

are picked and subjected to a Bayesian test (Boender & Kan, 1987), or if a deterministic solution is required, the use of interval analysis (Hansen, 1980). This latter option, although desirable, is not critical since substantial benefit could be accrued by reducing the number of molecules that are synthesized and found to be ineffective without attempting to eliminate entirely, within the confines of the model, the possibility of redundant effort.

The important conclusion to be drawn is that two very similar arrangements (P_1 and P_2) were selected from nonobserved starting positions and that if they had been taken as indicating whether synthesis should be carried out the material would, correctly, not have been produced.

This is the first step in developing a screening procedure capable of selecting candidate molecules for synthesis.

This work was performed as part of ESPRIT contract No. 443 on molecular engineering for optoelectronics. I would like to thank N. C. J. Stutchbury for the potential-derived charge routine and A. Charlton for the heat of sublimation measurement.

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Theoretical Study of Polymorphism in (*E*)-2-Dimethylamino-1-(5-nitro-2-furyl)ethylene

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(Received 25 July 1988; accepted 13 April 1989)

Abstract

Lattice-energy calculations for polymorphic crystal modifications of (*E*)-2-dimethylamino-1-(5-nitro-2-furyl)ethylene are reported. The results demonstrate the information that can be gained from this type of calculation and its potential to provide a qualitative screening tool for the crystal forms of the organic molecules appropriate for $\chi^{(2)}$ applications.

Introduction

An investigation of the intermolecular potential-energy variation, calculated by the atom-atom summation technique (Williams, 1981), for the experimentally determined molecular conformations of a polymorphic crystal under both experimentally observed and hypothetical symmetry constraints is reported. The results and approach have application to the search for new organic second-order nonlinear optical materials.

Estimates can be made of the first hyperpolarizability, β (Morley, Docherty & Pugh, 1987),

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which indicate the great potential yet to be realized from organic molecules. The molecular arrangement in a crystal, a property sensitive to molecular modifications which need not significantly affect the hyperpolarizability (Zyss, Chemla & Nicoud, 1981), determines whether the individual molecular contributions combine to give a substantial second-order susceptibility, $\chi^{(2)}$, or cancel partially or entirely. This arrangement is currently determined by trial-and-error synthesis. This is not only time consuming and expensive because of the low success rate but excludes all the advantages that could be accrued from positive molecular design.

The work reported below demonstrates for one system that an examination of the potential energy can give a significant insight into the selection of particular molecular arrangements. This not only provides an excellent method of gaining more information on crystal form, but also demonstrates that the technique may provide a tool, albeit qualitative, which can select molecules with an enhanced probability of forming useful nonlinear optical crystals.

The results given below were obtained using the program *PCK83* (Williams, 1982). A Buckingham potential was used with the supplied parameter values for the interatomic interactions. The molecular coordinates were obtained from the Cambridge Structural Database (Allen *et al.*, 1979). No H-atom coordinates had been determined experimentally and they were therefore added at sp^2 or eclipsed sp^3 orientations with bond lengths of 1.08 or 1.09 Å respectively. This is a standard option available in the graphics display package used. The two conformations of (*E*)-2-dimethylamino-1-(5-nitro-2-furyl)ethylene will be referred to by their respective identifiers (ENVFUR/ENVFUR01) in the Cambridge database. The first (ENVFUR) forms monoclinic crystals with the space group $P112_1/b$, while the second (ENVFUR01) results in orthorhombic crystals with $Pbca$ symmetry.

The charges used in all the calculations reported below were obtained from a least-squares fit to the electrostatic potential calculated for each of the conformations from an *ab initio* wavefunction (Cox & Williams, 1981) at the STO-3G level of approximation. The *ab initio* calculations showed ENVFUR to have an energy of formation of -633.83 a.u., while ENVFUR01 has an energy of formation 0.02 a.u. less at -633.85 a.u. This energy change is associated with a change in the respective dipole moments, found by population analysis, from 9.05 to 8.18 debye (30.2 – 27.3×10^{-30} C m). After the calculation of the point charges from the molecular wavefunctions, the C–H bond lengths of the molecules were reduced by 0.07 Å before the point charges were assigned. This reduction takes account of the displacement of electron density into the bond not reflected in STO-3G calculations (Williams, 1981). The magnitudes of the individual

charges were also increased by a factor of 1.13 to correlate with results on simpler molecules at the 6-31G** level (Cox & Williams, 1981).

(*E*)-2-Dimethylamino-1-(5-nitro-2-furyl)ethylene can be seen from Fig. 1 to conform to the general class of molecules examined for their second-order nonlinear optical properties, since it has donor and acceptor groups at either end of a conjugated pathway (Chemla & Zyss, 1987).

The distance between the N atoms is slightly less for ENVFUR than for ENVFUR01, but the individual bond lengths are more significantly shorter. For example, the bond length from the N atom of the nitro group to the ring is 1.39 Å in ENVFUR compared with 1.43 Å in ENVFUR01. The overall contraction is, however, reduced by the appropriate bond angles being closer to 120° in the former system, as shown in Fig. 1. Hence, the greater conjugation of ENVFUR produces more charge separation, a larger dipole moment and a decrease in the magnitude of the formation energy. As would be expected as a result of the above, the more-conjugated molecule forms dark-violet crystals with a metallic luster, whereas the orthorhombic crystals are dark red (Kusa, Polynova, Porai-Koshits, Kovac & Vegh, 1979).

Results

The results of packing-energy calculations on ENVFUR, in which various space-group symmetries have been imposed, are shown in Table 1. It is appropriate to note some of the basic features before the alternative set of results for ENVFUR01 are introduced, and the discussion and comparison of the findings begins.

Three alternative forms can be seen to have been selected out from the general background, two of which correspond to experimentally determined space-group

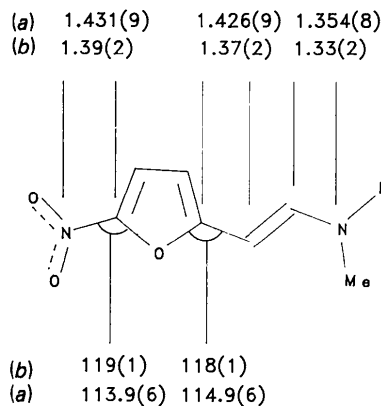


Fig. 1. The (*E*)-2-dimethylamino-1-(5-nitro-2-furyl)ethylene molecule showing the major conformational differences between the orthorhombic (a) and monoclinic (b) derived forms.

Table 1. Results for ENVFUR

Observed structure $P112_1/b$, volume = 904 Å³.

Model PD-STO-3GN-FS

Form	Energy (kJ mol ⁻¹)	Volume (Å ³)	Fig.
$P\bar{1}$	-101.65	465	2(a)
$P\bar{1}^{ROT}$	-102.78	454	3(a)
$P2_1$	-89.77	480	2(b)
Cc	-85.75	990	2(c)
$P112_1/b$	-101.34	936	2(d)
$P112_1/b^{ROT}$	-105.20	921	3(b)
$C2/c$	-95.96	1909	2(e)
$P2_12_1$	-97.35	931	2(f)
$Pbca$	-100.43	1809	2(g)
$Pbca^{CB}$	-101.40	1854	7

Notes: PD indicates potential-derived charges least-squares fitted to the electrostatic potential calculated directly from the molecular wavefunction. STO-3G indicates the quantum-mechanical level of approximation used in the charge calculation. N indicates that the charges have been rescaled by a factor of 1.13. This is an approximate factor relating results calculated at the STO-3G level of approximation with corresponding results at the 6-31G** level. FS indicates that the C-H bond lengths have been foreshortened by 0.07 Å. ROT indicates that the methyl groups have been rotated by 60°. CB indicates that the calculation was started with the asymmetric unit in the position of the alternative polymorph's asymmetric unit and with the appropriate unit-cell parameters.

symmetries for the compound, *i.e.* $P112_1/b$ and $Pbca$, while the third is the centrosymmetric triclinic form ($P\bar{1}$).

The two $Pbca$ results in Table 1 show the effect of finding a subminimum. The most positive energy form was started with the asymmetric unit initially in the observed monoclinic orientation and with a cubic unit

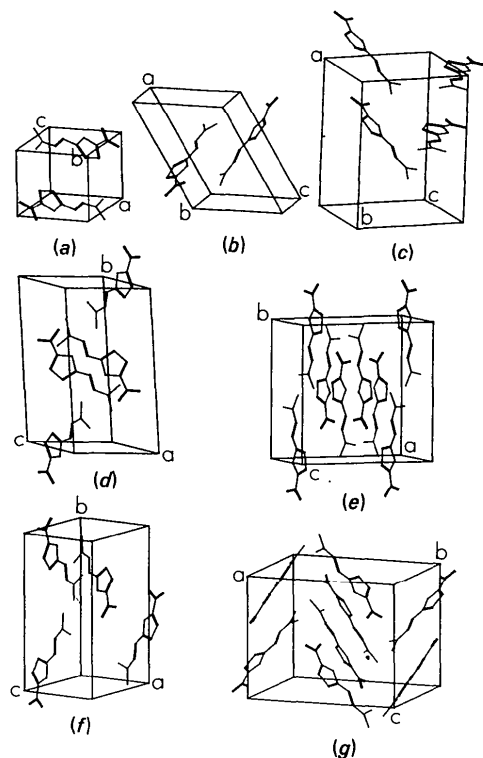


Fig. 2. Structures (a) to (g) of Table 1.

Table 2. Results for ENVFUR01

Observed structure $Pbca$, volume = 1773 Å³.

Model PD-STO-3GN-FS

Form	Energy (kJ mol ⁻¹)	Volume (Å ³)	Fig.
$P\bar{1}$	-97.52	465	4(a)
$P2_1$	-95.72	484	4(b)
Cc	-77.12	1064	4(c)
$P112_1/b$	-96.50	966	4(d)
$P112_1/b^{CA}$	-96.62	958	
$C2/c$	-85.29	2071	4(e)
$P2_12_1$	-87.94	975	4(f)
$Pbca$	-100.73	1826	4(g)
$Pbca^{ROT}$	-99.36	1852	5

Notes: the calculation with the asymmetric unit started in the position of the alternative polymorph's asymmetric unit and with the appropriate unit-cell parameters is indicated by CA. For an explanation of the other abbreviations used in this table, see Table 1.

cell of side 10 Å, while the more negative energy arrangement was started from the orientation, and with the cell settings, of the observed $Pbca$ modification. This result demonstrates that finding a subminimum with a very different appearance to that of the observed form (see Figs. 2g and 4g) does not necessarily lead to a wrong assignment of the importance of likelihood of a particular space-group arrangement if the energy of the subminimum is sufficiently close to that of the global minimum. In any screening exercise using molecules with unknown crystal structures in an assessment of their $\chi^{(2)}$ potential, only the energy and volume will be available to rank the arrangements derived under the constraints applied. It is therefore encouraging that the possibility of polymorphic forms is manifested in the results and that the ranking of subminima for this compound, on the limited evidence, is more satisfactory than might be envisaged.

The results found for ENVFUR01 are given in Table 2, from which it can be seen that the three most negative energy space groups predicted by the calculations are the same as those found for the ENVFUR molecule in Table 1. In this case, however, there is a comparatively large energy separation between the orthorhombic space group found experimentally, and the monoclinic and triclinic selections. The discussion below examines in more detail the relationship between the molecular size and conformation and the selected crystal form.

Discussion

The results of Tables 1 and 2 show the possibilities and problems of lattice-energy minimization. Some of these features are highlighted in the following discussion and attention drawn to the consequences they have on the development of a quasi-predictive tool for active materials.

The monoclinic derived molecule (ENVFUR) produces good discrimination between three of the space groups and the rest of the sample set, a result which is

repeated if not as emphatically in Table 2. The three most-favoured forms found by the calculations are all centrosymmetric and one of the forms in each case is an experimentally observed symmetry group started from a nonobserved asymmetric unit position, *i.e.* the asymmetric unit orientation of the alternative observed form. Thus, the wrong molecule started from the wrong place nevertheless provides useful information.

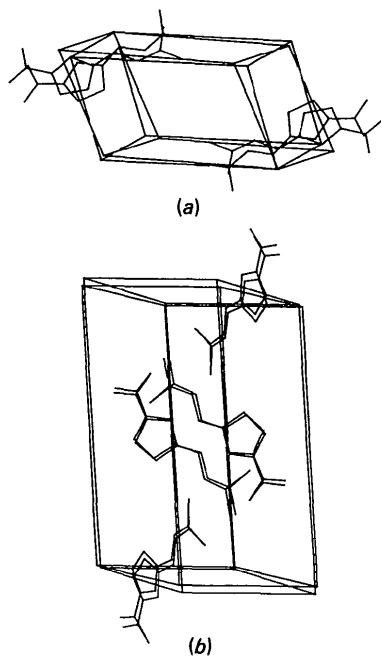


Fig. 3. Methyl-rotated form of ENVFUR superimposed on the standard form for space groups (a) $P\bar{1}$ and (b) $P112_1/b$.

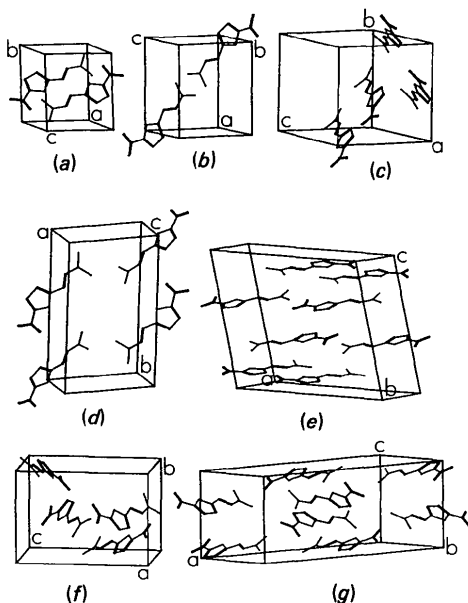


Fig. 4. Structures (a) to (g) of Table 2.

An examination of the effect of methyl-group rotation on the predicted $P\bar{1}$ and $P112_1/b$ space groups was carried out for the monoclinic derived molecule of Table 1. As with previous calculations of this kind (Higgins, 1989) a rotation of 60° was made to each of the methyl groups to mimic a form which will exist in solution. The argument is that a crystal form will be precipitated out of solution not simply because it has the most negative energy giving rise to the tightest binding, but because it is resilient to conformational changes in the molecule such as methyl-group rotation. It can be seen from Table 1 and Fig. 3 that the energy-relaxed structure given by the packing calculations of the experimentally observed monoclinic form is predicted to be the more stable with respect to this change.

A similar procedure to the above was carried out on the energy-relaxed experimentally observed orthorhombic form of Table 2 which, as shown in Fig. 5, is quite seriously perturbed by the methyl-group rotations. The energy, however, remains significantly below that of the alternative space groups examined. This result can be used to interpret the finding of Table 1, in which the monoclinic molecule refined from the observed orthorhombic settings is predicted, by a slight margin, to be the most favoured of the two observed forms. In the monoclinic molecular conformation this slight energy advantage, or equivalence within any reasonable assumption of the accuracy of the model, of the $Pbca$ form is insufficient to overcome the instability of the form to methyl-group rotation. Changing the conformation of the molecule and thereby reducing the crystals energy affects the monoclinic and triclinic forms disproportionately and allows the $Pbca$ form to be realized. There is, unfortunately, no experimental evidence currently available which would provide an independent check on the above argument.

An important aspect of the above results is the reduction in the lattice energy shown in Table 2 for the triclinic system when compared with the analogous entry in Table 1. The results in Table 2 are produced by minimizing from a cubic lattice using the orthorhombic form of the molecule started from the experimentally observed orthorhombic orientation. Similarly, the results in Table 1 use the same size of initial cube but with the monoclinic form of the molecule started at its observed orientation. Fig. 6 is a comparison of the results obtained by these independent calculations. It

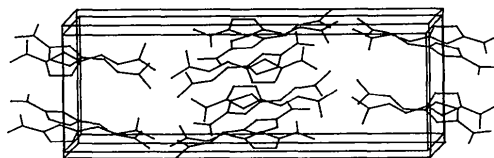


Fig. 5. Methyl-rotated form of ENVFUR01 superimposed on the standard form for space group $Pbca$.

demonstrates that the two forms are essentially identical and that a comparison of the energies is therefore meaningful. In this case subminima do not distort the comparison and it is probable that the results represent the global minimum for that space-group symmetry.

The $P112_1/b$ result of Table 2 when the molecule ENVFUR01 is given the observed orientation and cell size of ENVFUR, by least-squares fitting the heavy atoms to the locations found for the corresponding ENVFUR conformation, also supports the case of the energy difference produced by the use of the alternative polymorphic forms being a real effect and not the result of finding a subminimum. Table 3 below illustrates the changes in energy produced by the alternative conformations before and after relaxation from the observed $P112_1/b$ arrangement to the calculated energy minimum.

Comparing the results for the experimentally observed lattice in the above table demonstrates that the repulsive interaction is the most important energy term. Hence, it is the change in size of the molecule which is the dominant factor in the packing-energy difference found. This is a consequence of the approximately 3% shorter molecular bonds of ENVFUR compared with ENVFUR01 (see Fig. 1) allowing all the molecules of the ENVFUR crystal to be closer together and thereby increasing the magnitude of the binding energy. It should be recalled that the repulsive-energy contribution falls off exponentially with distance, whereas the attractive-dispersive term has an inverse sixth-power dependence (Williams, 1981). The above system also has an additional electrostatic contribution acting in the same sense, which is produced by the decrease in the dipole moment despite the increase in the length of ENVFUR01.

The $Pbca$ crystal form of ENVFUR01 can compensate to a large extent for both of the above effects because the extension and twisting of the polyenic arm is sufficient to allow the furan ring of an adjacent

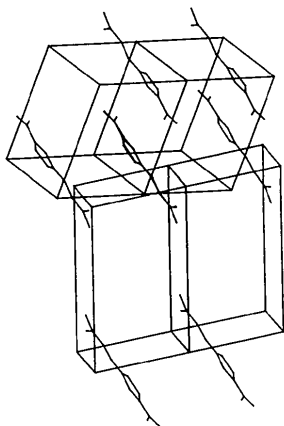


Fig. 6. Comparison of the $P1$ structures found using ENVFUR and ENVFUR01 started from their observed orientations.

Table 3. Energies for $P112_1/b$ symmetry

All energies quoted in kJ mol^{-1} .					
Molecule	Lattice	Dispersion	Repulsion	Coulomb	Total
ENVFUR	Observed	-145.89	77.33	-27.27	-95.83
	Relaxed	-135.23	59.28	-25.39	-101.34
ENVFUR01	Observed	-148.81	88.37	-25.28	-85.71
	Relaxed	-131.01	57.12	-22.72	-96.62

molecule to move into the previously excluded volume between the ring and the chain of the molecule (see Fig. 7). Hence, the unit-cell volume of the $Pbca$ form is less for the larger ENVFUR01 molecule than for the alternative ENVFUR system. The reverse situation, however, applies in the $P112_1/b$ case.

Concluding remarks

Although subminima were found they have not been seriously misleading for the particular space groups where a comparison is possible, because their energies are sufficiently close to the appropriate global minimum. This is an encouraging result from the point of view of developing a screening tool for crystals. More polymorphic systems need to be examined, however, before general conclusions can be drawn about the accuracy required of approximations to the global minimum.

The analysis of the results provided by the above discussion demonstrates the usefulness of lattice-energy calculations in understanding the factors influencing crystal form. A simple potential-energy model has provided a consistent framework within which this polymorphic system could be qualitatively understood.

This work was performed as part of ESPRIT contract No. 443 on molecular engineering for optoelectronics. I would like to thank N. C. J. Stutchbury for the potential-derived charge routine.

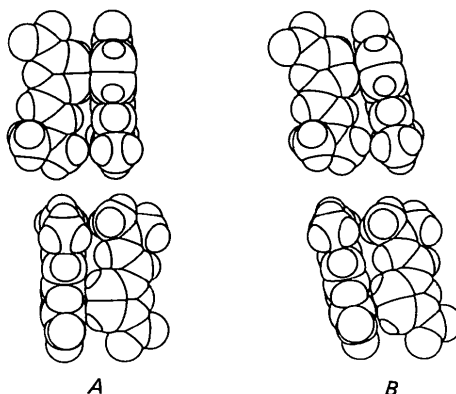


Fig. 7. Space-filling views of the $Pbca$ structure (A) of Table 2 and the $Pbca^{CB}$ structure (B) of Table 1. Molecules have been removed for clarity. Structure A can be seen to be better packed, with less excluded volume than structure B.

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Acta Cryst. (1989). B45, 447–452

The Structures of 1-Deoxy-(*N*-methyloctanamido)-D-glucitol (MEGA-8) and 1-Deoxy-(*N*-methylundecanamido)-D-glucitol (MEGA-11)

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(Received 21 November 1988; accepted 12 April 1989)

Abstract

1-Deoxy-(*N*-methyloctanamido)-D-glucitol, C₁₅H₃₁NO₆ (MEGA-8), crystallizes in space group *P*2₁2₁2₁, $M_r = 321.4$, $a = 4.865$ (1), $b = 9.186$ (3), $c = 39.097$ (9) Å, $V = 1747.24$ Å³, $Z = 4$, $D_x = 1.22$ Mg m⁻³, $\lambda(\text{Cu } K\alpha) = 1.5418$ Å, $\mu = 0.78$ mm⁻¹, $F(000) = 704$, $R = 0.035$ for 1318 reflections. 1-Deoxy-(*N*-methylundecanamido)-D-glucitol, C₁₈H₃₇NO₆ (MEGA-11), crystallizes in space group *P*1, $M_r = 363.5$, $a = 4.950$ (1), $b = 5.6027$ (8), $c = 19.162$ (4) Å, $\alpha = 83.19$ (2), $\beta = 89.76$ (2), $\gamma = 76.28$ (2)°, $V = 512.64$ Å³, $Z = 1$, $D_x = 1.18$ Mg m⁻³, $\lambda(\text{Mo } K\alpha) = 0.7107$ Å, $\mu = 0.093$ mm⁻¹, $F(000) = 200$, $R = 0.061$ for 1898 reflections. The glucitol C-atom-chain conformation is different in the two structures. In MEGA-8 it is fully extended, whereas in MEGA-11 it is bent. The alkyl C-atom chains are fully extended in both structures. The molecular packing is different. In MEGA-8 it is head-to-head bilayer with intercalating alkyl chains, whereas in MEGA-11 it is monolayer head-to-tail with non-intercalating alkyl chains. The hydrogen bonding in MEGA-8 is a finite chain; in MEGA-11 it includes a homodromic four-bond cycle.

Introduction

The alkylated saccharides constitute a large class of amphiphilic molecules, many of which form, or are expected to form, both thermotropic and lyotropic liquid crystals (Jeffrey, 1986). The rod-shaped mesogens consist of a carbohydrate head group and an *n*-alkyl chain which generally has to exceed six C

atoms. It is important that the head group contains sufficient unblocked hydroxyls to form the hydrogen-bonded 'core', which is responsible for the molecular association in the liquid-crystal phase (Jeffrey, 1984; Goodby, 1984). In the carbohydrates, the head group may be cyclic, *i.e.* a pyranosyl or furanosyl ring, bicyclic, as in a disaccharide, or acyclic, as in the derivatives of a sugar alcohol (Pffannemüller & Welte, 1985; Pffannemüller, Welte, Chin & Goodby, 1986; Baeyens-Volant, Cuvelier, Fornasier, Szalai & David, 1985). There are also 'disc-shaped' carbohydrate mesogens, where the central core is a pyranose or inositol ring around which there are substituted at least three and generally more alkyl chains (Kohne & Praefcke, 1984; Zimmermann, Jameson & Weiss, 1985). The functional groups, which attach the alkyl chains to the head group, appear to be irrelevant to the mesogeneity and hence great configurational variety is possible. The alkylated saccharides form lyotropic liquid crystals with water, when sufficiently soluble. Octyl β -D-glucopyranoside, a well known surfactant used in the crystallization of membrane proteins (Baron & Thompson, 1975), has classical lyotropic behavior forming laminar, cubic and hexagonal phases and a micellar solution with increasing dilution with water at room temperature (Chung & Jeffrey, 1989). The MEGA series of surfactants from MEGA-8 to MEGA-12 are also used in membrane protein crystallization (Hildreth, 1982).

This type of compound also occurs, with much more configurational complexity, in natural products, in the lipids of *Mycobacterium tuberculosis* in the form of 'cord-factors' (Asselineau & Asselineau, 1978), which