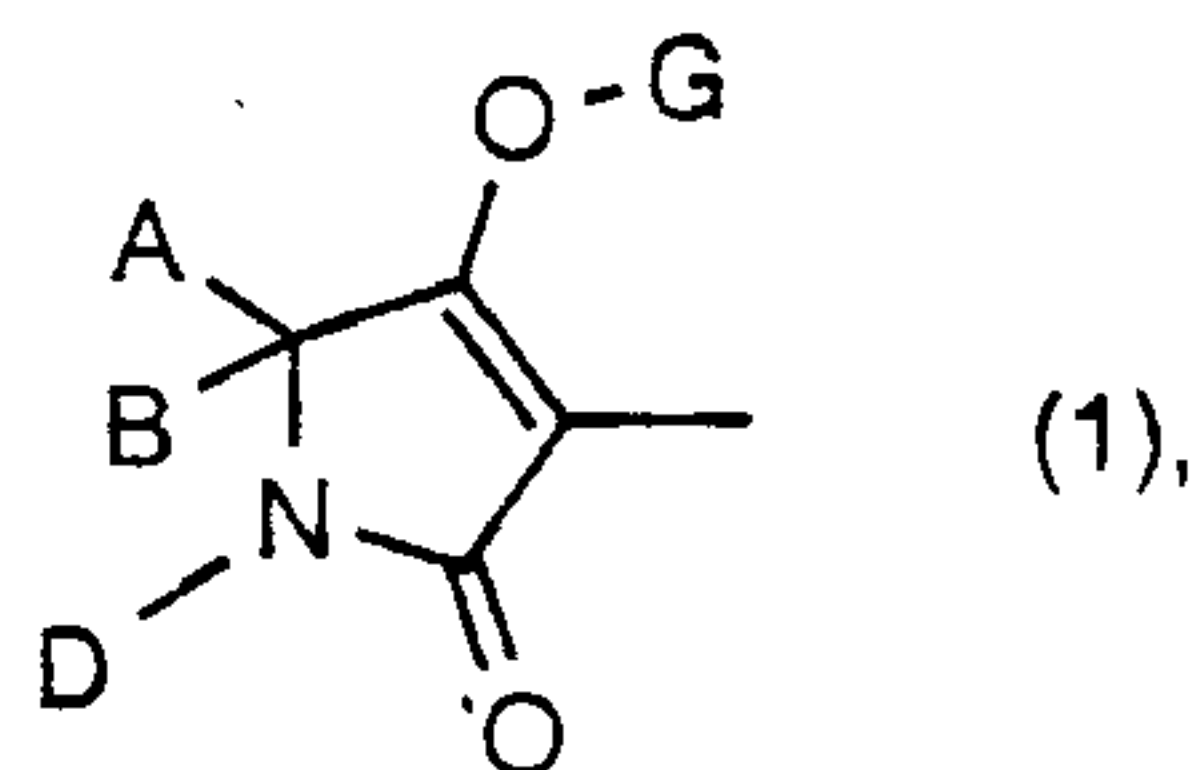
X represents alkyl, alkenyl or alkynyl,

Y represents hydrogen, methyl, ethyl, i-propyl, alkenyl or alkynyl,

5 Z represents hydrogen, alkyl, alkenyl or alkynyl,

with the proviso that at least one of the radicals W, X, Y or Z represents a chain having at least two carbon atoms,

10 CKE represents one of the groups



15

in which

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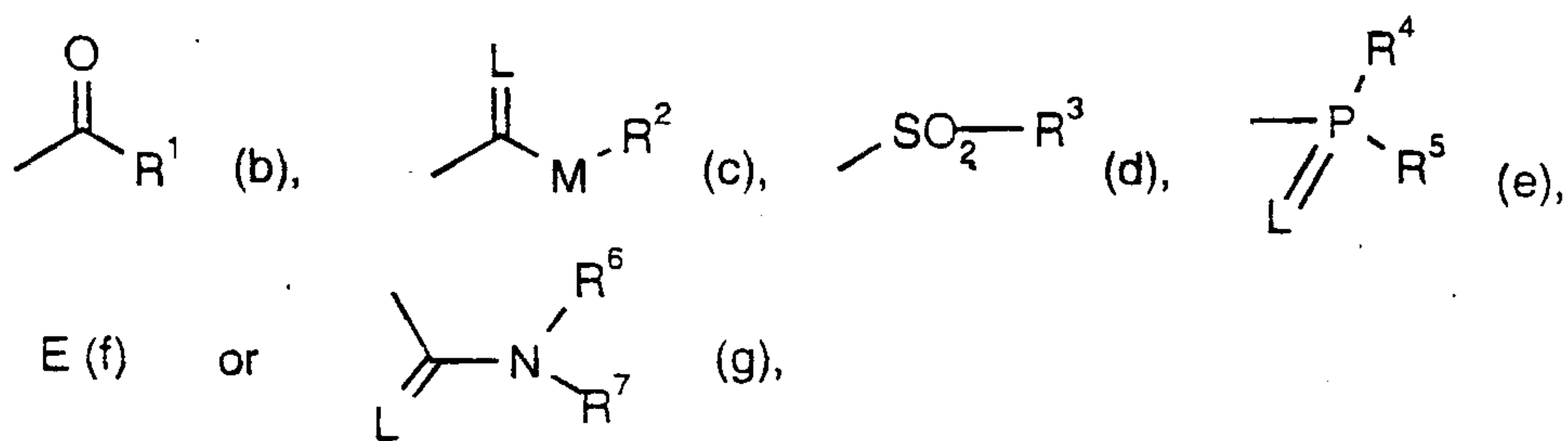
- 6 -

Q², Q⁴, Q⁵ and Q⁶ independently of one another each represent hydrogen or alkyl,

Q³ represents hydrogen, alkyl, alkoxyalkyl, alkylthioalkyl, optionally substituted cycloalkyl (in which optionally one methylene group is replaced by oxygen or sulphur) or optionally substituted phenyl, or

Q³ and Q⁴ together with the carbon atom to which they are attached represent a saturated or unsaturated unsubstituted or substituted cycle which optionally contains a heteroatom,

G represents hydrogen (a) or represents one of the groups



in which

E represents a metal ion equivalent or an ammonium ion,

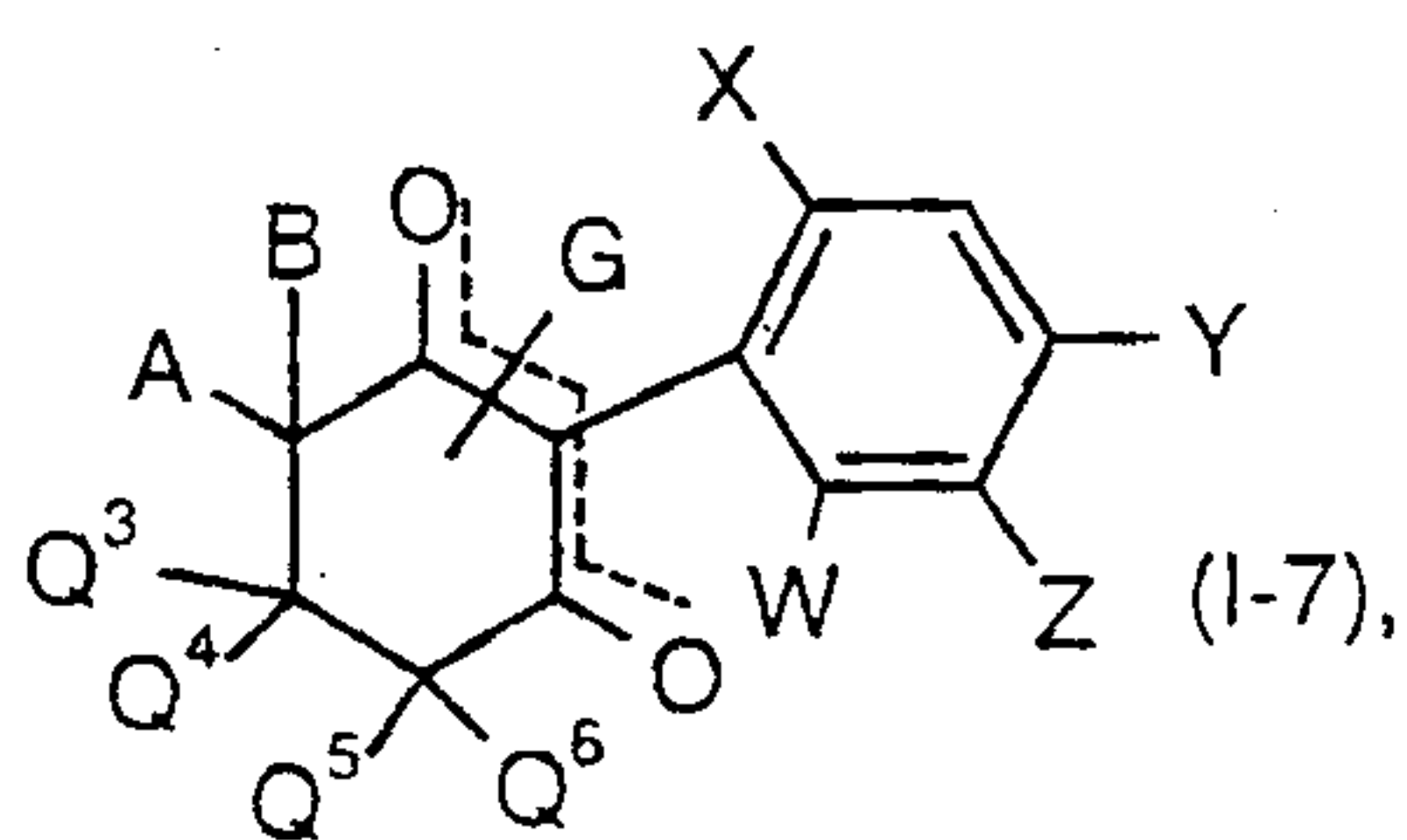
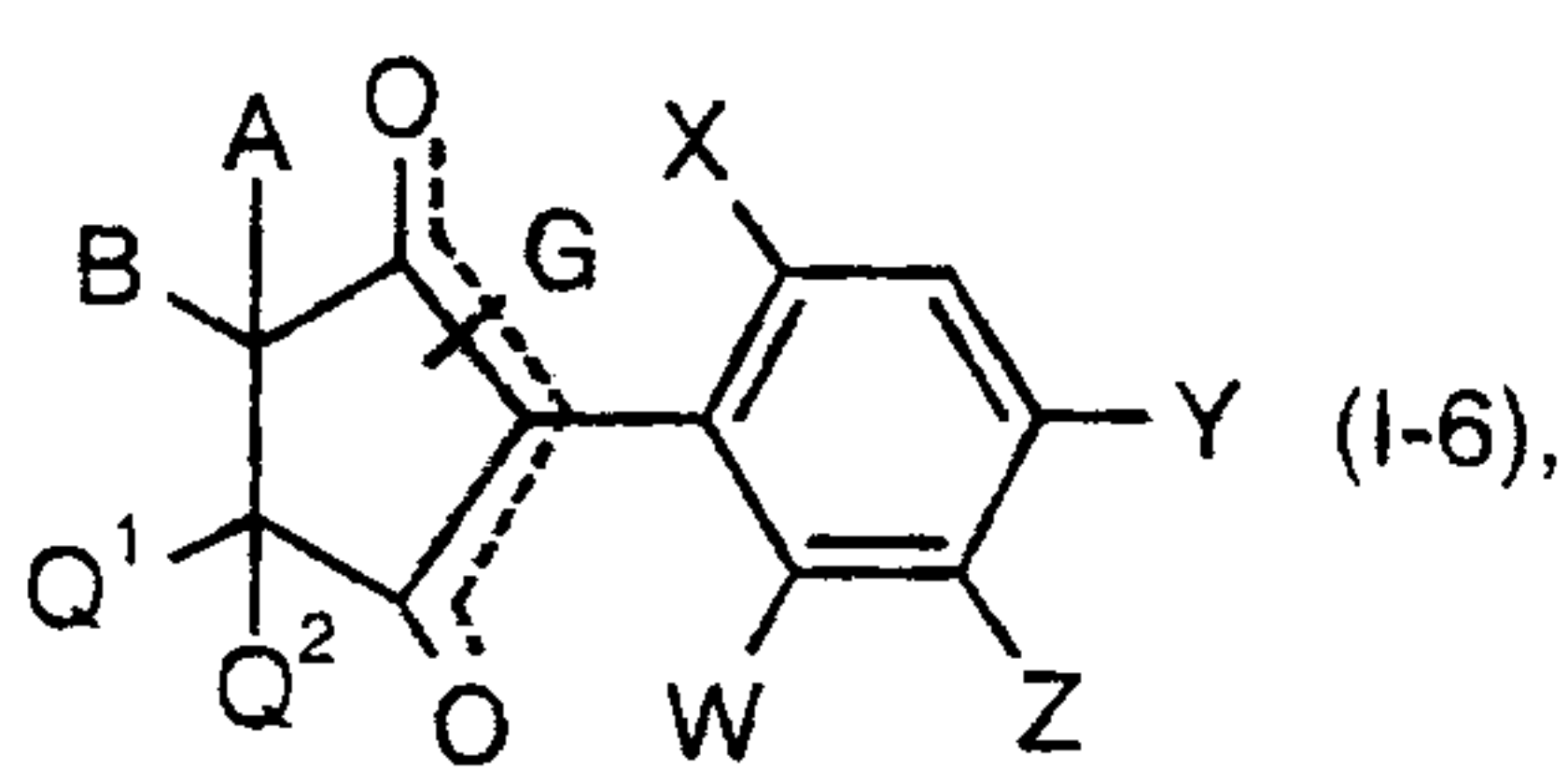
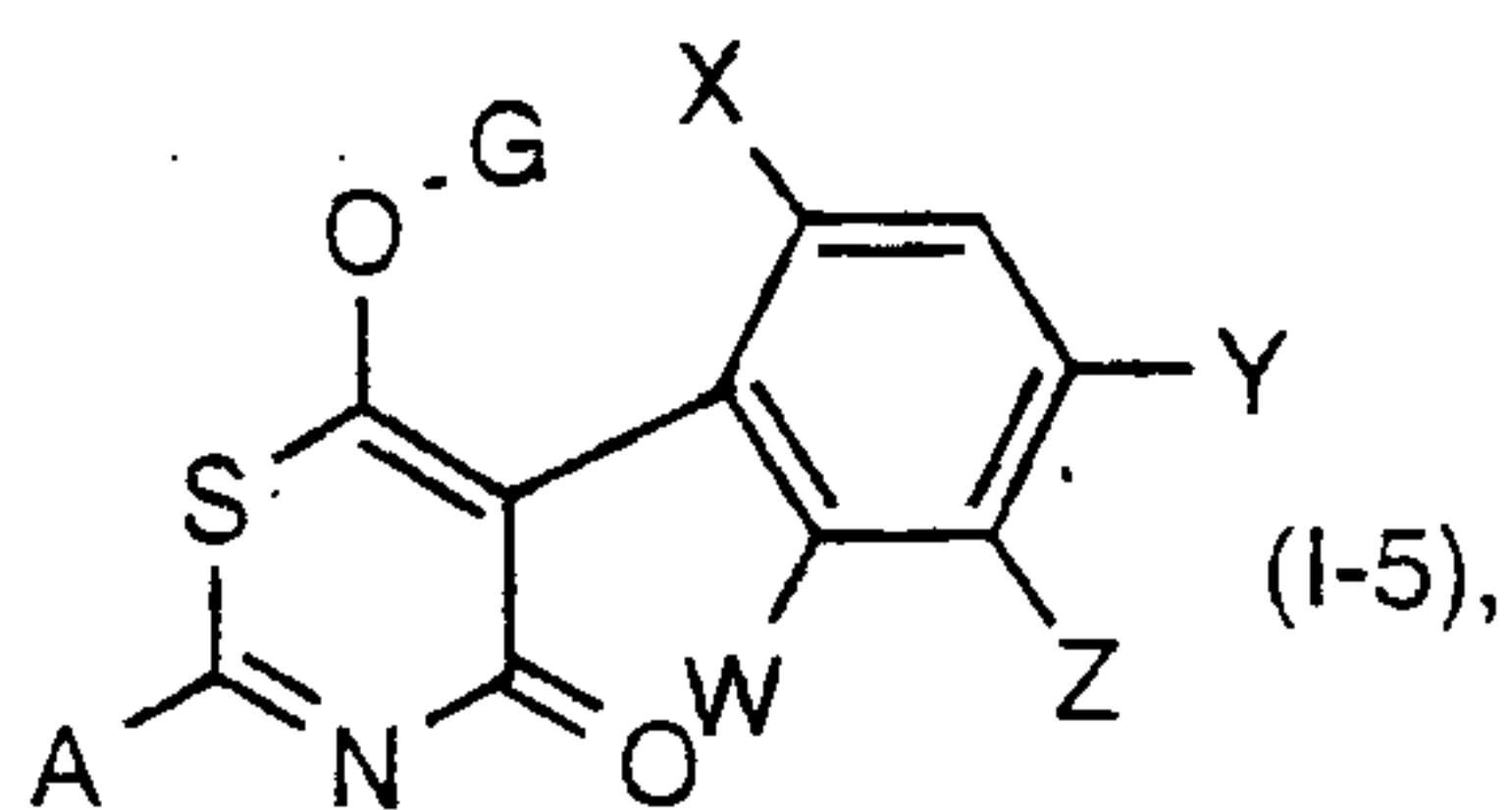
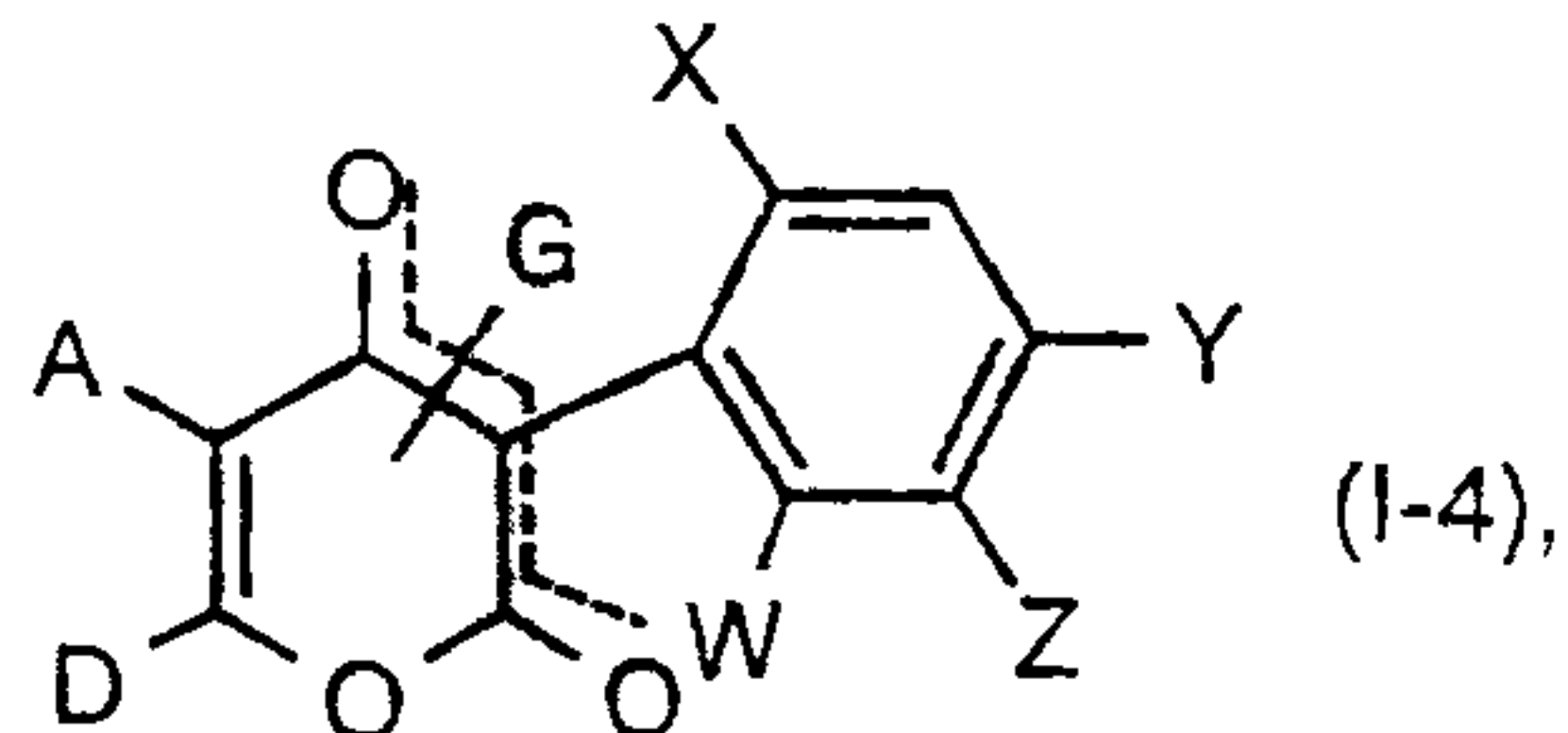
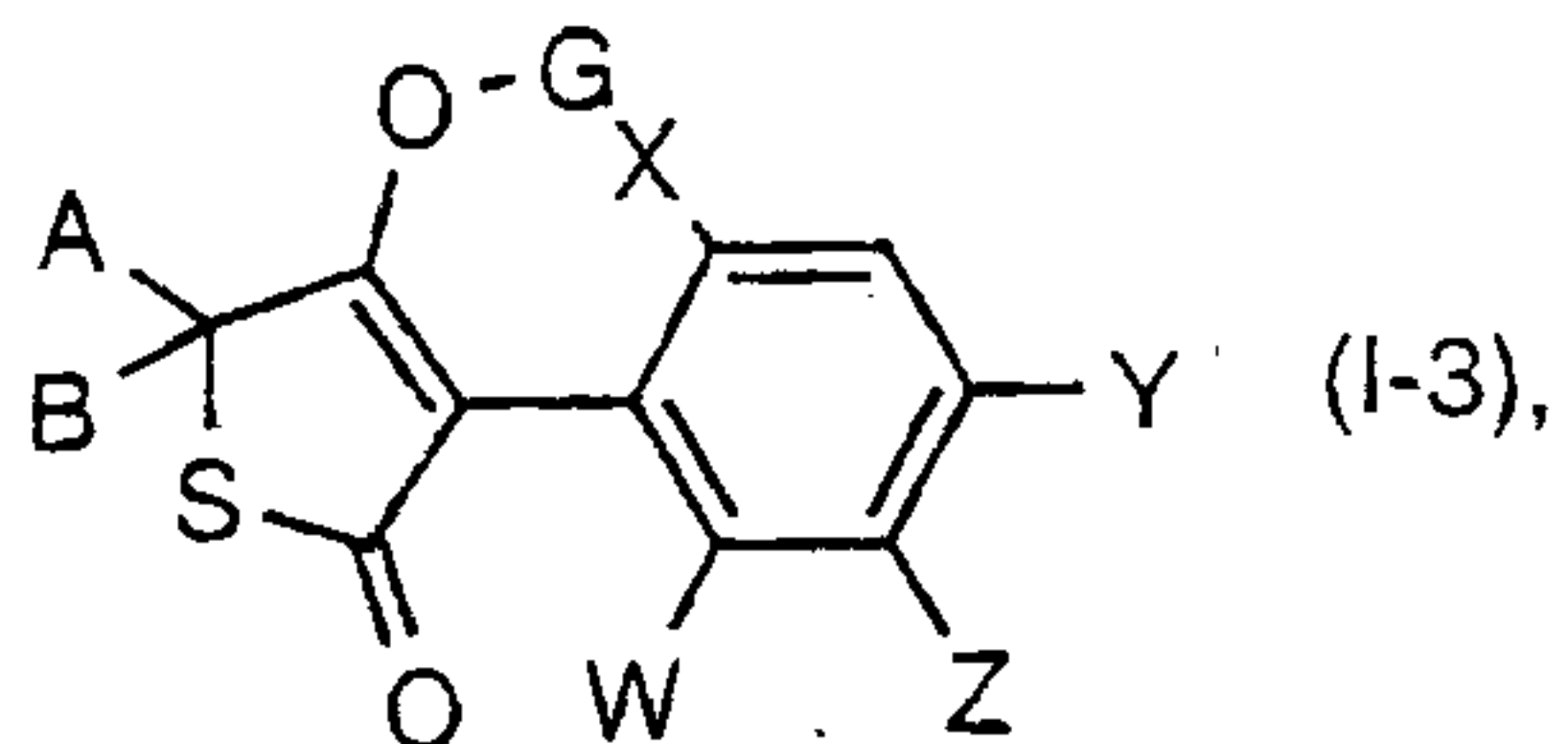
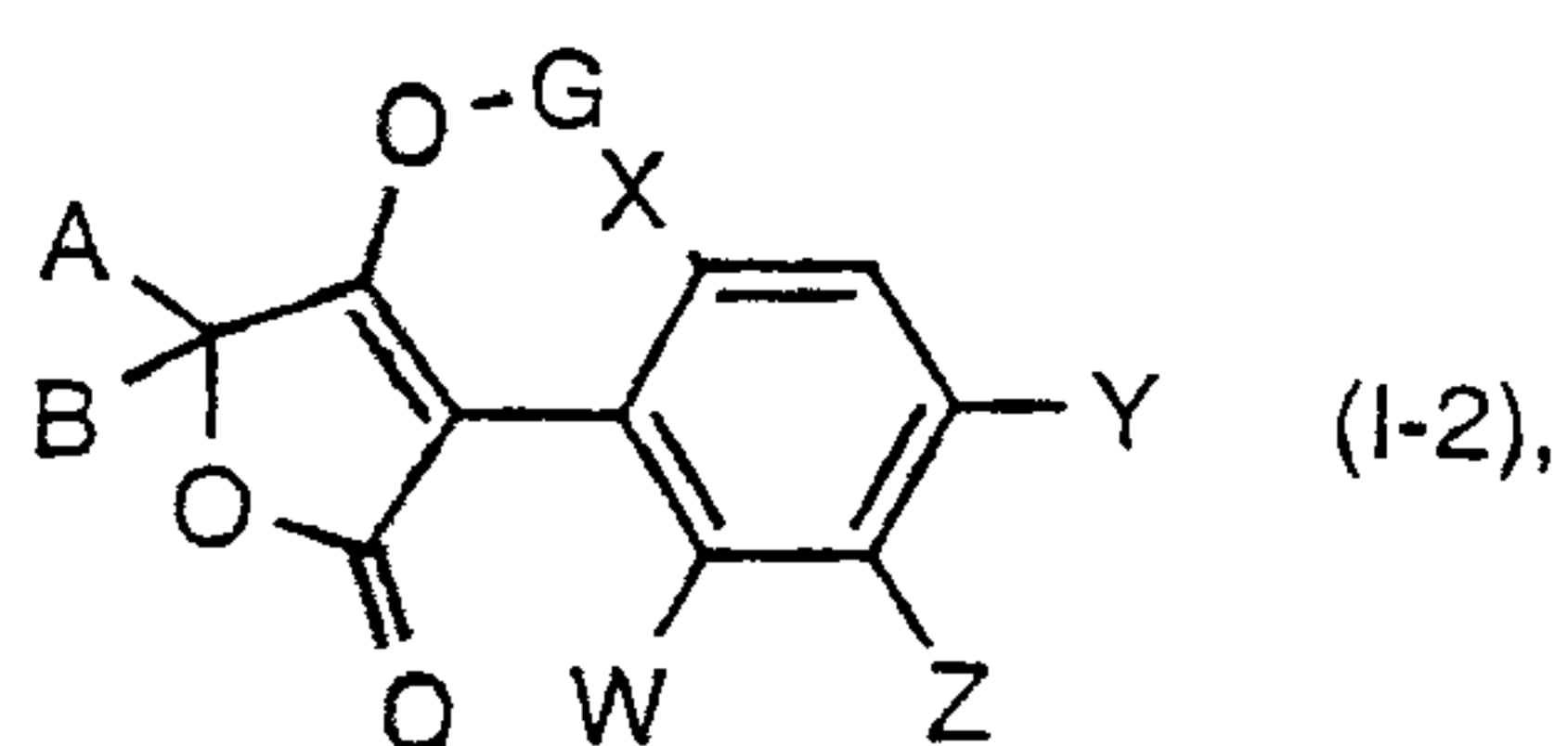
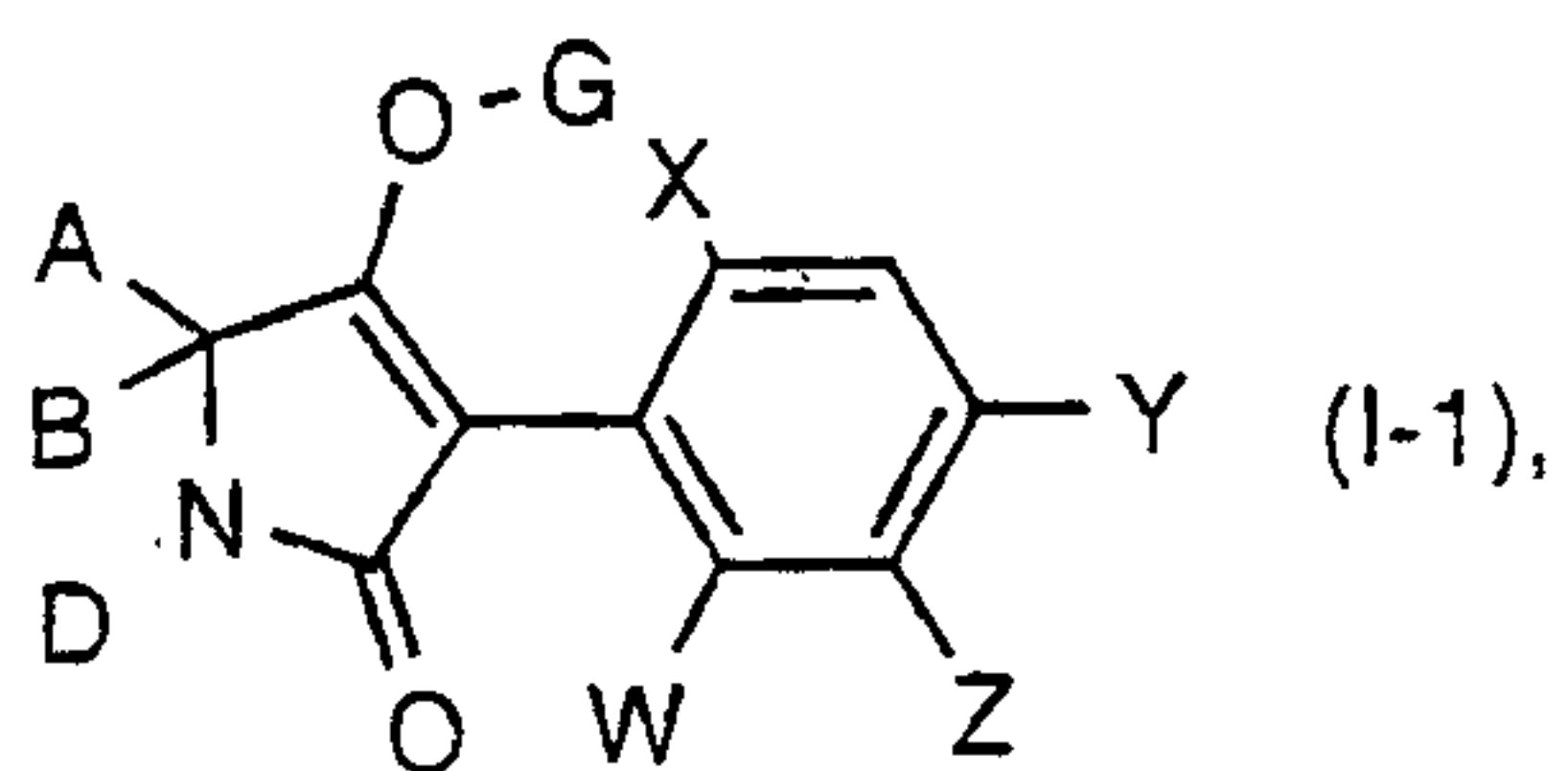
L represents oxygen or sulphur,

M represents oxygen or sulphur,

R¹ represents in each case optionally halogen-substituted alkyl, alkenyl, alkoxyalkyl, alkylthioalkyl, polyalkoxyalkyl or optionally halogen-, alkyl- or alkoxy-substituted cycloalkyl which may be interrupted by at least one heteroatom, in each

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5

in which

A, B, D, G, Q¹, Q², Q³, Q⁴, Q⁵, Q⁶, W, X, Y and Z are each as defined above.

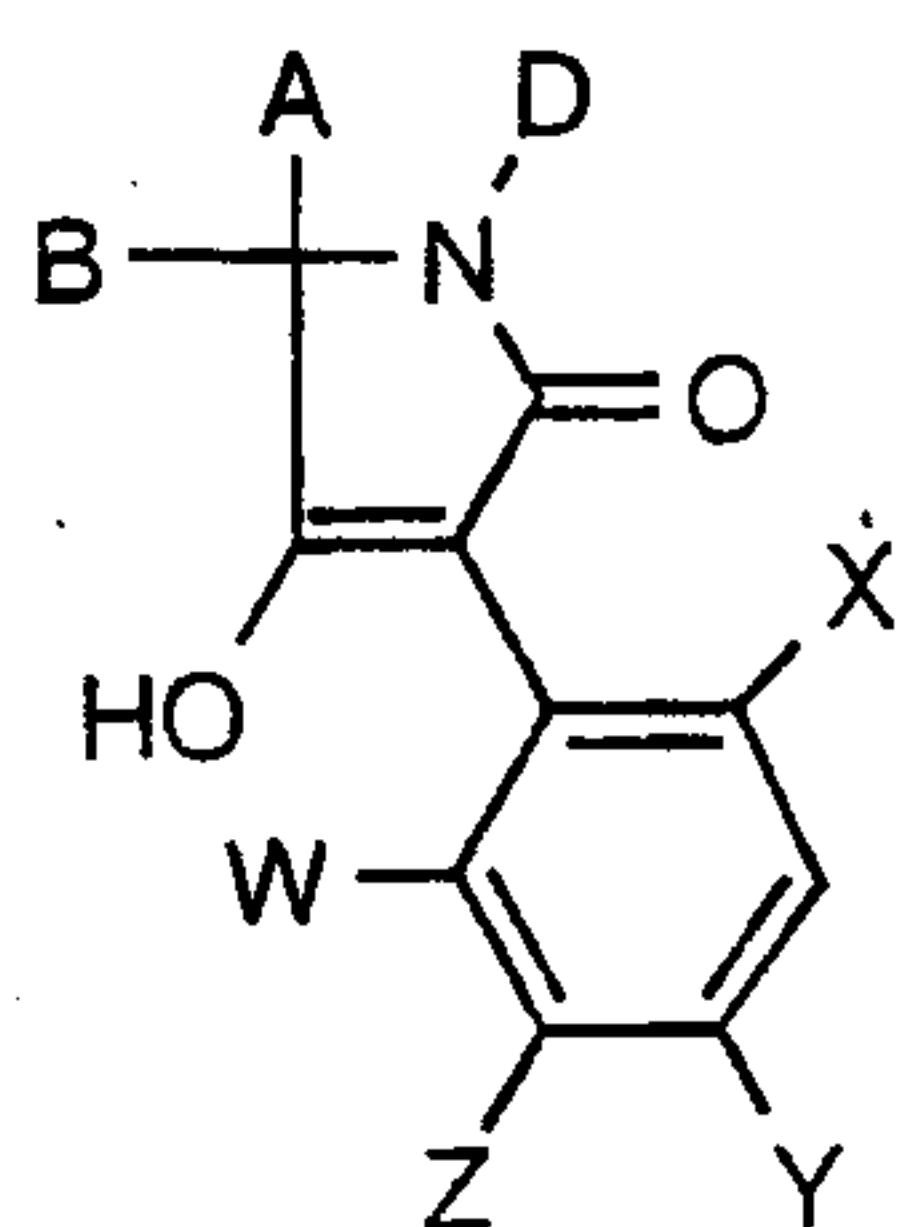
10

Including the various meanings of (a), (b), (c), (d), (e), (f) and (g) of the group G, the following principal structures (I-1-a) to (I-1-g) result if CKE represents the group (1):

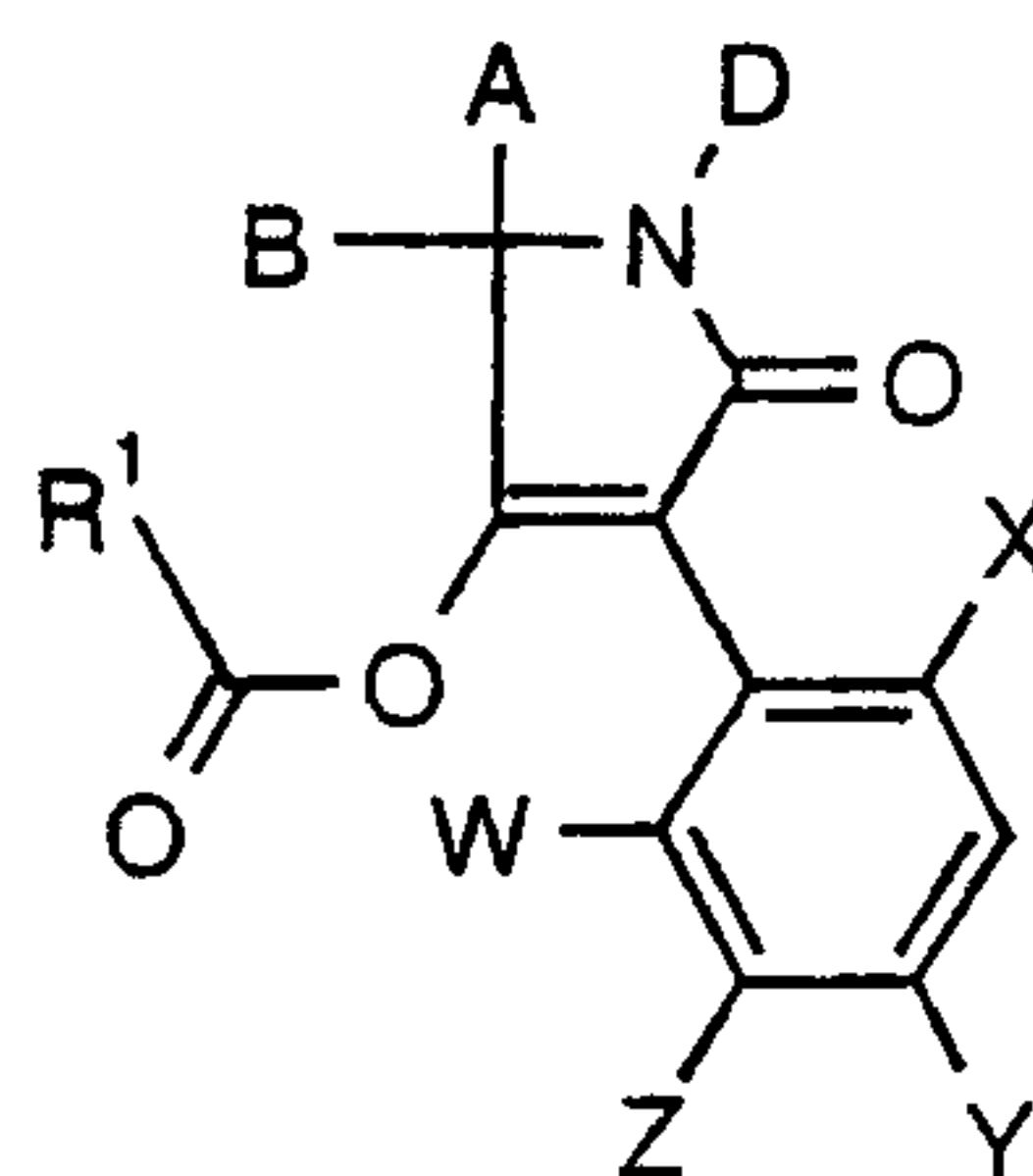
30725-1106D1

- 9 -

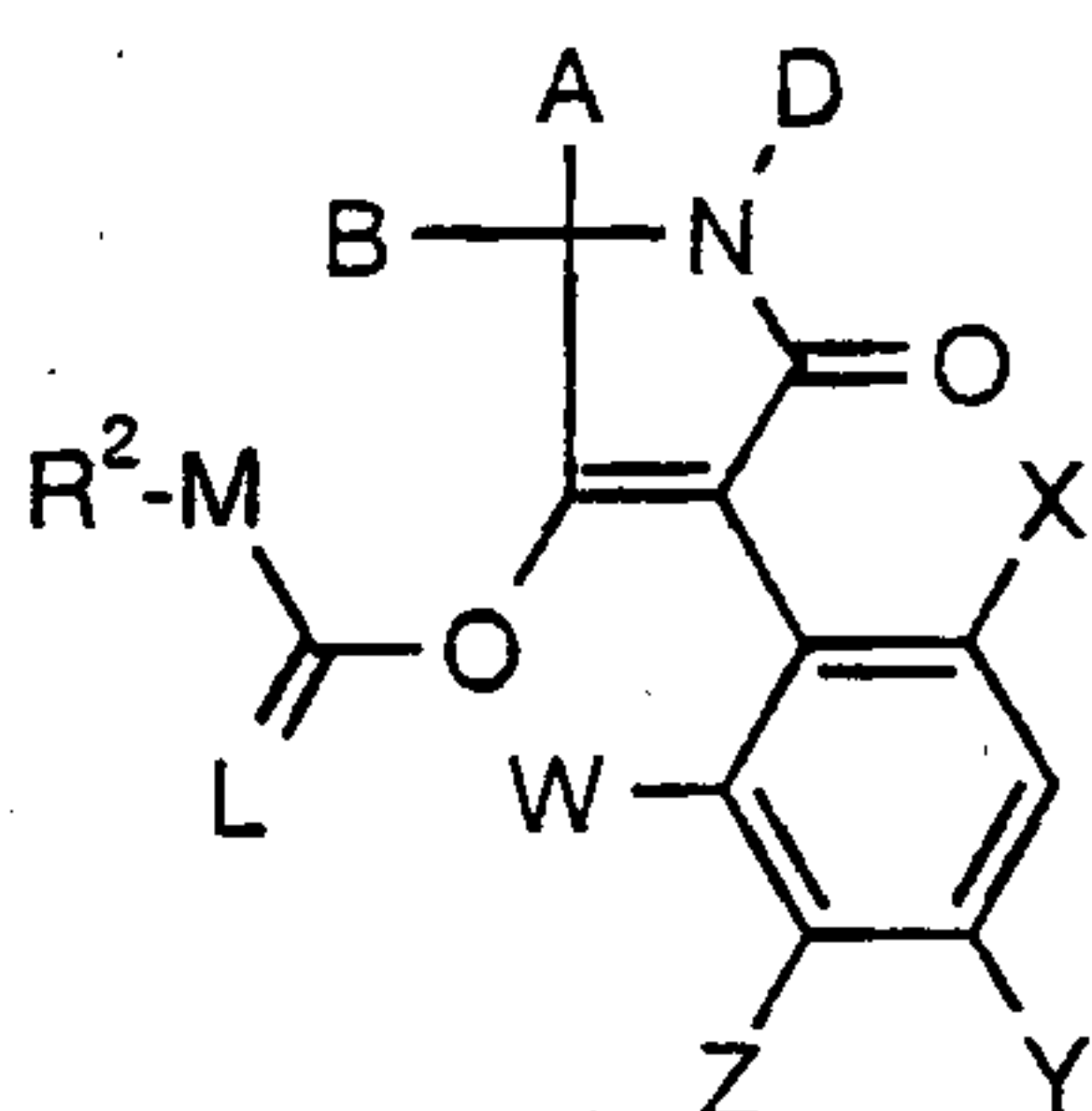
(I-1-a):



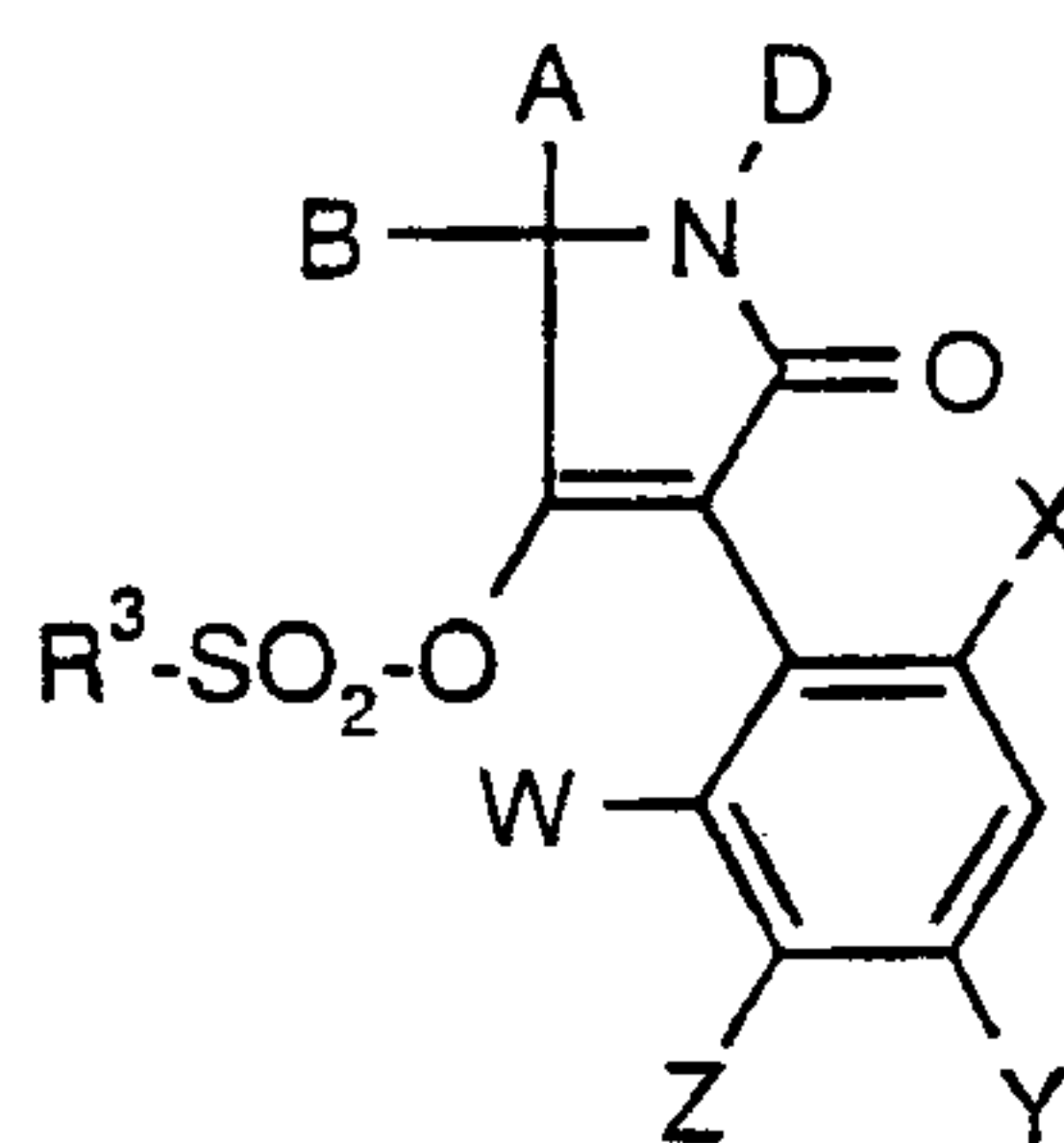
(I-1-b):



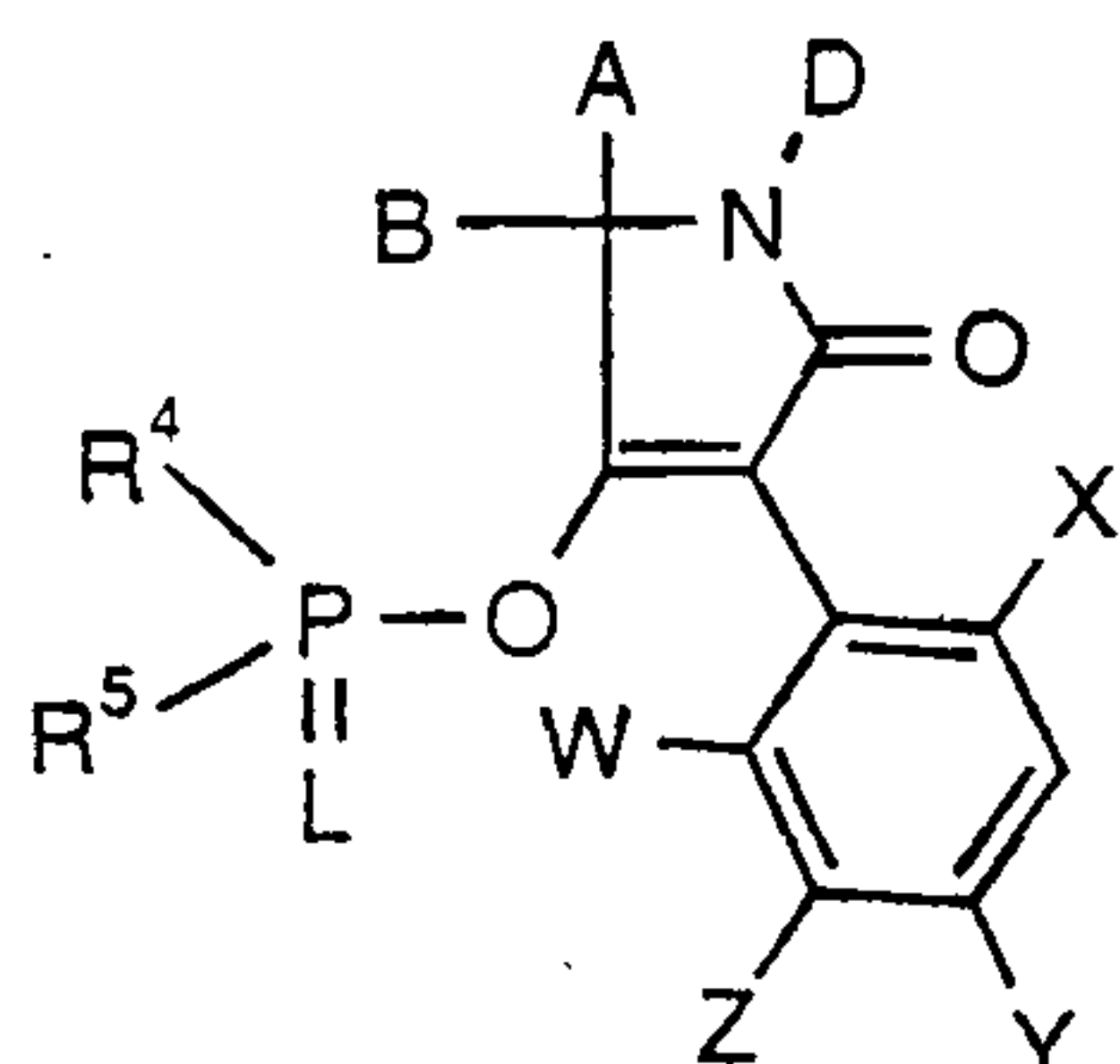
(I-1-c):



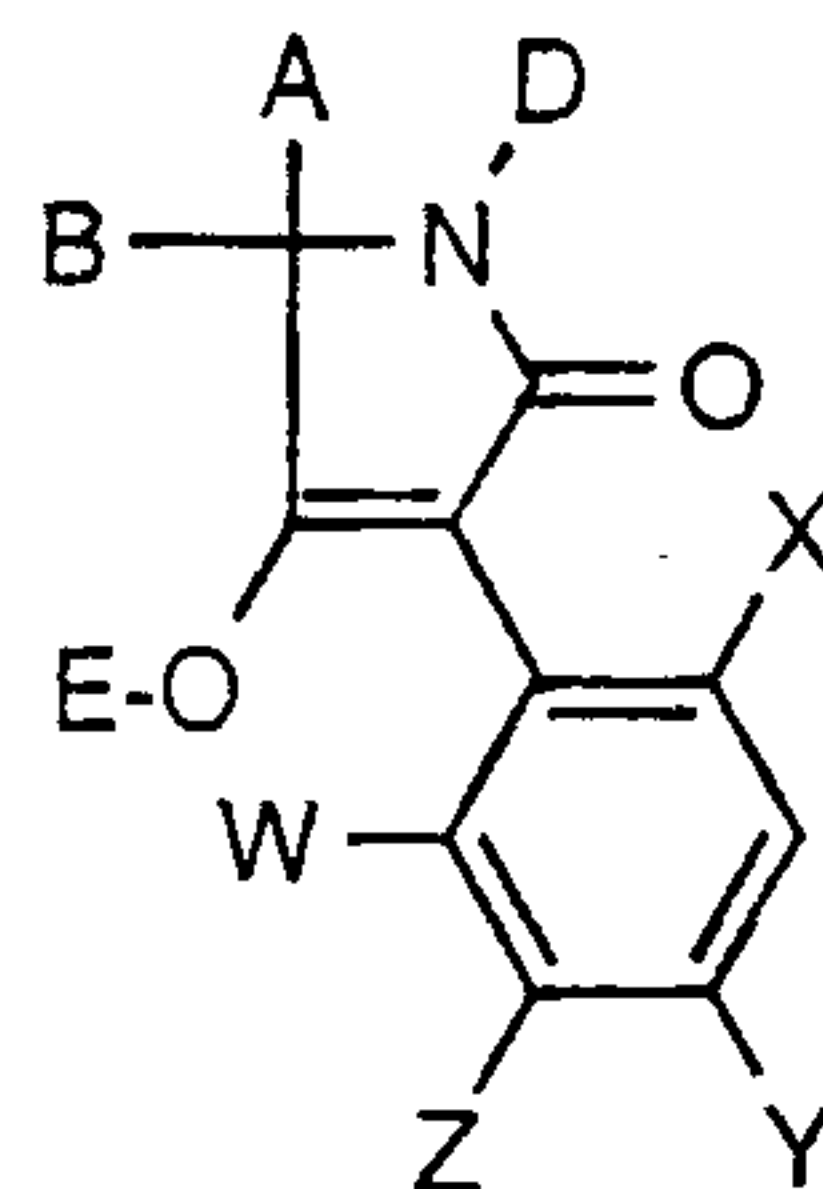
(I-1-d):



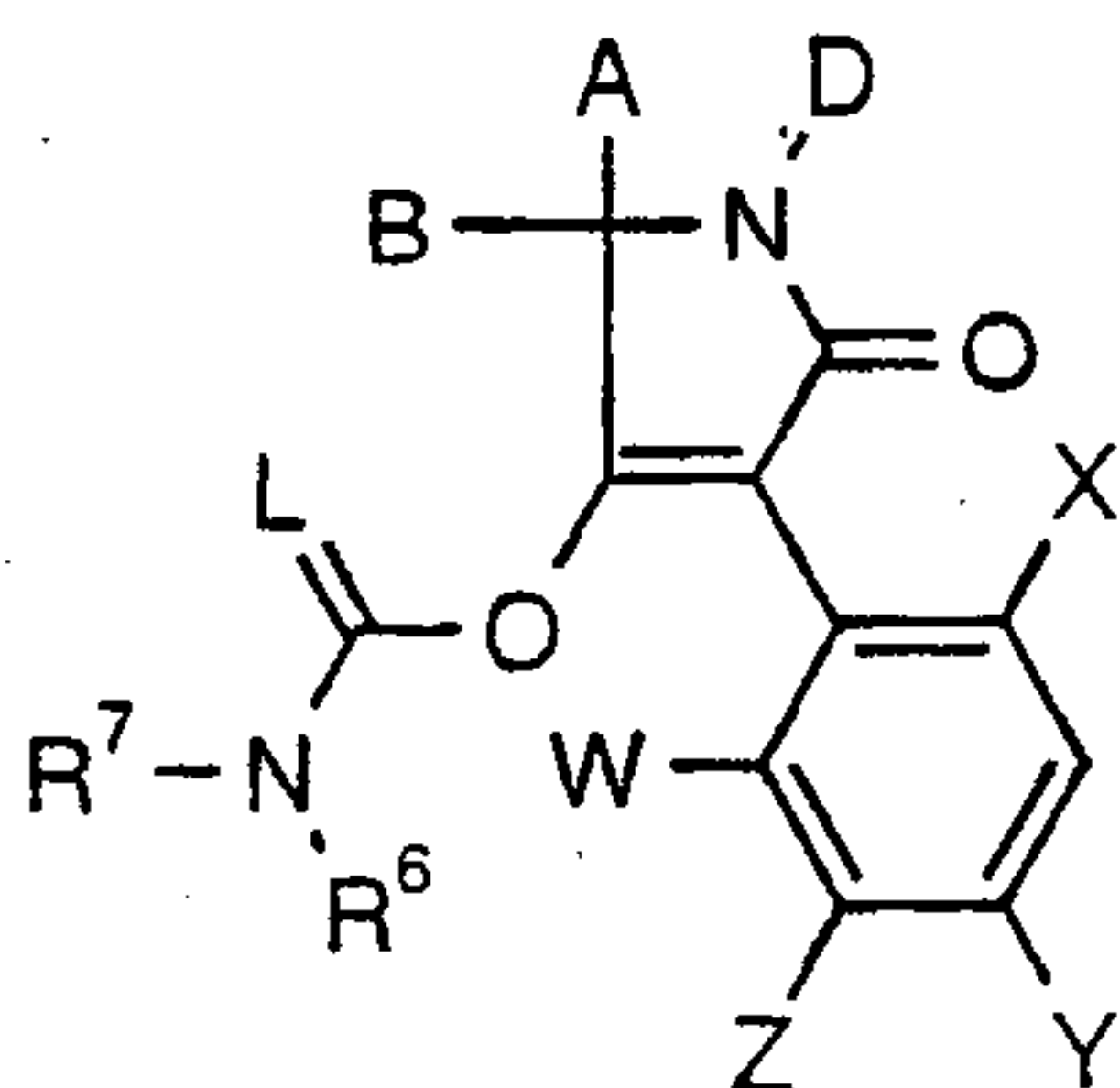
(I-1-e):



(I-1-f):



(I-1-g):



in which

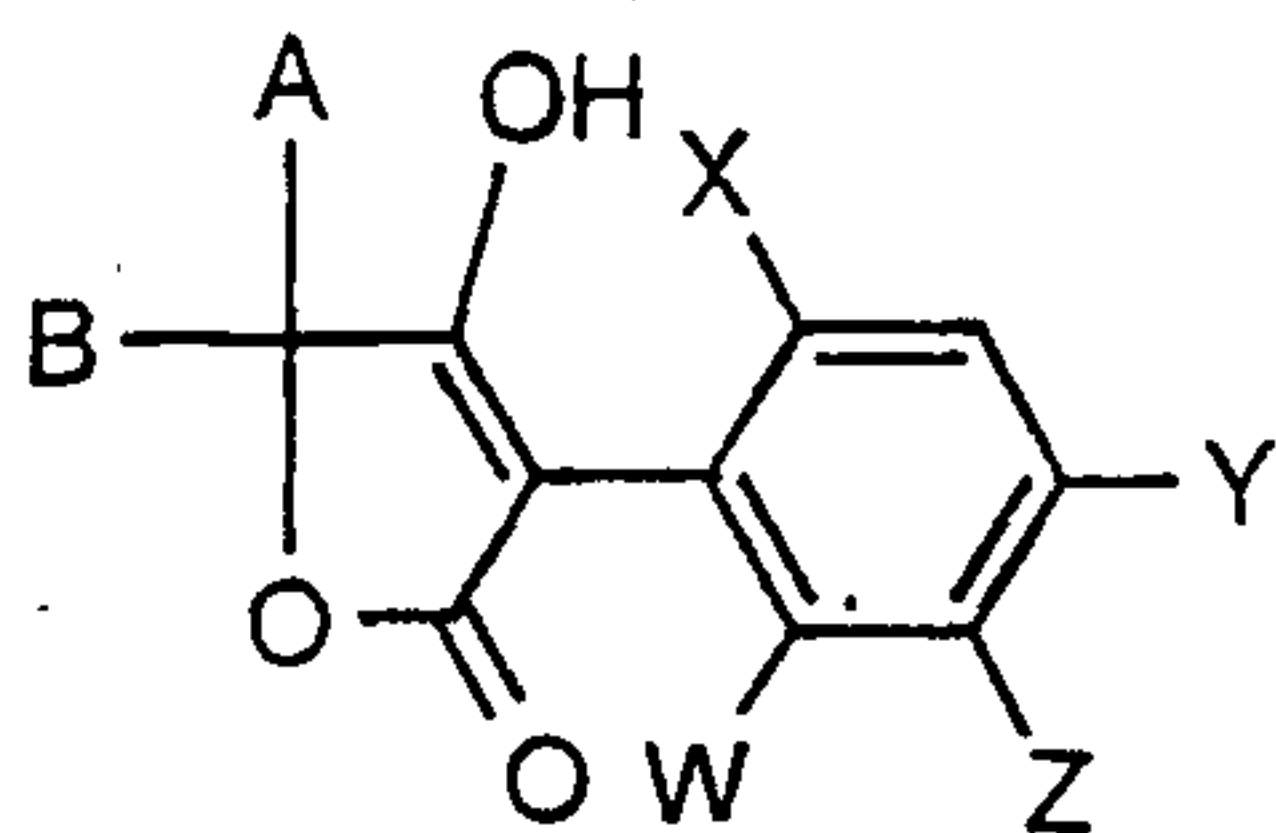
A, B, D, E, L, M, W, X, Y, Z, R¹, R², R³, R⁴, R⁵, R⁶ and R⁷ are each as defined above.

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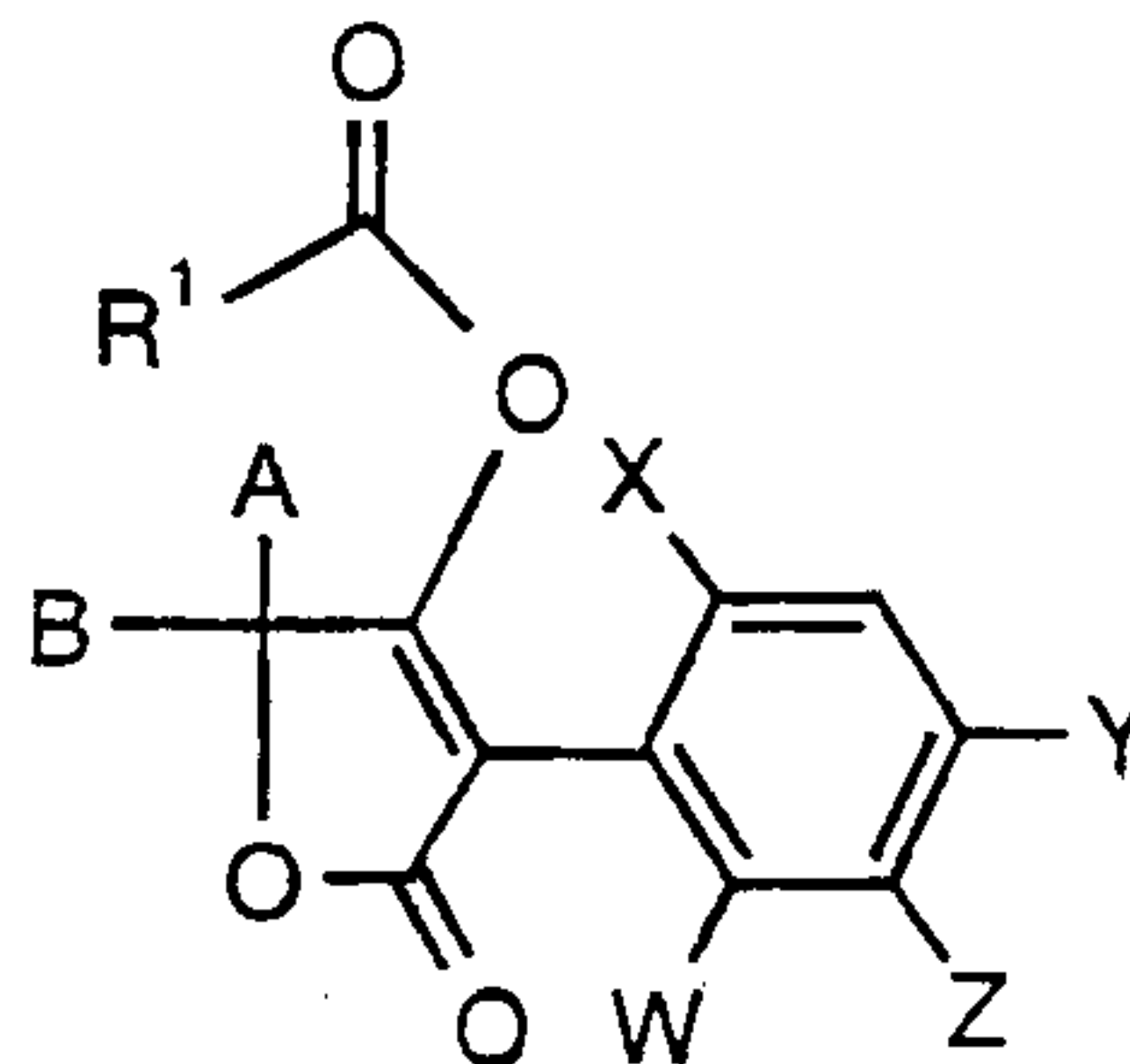
- 10 -

Including the various meanings (a), (b), (c), (d), (e), (f) and (g) of the group G, the following principal structures (I-2-a) to (I-2-g) result if CKE represents the group (2):

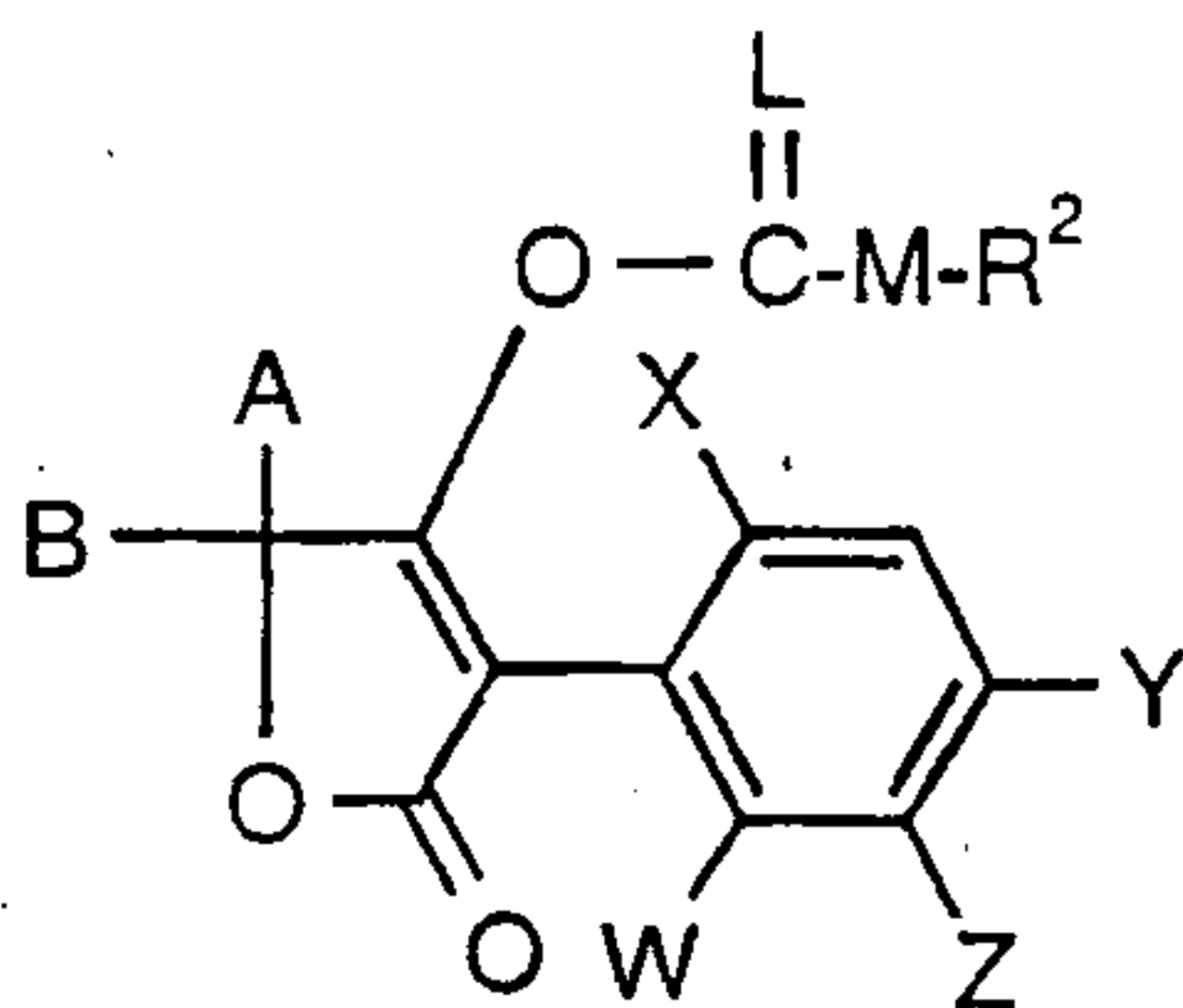
(I-2-a):



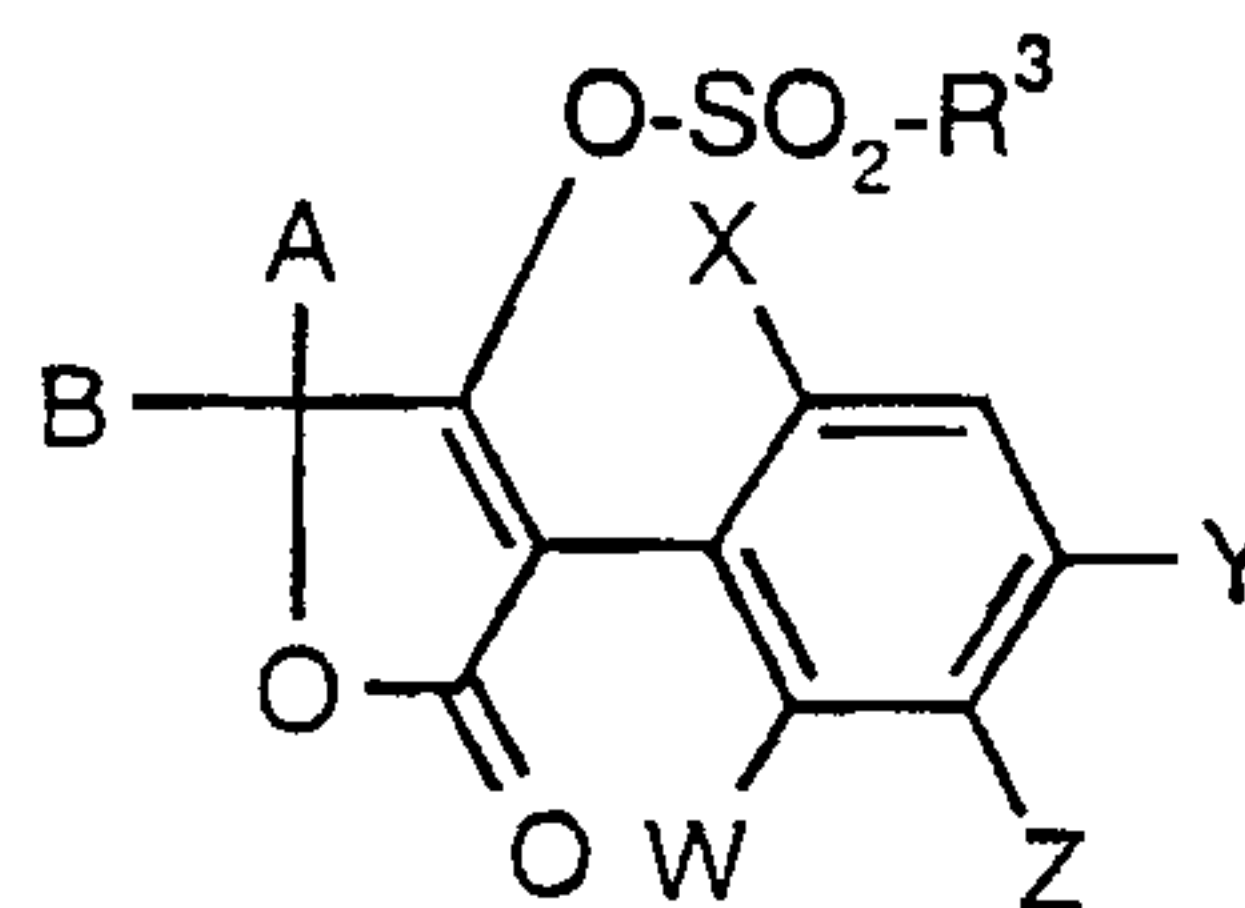
(I-2-b):



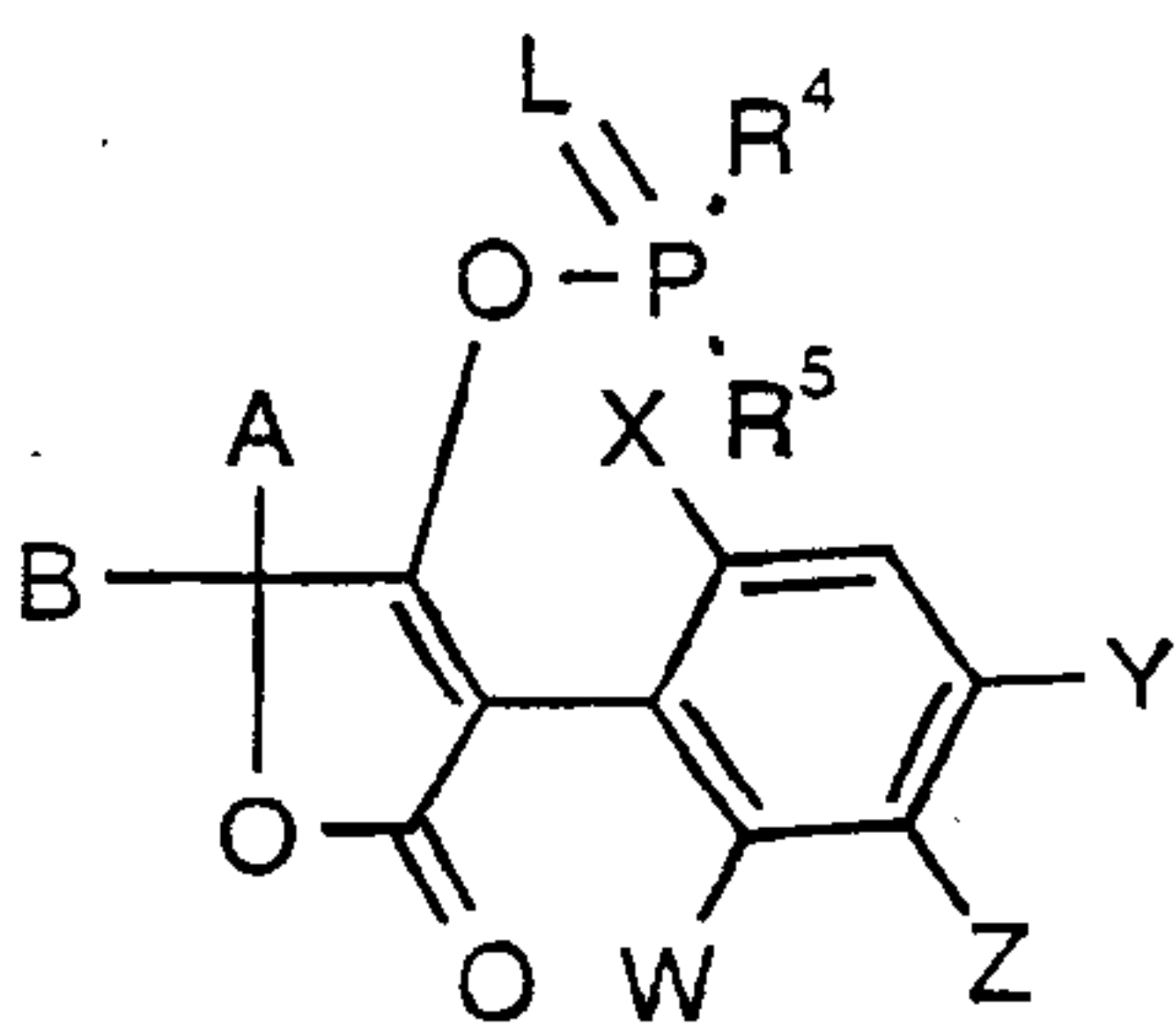
(I-2-c):



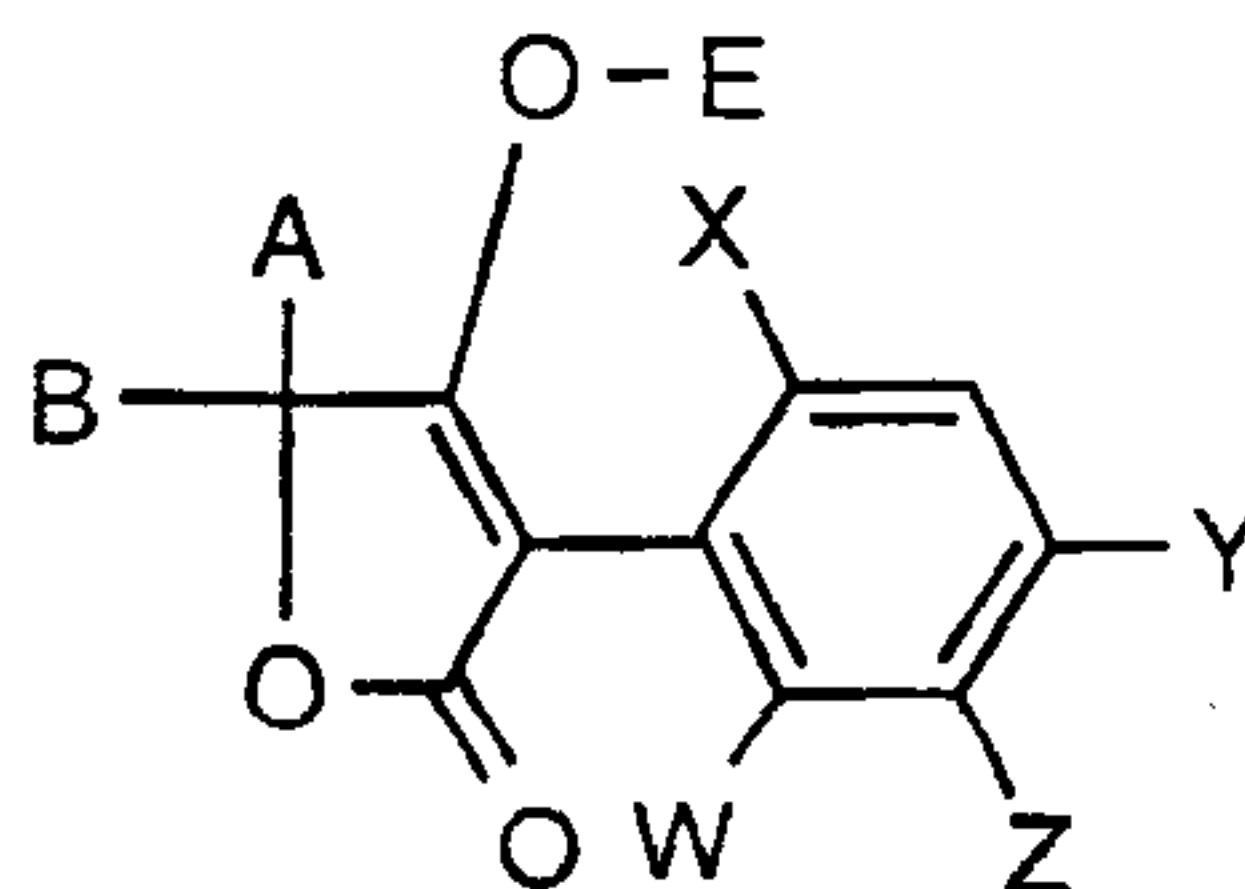
(I-2-d):



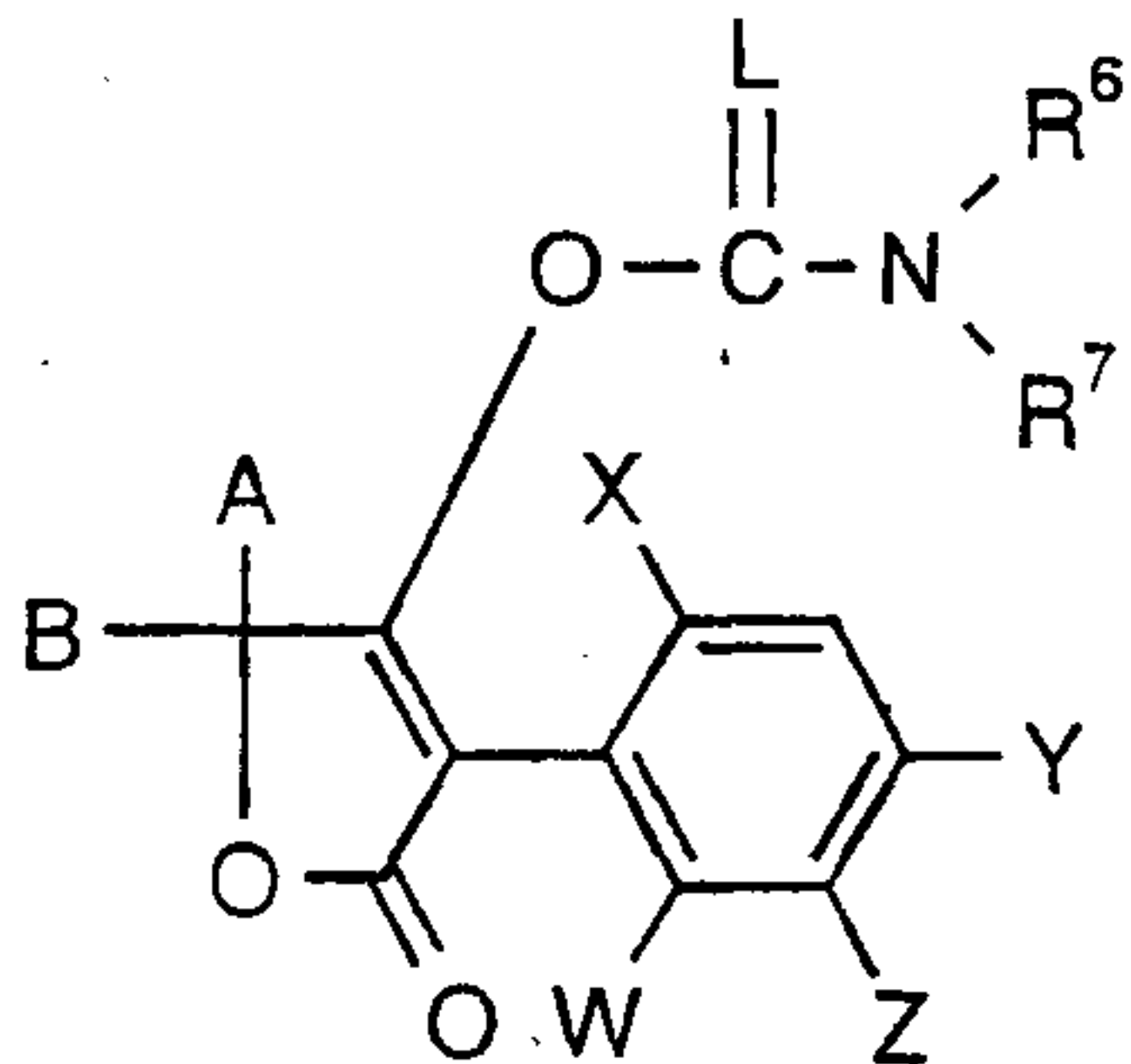
(I-2-e):



(I-2-f):



(I-2-g):



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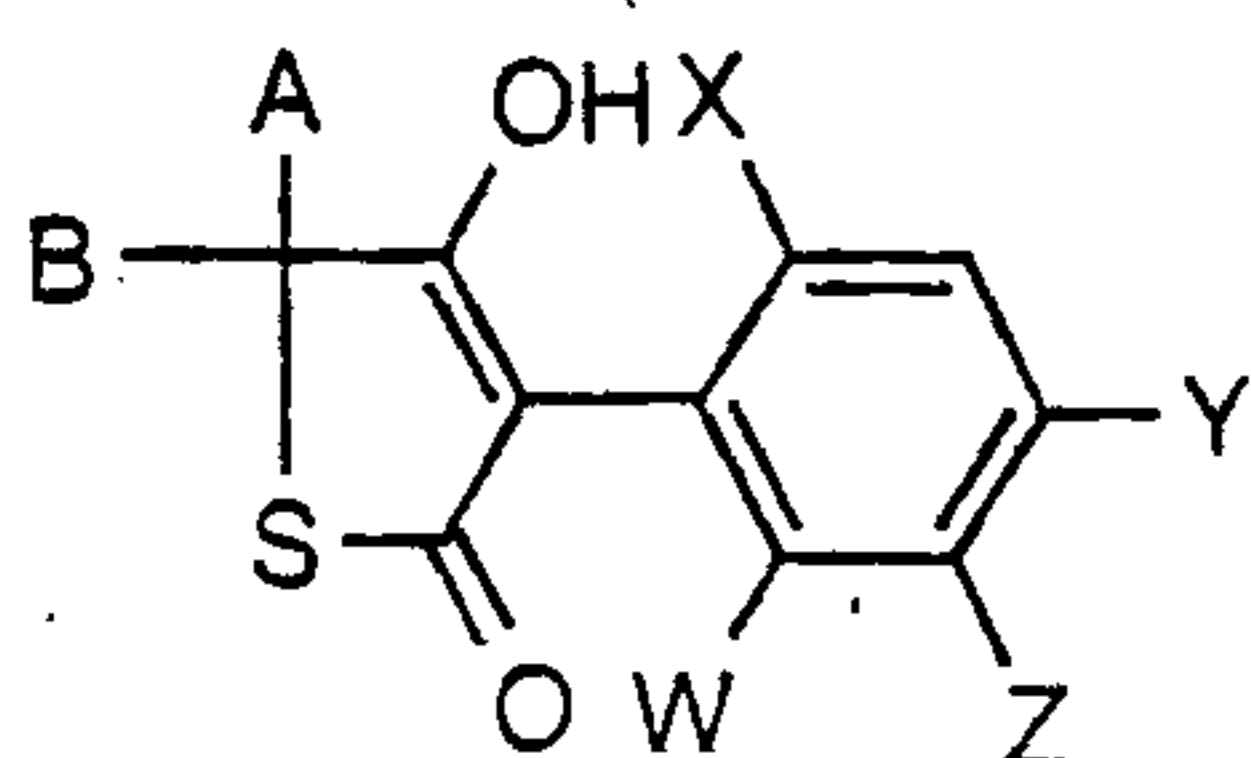
- 11 -

in which

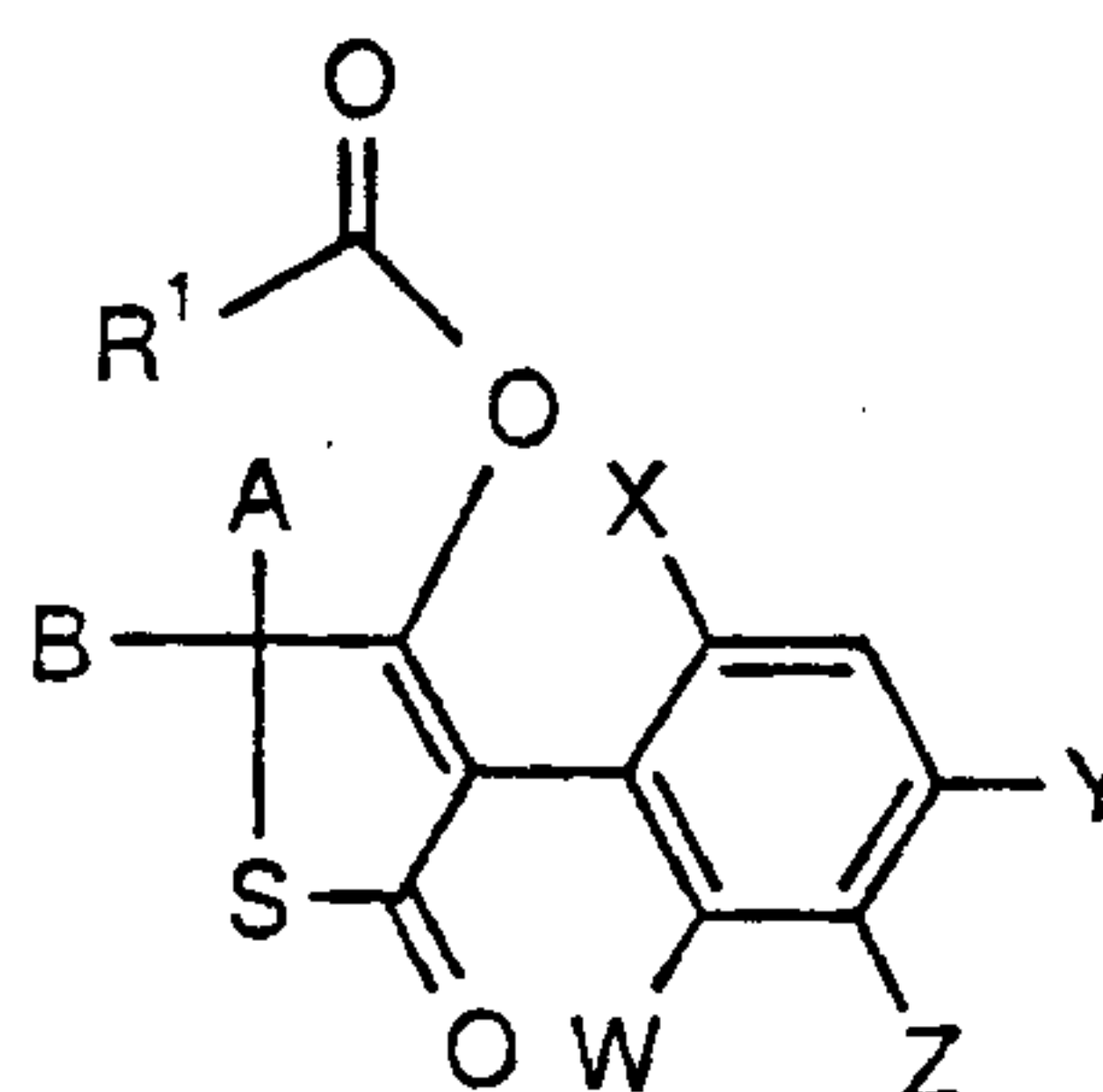
A, B, E, L, M, W, X, Y, Z, R¹, R², R³, R⁴, R⁵, R⁶ and R⁷ are each as defined
5 above.

Including the various meanings (a), (b), (c), (d), (e), (f) and (g) of the group G, the following principal structures (I-3-a) to (I-3-g) result if CKE represents the group (3):

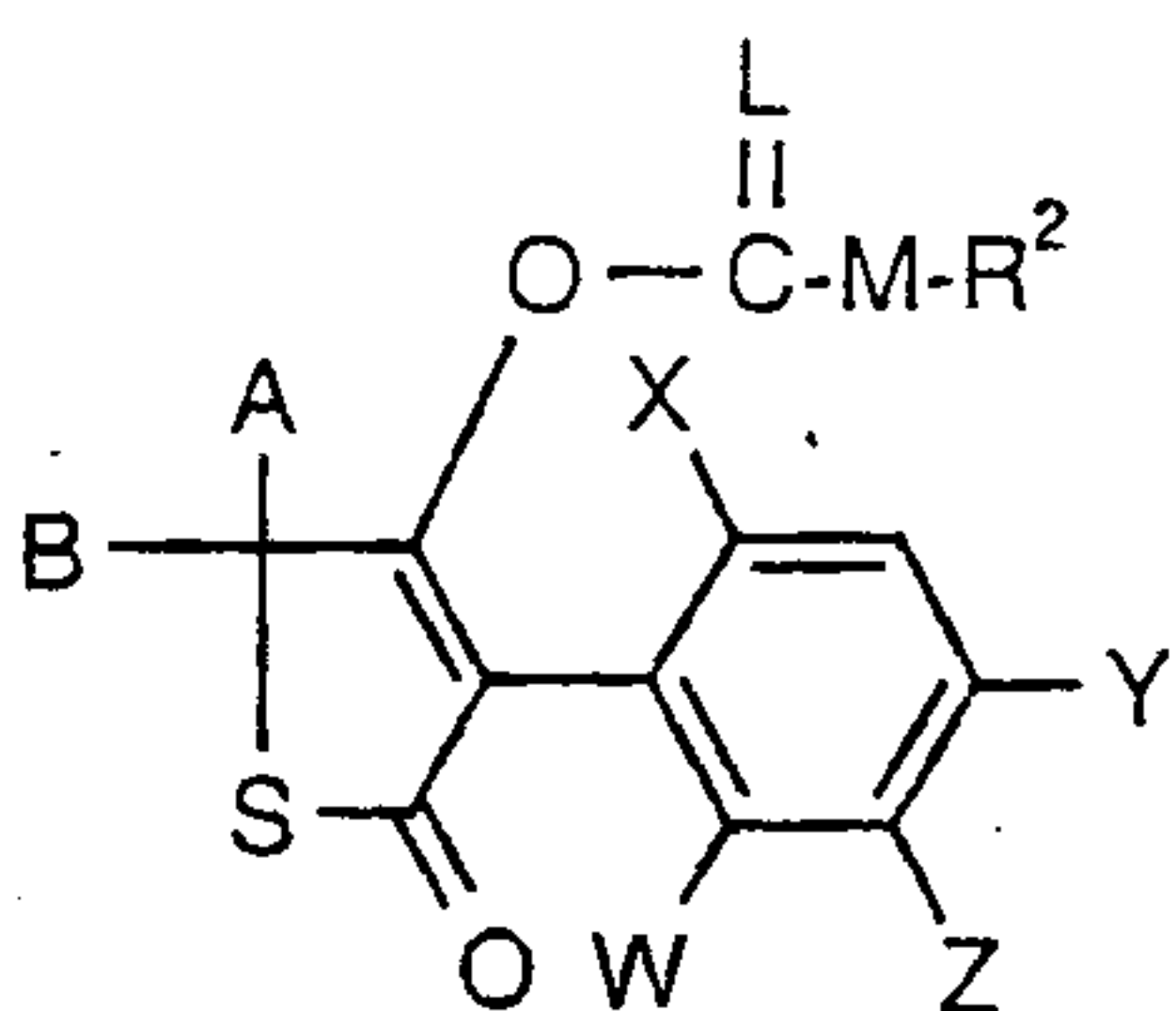
(I-3-a):



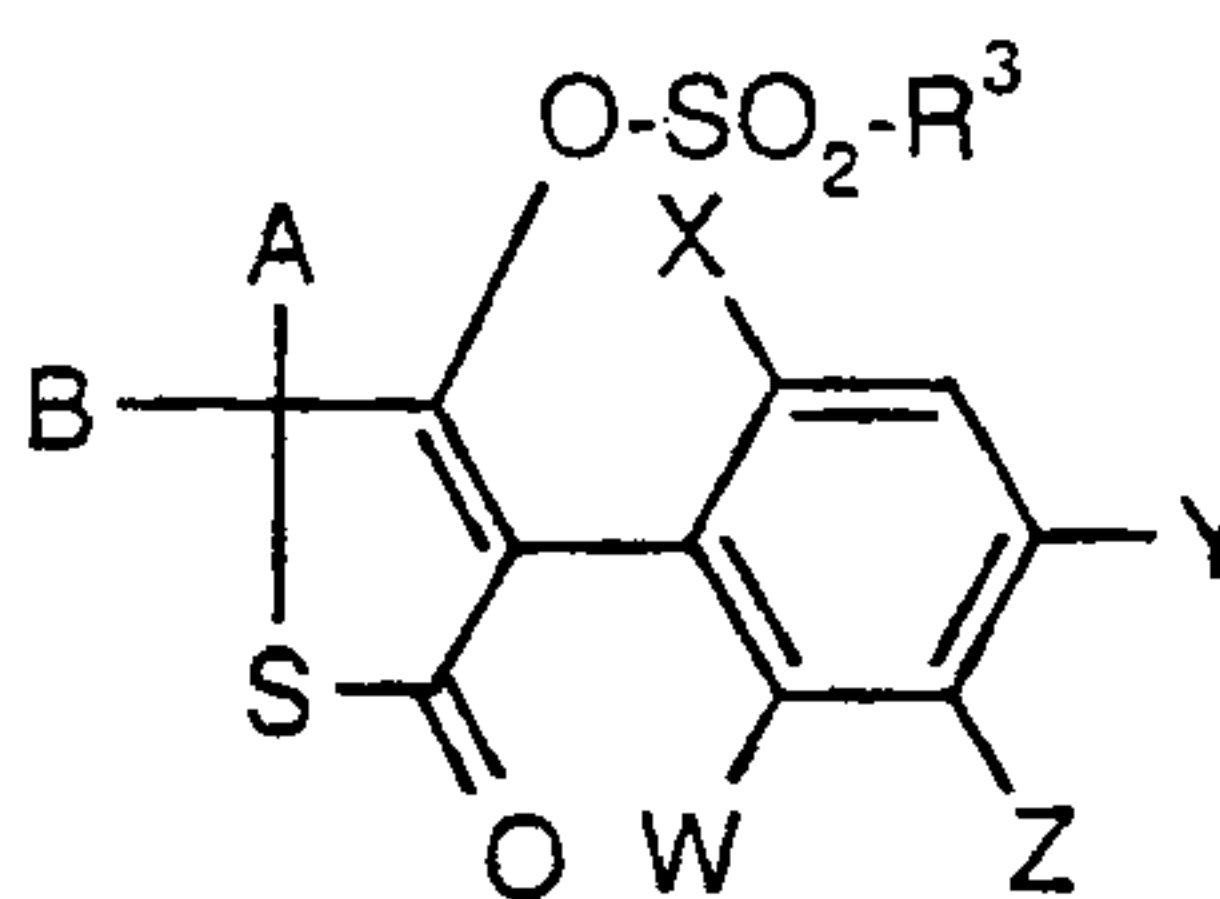
(I-3-b):



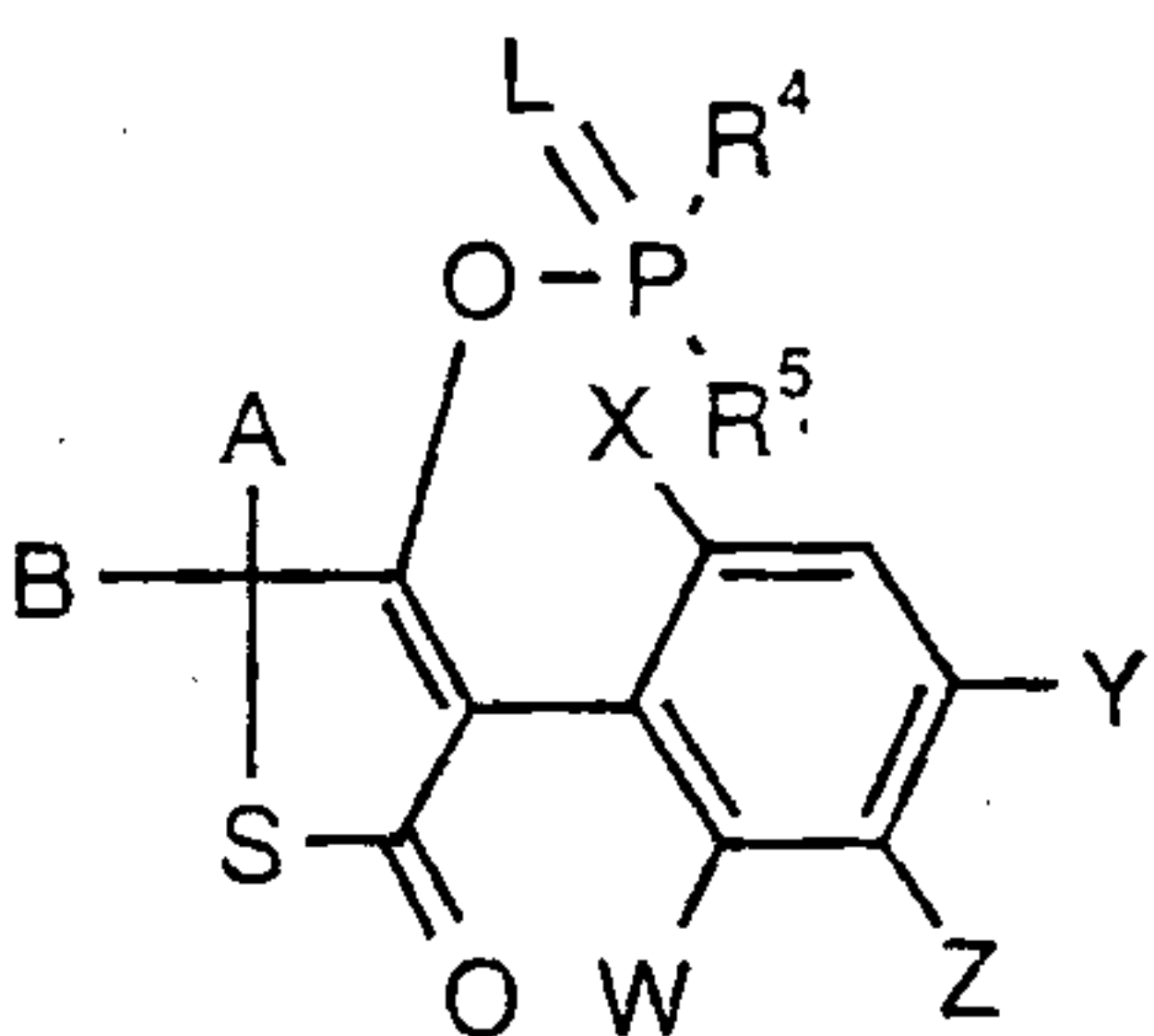
(I-3-c):



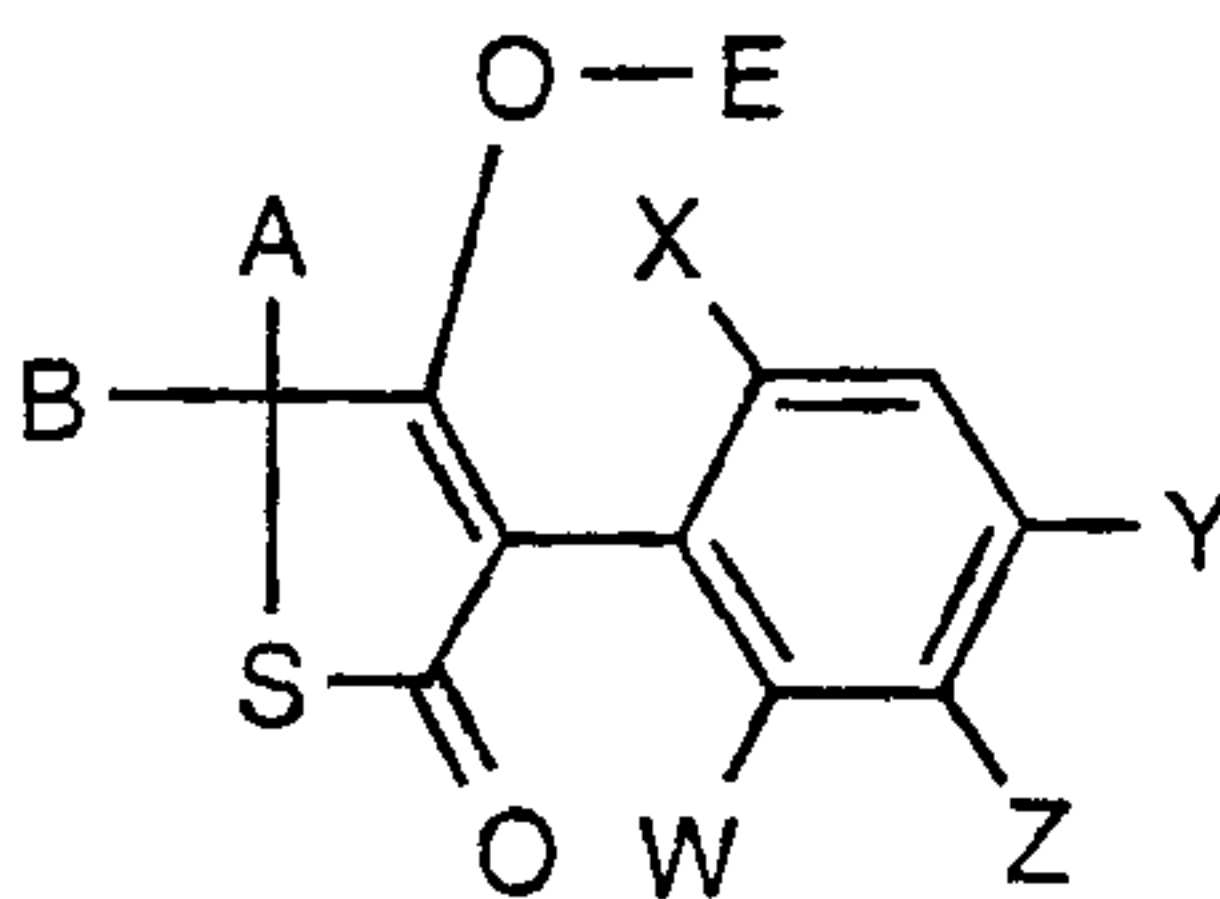
(I-3-d):



(I-3-e):



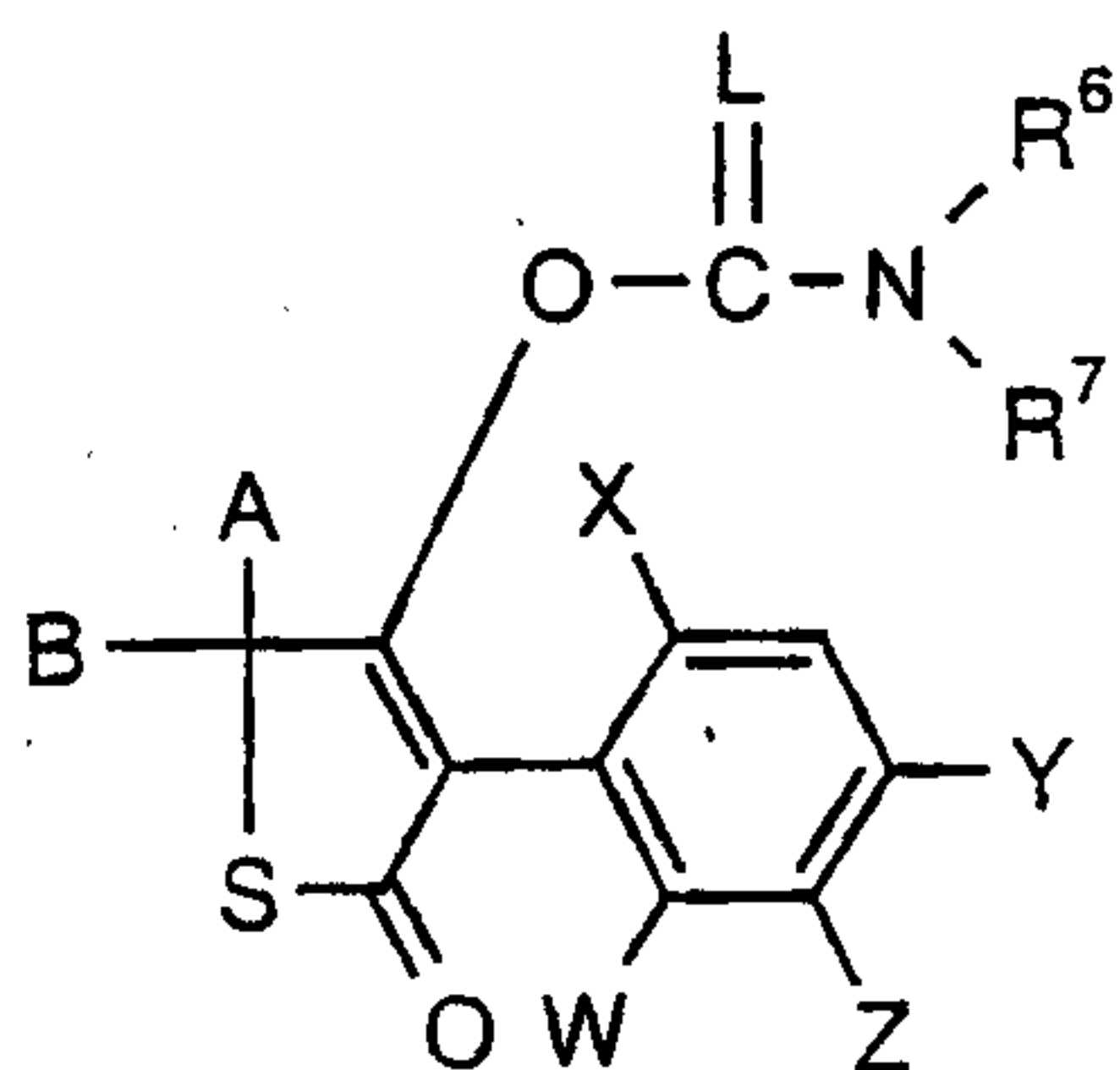
(I-3-f):



(I-3-g):

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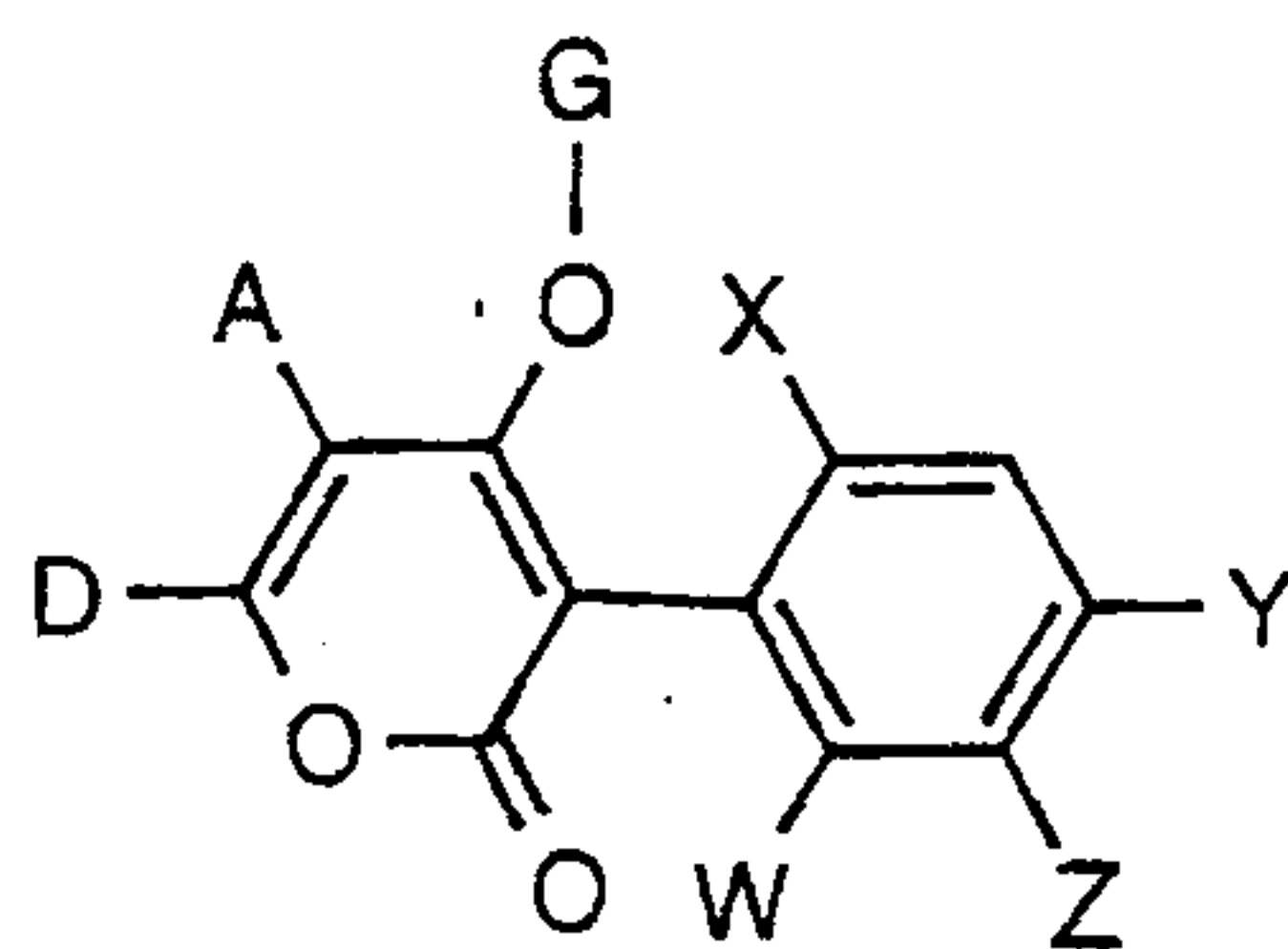
- 12 -



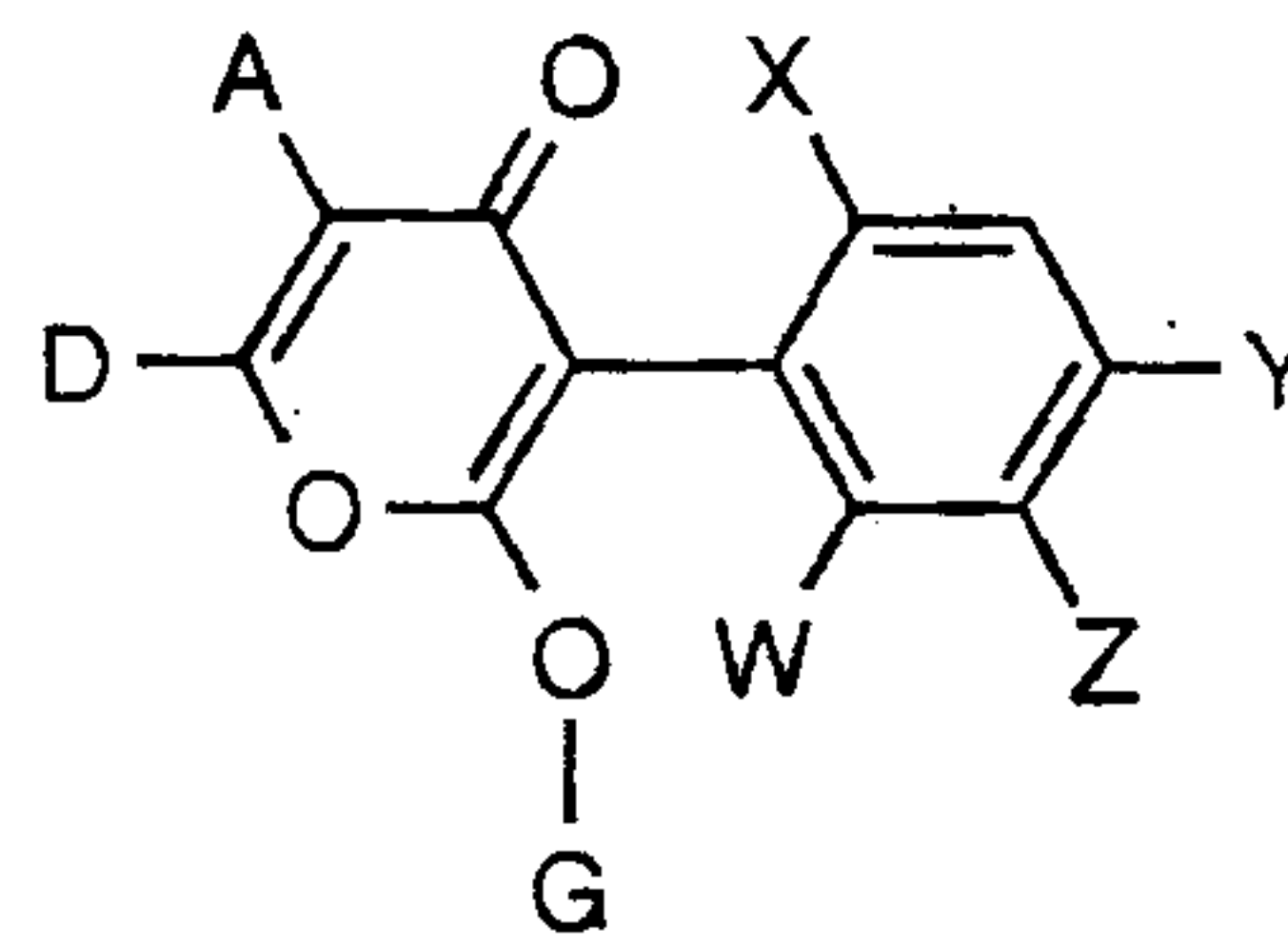
in which

5 A, B, E, L, M, W, X, Y, Z, R¹, R², R³, R⁴, R⁵, R⁶ and R⁷ are each as defined above.

Depending on the position of the substituent G, the compounds of the formula (I-4) can be present in two isomeric forms of the formulae (I-5-A) and (I-5-B)



(I-4-A)



(I-4-B)

10

which is meant to be indicated by the dashed line in the formula (I-4).

The compounds of the formulae (I-4-A) and (I-4-B) can be present both as mixtures and in the form of their pure isomers. Mixtures of the compounds of the formulae (I-4-A) and (I-4-B) can be separated, if desired, in a known manner by physical methods, for example by chromatographic methods.

15

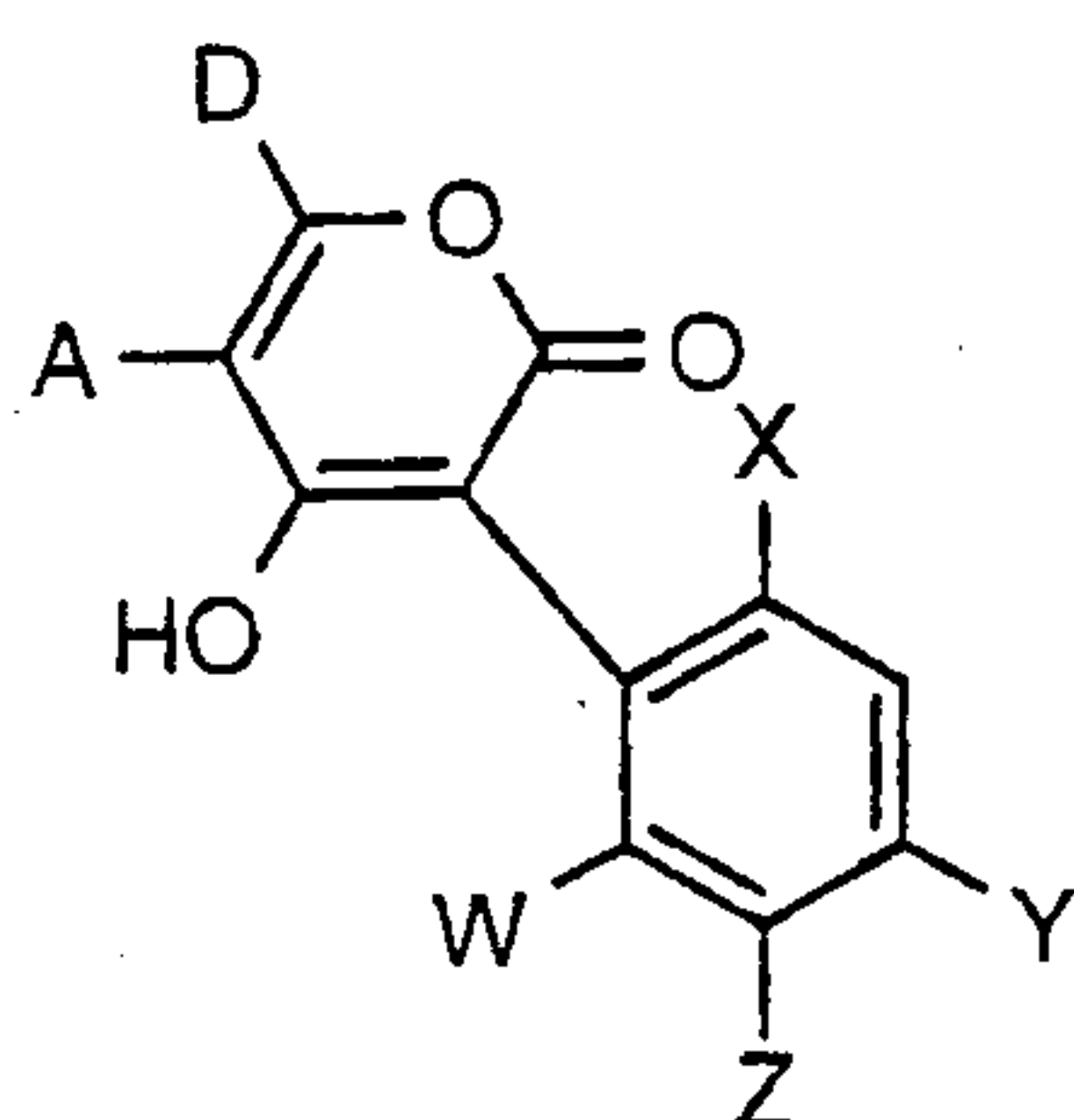
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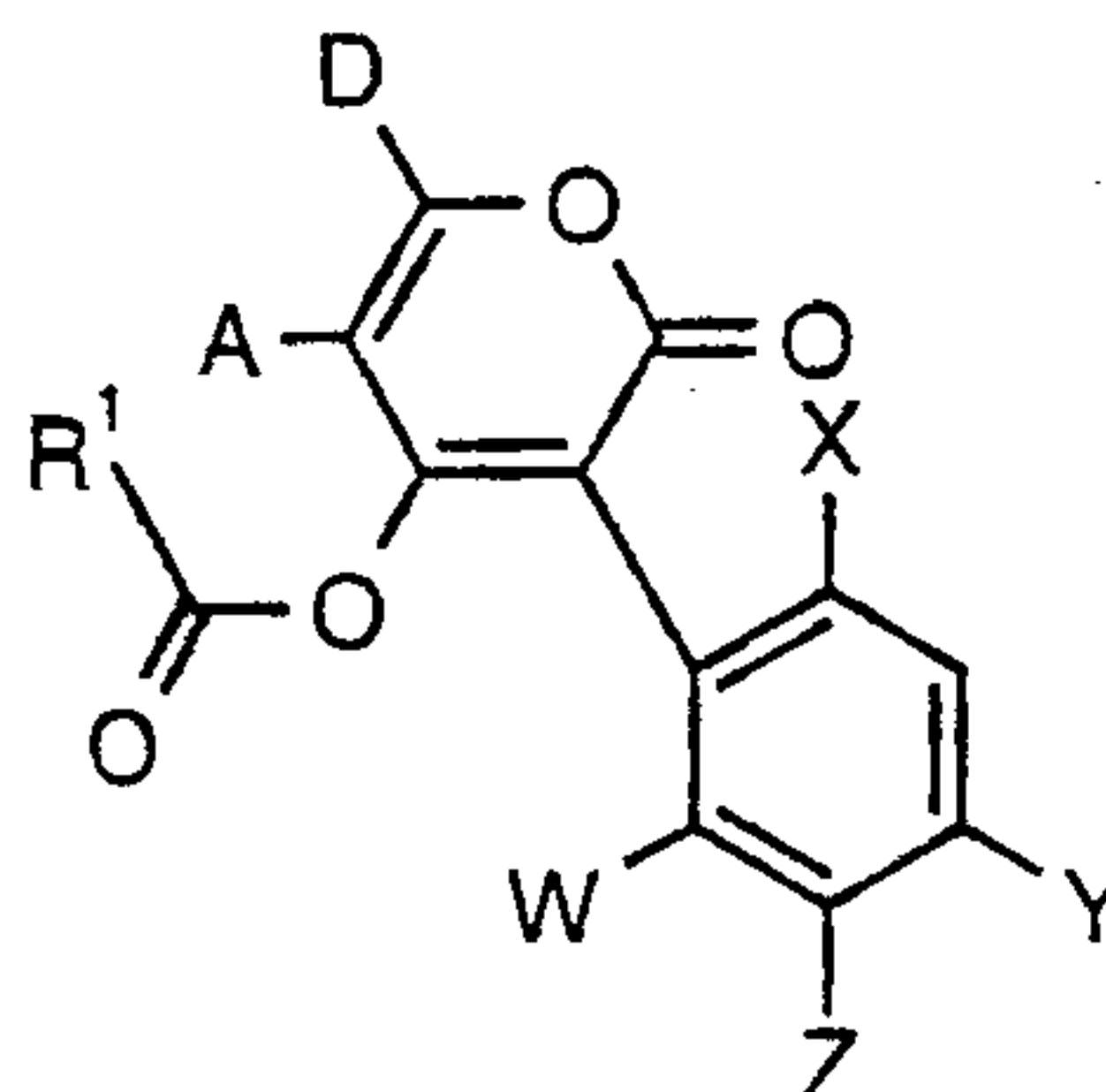
For reasons of clarity, hereinbelow in each case only one of the possible isomers is given. This does not exclude that the compounds may, if appropriate, be present in the form of the isomer mixtures or in the respective other isomeric form.

- 5 Including the various meanings (a), (b), (c), (d), (e), (f) and (g) of the group G, the following principal structures (I-4-a) to (I-4-g) result if CKE represents the group (4):

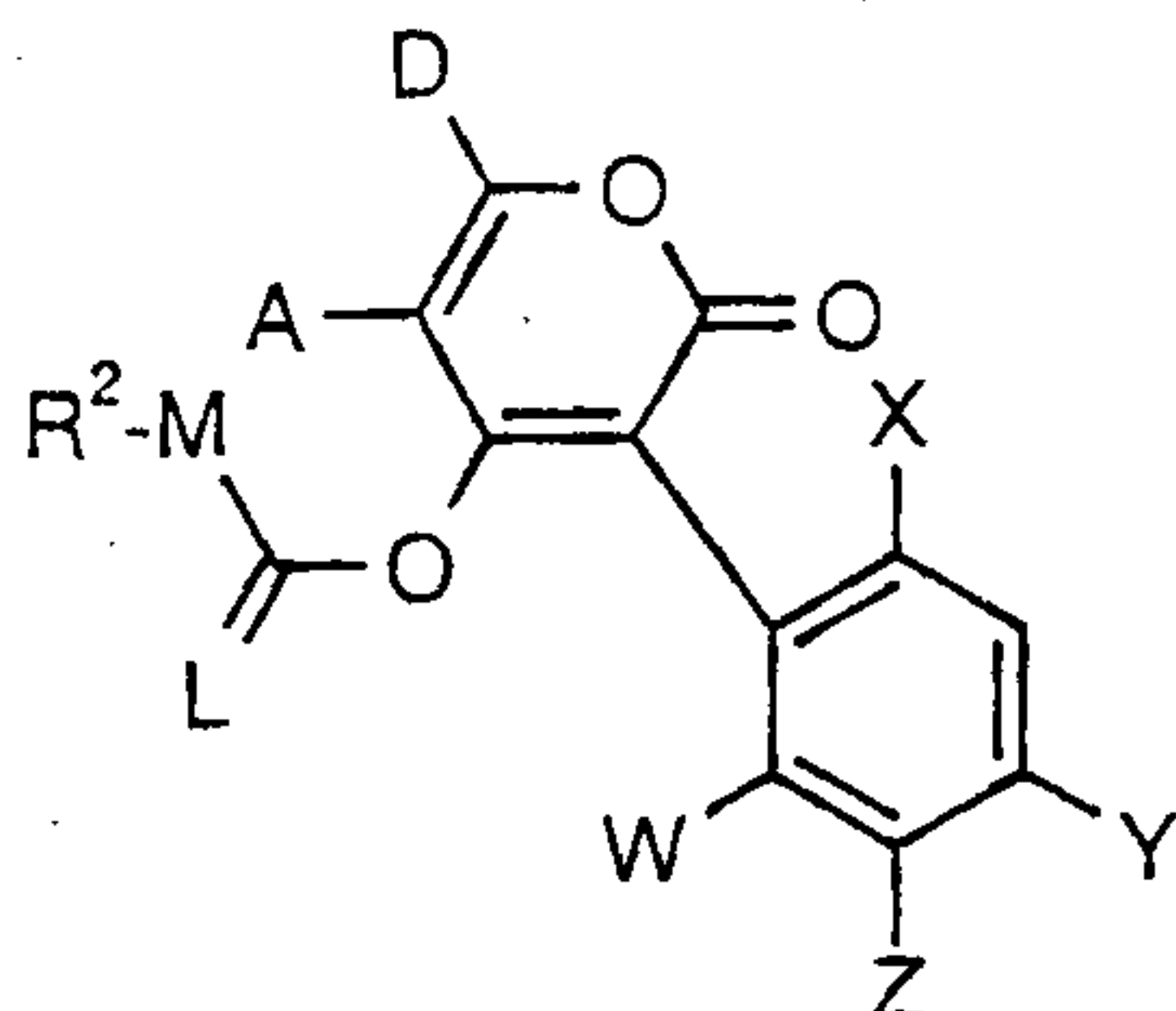
(I-4-a):



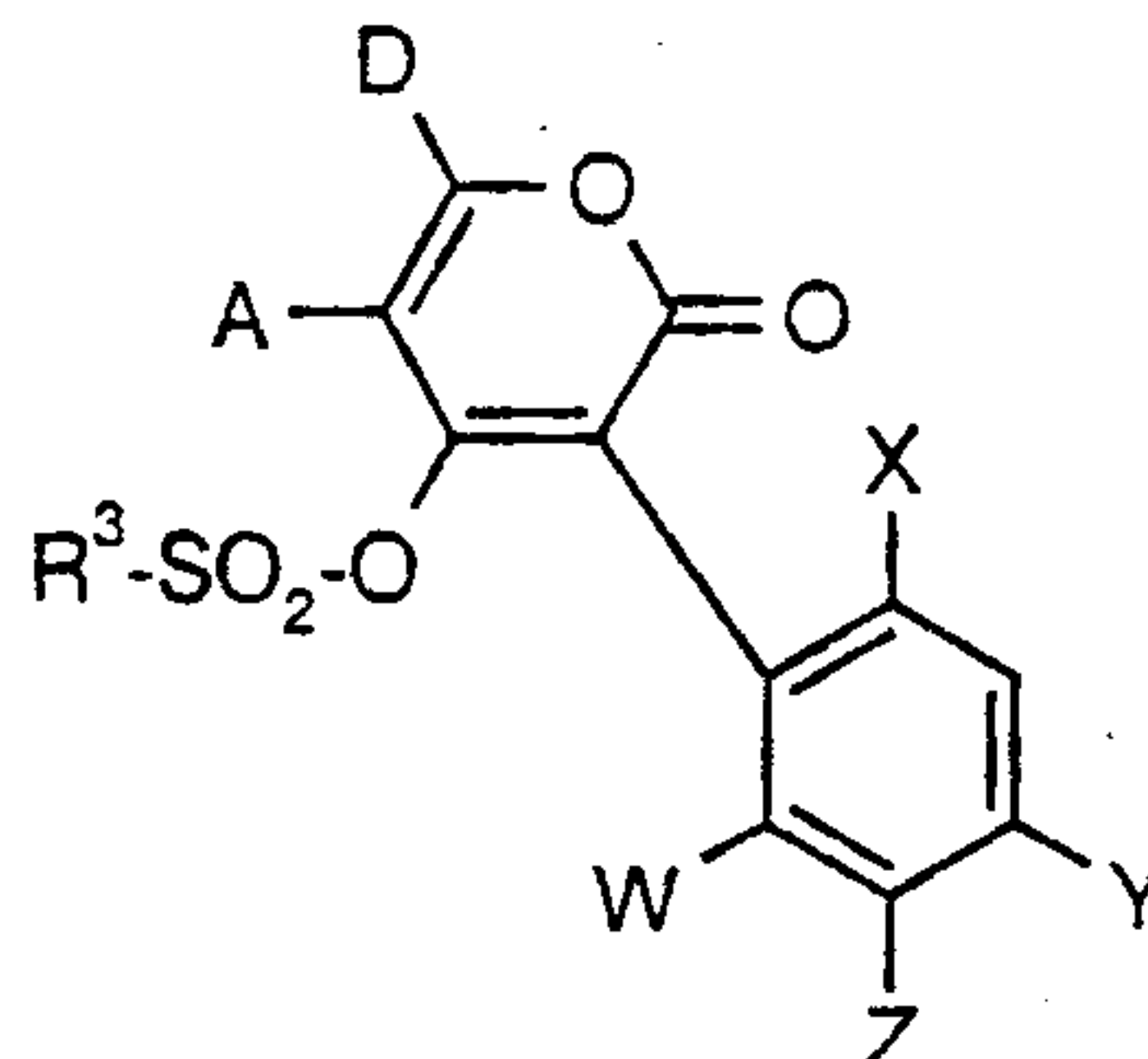
(I-4-b):



(I-4-c):



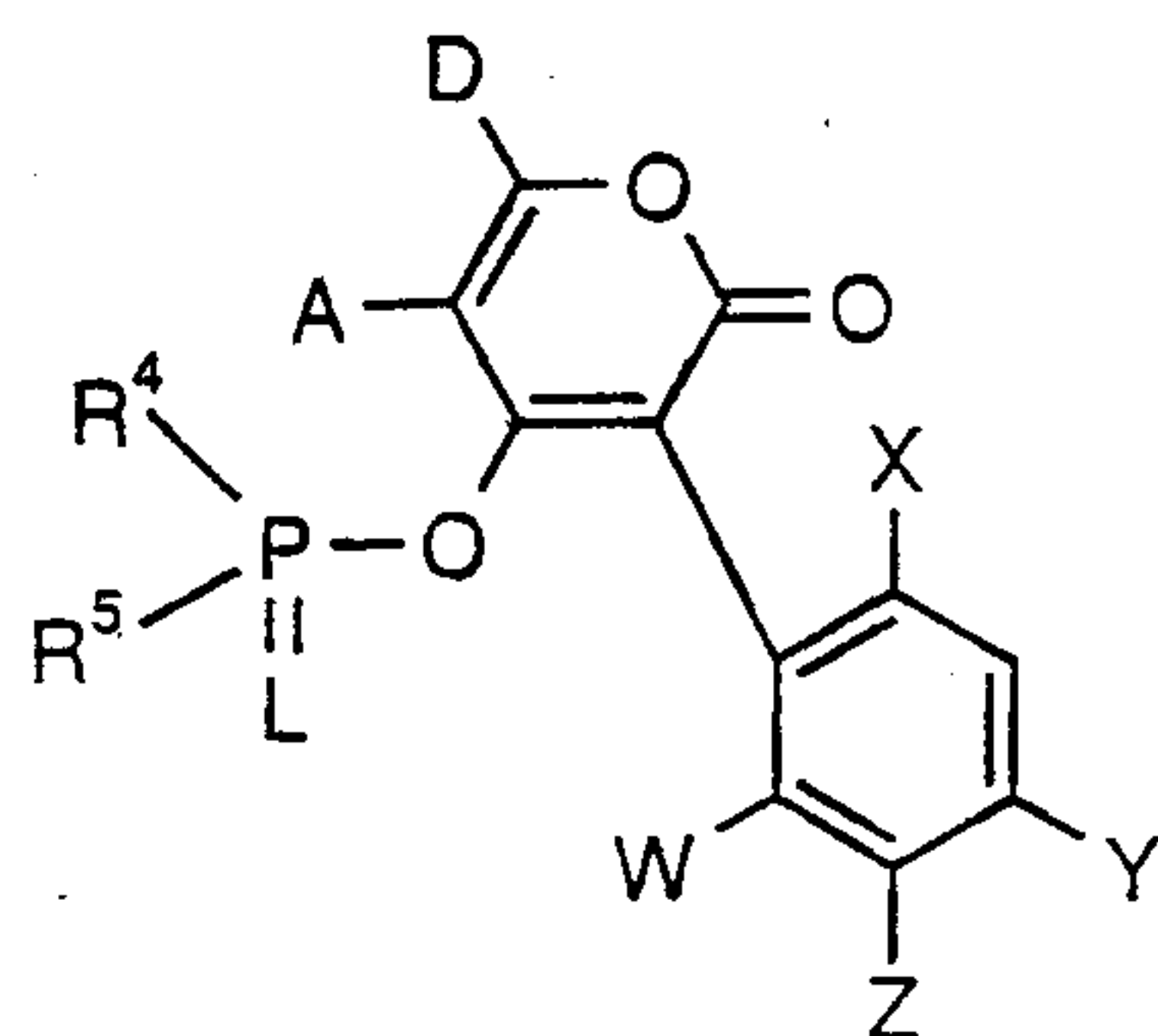
(I-4-d):



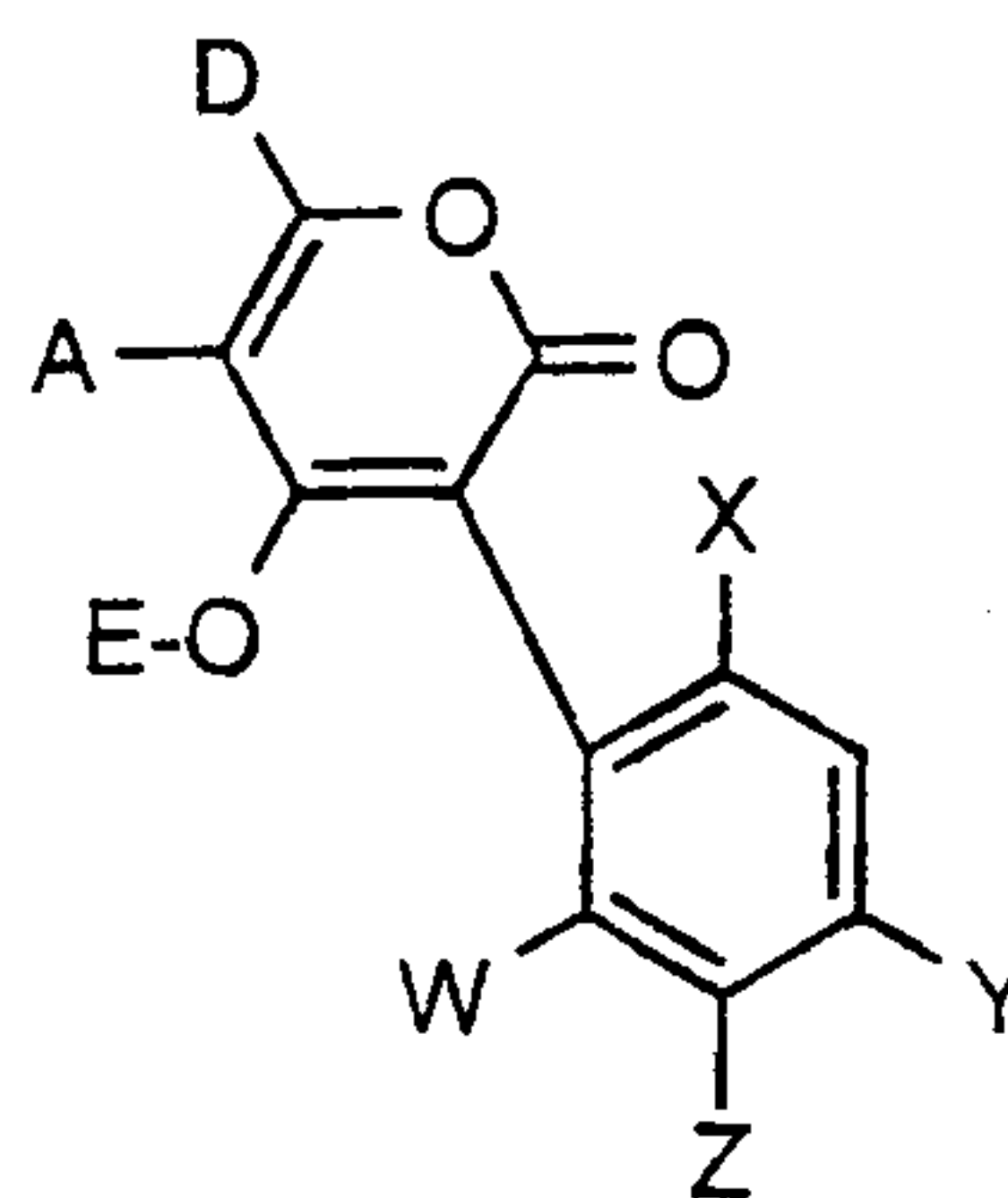
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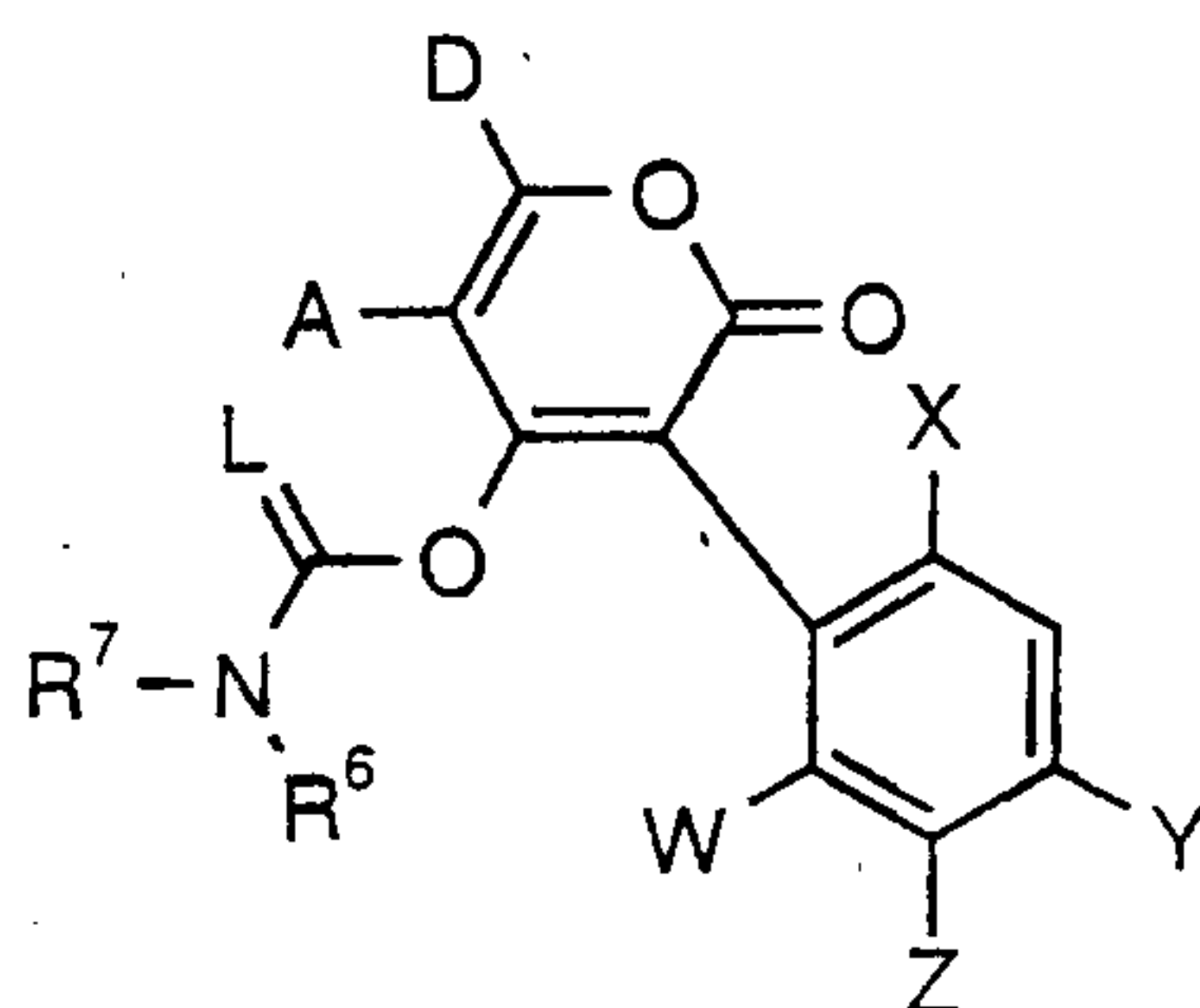
(I-4-e):



(I-4-f):



(I-4-g):



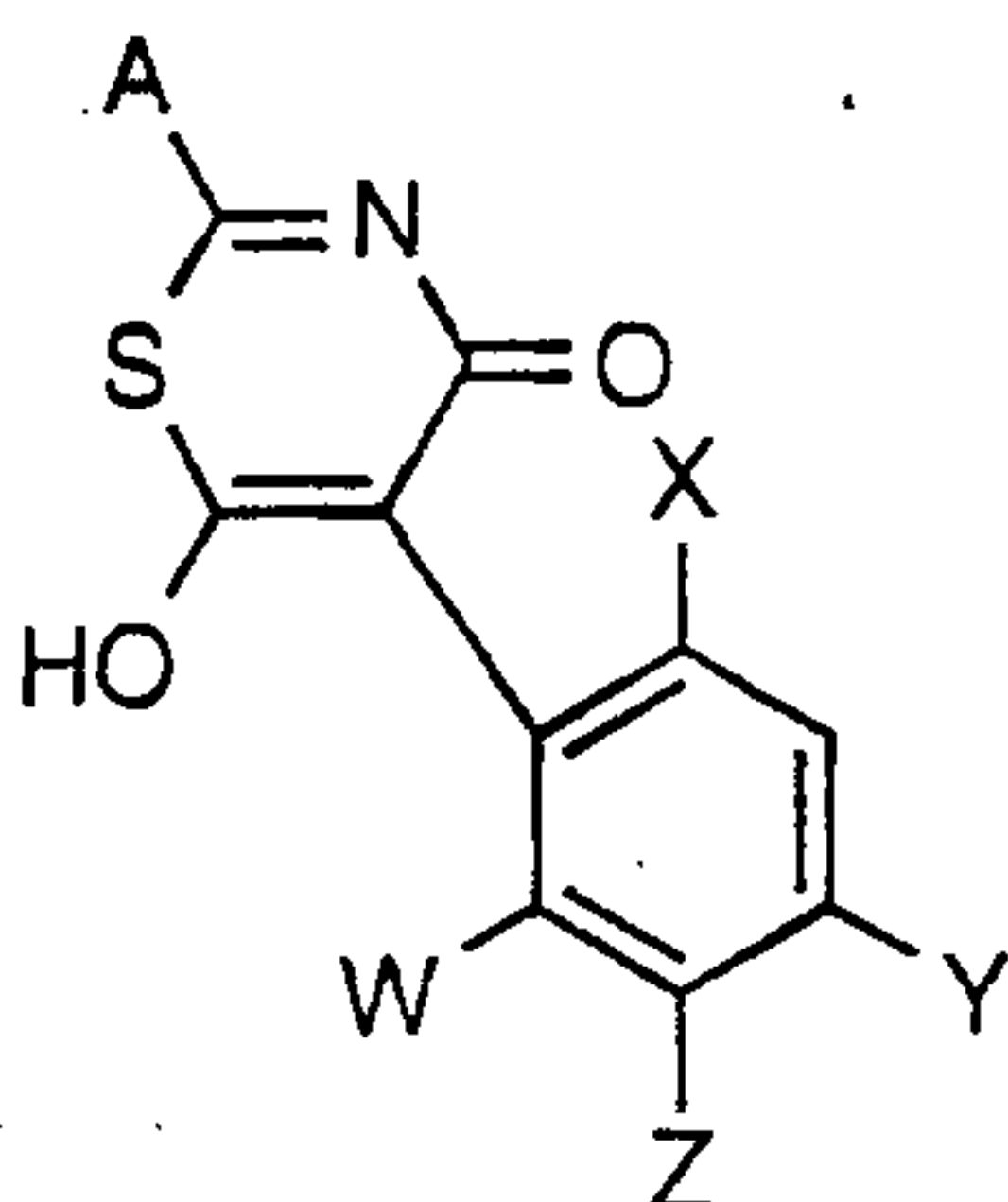
in which

A, D, E, L, M, W, X, Y, Z, R¹, R², R³, R⁴, R⁵, R⁶ and R⁷ are each as defined above.

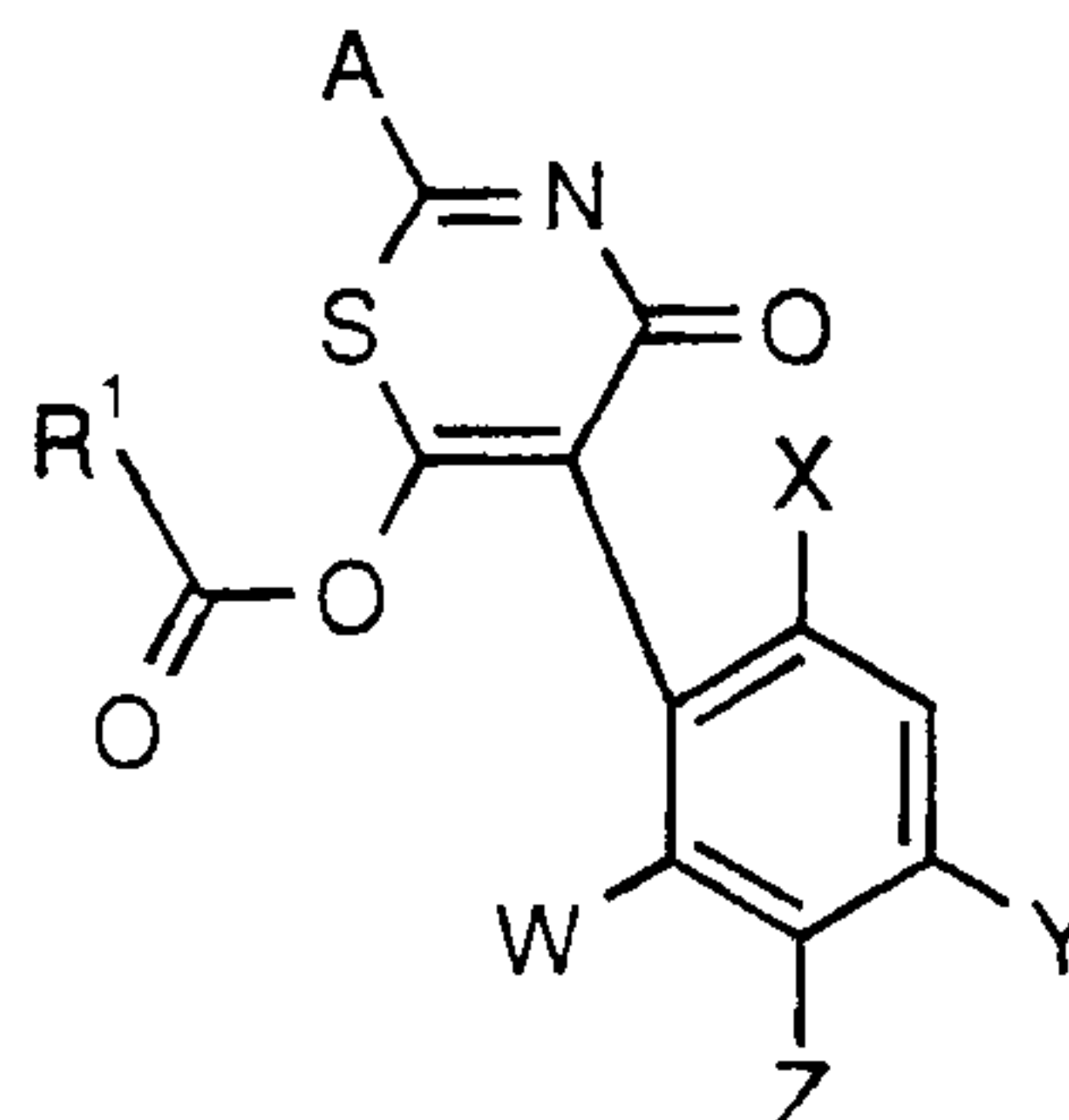
5

Including the various meanings (a), (b), (c), (d), (e), (f) and (g) of the group G, the following principal structures (I-5-a) to (I-5-g) result if CKE represents the group (5):

(I-5-a):



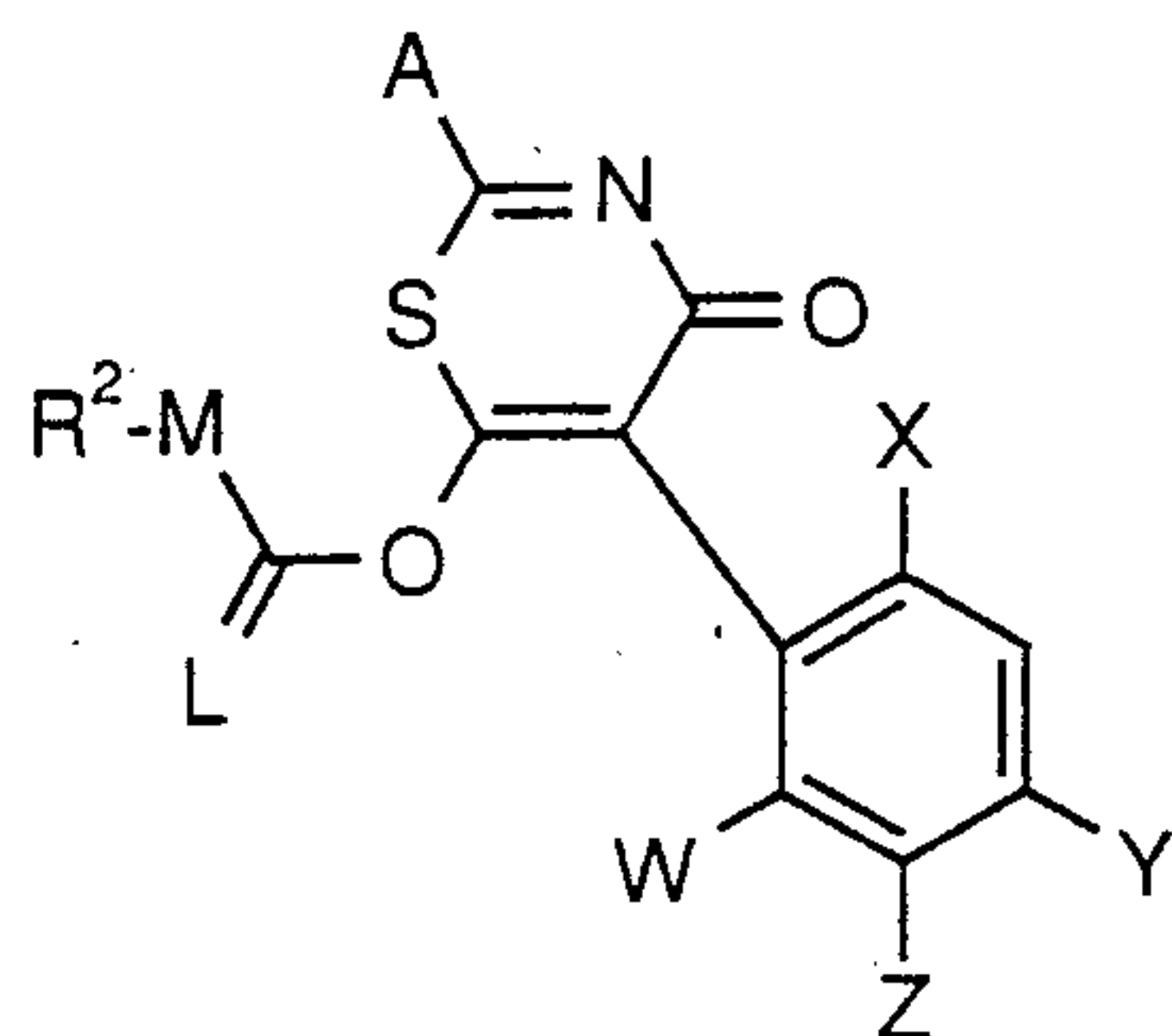
(I-5-b):



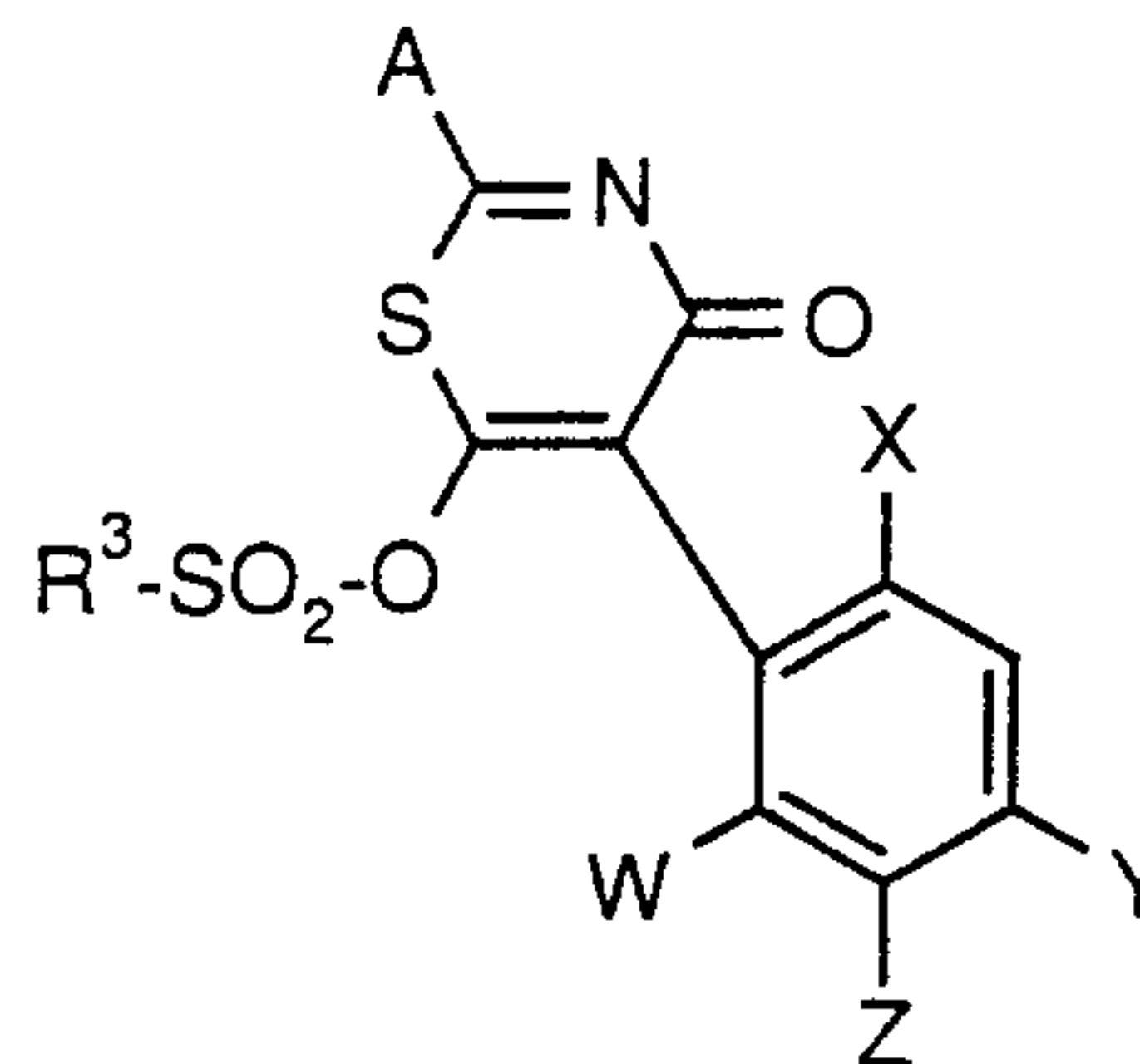
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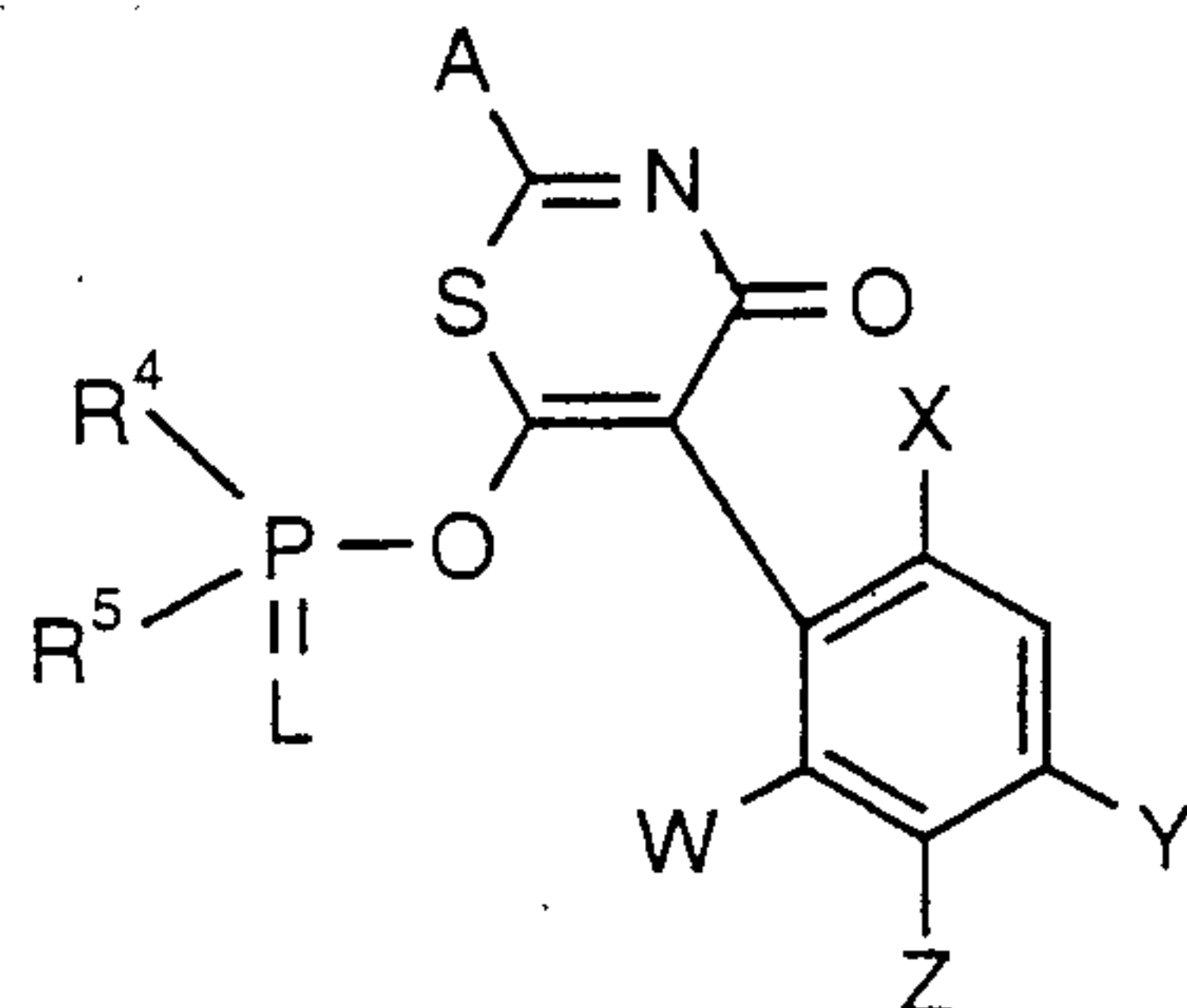
(I-5-c):



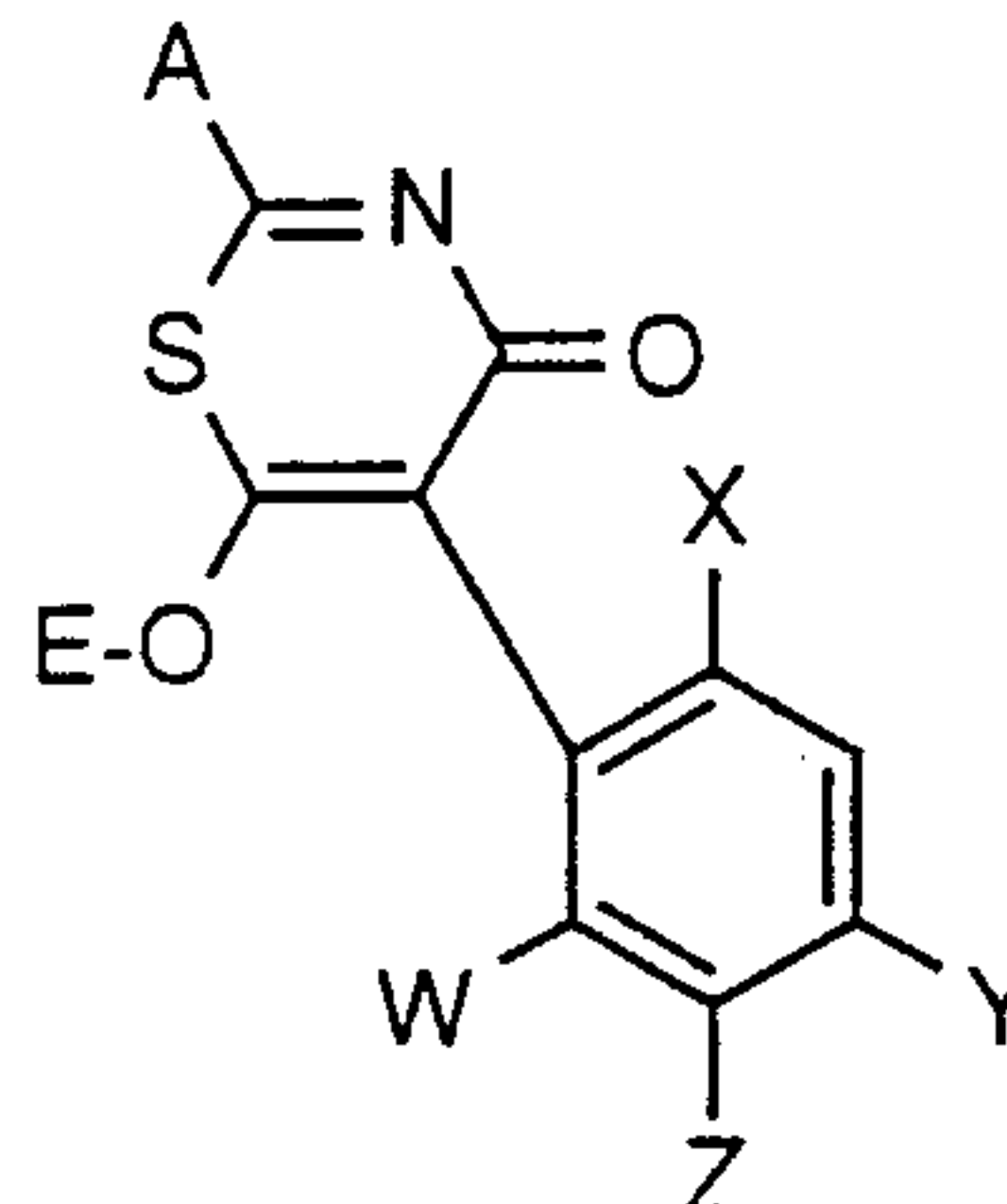
(I-5-d):



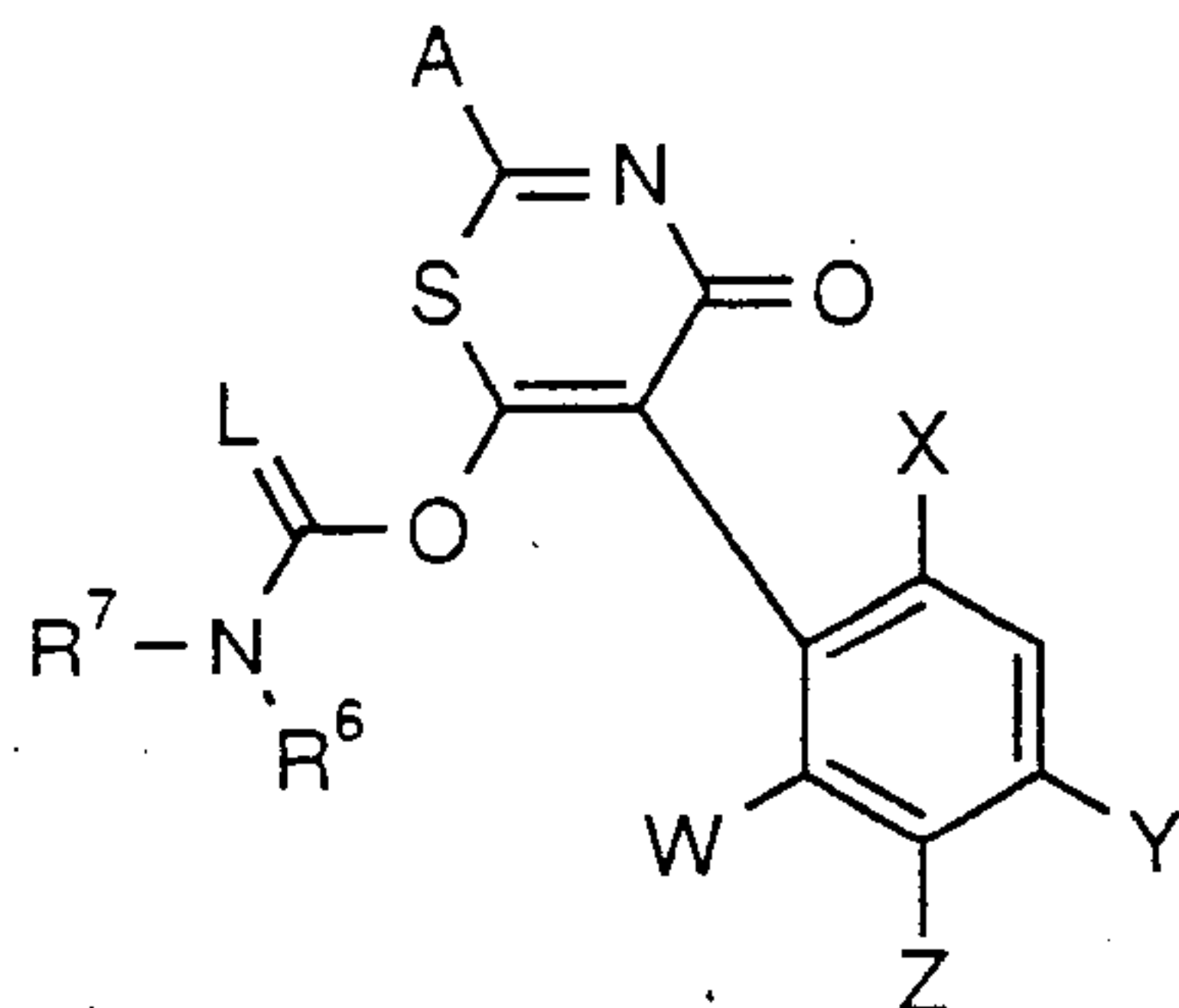
(I-5-e):



(I-5-f):



(I-5-g):



in which

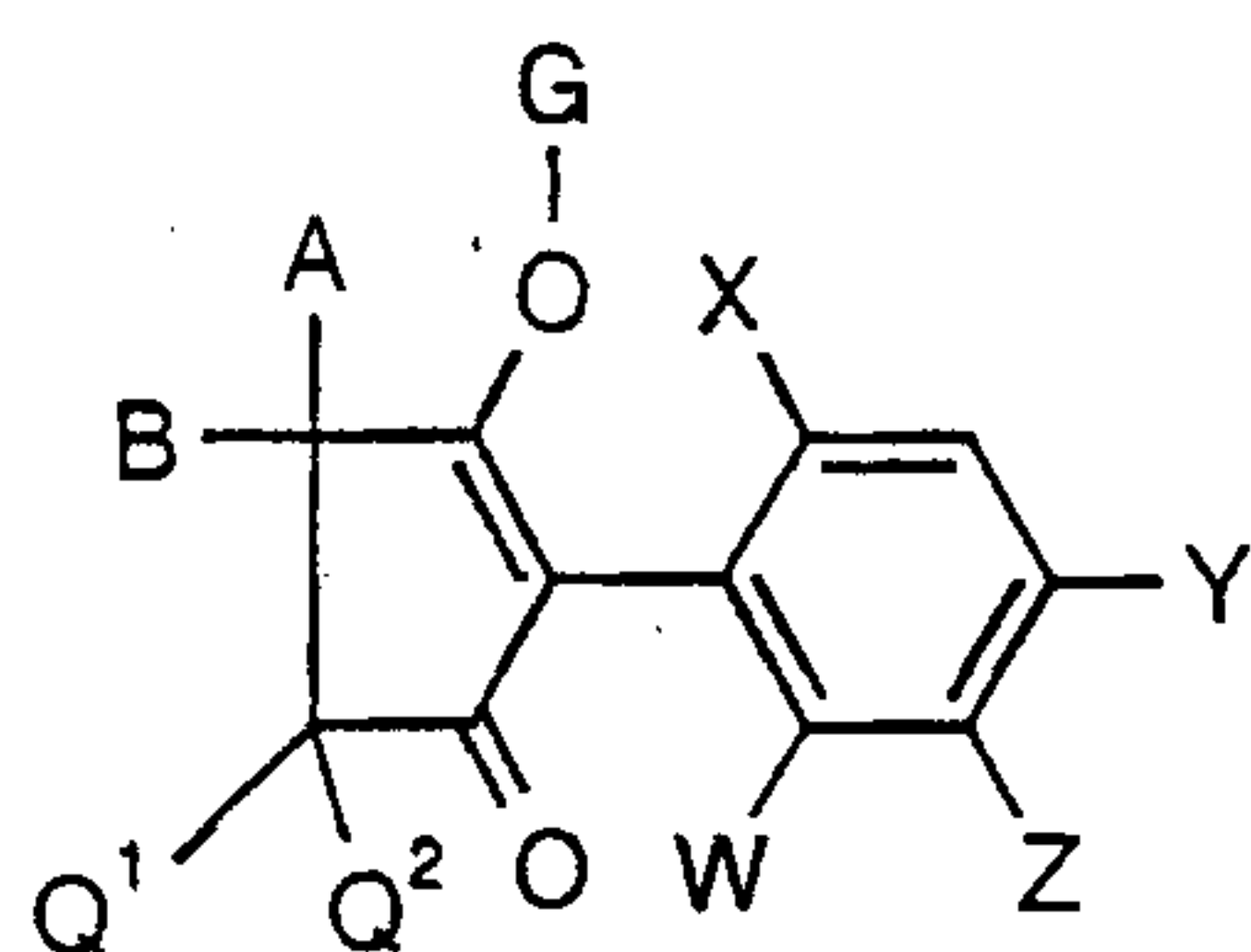
A, E, L, M, W, X, Y, Z, R¹, R², R³, R⁴, R⁵, R⁶ and R⁷ are each as defined above.

5

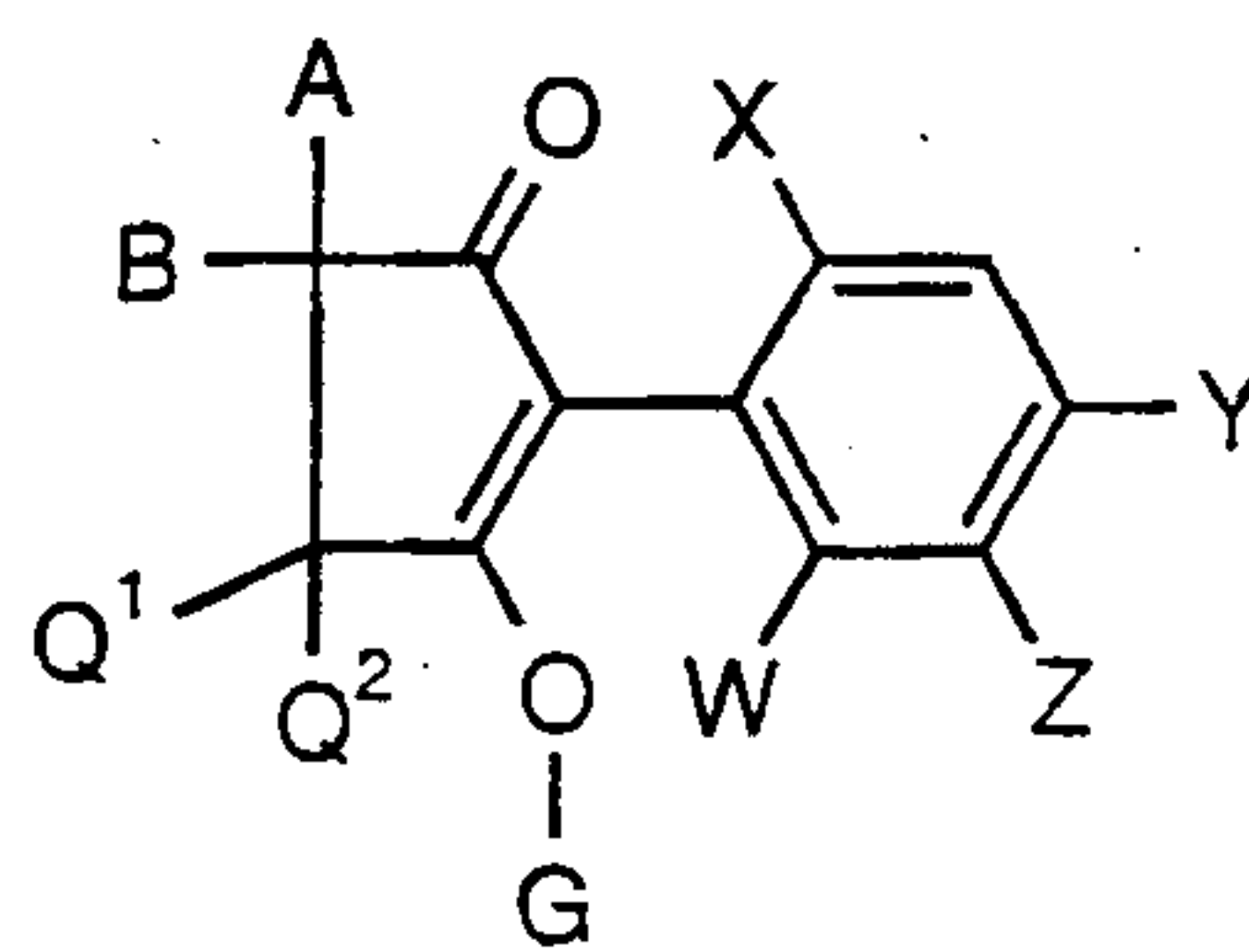
Depending on the position of the substituent G, the compounds of the formula (I-6) can be present in the two isomeric forms of the formula (I-6-A) and (I-6-B)

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(I-6-A)



(I-6-B)

which is meant to be indicated by the dashed line in the formula (I).

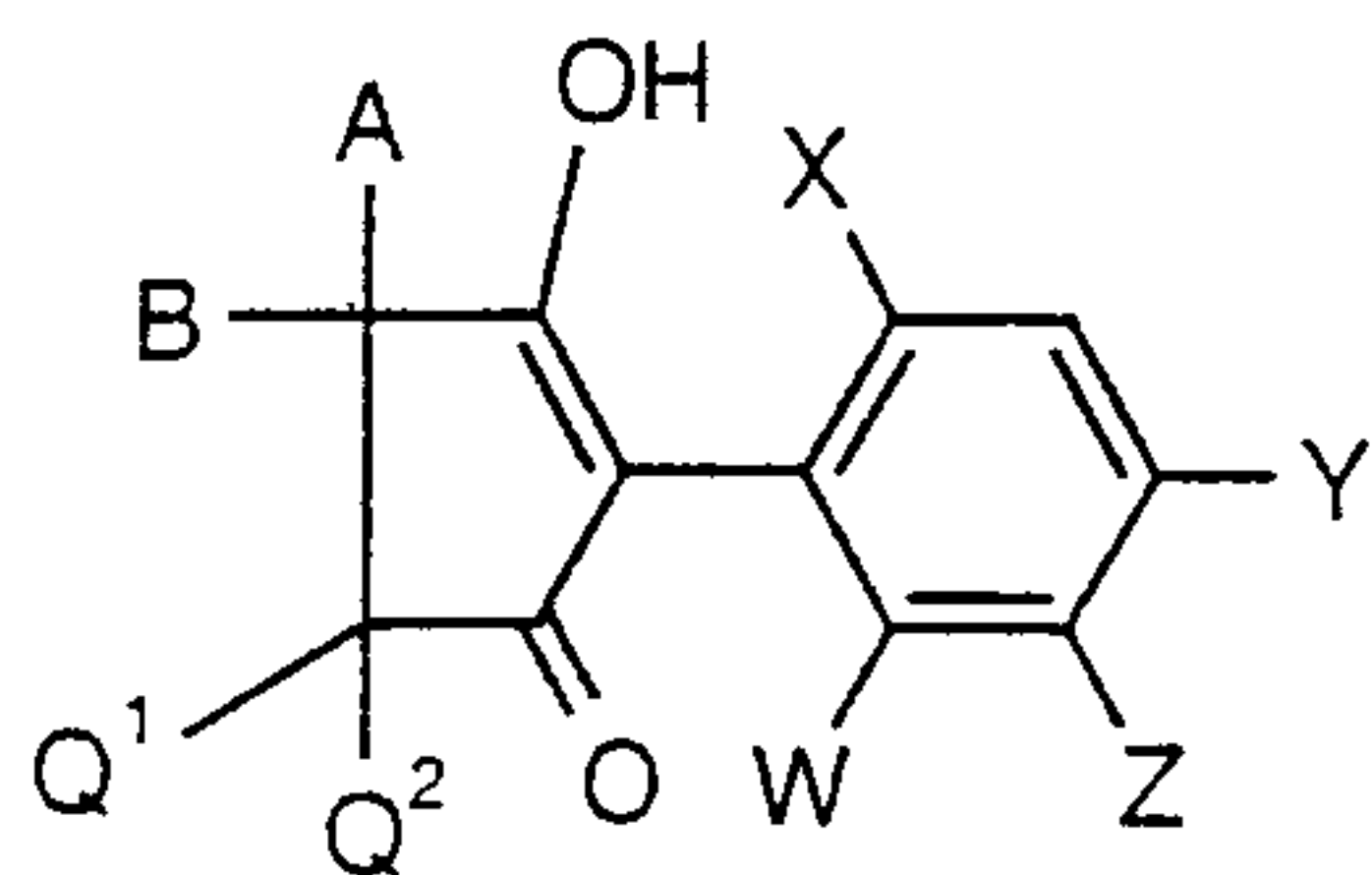
- 5 The compounds of the formulae (I-6-A) and (I-6-B) can be present both as mixtures and in the form of their pure isomers. Mixtures of the compounds of the formulae (I-6-A) and (I-6-B) can be separated, if desired, by physical methods, for example by chromatographic methods.
- 10 For reasons of clarity, hereinbelow in each case only one of the possible isomers is given. This does not exclude that the compounds may, if appropriate, be present in the form of the isomer mixtures or in the respective other isomeric form.

15 Including the various meanings (a), (b), (c), (d), (e), (f) and (g) of the group G, the following principal structures (I-6-a) to (I-6-g) result:

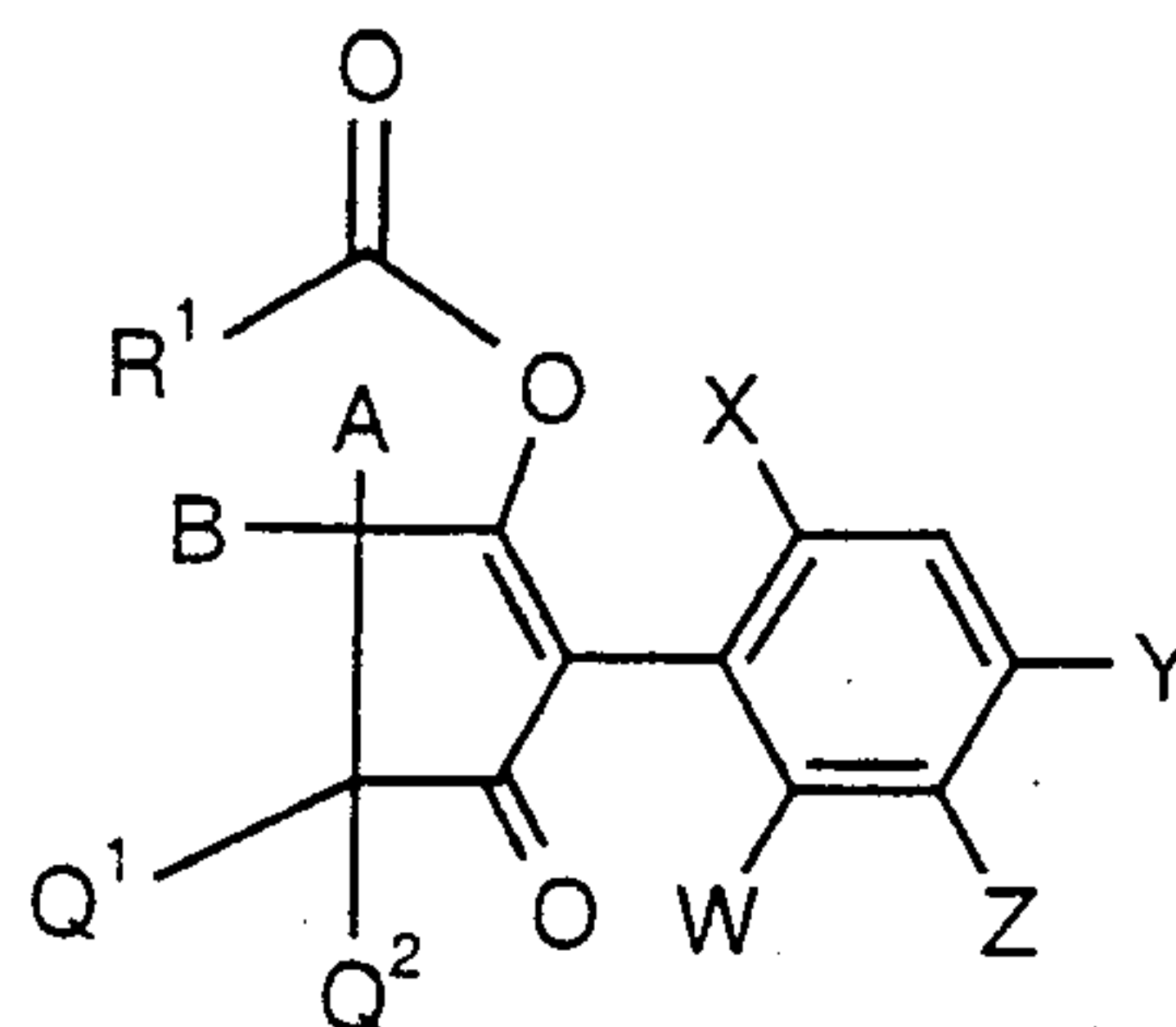
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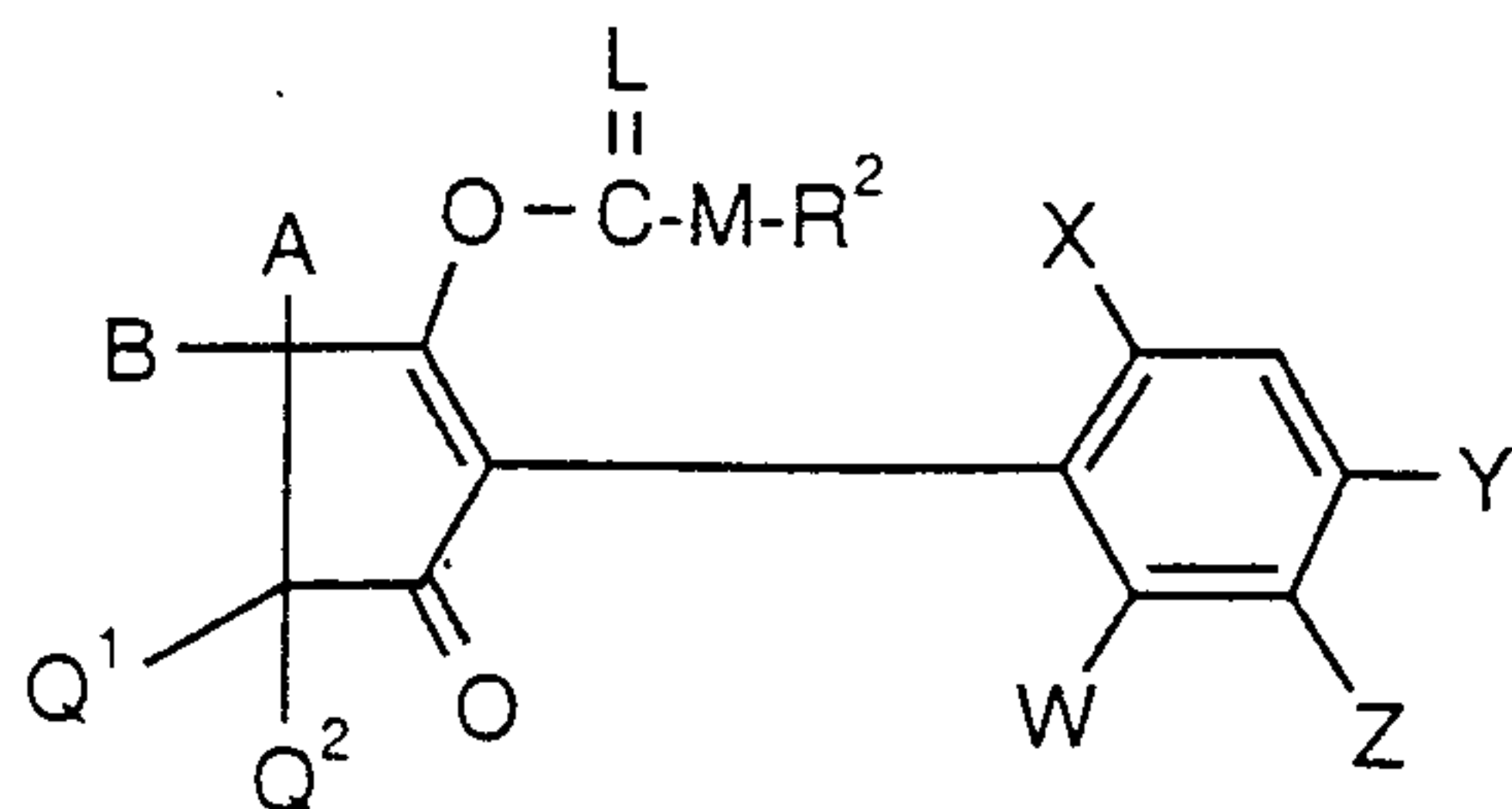
(I-6-a):



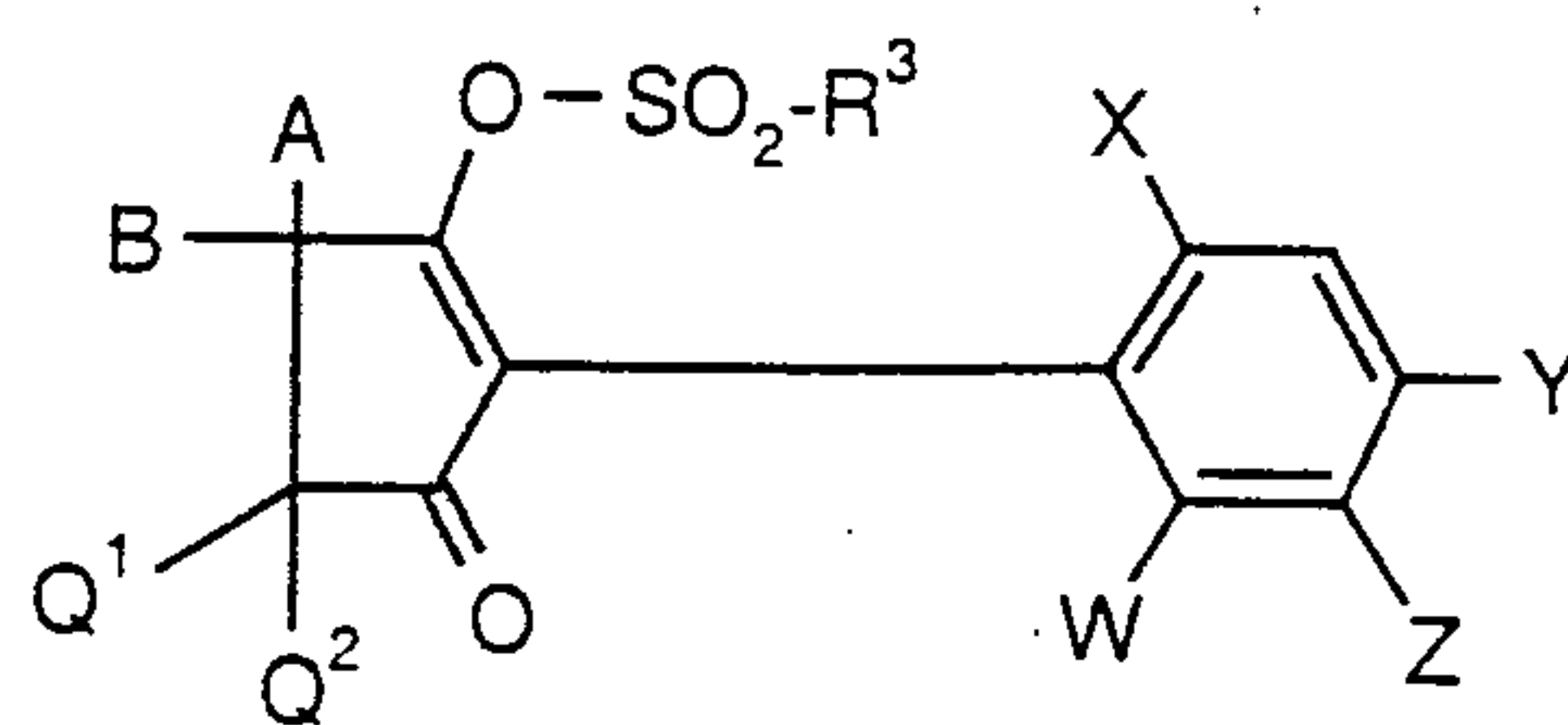
(I-6-b):



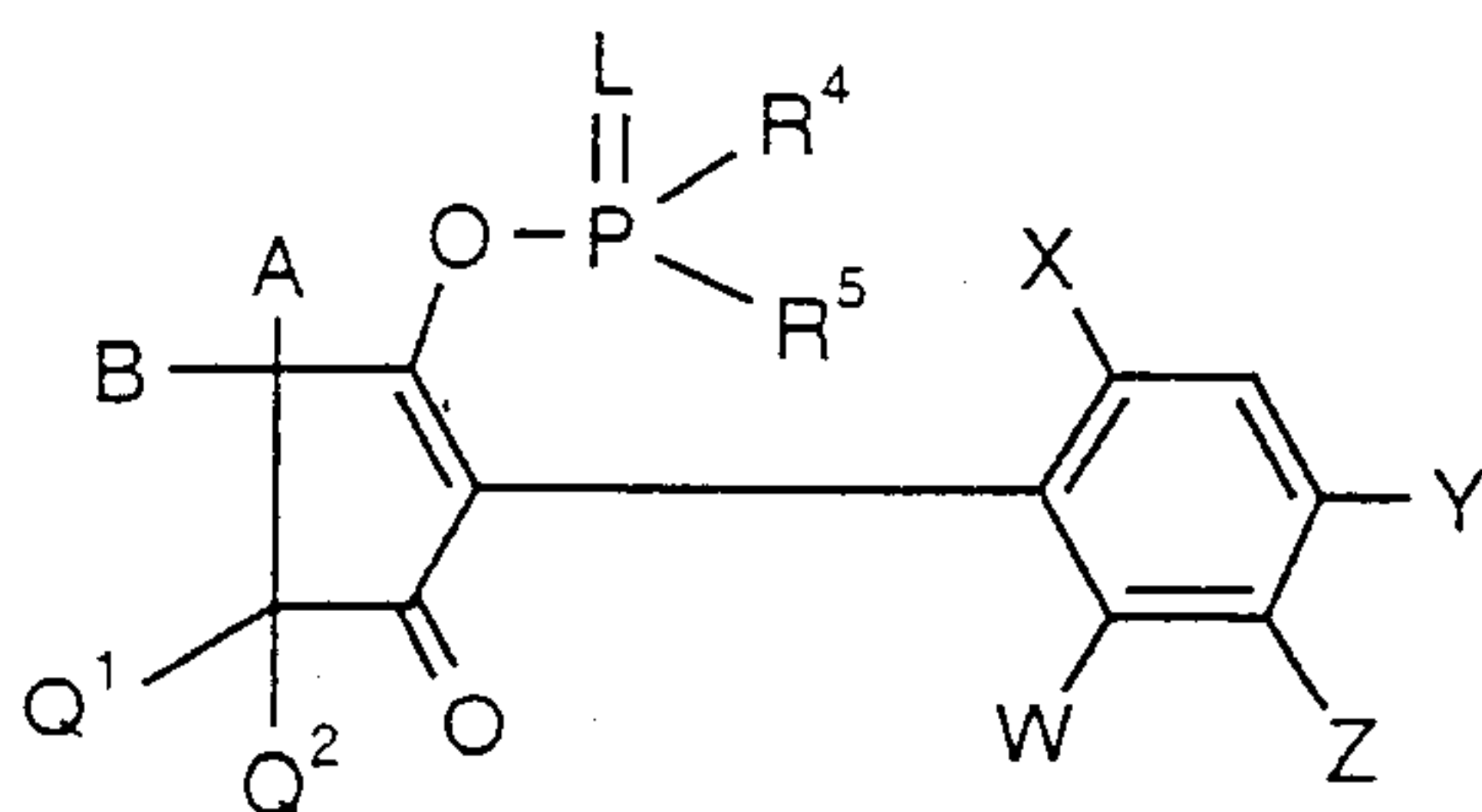
(I-6-c):



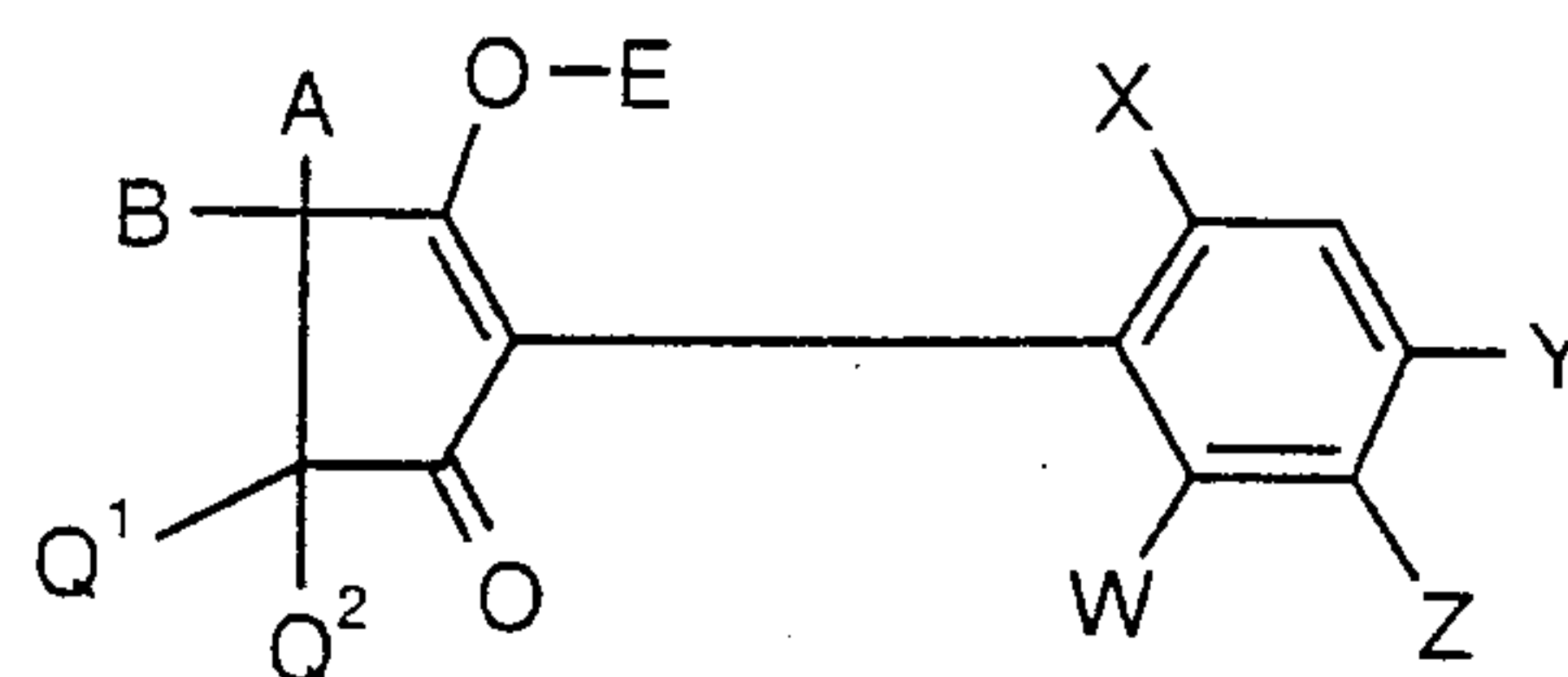
(I-6-d):



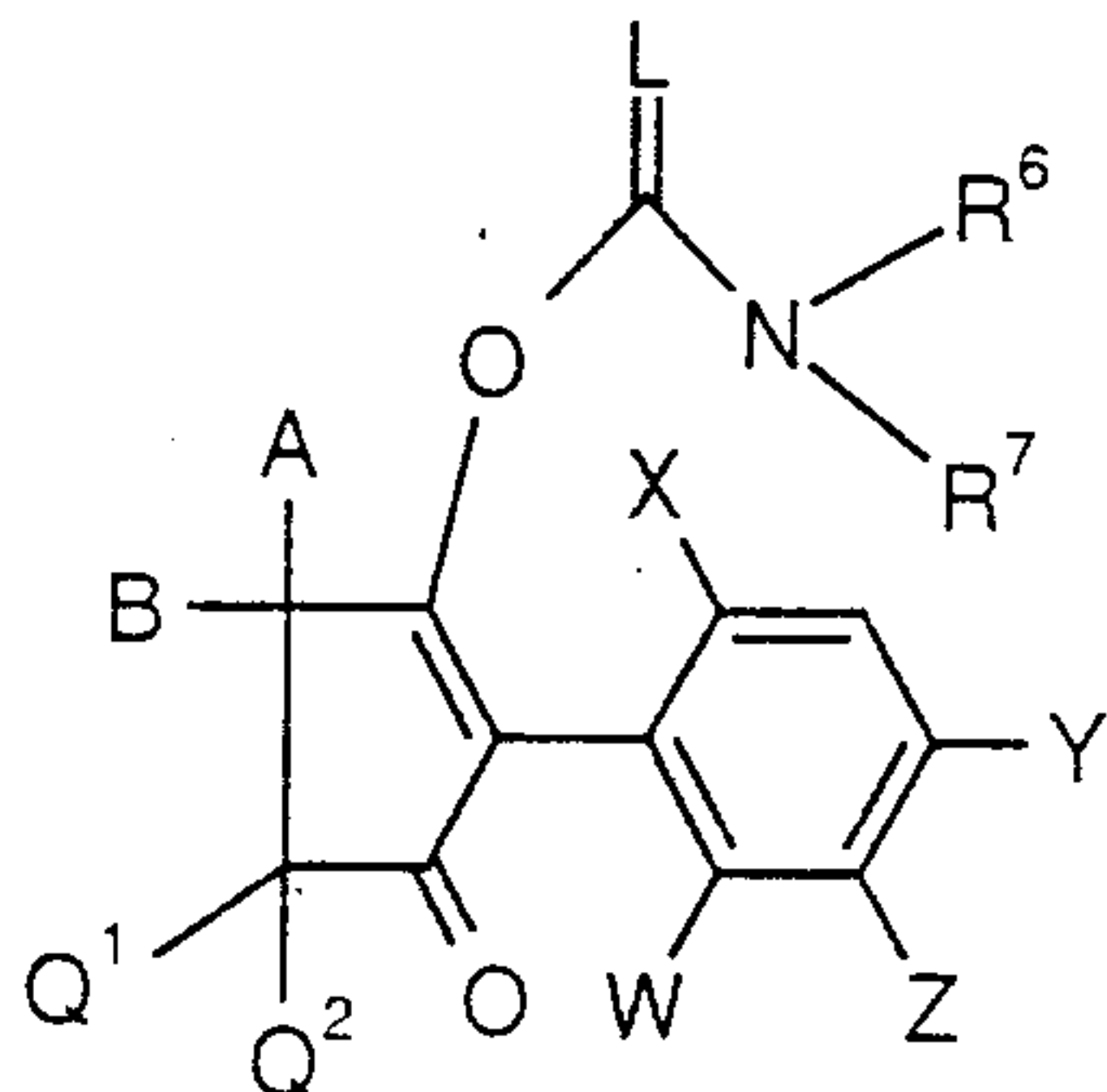
(I-6-e):



(I-6-f):



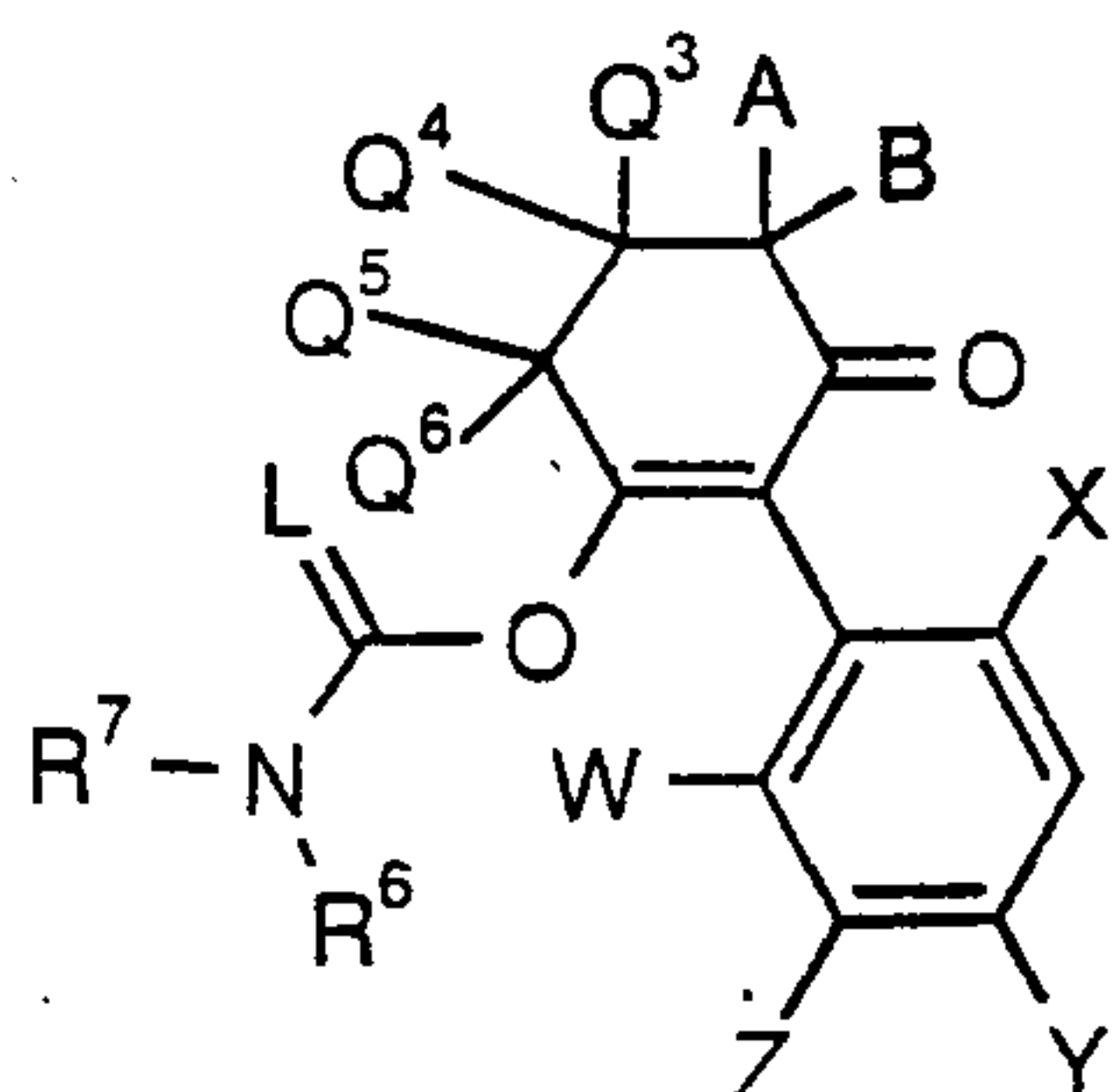
(I-6-g):



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(I-7-g):

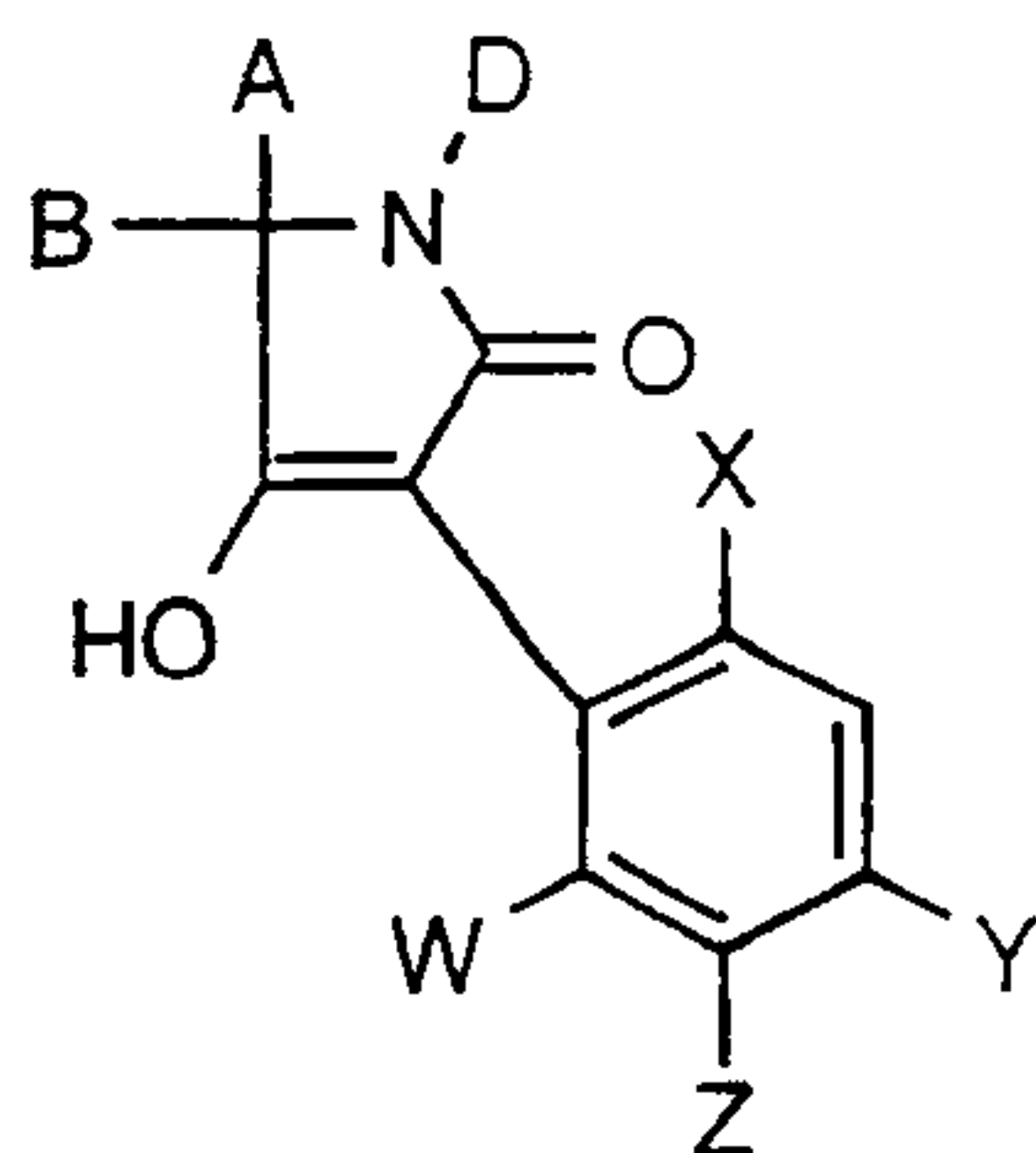


in which

5 A, B, E, L, M, Q³, Q⁴, Q⁵, Q⁶, W, X, Y, Z, R¹, R², R³, R⁴, R⁵, R⁶ and R⁷ are each as defined above.

Furthermore, it has been found that the novel compounds of the formula (I) are obtained by one of the processes described below:

10 (A) Substituted 3-phenylpyrrolidine-2,4-diones or their enols of the formula (I-1-a)



(I-1-a)

in which

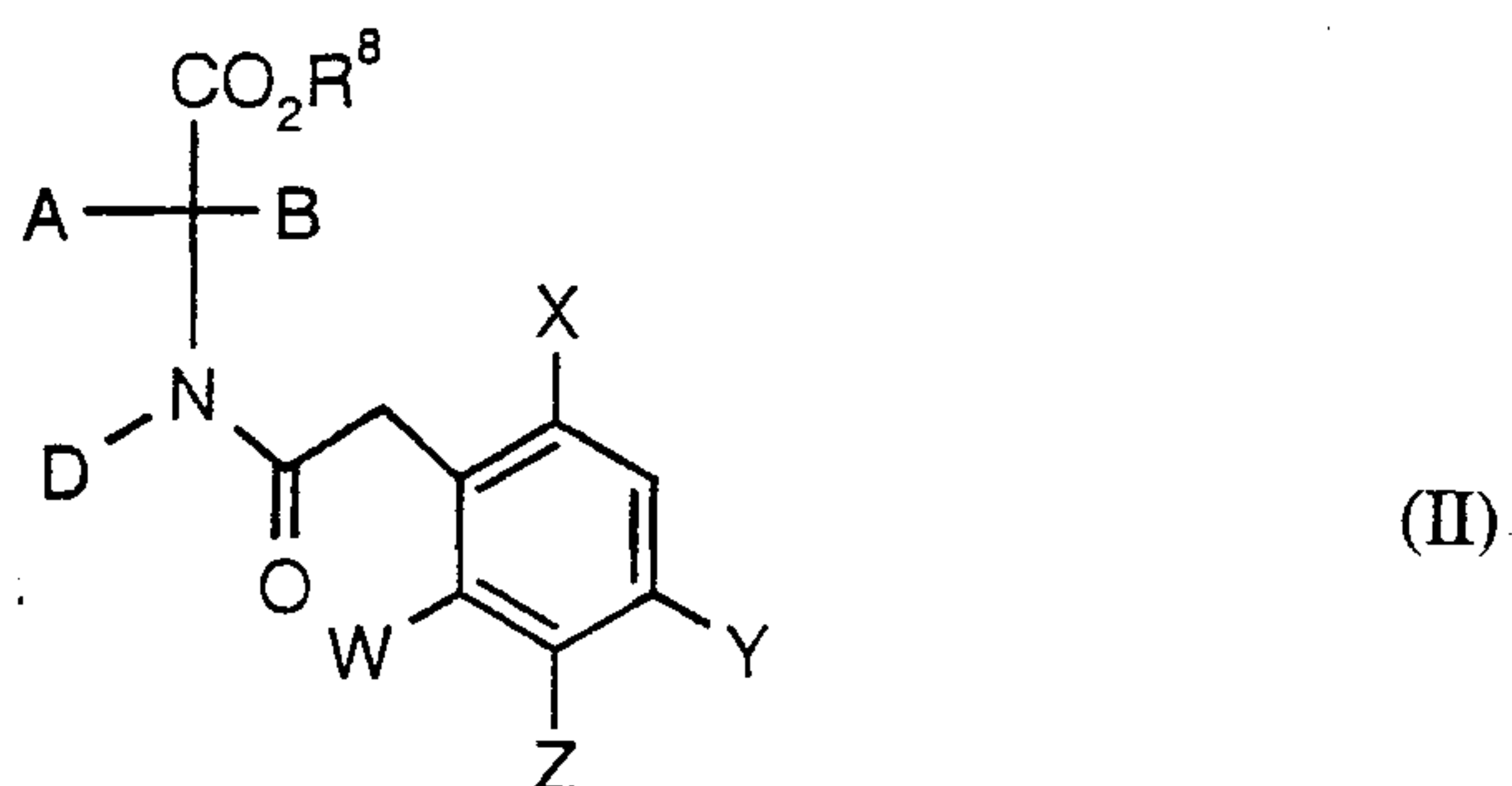
15 A, B, D, W, X, Y and Z are each as defined above

are obtained when

N-acylamino acid esters of the formula (II)

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in which

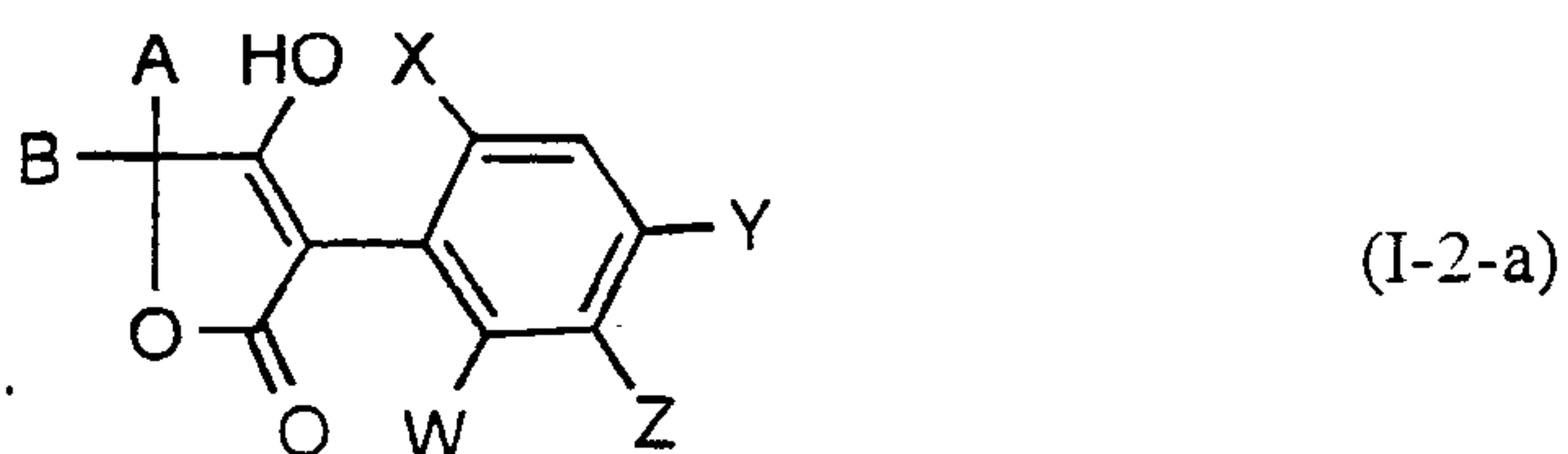
5 A, B, D, W, X, Y and Z are each as defined above

and

10 R⁸ represents alkyl (preferably C₁-C₆-alkyl),

are condensed intramolecularly in the presence of a diluent and in the presence of a base.

15 (B) Moreover, it has been found that substituted 3-phenyl-4-hydroxy- Δ^3 -dihydrofuranone derivatives of the formula (I-2-a)



in which

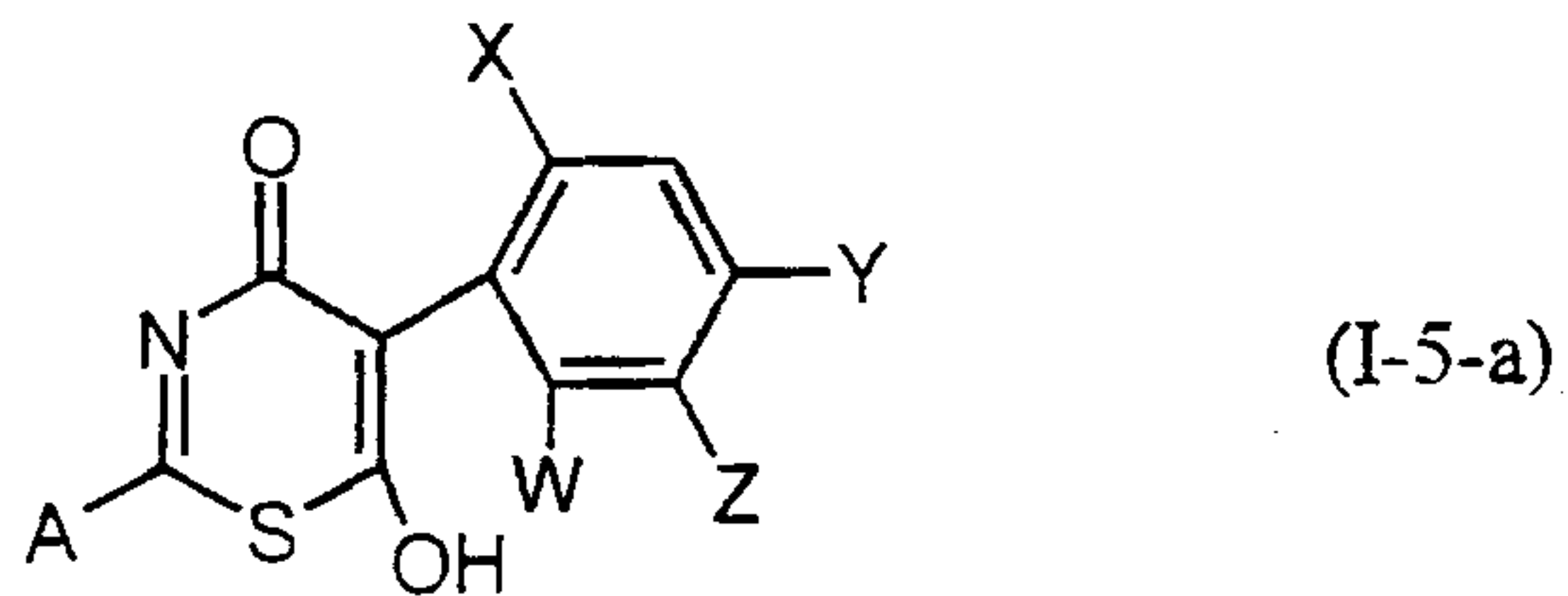
20 A, B, W, X, Y and Z are each as defined above

are obtained when

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(E) that the novel substituted phenyl-1,3-thiazine derivatives of the formula (I-5-a)



in which

5

A, W, X, Y and Z are each as defined above

are obtained when

10

thioamides of the formula (VII)

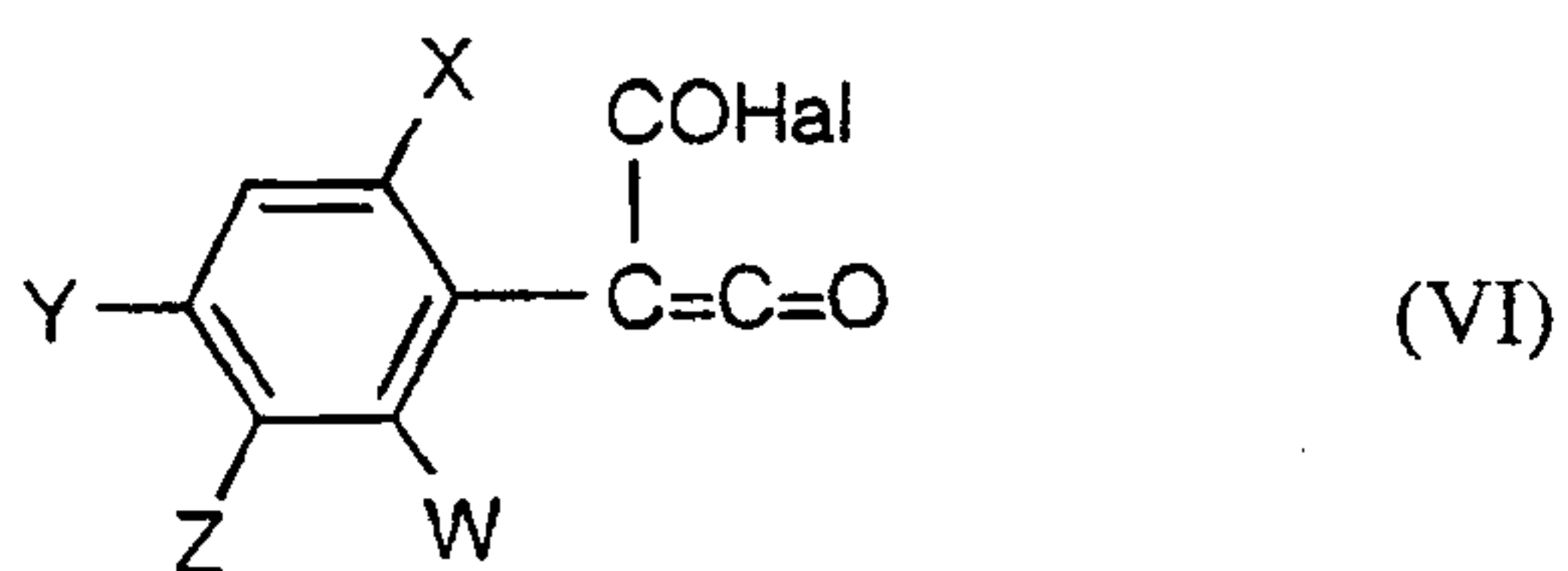


in which

A is as defined above

15

are reacted with ketene acid halides of the formula (VI)



in which

20

Hal, W, X, Y and Z are each as defined above,

