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Edited by D. PETERMAN

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应用微生物学进展 第21卷

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Production of Polyene Macrolide Antibiotics

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I. Introduction

A. HISTORY OF THE DISCOVERY OF NEW POLYENE MACROLIDE ANTIBIOTICS

Since the early 1950s, when the first polyene macrolide antibiotics were reported, more than 90 different members of this group have been described, and more are being discovered each year. Sixteen polyene macrolides were known in 1957 (Dutcher, 1957), 41 in 1960 (Vining, 1960), 57 in 1963 (Oroshnik and Mebane, 1963), and 84 in 1973 (Hamilton-Miller, 1973). Several more, which have been reported recently, are included in Table I. However, one must be cautious in considering many of these antibiotics to be new, since the question of the identity of many remains open. Most polyene macrolides have not been obtained in pure form, and the chemical compositions of most of them are unknown. Thus, tennecitin and pimaricin have been found to be the same; also lagosin and fungichromin. Polifungin A is identical with nystatin (Porowska *et al.*, 1972; Roszkowski *et al.*, 1972), but polifungin B is different from the latter. The pentaenes mycoticins A and B are identical, respectively, with the minor and major components of flavofungin and flavomycoin (Bognar *et al.*, 1970; Uri and Bekesi, 1958). It appears also that the aromatic heptaene macrolides candicidin, trichomycin, levorin, and hamycin are in fact mixtures in different proportions of the same components rather than separate entities. It has been suggested that hamycin and trichomycin are identical (Divekar *et al.*, 1966) and that candicidin is identical with trichomycin (Khokhlova *et al.*, 1963). Pyrolysis gas chromatography of these heptaene macrolides suggests that they are mixtures, with a single identical main component and varying proportions of similar minor components (Burrows and Calam, 1970; Calam, 1974). Countercurrent distribution studies indicated that of the four components of the levorin complex (A_0 , A_1 , A_2 , A_3), A_2 , the main component of the complex, is not distinguishable from the main component of candicidin (Bosshardt and Bickel, 1968). Khokhlova *et al.* (1963) indicated that strains of *Streptomyces griseus* which produce candicidin, *Streptomyces canescens* (the producer of ascosin), and *Streptomyces levoris* (the producer of levorin) are the same species, but

that *Streptomyces hachijoensis* (the producer of trichomycin) is different. A new polyene antibiotic isolated from *Streptomyces helvolioviolaceus* is identical with component A₃ of the levorin complex (Konev *et al.*, 1973).

B. CHEMICAL CHARACTERISTICS OF POLYENE MACROLIDE ANTIBIOTICS

The polyene macrolides form a subdivision of the macrolide antibiotics containing hydroxylated macrocyclic lactone rings and usually one or more sugars. Biogenetically the macrolides are a homogeneous group, being synthesized from acetate and propionate via the polyketide pathway (Bu'Lock, 1967). The macrolide antibiotics are divided into two subgroups: (a) polyene antifungal antibiotics and (b) nonpolyene antibacterial antibiotics. The polyene subgroup has a system of conjugated double bonds, or chromophore, in the macrolactone ring. This results in an amphipathic molecule containing both a rigid planar lipophilic portion and a flexible hydrophilic polyhydroxylated region. The chromophore accounts for some of the characteristic physical and chemical properties of the polyenes (strong light absorption, photolability, and poor solubility in water) and appears to be responsible for the differences in the biological modes of action of the polyene and the nonpolyene macrolide subgroups. The chromophore gives a typical multippeak ultraviolet-visible light absorption spectrum which permits a rapid characterization and division of the polyene macrolides into dienes, trienes, tetraenes, pentaenes, hexaenes, and heptaenes according to the number of conjugated double bonds in the chromophore. A classification of the existing polyenes is given in Table I. The polyene macrolides have lactone rings of 26–38 atoms, which are much larger than those of the nonpolyene macrolides (e.g., a 14-membered lactone in erythromycin). The aminosugars and aromatic moieties found in polyene macrolide antibiotics attached to the macrolide rings are shown in Fig. 1.

Recently, a new class of nonpolyene antifungal macrolides, the axenomycins, with large 34-member lactone rings has been reported. This group appears to be closely related in structure and biological activity to the polyene macrolides in spite of their nonpolyene character (Bianchi *et al.*, 1974).

Although purification of polyene macrolide antibiotics is difficult because of their low solubility and instability to heat and light, considerable progress has been achieved in recent years in the determination of the complex chemical structure of these compounds by utilizing sensitive analytical methods, such as electron impact and field desorption mass spectrometry (Rinehart *et al.*, 1974), proton magnetic resonance, and X-ray structure analysis of single crystals for absolute configuration determination

TABLE I
POLYENE MACROLIDE ANTIBIOTICS (CODE NUMBER 22)

Berd's classification ^a Code number Name (alternative names)	Producer strain	Amino sugar moiety	Nitrogen	Ionic character	Aromatic moiety
221 TRIENES					
(λ maxima 262, 272, 283 nm)					
2211 <i>Trienin type</i>					
Mycotrienin	<i>Streptomyces</i> sp.	ND ^b	Yes	—	—
Trienine	<i>Streptomyces</i> sp.	ND	Yes	—	—
Antibiotic MM-8	<i>Streptomyces</i> sp.	None	Yes	Neutral	—
Triene	<i>Chainia minutisclerotina</i>	ND	ND	—	—
2212 <i>Other trienes</i>					
Resistaphyllin	<i>S. antibioticus</i>	ND	Yes	—	—
222 TETRAENES					
(λ maxima 291, 304, 308 $\pm$ 2 nm)					
2221 <i>Pimaricin type</i>					
Aeromycin B (P-42-E)	<i>S. tumemomaceras</i> var. <i>griseoarenicolor</i>	Mycosamine ^c	Yes	—	—
	<i>S. lucensis</i>	Mycosamine	Yes	Amphoteric	—
Etruscomycin (lucensomycin) (1163 FI)					
Pimaricin (tennecetin)	<i>S. natalensis</i> , <i>S. chattanoogaensis</i> , <i>S. gilveosporus</i>	Mycosamine	Yes	Amphoteric	—
	<i>S. noursei</i> var. <i>jenensis</i>				
Tetramycin	<i>S. noursei</i> var. <i>jenensis</i>	Mycosamine	Yes	—	—
Tetrin A, B	<i>Streptomyces</i> sp.	Mycosamine	Yes	Amphoteric	—
Antibiotic PA-166	<i>Streptomyces</i> sp.	Mycosamine	Yes	Amphoteric	—

2222 <i>Rimocidin type</i>					
Akitamycin	<i>S. akitaensis</i> ,	ND	Yes	Amphoteric	—
(toyamycin)	<i>S. toyamaensis</i>				
Albotetraen	<i>S. albus sterilis</i>	ND	Yes	—	—
Rimocidin	<i>S. rimosus</i>	Mycosamine	Yes	Basic	—
(PA-86)					
Antibiotic RP-9971	<i>S. gascariensis</i>	ND	Yes	Amphoteric	—
2223 <i>Nystatin type</i>					
Amphotericin A	<i>S. nodosus</i>	Mycosamine	Yes	Amphoteric	—
Nystatin A ₁ , A ₂ , A ₃	<i>S. noursei</i> , <i>S. albulus</i>	Mycosamine	Yes	Amphoteric	—
(fungicidin)					
(polifungin)					
Plumbomycin A	<i>S. plumbeus</i>	ND	Yes	—	—
Plumbomycin B	<i>S. plumbeus</i>	ND	Yes	—	—
2224 <i>Other less-known tetraenes</i>					
Antimycin A	<i>S. aureus</i>	ND	—	—	—
Chromin	<i>S. chromogenes</i>	ND	Yes	Acid	—
Endomycin A	<i>S. endus</i>	ND	Yes	Amphoteric	—
(helixin A)					
Flavoviridomycin	<i>S. flavoviridus</i>	ND	Yes	—	—
	var. <i>fungicidicus</i> ,				
	<i>S. hygroscopicus</i>				
	var. <i>enhygrus</i>				
	<i>S. erumpens</i> ,				
	<i>S. ornatus</i>				
Ornamycin (17731)	<i>Streptomyces</i> sp.	ND	Yes	Amphoteric	—
Protocidin	No. 964-A				
Sistomycosin	<i>S. viridosporus</i>	ND	Yes	—	—
Tetraen A, B	<i>Chainia cinnamonea</i>	ND			
Tetramedyn	<i>S. medicicidicus</i>	ND	Yes	—	—
Tetramycin A, B	<i>Chainia grisea</i> ,	ND			
	<i>Chainia grisea</i> var. <i>fusca</i>				

(Continued)

TABLE 1 (Continued)

Berdy's classification ^a Code number Name (alternative names)	Producer strain	Amino sugar moiety	Nitrogen	Ionic character	Aromatic moiety
Unamycin A	<i>S. fungicidicus</i>	Mycosamine	Yes	Acid	—
Antibiotic A-5283	<i>Streptomyces</i> sp. A5283	ND	Yes	Amphoteric	—
Antibiotic AC ₂ -435	<i>Streptomyces</i> sp. AC ₂ -435	ND	No	—	—
Antibiotic J ₁ -B	<i>Streptomyces</i> sp. (<i>S. fungicidicus</i>)	ND	No	—	—
Antibiotic RP-7071	<i>Streptomyces</i> sp.	ND	Yes	Basic	—
Antibiotic LIA 0777	<i>Streptomyces</i> sp. 0777	Mycosamine	Yes	Amphoteric	—
223 PENTAENES					
2231 <i>Methylpentaenes</i> (<i>aldopentaenes</i>) (λ maxima 327, 340, 357 $\pm$ 2nm)					
Aurenin	<i>S. aureorectus</i>	None	No	—	—
Cabacidin	<i>S. gougeroti</i>	None	No	Neutral	—
Chainin	<i>Chainin</i> sp. 3047	None	No	Neutral	—
Filipin complex (durhamycin)	<i>S. filipensis</i>	None	No	Neutral	—
Fungichromin	<i>S. cinnamoneus</i> var. <i>cinnamoneus</i> ,	None	No	Neutral	—
(moldicidin B)	<i>S. roseoluteus</i> ,				
(pentamycin)	<i>S. cellulosa</i> ,				
(Glaxo-A246)	<i>S. pentaticus</i>				
(Lagosin)	<i>S. cellulosa</i>				
Fungichromatin	<i>Streptovorticillium</i> sp.	None	No	—	—
Pentaneicin	<i>S. sanguineus</i>	None	No	—	—
Pentaene	<i>Streptomyces</i> sp.	None	No	Acidic	—
Neopentaene	<i>S. rubrochlorinus</i>	None	No	—	—
Rubrochlorin	<i>S. xantholiticus</i>	None	No	—	—
Xantholicin B	<i>Streptovorticillium</i>	None	No	—	—
Antibiotic HA-106	<i>cinnamoneum</i> var. <i>sparsum</i>	None	No	—	—

Antibiotic HA-135	<i>Streptoverticillium sporiferum</i>	None	No	—	—
Antibiotic HA-145	<i>Streptoverticillium cinnamomeum</i> var. <i>alboporum</i>	None	No	—	—
Antibiotic HA-176	<i>Streptoverticillium cinnamomeum</i> var. <i>lanosum</i>	None	No	—	—
2232 "Normal" pentaenes (amphoteric pentaenes with amino sugar)(λ maxima 317, 331, 350 $\pm$ 2 nm)					
Alomycin	<i>S. acidomyceticus</i>	ND	Yes	—	—
Distamycin C	<i>S. distallicus</i>	ND	—	—	—
Eurocidin A	<i>S. eurocidicus</i> , <i>S. albireticuli</i>	Mycosamine	Yes	Amphoteric	—
Eurocidin B	<i>S. albireticuli</i>	Mycosamine	Yes	Amphoteric	—
Fumanomycin	<i>S. lavendobrunneus</i>	ND	Yes	—	—
Moldcidin A	<i>S. griseofuscus</i> , <i>Streptomyces</i> sp.	Mycosamine	Yes	Acidic	—
Onomycin I	<i>S. sp. J-4 (S. fungitidicus)</i>	ND	Yes	Acidic	—
Pentafungin	<i>S. antimycoticus</i>	ND	Yes	Amphoteric	—
Pentaene G-8	<i>S. anandensis</i>	ND	—	—	—
Quinquamycin	<i>S. lavendulae</i> E-20-27	ND	—	—	—
Antibiotic A-228	<i>Streptomyces</i> sp.	ND	Yes	Neutral	—
Antibiotic PA-153	<i>Streptomyces</i> sp.	ND	Yes	Amphoteric	—
Antibiotic 17-41 B	<i>Streptomyces</i> sp. 17-41	ND	—	—	—
Antibiotic 0371	<i>Streptoverticillium jenensis</i> krissii	—	Yes	—	—
2233 "Normal" pentaenes (Capacidin type)					
Capacidin	<i>S. noursei</i> (variant)	—	Yes	Basic	—
2234 Gangtokumycin type					
Gangtokumycin	<i>S. gangtokensis</i> (<i>S. hygroscopicus</i>)	—	Yes	—	—

(Continued)

TABLE I (Continued)

Berdy's classification ^a Code number Name (alternative names)	Producer strain	Amino sugar moiety	Nitrogen	Ionic character	Aromatic moiety
Genimycin 2235 <i>Carbonyl pentaenes</i> (λ maxima 364 nm; broad peak)	<i>Actinosporangium</i> sp.	—	Yes	—	—
Flavofungin (mycoticin A)	<i>S. ruber</i> , <i>S. flavofungini</i>	None	No	Neutral	—
Flavomycin	<i>S. roseoflavus</i> var. <i>jenensis</i>	None	No	—	—
Mycoticin B Surgomycin 224 HEXAENES (λ maxima 340, 358, 380 $\pm$ 2 nm)	<i>S. flavofungini</i> <i>S. surgutus</i>	None ND	No	Neutral	—
2241 Probably macrocyclic hexaenes					
Candihexin A, B	<i>S. viridoflavus</i> var. 18A2	Mycosamine	Yes	—	—
Candihexin E, F	<i>S. viridoflavus</i> var. 18A2	None	Yes	—	—
Cryptocidin	<i>Streptomyces</i> sp. 963, <i>S. bulgaricum</i>	ND	Yes	Acid	—
Hexin	<i>Streptomyces</i> sp.	ND	—	—	—
Endomycin B (helixin B)	<i>S. endus</i> , <i>S. hygroscopicus</i> var. <i>enhygrus</i>	ND ND	— —	— —	— —
Flavacid	<i>S. flavus</i>	None	Yes	Acid	—
Mediocidin	<i>S. mediodictus</i>	ND	Yes	Amphoteric	—
Tetrahexin (tetraesin)	<i>Streptomyces</i> sp. ATCC 14972	ND	Yes	Amphoteric	—
2242 <i>Fradicin</i> type (unknown structures)					
Fradicin	<i>S. fradiae</i>	ND	Yes	Basic	—
Mycelin	<i>S. roseoflavus</i> , <i>S. diastatochromogenes</i> , <i>S. fradiae</i>	ND	No	—	—

Myceclin IMØ	<i>S. diastatochromogenes</i>	ND	Yes	—	—
Antibiotic A-1404	<i>S. fradiae</i> A-1404, <i>S. diastatochromogenes</i> 207	ND			
2243 Carbonyl hexaenes (λ maxima 385 nm; broad peak)					
Dermostatin (viridofulvin)	<i>S. viridogriseus</i>	None	No	Neutral	—
225 HEPTAENES					
2251 Aromatic heptaenes					
22511 Containing p-aminoacetophenone					
Antifungin 4915	<i>S. paucisporogenes</i>	ND	Yes	ND	ND
Ascosin	<i>S. canescens</i>	Mycosamine	Yes	Amphoteric	p-Aminoacetophenone
Ayfactin, (AYF), (AE-56), (Aureofacin)	<i>S. aureofaciens</i> , <i>S. viridofaciens</i>	Mycosamine	Yes	Acid	p-Aminoacetophenone
Aureofungin	<i>S. cinnamomeus</i> var. <i>terricola</i>	Mycosamine	Yes	Amphoteric	p-Aminoacetophenone
Azacolutin (F-17-C)	<i>S. cinnamomeus</i> var. <i>azacoluta</i>	ND	Yes	Amphoteric	p-Aminoacetophenone
Candidicin (G-252) (PA-150)	<i>S. griseus</i>	Mycosamine	Yes	Amphoteric	p-Aminoacetophenone
Eurotin A	<i>S. griseus</i> H-5592	ND	Yes	Basic	p-Aminoacetophenone
Gerobricin	<i>S. fujuy</i> ATCC 13670	ND	Yes	ND	ND
Hamycin	<i>S. primiprina</i>	Mycosamine	Yes	Amphoteric	p-Aminoacetophenone
Heptaefungin A	<i>S. longisporolavendulae</i>	Mycosamine	Yes	Amphoteric	p-Aminoacetophenone
Levorin A ₆ , A ₁ , A ₂ , A ₃ , B (26/1)	<i>S. levoris</i>	Mycosamine	Yes	Amphoteric	p-Aminoacetophenone

(Continued)