Synthesis and Biological Activities of 4-Arylmethylidene-1-aryl-2-(selenomorpholin-4-yl)-2-imidazolin-5-one

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In a search for novel agrochemical with high activity and low toxicity, a series of substituted 4,5-dihydro-imidazol-5-one containing selenomorpholine group were synthesized by a three-step synthetic route starting from 2-azido-3-aryl-acrylic acid ethyl ester. The structures of title compounds were confirmed by ^{1}H NMR, IR, mass spectroscopy and elemental analysis. The preliminary bioassay against *Gibberella zeae*, *Fusarium oxysporum*, *Pellicularia sasakii*, *Physalospora piricola* and *Cercospora beticola* indicated that most title compounds displayed fungicidal activity at the concentration of 50 ppm and compounds 4l, 4n, 4o were found to have particularly high activities against *Physalospora piricola*. A further *in vivo* test showed that compounds 4l, 4n and 4o possessed better fungicidal activity against *Physalospora piricola* at a concentration of 100 ppm than Carbendazim. To our knowledge, this is first report that 4,5-dihydro-imidazol-5-one containing selenomorpholine group display fungicidal activity against *Physalospora piricola*.

Keywords dihydro-imidazol-5-one, selenomorpholine, heterocyclic compound, bioactivity

Introduction

The bioactivity of 4,5-dihydro-imidazol-5-one derivatives has been of considerable interest for many years.[1-3] In 1980s, Cyanamid Company found some imidazolinone to be excellent herbicides. Since then, various derivatives have been used in pharmaceutical, agricultural and other areas. Many 4,5-dihydro-imidazol-5-one derivatives, such as Imazethapyr, Imazapyr, Imazaquin and Imazamethabenz-methyl, have shown excellent herbicidal and fungicidal activities.[4-6] Meanwhile, a wide range of biological activities has been attributed to the compounds containing cycle. In our previous papers, we found that some compounds containing selenomorpholine have good antibacterial activity and system activity.[7-13] We reasoned that if a heterocycle were to be introduced into the 4,5-dihydro-imidazol-5-one ring, the linked heterocyclic compound may enhance the activity and broaden the bioactive spectrum. As a continuation of our research work aimed at searching for novel agrochemicals, our interest in the heterocyclic compound containing selenomorpholine let us to synthesize a number of title compounds.

Experimental

Melting points were determined with a model X4 apparatus and were uncorrected. ^{1}H NMR spectra were taken on an XL-200 spectrometer in $CDCl_3$ using TMS as an internal standard. Mass spectra were recorded on a Hewlett-Packard 5988A instrument. Elemental analyses were carried on a Perkin-Elmer 2400 CHN instrument. All solvents and materials were of reagent grade and purified as required. The intermediate, 3-aryl-2-azido acrylic acid ethyl ester, was prepared according to the reported method.[14] The purity of the products were tested by TLC and some characteristic data are listed in Table 1, Table 2 and Table 3, respectively.

General procedure for the preparation of 3-*aryl*-2-*triphenylphosphiniminyl acrylic acid ethyl ester* (**2**)

To a stirred mixture of 10 mmol of 3-aryl-2-azido acrylic acid ethyl ester (**1**)[15] and 100 mL of anhydrous methylene dichloride, a solution of 2.62 g (10 mmol) triphenylphosphine in 90 mL of anhydrous methylene dichloride was added dropwise at 0 ℃ and the reaction was allowed to proceed for 3—4 h. After the completion of reaction, the solvent was removed under reduced pressure, ethyl ether was added to dissolve the residue, then petroleum ether was added till crystals precipitated, and the crystals were collected by filtration, washed with petroleum ether and dried under reduced pressure, yield 71%—88%.

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Received May 6, 2003; revised June 30, 2003; accepted August 11, 2003.

General procedure for the preparation of 3-*aryl*-2-*aryliminomethylidene amino acrylic acid ethyl ester* **(3)**

To a solution of 6 mmol of 3-aryl-2-triphenylphosphiniminyl acrylic acid ethyl ester **(2)** in 18 mL of anhydrous methylene dichloride was added the aromatic isocyanate (6 mmol) under nitrogen at room temperature. After stirring for 3—6 h, the solvent was removed under reduced pressure and the mixture solvent of ether and petroleum ether (1∶2, 25 mL) was added to precipitate the triphenylphosphine oxide. The precipitate was removed by filtration, the filtrate was collected and solvent removed to give carbodiimide **(3)**, which was used directly without further purification.

General procedure for the preparation of the title compounds **(4)**

To a solution of 5 mmol of carbodiimide **(3)** prepared above in 20 mL of acetonitrile was added 5 mmol of selenomorpholine and the mixture was stirred for 15 min. The solvent was removed under reduced pressure, the residue was recrystallized from ethyl ether and methylene dichloride to give 4-arylmethylidene-1-aryl-2-(selenomorpholin-4-yl)-4,5-dihydro-imidazol-5-one. From compound **2** to compound **4**, the total yield is 21.6%—57.7%.

Results and discussion

Synthesis and characterization of the title compounds

The synthetic pathway for the title compounds is outlined in Scheme 1. The synthesis started with 2-azido-3-aryl-acrylic acid ethyl ester **(1)**, which reacted with triphenylphosphine under mild condition to give intermediate **(2)** in yield of 71%—88%. Compounds **(2)** reacted with aromatic isocyanates to produce the key intermediates, 3-aryl-2-aryliminomethylidene amino acrylic acid ethyl ester **(3)**, which were allowed to react with selenomorpholine to afford the title compounds in yield of 21.6%—57.7%. The formation of **4** can be rationalized in terms of an nucleophilic addition of selenomorpholine to 3-aryl-2-aryliminomethylidene amino acrylic acid ethyl ester **(3)** to produce the intermediate 3-aryl-2-[1-arylamino-1-(selenomorlin-4-yl)methylidene amino] acrylic acid ethyl ester which cyclized fast to give **4**. The products **(4)** were purified by recrystallization from ether/methylene dichloride except **4e**, **4j** and **4t**. The structures of **4** were confirmed by ^{1}H NMR and MS spectroscopy as well as elemental analysis. Taking **4d** as a representative example, its ^{1}H NMR spectrum showed four methylene protons at δ 2.62 ($SeCH_2$) and 3.70 (NCH_2) as triplets respectively, the methylene protons at phenyl and proton at vinyl displayed two singlets at δ 5.98 and 6.76 respectively. The aromatic protons displayed a multiplet at δ 6.82—8.94. In addition, the EI-MS spectra of **4** demonstrate the existence of the molecular ion peaks, all fragmentation ions are consistent with their structures and can be clearly assigned, for example, compound **4d**, under electron impact, gives the molecular ion peak m/z (%) of 441 (5.8), the other ion peaks being 320 (12.2), 150 (6.5), 135 (19.0), 107 (14.6), and 77 (100).

Fungicidal activity

The fungicidal activity of the title compounds was measured according to the modified method described previously.[16]

The five fungi used, *Gibberella zeae*, *Fusarium oxysporum*, *Pellicularia sasakii*, *Physalospora piricola* and *Cercospora beticola*, belong to the group of field fungi and were isolated from various crops. The effect of title compounds on growth of the test fungi was investigated in solid medium. The medium contained potato, 200 g; glucose, 15 g; agar, 20 g in 1 L distilled water. Samples were dissolved in dimethyl formamide, so that the final concentration of solvent in the medium was 10 mL/L. Medium (20 mL) including the particular sample in the required concentrations was transferred to Petri dishes (90 mm) and inoculated with a small piece of mycelium (2 mm in diameter). The dishes were incubated at 27 ℃ in the dark. Mycelial diameters were measured after 72 h. Evaluation was carried out by measuring the diameter of the colonies three times at different site. The mean value of three repetitions for each fungus was used for calculation. The data were evaluated by analysis of variance. The probability of single difference was calculated at the 5% level. All data were statistically significant at this level. The results indicated that most title compounds displayed good fungicidal activity against *Physalospora piricola* at 50 ppm, but they were poorly active against *Gibberella zeae*, *Fusarium oxysporum*, *Pellicularia sasakii* and *Cercospora beticola* (Table 4). The further *in vivo* bioassay carried out by the conventional method indicated that compounds **4l**, **4n** and **4o** possessed better fungicidal activity against *Physalospora piricola* at the concentration of 200 ppm than Carbendazim did at the concentration of 100 ppm (Table 5), which is well known for good fungicidal activity against *Physalospora piricola*. In gerneral, when Ar^2 is 4-chloride phenyl and 3-methyl phenyl, compounds **4** have better fungicidal activity against *Physalospora piricola*, when Ar^2 is 3-methyl phenyl, compounds **4** have bad fungicidal activity. Although 4,5-dihydro-imidazol-5-one derivatives have been reported to possess various biological activities, as far as we know there is no report on their fungicidal activity against *Physalospora piricola*, further studies on the structure-activity relationships and structural modification of these compounds are being pursued.

Scheme 1

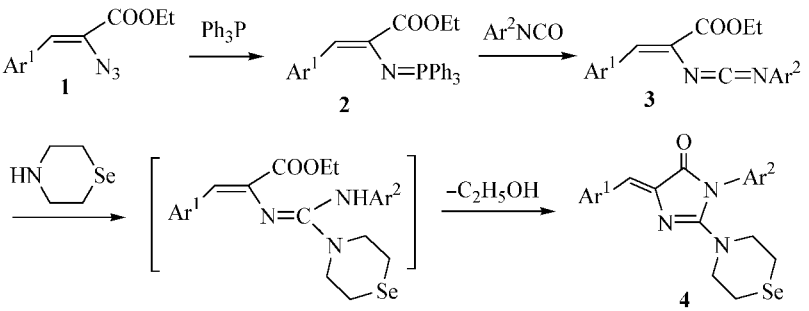

Table 1 Experimental data of compounds **4**

No.	Ar^1	Ar^2	Formula	m.p. (℃)	Yield[a] (%)	Elemental analysis (Calcd/Found, %)		
						C	H	N
4a	C_6H_5	C_6H_5	$C_{20}H_{19}N_3OSe$	209—210	50.2	60.61/60.56	4.83/4.72	10.60/10.49
4b	4-$CH_3OC_6H_4$	C_6H_5	$C_{21}H_{21}N_3O_2Se$	202—204	48.5	59.16/59.28	4.96/5.02	9.86/9.73
4c	4-ClC_6H_4	C_6H_5	$C_{20}H_{18}ClN_3OSe$	219—221	49.0	55.76/55.67	4.21/4.17	9.75/9.81
4d	3,4-$OCH_2OC_6H_3$	C_6H_5	$C_{21}H_{19}N_3O_3Se$	209—211	57.7	57.28/57.15	4.35/4.42	9.54/9.47
4e	4-$Me_2NC_6H_4$	C_6H_5	$C_{22}H_{24}N_4OSe$	—[b]	30.2	60.13/59.88	5.51/5.67	12.75/12.98
4f	C_6H_5	3-ClC_6H_4	$C_{20}H_{18}ClN_3OSe$	169—170	31.1	55.76/55.64	4.21/4.36	9.75/9.66
4g	4-$CH_3OC_6H_4$	3-ClC_6H_4	$C_{21}H_{20}ClN_3O_2Se$	203—204	45.8	54.73/54.81	4.37/4.32	9.12/9.25
4h	4-ClC_6H_4	3-ClC_6H_4	$C_{20}H_{17}Cl_2N_3OSe$	188—189	42.2	51.63/51.69	3.68/3.78	9.03/9.14
4i	3,4-$OCH_2OC_6H_3$	3-ClC_6H_4	$C_{21}H_{18}ClN_3O_3Se$	208—210	44.3	53.12/52.89	3.82/3.67	8.85/8.72
4j	4-$Me_2NC_6H_4$	3-ClC_6H_4	$C_{22}H_{23}ClN_4OSe$	—	28.1	55.76/55.51	4.89/4.75	11.82/12.03
4k	C_6H_5	4-ClC_6H_4	$C_{20}H_{18}ClN_3OSe$	197—199	45.0	55.76/55.83	4.21/4.34	9.75/9.68
4l	4-$CH_3OC_6H_4$	4-ClC_6H_4	$C_{21}H_{20}ClN_3O_2Se$	212—214	47.2	54.73/54.62	4.37/4.41	9.12/9.10
4m	4-ClC_6H_4	4-ClC_6H_4	$C_{20}H_{17}Cl_2N_3OSe$	222—223	33.0	51.63/51.57	3.68/3.59	9.03/8.87
4n	3,4-$OCH_2OC_6H_3$	4-ClC_6H_4	$C_{21}H_{18}ClN_3O_3Se$	240—241	21.6	53.12/53.08	3.82/3.76	8.85/8.79
4o	4-$Me_2NC_6H_4$	4-ClC_6H_4	$C_{22}H_{23}ClN_4OSe$	245—247	39.1	55.76/55.70	4.89/4.81	11.82/11.75
4p	C_6H_5	3-$CH_3C_6H_4$	$C_{21}H_{21}N_3OSe$	159—160	52.8	61.46/61.52	5.16/5.10	10.24/10.27
4q	4-$CH_3OC_6H_4$	3-$CH_3C_6H_4$	$C_{22}H_{23}N_3O_2Se$	178—180	55.7	60.00/59.87	5.26/5.21	9.54/9.49
4r	4-ClC_6H_4	3-$CH_3C_6H_4$	$C_{21}H_{20}ClN_3OSe$	240—243	43.6	56.70/56.82	4.53/4.50	9.45/9.51
4s	3,4-$OCH_2OC_6H_3$	3-$CH_3C_6H_4$	$C_{22}H_{21}N_3O_3Se$	214—216	42.0	58.15/58.27	4.66/4.72	9.25/9.31
4t	4-$Me_2NC_6H_4$	3-$CH_3C_6H_4$	$C_{23}H_{26}N_4OSe$	—	32.8	60.92/60.81	5.78/5.81	12.36/12.39

[a] Yield of the final step; [b] syrupy liquid.

Table 2 ^1H NMR spectroscopic data of compounds **4**

No.	^1H NMR (δ, $CDCl_3$, TMS)
4a	2.63 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.75 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 6.82 (s, 1H, =CH); 7.22—8.08 (m, 10H, Ar)
4b	2.63 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 3.83(s, 3H, OCH_3); 6.82 (s, 1H, =CH); 7.01—8.07 (m, 9H, Ar)
4c	2.63 (t, $^3J_{HH}$ = 6.6 Hz, 4H, $SeCH_2$); 3.74 (t, $^3J_{HH}$ = 6.8 Hz, 4H, NCH_2); 6.74 (s, 1H, =CH); 7.22—8.02 (m, 9H, Ar)
4d	2.62 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.70 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 5.98 (s, 2H, OCH_2O); 6.76 (s, 1H, =CH); 6.82—8.94 (m, 8H, Ar)
4e	2.60 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.01 (s, 6H, NCH_3); 3.66 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 6.82 (s, 1H, =CH); 7.21—8.00 (m, 9H, Ar)
4f	2.66 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.74 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 6.82 (s, 1H, =CH); 7.20—8.08 (m, 9H, Ar)
4g	2.65 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.4 Hz, 4H, NCH_2); 3.84 (s, 3H, OCH_3); 6.80 (s, 1H, =CH); 7.21—8.06 (m, 8H, Ar)

Continued

No.	^{1}H NMR (δ, $CDCl_3$, TMS)
4h	2.63 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.74 (t, $^3J_{HH}$ = 7.4 Hz, 4H, NCH_2); 6.75 (s, 1H, =CH); 7.22—8.02 (m, 8H, Ar)
4i	2.65 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 5.99 (s, 2H, OCH_2O); 6.77 (s, 1H, =CH); 7.20—7.94 (m, 7H, Ar)
4j	2.67 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.03 (s, 6H, NCH_3); 3.68 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 6.66—8.01 (m, 8H, Ar)
4k	2.66 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.74 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 6.82 (s, 1H, =CH); 7.20—8.08 (m, 9H, Ar)
4l	2.65 (t, $^3J_{HH}$ = 7.2 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 3.83 (s, 3H, OCH_3); 6.82 (s, 1H, =CH); 6.88—8.06 (m, 8H, Ar)
4m	2.63 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.75 (t, $^3J_{HH}$ = 7.4 Hz, 4H, NCH_2); 6.75 (s, 1H, =CH); 7.20—8.02 (m, 8H, Ar)
4n	2.65 (t, $^3J_{HH}$ = 7.4 Hz, 4H, $SeCH_2$); 3.73 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 5.99 (s, 2H, OCH_2O); 6.77 (s, 1H, =CH); 7.01—7.94 (m, 7H, Ar)
4o	2.66 (t, $^3J_{HH}$ = 7.2 Hz, 4H, $SeCH_2$); 3.03 (s, 6H, NCH_3); 3.70 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 6.85—8.01 (m, 8H, Ar)
4p	2.41 (s, 3H, CH_3); 2.65 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.75(t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 6.80 (s, 1H, =CH); 7.07—8.09 (m, 9H, Ar)
4q	2.40 (s, 3H, CH_3); 2.63 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 3.83 (s, 3H, OCH_3); 6.80—8.06 (m, 8H, Ar)
4r	2.41 (s, 3H, CH_3); 2.63 (t, $^3J_{HH}$ = 7.0 Hz, 4H, $SeCH_2$); 3.75 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 6.72 (s, 1H, =CH); 7.01—8.02 (m, 8H, Ar)
4s	2.40 (s, 3H, CH_3); 2.62 (t, $^3J_{HH}$ = 7.2 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.2 Hz, 4H, NCH_2); 5.98 (s, 2H, OCH_2O); 6.74—7.95 (m, 7H, Ar)
4t	2.39 (s, 3H, CH_3); 2.62 (t, $^3J_{HH}$ = 6.8 Hz, 4H, $SeCH_2$); 3.72 (t, $^3J_{HH}$ = 7.0 Hz, 4H, NCH_2); 3.01(s, 6H, NCH_3); 6.84 (s, 1H, =CH); 6.66—8.02 (m, 7H, Ar)

Table 3 IR and EI-MS data of compounds **4**

No.	IR, ν/cm^{-1}		m/z (%)
	$\nu_{C=O}$	$\nu_{C=C}$	
4a	1720	1641	397 (M^+, 5.38), 276 (22.06), 150 (1.46), 136 (2.29), 77 (100)
4b	1725	1640, 1600	427 (M^+, 52.55); 306 (100); 107 (13.87); 77 (87.64)
4c	1730	1648	431 (M^+, 2.58); 310 (11.20); 150 (32.32); 135 (16.58); 109 (11.53); 77 (100)
4d	1716	1643	441 (M^+, 5.79); 320 (12.13); 150 (6.54); 135 (19.03); 107 (14.59); 77 (100)
4e	1711	1635	440 (M^+, 8.16); 320 (12.03); 159 (100); 135 (3.82); 108 (6.51); 77 (59.99)
4f	1717	1648	431 (M^+, 5.30); 311 (23.51); 151 (12.18); 116 (100)
4g	1716	1642, 1603	461 (M^+, 3.96); 341 (13.67); 165 (41.29); 151 (12.85); 146 (100); 107 (24.94)
4h	1726	1646	465 (M^+, 3.50); 345 (11.40); 165 (46.46); 150 (79.96); 111 (100)
4i	1714	1645	475 (M^+, 4.22); 355 (16.38); 160 (87.29); 151 (13.49); 111 (100); 107 (29.74)
4j	1722	1608	474 (M^+, 3.95); 165 (13.59); 159 (100); 135 (4.86); 110 (32.09)
4k	1725	1640	431 (M^+, 4.60); 310 (16.81); 165 (27.35); 151 (22.57); 116 (100)
4l	1715	1642, 1602	461 (M^+, 6.27); 341 (19.17); 165 (39.51); 146 (100); 107 (21.70)
4m	1726	1641	465 (M^+, 7.43); 345 (23.46); 165 (55.39); 150 (100)
4n	1719	1642	476 (M^+, 58.46); 354 (80.98); 164 (56.76); 160 (100); 151 (31.67); 107 (22.18)
4o	1721	1606	475 (M^+, 3.62); 353 (4.01); 165 (13.62); 160 (100); 151 (11.38); 111 (24.54)
4p	1723	1643	411 (M^+, 5.59); 200 (29.23); 159 (28.07); 116 (77.43); 107 (14.63); 91 (100)
4q	1727	1642, 1603	441 (M^+, 3.82); 321 (18.27); 159 (29.86); 107 (15.33); 91 (100)
4r	1729	1645	445 (M^+, 23.26); 324 (46.15); 150 (52.74); 107 (16.27); 91 (83.36)
4s	1725	1649, 1613	455 (M^+, 3.17); 334 (14.48); 160 (52.63); 107 (17.13); 91 (100)
4t	1718	1637	Undetected

Table 4 Fungicidal activity of compounds **4** (50 ppm, relative inhibition rate, %)

No.	*Gibberella zeae*	*Cercospora beticola*	*Fusarium oxysporum*	*Pellicularia sasakii*	*Physalospora piricola*
4a	60.0	75.0	44.4	2.9	88.9
4b	53.3	40.0	66.7	14.3	83.3
4c	0	45.0	0	20.0	88.9
4d	66.7	60.0	55.6	14.3	38.9
4e	20.0	70.0	0	28.6	44.4
4f	33.3	45.0	0	0	55.6
4g	46.7	25.0	0	0	55.6
4h	53.3	25.0	0	0	72.2
4i	80.0	50.0	44.4	42.9	61.1
4j	86.7	50.0	83.3	48.6	88.9
4k	33.3	45.0	44.4	28.6	83.3
4l	33.3	70.0	72.0	57.1	94.4
4m	46.7	50.0	38.9	54.3	72.2
4n	66.7	50.0	44.4	42.9	94.4
4o	6.0	65.0	66.7	71.4	100
4p	86.7	50.0	61.1	60.0	88.9
4q	46.7	75.0	55.6	85.7	72.2
4r	20.0	90.0	16.7	42.8	88.9
4s	66.7	75.0	61.1	37.1	83.3
4t	66.7	55.0	72.2	57.1	77.8

Table 5 Fungicidal activity against *Physalospora piricola* of compounds **4** (*in vivo*, %)

	Concentration (ppm)	**4l**	**4n**	**4o**	Carbendazim
The test compounds were added to the medium before infection by the fungus	50	43	45	48	Undetected
	100	67	62	68	42
	200	69	74	81	
The test compounds were added to the medium after infection by the fungus	50	65	68	72	
	100	72	73	78	47
	200	73	75	78	

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(E0305063 CHENG, B.)