

Review Article

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Advancement of Research on Neonicotinoid Insecticides

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Abstract

the authors summarized the research progress of neonicotinoid insecticides, including the important variety of this kind of pesticides and the resistant condition of pest to neonicotinoid pesticides.

Introduction

Neonicotinoid insecticides are a relatively new group of active ingredients with novel modes of action (Tomlin 2000). They act as antagonists by binding to postsynaptic nicotinic receptors in the insect's central nervous system. This leads to the accumulation of acetylcholine, resulting in the paralysis and death of insects (Iwasa 2004). Neonicotinoids are active against many sucking and biting insects including aphides and some Lepidoptera species. This class of insecticides is commonly used on rice, maize, sunflowers, rapeseed, potatoes, sugar bees, vegetables, and fruit crops (Biever et al. 2003). In 1978, Soloway (Soloway 1978) reported nitro methylene heterocyclic compound with insecticidal activity, such as the heartland of methyl nitrate heterocyclic compound, among them nithiazine is the highest active, these is the first generation of new neonicotinoid insecticides. Because 2-phosphite methyl nitrate in the light is not stable, nithiazine is not use in the service of agriculture. In the 1980 s, Bayer based on the guideline of the first generation of new neonicotinoid compounds structure, with the introduction of aromatic heterocyclic methyl groups to 2-nitroiminoimidazolidine derivatives to synthesize a series of chlorinated neonicotinoid compounds, this is the second generation of new neonicotinoid insecticides.

Imidacloprid was the first successful development of new neonicotinoid insecticides. It was the unique mechanism of action of pesticides, no interaction with conventional pesticide resistance, high efficiency, broad spectrum and root systemic good, contact and stomach action, low toxicity to animals and safe for the environment. It can effectively control with homoptera, coleoptera, diptera and lepidoptera pests, as well as to the pest which resistant to conventional pesticides. It can be used to deal with the stem leaf, also can be used to deal with the soil and seeds. As a result, neonicotinoid has attracted widespread attention. The commercialization are: imidacloprid, acetamiprid, thiamethoxam, clothianidin, thiacloprid, dinotefuran, nitenpyram, imidacloprid, Cycloxaprid (Iwasa 2004).

1. The structure characteristics and commercialization varieties of neonicotinoid

1.1 Structure characteristics

As is known to all, imidacloprid was the first development neonicotinoid insecticides, which has developed three generations. People called the nicotine containing chlorinated pyridyls the first generation of

neonicotinoid insecticides, such as imidacloprid, acetamiprid, *et al.* Containing thiazolyl was called second generation neonicotinoid insecticides, such as thiamethoxam and clothianidin; Containing

tetrahydrofuran was called third generation of neonicotinoid insecticides. Figure 1 is the structure characteristic of neonicotinoid insecticides.

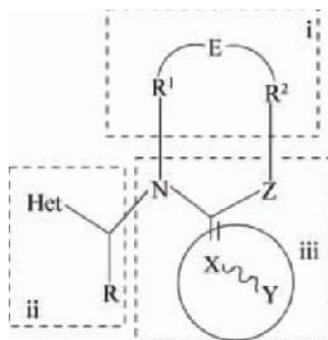


Figure 1 The structure characteristic of noenicotinoid

Figure 1 showed, neonicotinoid insecticides structure is mainly composed of four parts: the picture I is heterocyclic group (Het), such as pyridine ring, thiazole ring and furan ring; picture II is the chain for bridge, mainly such as methylene; picture III is the reactive group, that is - T - C (Z) = X - Y, mainly include the nitrylmethylene (C = C - NO₂), nitrylimino group (C = N - NO₂) and cyanoimino group (C = N - CN). Picture IV is the Nitrogen-Containing ring or open ring group. Ring group usually is constituted of five or six ring.

In China, there are three neonicotinoid insecticides in market. They are Paichongding, imidaclothiz, Cycloxaprid. Cycloxaprid is registered by Shanghai Shengnong Pesticide Co., Ltd. Considered the structure, it belongs to the first generation of neonicotinoid insecticides. The action target is nicotinic acetylcholine esterase receptors, but the role site is different, so it is effective to the resistant pest to imidacloprid.

2. The commercialization of neonicotinoid insecticides

(Zhang *et al.* 2012; Girolami *et al.* 2012; Wu *et al.* 2009; Pu *et al.* 2015; Fan *et al.* 2012; Zhou *et al.* 2006)

2.1 Imidacloprid

Imidacloprid is developed by Germany's Bayer in 1984. It is an international large products, market share in the head of neonicotinoid varieties (36.9%), 2010 sales is \$980 million. Imidacloprid is not only has excellent systemic, but also has high efficiency, wide insecticidal spectrum, long period of its effect, low toxicity to mammals. Due to its good systemic, often used as seed treatment and soil treatment agent.

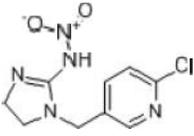
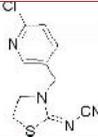
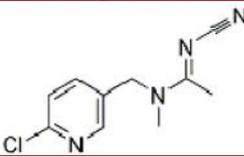
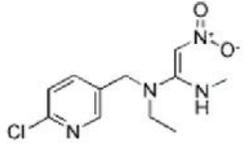
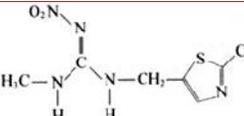
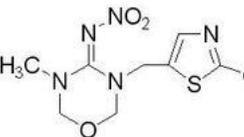
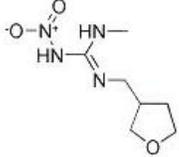
It has very good effect to Homoptera, coleoptera, diptera. Products are mainly used in sugar beet, grain, corn, sunflower, and cotton seed. Also can be used in animal to control sanitary pests. In United States, imidacloprid was used to control the pest of turf of playground and golf course.

In recent 10 years, as the alternative varieties of high toxic pesticides, the demand of Imidacloprid is growing gradually. In china it is mainly used to control rice planthopper. However, due to using a single pesticide for a long time in rice, imidacloprid produced a strong resistance. So in 2007, the ministry of agriculture issued to suspend the use of imidacloprid for prevention and control of rice plant hopper. The problem of pest resistant to imidacloprid has caused widespread concern. In order to prolong the service life of imidacloprid, delay the produce of pest resistance to it's, many scholars at home and abroad have studied the resistance development and mechanism of imidacloprid deeply, and raised the reasonable and effective control measures.

2.2 Thiamethoxam

Thiamethoxam is oxazinesyngenta listed products in 1997, is the world's second largest after imidacloprid pesticide, belong to the second generation of neonicotinoid insecticides. Not only has the tag, stomach poison, inside absorb activity, but also has higher activity, better security, broader insecticidal spectrum and speed, the effective length, etc. For with wings mesh, diptera and lepidoptera pests such as high activity, can effective prevention and control of various kinds of aphids, leafhoppers, planthoppers, mealworms, potato beetle, jump, nematodes, leaf moth and other pests and pests becoming resistant to multiple types of chemical pesticide.

Table 1 The structure and company of neonicotinoid

Name	Structure	Development company
imidacloprid		Bayer
Thiacloprid		Bayer
Acetamiprid		Nippon Soda
Nitenpyram		Sumitomo Chemical
Clothianidin		Sumitomo Chemical, Bayer
thiamethoxam		syngenta
Dinotefuran		Mitsui chemicals

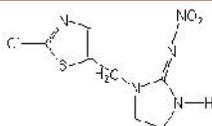
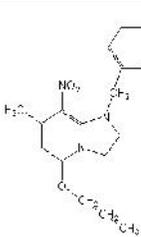
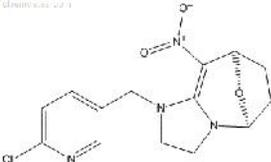
2.3 Clothianidin

In 2002, Takeda, Japan (now Sumitomo) developed clothianidin. It is mainly used for rice and lawn. Then Bayer also developed this product, code TI-345. Clothianidin can be used as seed treatment or leaf treatment agent. Bayer registered as seed treatment in 2005 in the United States, trade name was Poncho. In Europe, the mixture of clothianidin and Beta-Cypermethrin were used as seed treatment to control the pest of sugarbeet. In 2010, the sales up to 380 million, became the third largest neonicotinoid insecticides. Considered the harm to bees, the United States environment protection agency working with regulators in California and Canada registered clothianidin again on December 2011.

2.4 Acetamiprid

Acetamiprid was developed by Nippon Soda Co., Ltd. In 1996. The trade name is Mospilan. It was always used to prevent fruits, vegetables, citrus and apple from a variety of insects, including aphids, thrips, diamond back moth and leaf mites. It can be used as soil treatment, seed treatment and leaf spraying. From 1998 to the end of 2011, there are 42 companies to register the product at home and abroad, including 1 foreign company. Actually imidacloprid and acetamiprid use a set of production device, only one product can be produce. Due to the attitude against high toxic pesticides in Brazilian became more and more strongly, dichlorvos and other high toxic pesticides in foreign market is shrinking. So imidacloprid and acetamiprid became the best alternative products. With the increased demand in export, the prices have increased.

Table 2 The neonicotinoids of China

Name	Structure	Development company
Imidaclothiz		Jiangsunantong
Paichongding		Jiangsu Kwin co., LTD
Cycloxaprid		East China University of Science and Technology and Shanghai Shengnong

2.5 Thiacloprid

This product is developed by Bayer Company in 1997. It has a good, rapid and long-term activity against sucking and biting pests, especially against the aphids in apple. Because of systemic, it can be used as seed treatment and leaf spraying.

2.6 Dinotefuran

Dinotefuran is developed by Mitsui Chemicals, Inc. Its chemical structure is similar with the existing neonicotinoids. Tetrahydrofuran replaced the chlorinated pyridine, and chlorinated thiazolyl. There did not contain halogen elements in its characteristic. At the same time, it is different from the nicotine performance, so people called it "furan nicotine". Dinotefuranis broad spectrum, and is safety to human, animal and environment. It also has different application methods. This pesticide is expected to become a large-scale pesticide in the world.

2.7 Nitenpyram

Nitenpyram is developed by Takeda, Japan, trade name is Bestguard. It has a good, rapid, long-term activity against aphids, leafhopper, whitefly. It was low toxicity to animal. Mixed with such asorganophosphate, carbamate, nereistoxin toxins it could produce synergy and was effective to mites. Nitenpyramcan be used as leaf treatment and soil treatment.

2.8 Paichongding

Paichongding was developed by the East China University of Science and Technology and Jiangsu Kwin co., LTD together. It has the characteristic of low toxicity to mammals, non-toxic to bees and low toxicity to the fish, and has better insecticidal activity to brown plant hopper which has produce resistance to imidacloprid. It was patented in 2004, is mainly used for controlling the Homoptera insect pest. It can control rice plant hopper effectively and the control effect is 90% above; it can also control the vegetable aphids and the control effect of 94% above, which is better than imidacloprid. It can be widely used in fruit trees, wheat, beans, vegetables, rice and corn and other crops to control pest. Currently registered technical material content is 95% and the preparation formulations of 10% paichongding suspending agent has been sold in market.

2.9 Imidaclothiz

Imidaclothiz is developed by Nantong Jiangshan Agrochemical and Chemicals co., LTD. It is a kind of neonicotinoid insecticides, with low toxicity, systemic strong, high activity, low cost. The preparation formulation is not restricted by temperature, no cross resistance. It can be used for a variety of crops to control leafhoppers, plant hoppers and thrips effectively, especially to control rice two moths and three moths. Currently registered technical material content is 95%.The preparation formulations is 10% wetttable powder and 40% water dispersible granule, which has been sold in market.

2.10 Cycloxaprid

Cycloxaprid is the achievement of the project of “based on the target of new synthetic chemical pesticides design, optimization and products created” taken the lead by East China University of Science and Technology and Shanghai Shengnong Pesticide Co., Ltd supported by 863 plants. It is created by our country itself. It was wide insecticidal spectrum, high toxicity, no cross-resistance, no phytotoxicity to crops, low toxicity, low residue, and the activity to pests is better than that of imidacloprid. It was used in rice, vegetable, fruit tree, corn, cotton and wheat. The field test has been conducted in 14 provinces. Especially the control efficiency to plant hopper and rice leaf roller was tested. In 2014, the process has been optimized and made a great progress, cost reduced from the original 800 thousand yuan per ton to 300 yuan per ton. Researchers developed 25% Cycloxaprid wettable powder and 50% Cycloxaprid water dispersible granule. 25% Cycloxaprid oil suspending agent is under development.

3. Resistance station of neonicotinoid insecticides to pest

3.1 Resistance station of neonicotinoid insecticides to aphid

In Zimbabwe, the United States, southern Europe, northern Europe and other places, the highest resistance level of imidacloprid to peach aphid is 18 times (Cox *et al.* 2001; Foster *et al.* 2003), at the same time found that they have cross resistance between nitenpyram, acetamiprid, nicotine and imidacloprid (Foster *et al.* 2003). Also found that the resistance of peach aphid which body was red due to eating tobacco to new neonicotinoid insecticides is low (Nauen *et al.* 1998). In 1997-2000, Schaub L (Schaub *et al.* 2001) found the effectiveness of imidacloprid to *Dysaphis plantaginea* become low, suggested the possibility of resistance development. In 2000, Wang Kai-yun (Wang *et al.* 2000) measured the resistance level of imidacloprid to cotton aphid at 4 areas using drop method, found that cotton aphid has already developed different degree resistance to imidacloprid, among them the resistance level of Liaocheng population is 9.6 times. Pan Wen-liang (Pan *et al.* 2002) monitored that the resistance of apple meadow sweet aphid to imidacloprid in Hebei region is 2 to 4 times. Guo Tian-feng (Guo *et al.* 2012) evaluate the resistance of imidacloprid and acetamiprid to Xinjiang cotton aphid population using immersion method, the results showed that the sensitivity of cotton aphid to

imidacloprid is higher than to acetamiprid, cotton aphid has produced low levels of resistance to acetamiprid in southern and northern Xinjiang.

3.2 Resistance station of neonicotinoid insecticides to rice planthopper

Neonicotinoid insecticides were used to control rice planthopper in Japan and Southeast Asia. In the early 1990s, Japan found that rice planthopper to imidacloprid are sensitive in 9 districts. After breeding with malathion and propoxur inside, one of the populations which was not in contact with imidacloprid was found the resistance level to imidacloprid was 18 times (Sone *et al.* 1995). Liu (Liu *et al.* 2003; Zhuang *et al.* 2000) revealed that, after 25 generations of breeding with imidacloprid, the *Nilaparvata lugens* collected from the field produced more than 70 times resistance, after 35 generations the resistance increased to 250 times. In 1996-1999, Zhuang Yong-lin (Zhuang *et al.* 2000) detected the sensitivity change of imidacloprid to Brown planthopper (*Nilaparvata lugens* (Stål)) in Nanning, Guilin (Guangxi province), Anqing (Anhui province), Jiangpu, Yizheng and Nantong (Jiangsu province). The results indicated that the resistance level of imidacloprid to field populations was 1.2-6.3 times. It revealed that the resistance level of imidacloprid to Brown planthopper (*Nilaparvata lugens* (Stål)) collected from Nanjiang (Jiangsu province), changed (Hunan province), Nanning, Guilin (Guangxi province) were 475, 179, 178, and 70 times respectively, but these populations were no obvious cross resistance to thiamethoxam. Shen also measured the resistance level of imidacloprid and thiamethoxam to rice planthopper collected from Wuxi, Jianhu and Baimahu farm (Jiangsu province) and Huzhou (Zhejiang Province). Jian Hu, White Horse Lake farm rice planthopper of oxazine resistance. They found the LC_{50} of imidacloprid to population of Huzhou (Zhejiang province) and Jianhu (Jiangsu province) was 20-15 times than the population of Baimahu farm (Jiangsu province), the population of Wuxi (Jiangsu province) was only 1.2 times than the population of Baimahu farm (Jiangsu province). And all the populations were no resistance to thiamethoxam. Liu Ze-wen (Liu *et al.* 2002) measured the resistance of imidacloprid to Brown planthopper (*Nilaparvata lugens* (Stål)) and white back planthopper collected from Anqing, the resistance level were 6-9 times and belong to medium resistance level. In 2011-2013, Wang Peng (Wang *et al.* 2013) monitored the resistance level of 19 populations from China's main rice region to commonly used pesticides. The results revealed that the resistance level of the

19 monitored population were 82.3-1935.8 times and belong to high and extreme high level. Compared with the results of 2006-2009, the resistance level of imidacloprid had a tendency to rise again. The resistance level of thiamethoxam to Brown planthopper (*Nilaparvata lugens* (Stal)) was low or medium (6.1-14.4) in 2010, but to 2011, except Guilin (Guangxi province) the resistance level of other 8 population was medium or high, there were obvious increase than the last year.

3.3 Resistance station of neonicotinoid insecticides to potato beetle

Due to the resistance problem, the United States used imidacloprid to prevent and control potato beetle since 1995. Through monitoring the larvae of potato beetle in North America and Europe in 2001, the results showed that there was about 30 times difference among different populations (Olsen *et al.* 2000). Zhao (Zhao *et al.* 2000) revealed that the resistance level of imidacloprid to potato beetle adult and larvae were 100 and 13 times respectively after using imidacloprid for a few years in Long Island. Hollingworth (Hollingworth *et al.* 2002) measured the resistance level of imidacloprid to potato beetle adults was 150 times using topical application, but the resistance level of thiamethoxam that was not used ever was far lower than that of imidacloprid, the results also showed that the potato beetle adults of Germany, Austria and Poland to imidacloprid was still sensitive. Liuping (Liu *et al.* 2011) monitored the changes of sensitivity of 9 field potato beetle population (Xinjiang Uyghur Autonomous Region) to imidacloprid, acetamiprid, thiamethoxam, thiacloprid using drop method in 2009 and 2010. They revealed that the resistance to imidacloprid and thiamethoxam decreased year by year. Three populations of six monitored in 2009 were low resistance to acetamiprid and thiamethoxam (5.0-10.0 times). All of the six monitored population in 2000 has produced resistance to thiamethoxam, three of them were middle resistance (10.1-40.0) and three were low resistance.

3.4 Resistance station of neonicotinoid insecticides to western flower thrips

Western flower thrips spread to China in 2003. Due to its short development period, large production and parthenogenesis, it often outbreak on production. Neonicotinoid pesticides was one of the effective prevention and control of pesticides in China. After breeding 32 generations of western flower thrips sensitivity population, the resistance time were 13.8

times. Breeding 12 generations gradually after stopping to use drugs screening, the resistance of western flower thrips to imidacloprid dropped, but it could not restored to the sensitivity level. It suggested that there were higher risk to prevent and control western flower thrips with imidacloprid (Wang *et al.* 2012).

3.5 Resistance station of neonicotinoid insecticides to other pests

In Arizona of United States, lygus *Hesperus* in cotton were 100 times resistance to imidacloprid (Dennehy *et al.* 1996); *Frankliniella occidentalis* and *Heliothis virescens* which has produce resistance to other pesticides were also resistance to imidacloprid (Zhao *et al.* 1995; Pedra *et al.* 2004). Treated with acetamiprid, the resistance of *Plutella xylostella* collected from the field were 110 times after breeding five generations. It were proved that the resistance is not stable, stopping breeding with pesticide for seven generations the resistance decreased from 110 times to 243 times (Ninsin *et al.* 2005). In addition, it was reported that housefly, german cockroach (Wen *et al.* 1997) and *Drosophila melanogaster* (Dabom *et al.* 2001) have produced a resistance to imidacloprid.

4. Conclusion and Prospect

Table 2 revealed that, there are 10 kinds of neonicotinoid commodity that sold in the market. Along with the use of neonicotinoid pesticides, the resistant problem appeared continuously. Among them the resistance of aphids and rice planthopper was particularly prominent. In order to effectively prevent and delay the resistance development of some new insecticide, to establish the sensitivity baseline of resistant insecticide and the dose of early resistance diagnosis is necessary. And to detect and monitor the resistance is also important. We should require the applicant to provide scientific and not easy to produce resistance method when they registered. Before using widely in the field, resistance breeding should be done the laboratory, in order to provide the related information of resistance development, and determine the resistance stability and provide a basis for reasonable rotation. Resistant spectrum should be determined to provide information of other insecticide resistance for reasonable rotation.

Resistance genetics is to research the rule of resistance development and is the base of resistance management. To study the resistant inheritance mode of insects to imidacloprid is very important to resistant

monitoring, risk assessment and resistance management. Neonicotinoid insecticides have broad application prospect, so studying resistant inheritance mode of target pests is significant to formulate reasonable and effective resistance management strategy and extend the service life. It is also have important reference value to other insects resistant research.

Acknowledgments

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Conflict of interest

The authors declare that they have no conflict of interest.

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