

专论与综述

微生物降解二甲酰亚胺类杀菌剂研究进展

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摘要: 二甲酰亚胺类杀菌剂具有高效、广谱等特性, 对多种果蔬的真菌性病害具有较好的防治效果, 但该类杀菌剂半衰期较长且具有内分泌干扰活性, 其残留给生态系统和食品安全带来严重危害。利用微生物及其酶降解农产品和环境中的农药残留是有效方法之一。本文概述了二甲酰亚胺类杀菌剂的基本性质和危害, 以及农产品中的残留现状, 综述了微生物降解二甲酰亚胺类杀菌剂的研究进展, 重点阐述了降解微生物种类及其降解途径。最后, 对利用酶及基因工程技术修复二甲酰亚胺类杀菌剂研究方向进行了展望, 以期对二甲酰亚胺类杀菌剂的污染防控提供研究思路。

关键词: 杀菌剂; 残留; 生物降解; 降解途径; 水解酶

Research progress in microbial degradation of dicarboximide fungicides

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Abstract: Dicarboximide fungicides with high efficacy and a broad antifungal spectrum have strong control effects on a variety of fungal diseases attacking fruits and vegetables. However, they have long half-lives and the effects of endocrine disruptors, with the residues bringing serious harm to ecosystems and food safety. Degrading fungicide residues in agricultural products and the environment by microorganisms and their enzymes is an effective method. This paper summarizes the basic properties, hazards, and residue status in agricultural products of

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dicarboximide fungicides. Furthermore, this paper reviews the research progress in microbial degradation of dicarboximide fungicides, with focuses on the microbial species and pathways of degradation. Finally, this paper prospects the research directions of the application of enzyme and genetic engineering to remove dicarboximide fungicides from food products, aiming to provide research ideas for the pollution prevention and control of dicarboximide fungicides.

Keywords: fungicide; residue; biodegradation; degradation pathway; hydrolase

杀菌剂是指在一定剂量或浓度下,能够杀死植物病原菌或抑制其生长萌发的农药,广泛应用于栽培植物以控制真菌感染,按作用方式和机制可分为保护性杀菌剂和治疗性杀菌剂^[1]。二甲酰亚胺类杀菌剂(dicarboximide fungicides, DF)是一类保护性杀菌剂,具有高效、广谱、快速、低残留等特点,主要品种有腐霉利(procymidone)、异菌脲(iprodione)、菌核净(dimetachlone)和乙烯菌核利(vinclozalin)等,其结构式中均含有 3,5-二氯苯基。

由于广泛和不合理的使用,DF 在农产品、土壤中被频繁检出。DF 残留及其转化产物可以通过空气、灰尘、水及食物等介质经皮肤、消化道、呼吸道等进入人体^[2],还会通过食物链发生生物富集,严重危害人的身体健康。例如,通过皮肤接触或吸入直接暴露于高浓度腐霉利会引发恶心、呕吐^[3],而长期食用含有腐霉利残留的果蔬后,会引起肝脏损伤^[4]、糖脂代谢紊乱^[5]及解毒功能障碍^[6]。因此,如何消除环境和食品中的 DF 已成为亟需解决的问题。利用微生物及其酶降解农产品和环境中的农药残留是有效方法之一。本文概述了二甲酰亚胺类杀菌剂的基本性质和危害,以及农产品中的残留现状,综述了微生物降解二甲酰亚胺类杀菌剂的研究进展,重点阐述了降解微生物种类及其降解途径。最后,对利用酶及基因工程技术修复二甲酰亚胺类杀菌剂的研究方向进行了展望,以期对二甲酰亚胺类杀菌剂的污染防控提供研究思路。

1 概述

1.1 二甲酰亚胺类杀菌剂简介

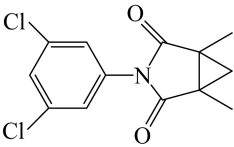
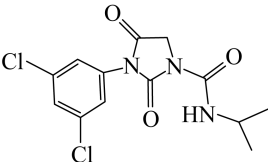
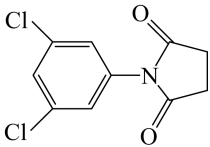
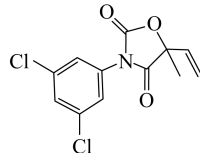
DF 具有很高的专化性,对灰葡萄孢(*Botrytis cinerea*)^[7]、核盘菌属(*Sclerotinia*)^[8]和链格孢属(*Alternaria*)^[9]等真菌引起的病害具有显著的防治效果,其中,对灰葡萄孢效果尤其好。DF 通过作用于真菌的双组分组氨酸激酶信号途径^[10],引起灰葡萄孢的甘油积累、膜质过氧化和质膜渗透,进而抑制灰葡萄孢的分生孢子萌发、菌丝生长和毒素产生^[11-12],被广泛应用于菌核病、灰霉病^[13-14]及黑斑病^[15-17]的防治。部分 DF 的理化性质如表 1 所示。其中,腐霉利和乙烯菌核利具有高疏水性,能够与土壤颗粒和有机物紧密结合,具有较长的半衰期和较低的土壤吸收率^[21];而异菌脲疏水性相对较低,在土壤环境中具有较强的移动性,易通过径流和浸出进入地下水和地表水中^[22]。DF 在土壤和水基质中的残留,不仅对非靶标生物造成毒性,也可通过食物链富集危害人体健康。

1.2 DF 毒性

许多证据表明,DF 对非靶标生物和人类具有潜在的肝毒性^[23]、肾毒性^[24]、心脏毒性^[25-26]和内分泌干扰作用^[27],且具有细胞毒性^[28-29]、基因毒性^[30]、致突变性^[28]和发育毒性^[31-33]。此外,腐霉利毒性易向胎儿转移^[34],导致男性胎儿生殖器畸形发育、肛门生殖器距离显著降低、乳头滞留增加^[35]、睾丸损伤^[36],以及女性胎儿卵巢^[37]和子宫发育不良^[38-39]等严重后果。长期暴露于菌核净会引起呼吸系统的严重病变^[40]及与内皮

表 1 部分二甲酰亚胺类杀菌剂的理化性质

Table 1 Some of the physical-chemical properties of dicarboximide fungicides

Item	腐霉利 Procymidone	异菌脲 Iprodione	菌核净 Dimetachlone	乙烯菌核利 Vinclozalin
CAS 号	32809-16-8	36734-19-7	24096-53-5	50471-44-8
CAS Number				
分子式	$C_{13}H_{11}Cl_2NO_2$	$C_{13}H_{13}Cl_2N_3O_3$	$C_{10}H_7Cl_2NO_2$	$C_{12}H_9Cl_2NO_3$
Molecular formula				
结构式				
Structural formula				
分子量	284.14	330.17	244.08	286.11
Molecular weight				
土壤中半衰期	14.3–24.1 ^[18]	7.8–12.2 ^[19]	8.2–14.6 ^[20]	9–16 ^[21]
Half-life in soil (d)				
水中溶解度	2.46	6.8	1630	3.4
Water solubility				
(20 °C, mg/L)				
地下水普遍性评分	3.3	0.43	—	1.98
Groundwater ubiquity score				
(GUS)				
正辛醇/水分配系数	3.3	2.99	1.4	3.02
Octanol-water partition				
coefficient				
吸附系数	378	700	—	300
Sorption potential				

资料来自农药特性数据库；—：无相关数据；下同。

Source: Pesticide Properties Database; —: No relevant data; The same below.

功能障碍相关的心血管疾病的发作^[41]。异菌脲已被列为可能的致癌物，可抑制人体干细胞的生长、影响男性精子质量和造成 DNA 的损伤，还会导致人体内分泌紊乱、神经系统功能失调、降低体内荷尔蒙分泌^[42]等。

1.3 DF 在农产品中的残留

DF 通过大气沉降、污水排放、雨水淋溶等途径进入环境^[43]，在农产品、土壤^[44]和水体^[45-46]等不同环境基质中被频繁检出，甚至在动物^[47]和人类尿液^[48]中也能检测到 DF，如异菌脲。在对市场中农产品进行农药残留抽检时，腐霉利的检出率最高(表 2)，其中，在韭菜上的最大残留量高达 11.270 mg/kg，远远超过了国家规定的

残留限量标准(5 mg/kg)。除此之外，在葡萄汁^[61]、中药^[62]、食用菌^[63-64]中也检测到腐霉利的残留。

2 可降解 DF 的微生物

生物、化学和光化学降解是在自然环境中减除 DF 的主要方式，而微生物降解在有害、有机外源性物质的消减中发挥着最积极的作用，作为一种生态友好、经济有效和反应温和的消除方法被优先使用^[65]。在可降解 DF 的微生物中(表 3)，目前有关异菌脲降解的报道研究较多，以细菌为主，包括节杆菌属(*Arthrobacter*)^[66]、微杆菌属(*Microbacterium*)^[69]、芽孢杆菌属(*Bacillus*)^[72]、无色杆菌属(*Achromobacter*)^[73]、

表 2 我国部分农产品中腐霉利的残留状况

Table 2 Residues of procymidone in some agricultural products in China

样品类型 Sample type	采样地点 Sampling site	检出率 Detection rate (%)	最大残留量 Maximum residual amount (mg/kg)	样品数量 Number of samples	残留限量标准 Residue limit standard (mg/kg)	参考文献 Reference
韭菜 Leek	陕西省 Shaanxi province	23.0	0.350	13	5	[49]
	山东省泰安市 Tai'an city, Shandong province	20.4	11.270	64		[50]
	湖南省长沙市 Changsha city, Hunan province	11.0	3.030	690		[51]
	河南省各市 Cities in Henan province	32.7	3.853	452		[52]
	芹菜 Celery	25.0	5.391	8	5	[53]
草莓 Strawberry	河北省各市 Cities in Hebei province	29.9	—	144		[54]
	重庆市 Chongqing	88.3	1.090	77	10	[55]
	浙江省杭州市 Hangzhou city, Zhejiang province	23.1	0.458	459		[56]
西红柿 Tomato	北京市昌平区 Changping district, Beijing	52.5	—	40	5	[57]
茄子 Eggplant	浙江省金华市 Jinhua city, Zhejiang province	30.9	0.875	97	5	[58]
黄瓜 Cucumber	浙江省各市 Cities in Zhejiang province	10.3	0.060	68	5	[52]
辣椒 Chili pepper	河南省各市 Cities in Henan province	8.25	0.625	303	5	[59]
葡萄 Grape	贵州省 Guizhou province	63.3	—	30	5	[60]
	广东省佛山市禅城区 Chancheng district, Foshan city, Guangdong province					

陕西省包括汉中、安康、商洛、西安、宝鸡、咸阳、渭南、铜川、延安、榆林 10 个市；重庆市包括忠县、万州、梁平、开州、巫山、巫溪、奉节、梁平、云阳 9 个区县；贵州省包括贵阳市、遵义市、毕节市、黔西南州、黔东南州、铜仁市和安顺市。

Shaanxi province includes 10 cities: Hanzhong, Ankang, Shangluo, Xi'an, Baoji, Xianyang, Weinan, Tongchuan, Yan'an and Yulin; Chongqing includes nine districts and counties of Zhongxian, Wanzhou, Liangping, Kaizhou, Wushan, Wuxi, Fengjie, Liangping and Yunyang; Guizhou province includes Guiyang city, Zunyi city, Bijie city, Qianxinan prefecture, Qiannan prefecture, Qiandongnan prefecture, Tongren city and Anshun city.

类节杆菌属(*Paenarthrobacter*)^[74]、苍白杆菌属(*Ochrobactrum*)^[76]、假节杆菌属(*Pseudarthrobacter*)^[77]及固氮螺菌属(*Azospirillum*)^[78]等。其次是降解腐霉利和菌核净的微生物，也多为细菌。如 Zhang 等^[85-86]从受污染土壤中筛选鉴定出斯氏普罗威登斯菌(*Providencia stuartii*) JD 和内藏山

短波单胞菌(*Brevundimonas naejangsanensis*) J3, 研究发现, 这 2 种细菌对大部分 DF 均具有较强的降解能力, 菌株 JD 和 J3 在最适条件下培养 7 d 后, 可使 50 mg/L 菌核净、异菌脲和腐霉利的去除率分别达到 82.91%和 80.83%、79.77%和 81.32%、80.78%和 80.76%。Zhang 等^[84]分

表 3 部分降解二甲酰亚胺类杀菌剂的微生物

Table 3 Microorganisms that partially degrade dicarboximide fungicides

底物 Substrate	微生物 种类 Microbial species	菌株 Strain	培养时间 Incubation time (d)	底物浓度 Substrate concentration	降解率 Degradation rate (%)	降解方式 Degradation manner	菌株来源 Source of strain	参考文献 Reference
异菌脲 Iprodione	细菌 Bacteria	节杆菌属 <i>Arthrobacter</i>						
		<i>Arthrobacter</i> sp. CQH	4.67	100 mg/L	100	矿化 Mineralization	土壤 Soil	[66]
		<i>Arthrobacter</i> sp. MA6	7	9.9 mg/L	86.7	矿化 Mineralization	土壤 Soil	[67]
		<i>Arthrobacter</i> sp. C1	10	60 mmol/L	100	共代谢 Co-metabolism	土壤 Soil	[68]
		微杆菌属 <i>Microbacterium</i>						
		<i>Microbacterium</i> sp. Y-20	0.5	100 mg/L	100	矿化 Mineralization	土壤 Soil	[69]
		<i>Microbacterium</i> sp. Y-29						
		<i>Microbacterium</i> sp. Y-32						
		<i>Microbacterium</i> sp. CQH-1	4	100 mg/L	100	矿化 Mineralization	土壤 Soil	[70]
		<i>Microbacterium</i> sp. YJN-G	1	100 mg/L	100	矿化 Mineralization	活性污泥 Activated sludge	[71]
		芽孢杆菌属 <i>Bacillus</i>						
		<i>Bacillus</i> sp. KMS-1	7	25 mg/L	41.4	矿化 Mineralization	土壤 Soil	[72]
		无色杆菌属 <i>Achromobacter</i>						
		<i>Achromobacter</i> sp. C2	10	60 mmol/L	100	共代谢 Co-metabolism	土壤 Soil	[73]
		类节杆菌属 <i>Paenarthrobacter</i>						
		<i>Paenarthrobacter</i> sp. YJN-5	3.3	1.5 mmol/L	95	矿化 Mineralization	土壤 Soil	[74]
		<i>P. nicotinovorans</i> Y-19	0.583	100 mg/L	100	矿化 Mineralization	土壤 Soil	[75]
		苍白杆菌属 <i>Ochrobactrum</i>						
		<i>Ochrobactrum</i> sp. YJN1-1	—	—	—	矿化 Mineralization	土壤 Soil	[76]
		假节杆菌属 <i>Pseudarthrobacter</i>						

(待续)

(续表 3)

底物 Substrate	微生物 种类 Microbial species	菌株 Strain	培养时间 Incubation time (d)	底物浓度 Substrate concentration	降解率 Degradation rate (%)	降解方式 Degradation manner	菌株来源 Source of strain	参考文献 Reference
		<i>Pseudarthrobacter</i> sp. Y-5 固氮螺菌属 <i>Azospirillum</i> <i>Azospirillum</i> sp. A1-3 普罗威登斯属 <i>Providencia</i> <i>P. stuartii</i> JD	3	300 mg/L	96.71	矿化 Mineralization	废水 Wastewater	[77]
		短波单胞菌属 <i>Brevundimonas</i> <i>B. naejangsanensis</i> J3 假单胞菌属 <i>Pseudomonas</i> <i>P. fluorescens</i> <i>Pseudomonas</i> sp. <i>P. paucimobilis</i>	4.5	27.96 mg/L	50.80	矿化 Mineralization	土壤 Soil	[78]
		接合酵母属 <i>Zygosaccharomyces</i> <i>Z. rouxii</i> DBVPG 6399	7	50 mg/L	80.83	矿化 Mineralization	土壤 Soil	[79]
		真菌 Fungi	7	50 mg/L	80.78	矿化 Mineralization	土壤 Soil	[79]
		细菌 Bacteria	1	8.25 mg/L	100	矿化 Mineralization	土壤 Soil	[80]
		普罗威登斯属 <i>Providencia</i> <i>P. stuartii</i> JD	9	1 mg/L	100	—	—	[81]
		短波单胞菌属 <i>Brevundimonas</i> <i>B. naejangsanensis</i> J3 甲基杆菌属 <i>Methylobacterium</i> <i>Methylobacterium</i> sp. T32-2 <i>Methylobacterium</i> sp. T32-1	7	50 mg/L	80.76	矿化 Mineralization	土壤 Soil	[79]
		青霉菌属 <i>Penicillium</i> <i>Penicillium</i> sp. T31-4-5	20	300 mg/L	66.40	矿化 Mineralization	土壤 Soil	[82]
			14	300 mg/L	77.20	矿化 Mineralization	土壤 Soil	[83]
			20	300 mg/L	77.20	矿化 Mineralization	土壤 Soil	[82]

(待续)

(续表 3)

底物 Substrate	微生物 种类 Microbial species	菌株 Strain	培养时间 Incubation time (d)	底物浓度 Substrate concentration	降解率 Degradation rate (%)	降解方式 Degradation manner	菌株来源 Source of strain	参考文献 Reference
菌核净 Dimetachlone	细菌 Bacteria	普罗威登斯属 <i>Providencia</i>						
		<i>P. stuartii</i> JD	7	50 mg/L	82.91	矿化 Mineralization	土壤 Soil	[79]
		短波单胞菌属 <i>Brevundimonas</i>						
		<i>B. naejangsanensis</i> J3	7	50 mg/L	81.32	矿化 Mineralization	土壤 Soil	[79]
		类节杆菌属 <i>Paenarthrobacter</i>						
		<i>Paenarthrobacter</i> sp. JH-1	3	300 mg/L	98.53	矿化 Mineralization	活性污泥 Activated sludge	[84]

离出一株菌核净高效降解菌类节杆菌(*Paenarthrobacter*) sp. JH-1, 该菌株能利用高浓度的菌核净作为唯一碳源进行生长, 在 3 d 内对 300 mg/L 菌核净的降解率为 98.53%。近期, 有研究利用所在实验室保存的细菌对腐霉利进行了降解实验, 初步发现地衣芽孢杆菌(*B. licheniformis*) B-1^[87]及枯草芽孢杆菌(*B. subtilis*) JF^[88]对腐霉利具有较高的降解能力; 菌株 B-1 及 JF 在最适条件下培养 5 d 可使 50 mg/L 腐霉利的降解率分别达到 85.30%和 91.09%。这 2 株菌对 DF 的降解能力还有待进一步研究。

研究发现, 除细菌外, 自然界中也存在具有降解 DF 能力的真菌。如 Zadra 等^[81]分离到一株鲁氏接合酵母(*Zygosaccharomyces rouxii*) DBVPG 6399, 将该菌株置于含 1 mg/L 异菌脲的培养基中培养 9 d 后, 能使异菌脲完全降解。高悦^[82]从长期受农药污染的蔬菜大棚土壤中筛选到能降解腐霉利的青霉菌(*Penicillium* sp.) T31-4-5, 该菌在最适条件下, 经 20 d 的培养可使液体培养基中 300 mg/L 的腐霉利下降到 100 mg/L 以下, 降解率达到 77.2%。

研究表明, 由于复合菌系具有更加丰富多

样的降解酶系, 菌株协同作用已成为提高 DF 降解效率的有效途径^[21]。Zhang 等^[79]研究发现, 相较于将斯氏普罗威登斯菌 JD 和短波单胞菌 J3 单独培养, 将 2 株菌共培养能增强对 DF 的降解效率; 通过分别调整菌株 JD 和 J3 的菌体浓度为 6×10^8 CFU/mL, 以 1:1 共培养 5 d 后, 使浓度为 50 mg/L 的菌核净、异菌脲和腐霉利的降解率分别达到 85.56%、83.44%和 84.68%, 7 d 后培养液中几乎检测不到 DF 底物, 比菌株单独培养提前了 1 d 以上。此外, 金护定等^[83]将甲基杆菌(*Methylobacterium* sp.) T32-1、T32-2 及青霉菌 T31-4-5 共培养得到混合菌群 T₃, 发现在最适条件下培养 8 d 后, 菌群 T₃对 300 mg/L 腐霉利的降解率达 90%, 是菌株 T32-1 单独培养的 1.165 倍。

3 DF 的微生物降解机制

3.1 DF 的微生物降解途径

微生物对 DF 的降解分为酶促降解和非酶促降解 2 种。酶促反应是指微生物所含的胞内酶或分泌的胞外酶直接作用于农药使其降解, 包括氧化、还原、水解和酯化等。非酶促反应

是指微生物通过代谢改变农药外环境的离子浓度或物理化学性质,从而促使农药降解,包括矿化作用和共代谢^[89]。其中,共代谢中微生物不能以农药作为唯一的碳源和能源,需要从其他化合物中获得能源^[90]。而矿化作用中微生物直接将农药作为生长底物,分解利用并生成无机物、CO₂和H₂O等^[91]。

研究发现,微生物对DF的降解主要是酶促反应,包括酶分子对DF的水解、氧化等反应,主要过程是其结构中的酰胺键(-CO-N=,包括肽键-CO-NH-)发生水解,3,5-二氯苯胺(3,5-dichloroanilin, 3,5-DCA)是微生物代谢DF的主要产物^[75]。3,5-DCA具有很强的肾毒性及神经毒性,其毒性和持久性比母体更高^[92-93],如其对大鼠的LD₅₀值约为腐霉利的10倍。

目前,关于微生物降解DF的研究大多还停留在降解菌筛选及降解特性等方面,对降解机理鲜有涉足。Zhang等^[79]研究后提出,微生物降解腐霉利主要通过水解、氧化等一系列反应完成,如在斯氏普罗威登斯菌JD和短波单胞菌J3共培养条件下,腐霉利被连续水解,其结构中的酰胺键首先发生水解^[75]生成促生代谢物2-(3,5-二氯苯基氨基甲酸酯)-1,2-二甲基环丙烷羧酸,然后进一步被水解为3,5-DCA和可能的1,2-二甲基环丙烷二羧酸,接着3,5-DCA降解为苯酚,随后进一步氧化为己二烯二酸(图1A)。Zhang等^[84]根据研究结果的分析,提出了菌株JH-1中菌核净的代谢途径,即菌核净通过两步连续水解反应降解为3,5-DCA和琥珀酸(图1B)。现有研究表明,异菌脲的微生物降解途径主要有2种路径(图1C),途径I主要存在于细菌中,异菌脲被连续水解降解为3,5-DCA和氨基甲酸。而途径II只发现存在于鲁氏接合酵母DBVPG 6399中,且降解效果较差^[42]。

3.2 DF的降解酶

在自然条件下,微生物的应用易受环境条件的限制,相比之下,降解酶具有更高的活性

和减除效率。目前,在二甲酰胺亚胺类杀菌剂中,仅对异菌脲降解途径中有关的分子基础研究进行了报道。Campos等^[73]认为酰胺水解酶在异菌脲降解过程中起作用,酰胺酶能够水解各种酰胺化合物的C-N键并产生相应羧酸^[94],在包括杀虫剂在内的外源物质的代谢中具有重要作用^[95]。Donoso-Piñol等^[96]从处理农药的生物净化系统中筛选出无色杆菌(*Arthrobacter* sp.) C1,菌株C1中含有445个仅对异菌脲的出现作出过表达反应的蛋白,表明其可能在异菌脲降解过程中起一定作用。Yang等^[74]分离得到一株异菌脲降解菌类节杆菌(*Paenarthrobacter* sp.) YJN-5,并对其进行了鉴定;分析显示,在该菌基因组中存在一种长1 410 bp、编码469个氨基酸的酰胺水解酶基因*ipah*;该水解酶Ipah含有高度保守的催化三联体Ser-Ser-Lys,负责菌株YJN-5中异菌脲的初始降解步骤,水解异菌脲的N-1酰胺键生成N-(3,5-二氯苯基)-2,4-二氧代咪唑烷。Zhang等^[97]分离并表征了另一株类节杆菌YJN-D,该菌株具有与菌株YJN-5相同的异菌脲代谢途径,并在菌株YJN-D中鉴定出脱乙酰酶基因*ddaH*以及水解酶基因*duaH*,*ddaH*负责裂解N-(3,5-二氯苯基)-2,4-二氧代咪唑烷的乙内酰胺环,*duaH*负责裂解(3,5-二氯苯基脲)乙酸的尿素侧链产生3,5-DCA。这些异菌脲代谢基因分布在菌株YJN-5的3个质粒上,并且在菌株YJN-5和YJN-D之间高度保守,表明其可能是异菌脲降解的关键基因。此后,Zhang等^[77]从农药厂废水样品中分离得到一株异菌脲降解菌假节杆菌(*Pseudarthrobacter* sp.) Y-5,利用染色体整合技术成功实现了该菌异菌脲降解胞内酶*ipah*基因在枯草芽孢杆菌中的胞外表达,构建了基因工程菌WB800-*ipah*;进一步研究表明,菌株Y-5和WB800-*ipah*均能在9 d内消除土壤中90%以上的5 mg/kg的异菌脲;相较于菌株Y-5,菌株WB800-*ipah*具有更强的降解性,并且接种量远低于菌株Y-5,具有更好的成本效益。

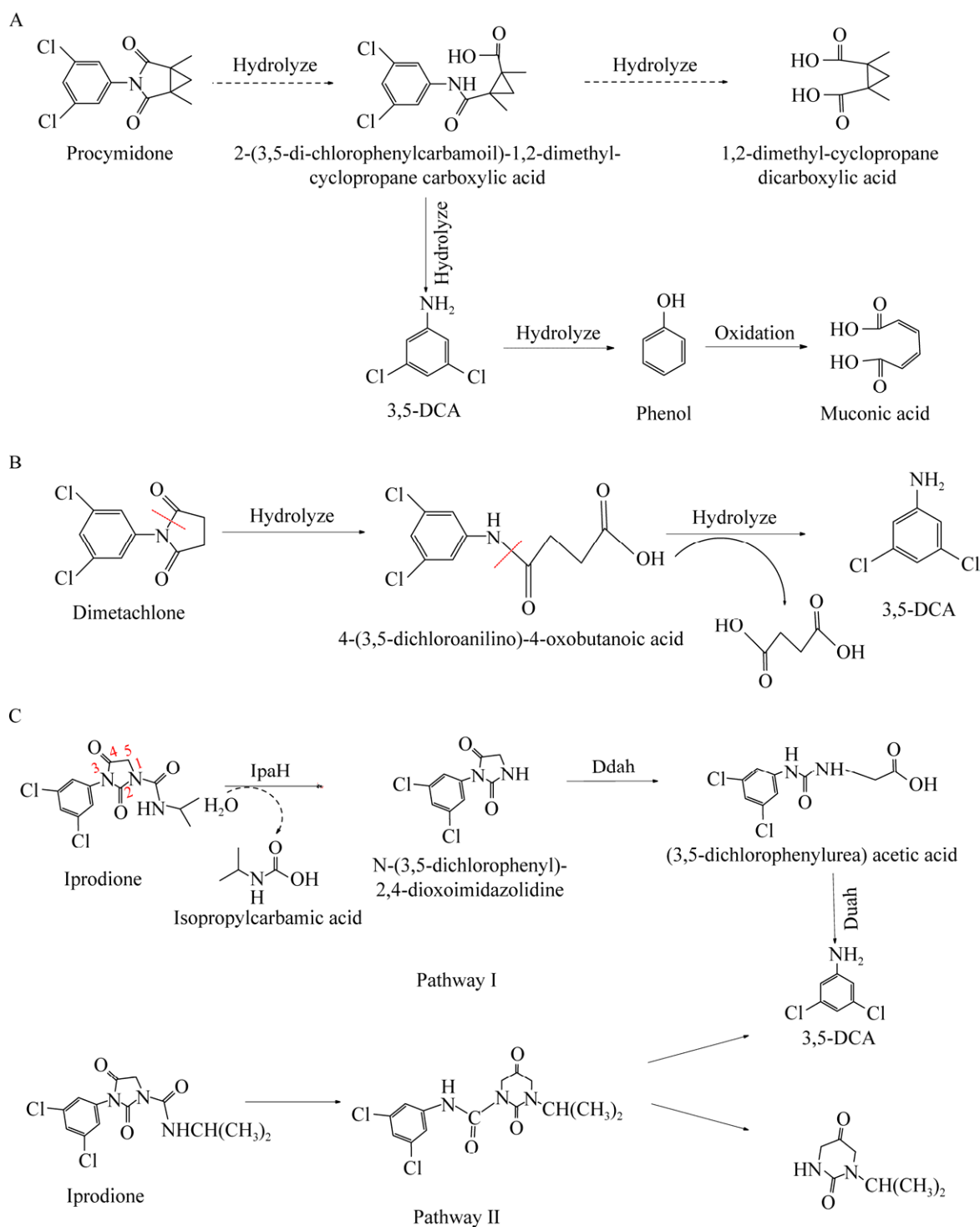


图 1 部分二甲酰亚胺类杀菌剂的微生物代谢途径 A: 腐霉利的微生物代谢途径; B: 菌核净的微生物代谢途径; C: 异菌脲的微生物代谢途径。

Figure 1 Microbial degradation pathways of some dicarboximide fungicides. A: Procymidone microbial metabolic pathway; B: Dimetachlone microbial metabolic pathway; C: Iprodione microbial metabolic pathway.

4 结语与展望

DF 的大量残留严重威胁到食品安全和人体健康,生物降解作为消除农药污染的有效手段之一,已在实际生产中得到应用。目前对 DF 降解菌的筛选及生物降解途径已有相关研究,筛选出了一些高效降解 DF 的菌株,并提出了部分 DF 的微生物降解途径,主要通过水解、氧化等一系列反应完成。但对 DF 降解的具体途径及酶的作用机理,特别是相关催化酶、催化机制、酶学特性等方面的深入研究鲜有涉及,仅在几株异菌脲降解菌中鉴定出了相关降解酶基因。此外,目前关于微生物修复食品中 DF 的研究尚为鲜见,降解 DF 的微生物大多来自被农药污染的土壤,菌株安全性难以保障,亟须筛选食品来源的高效降解菌。

针对微生物降解 DF 的具体分子机制尚不清楚等问题,未来还需要:(1) 筛选能够高效降解 DF 的优异菌株,尤其是食品来源的菌株;(2) 开展高效降解菌株代谢途径的分析、降解基因的筛选及降解酶特性分析等工作;(3) 借助基因组学、蛋白质组学、转录组学等方法,挖掘降解过程中涉及的酶及其编码基因,从而阐明降解机制;(4) 结合合成生物学和酶的定向选育等先进技术,构建功能强大的基因工程菌并获得重组酶,以期更有效地利用微生物修复农产品中残留的 DF。

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