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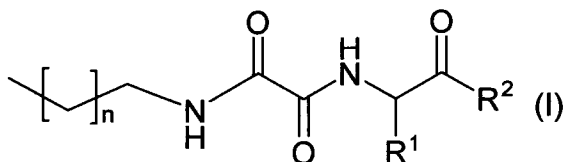
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(54) Title: AMPHIPHILIC OXALAMIDE ORGANOGELATORS DESIGNED FOR GELATION OF ORGANIC SOLVENTS, WATER AND HYDROCARBON COMMERCIAL FUELS



(57) Abstract: The compounds of general formula (I) wherein: R¹ is hydrogen, phenyl, C₁₋₆ alkyl which can be further substituted with R³ R³ is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups; R² is OR⁴, NHR⁴ R⁴ is hydrogen, C₁₋₆ alkyl or benzyl n is an integer from 1 to 12 or the salts of compounds mentioned which can form gels with commercial fuels, organic solvents and water.



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Amphiphilic oxalamide organogelators designed for gelation of organic solvents, water and hydrocarbon commercial fuels

Field of invention

The present invention relates to compounds which are capable of forming gels with commercial fuels, appropriate organic solvents and water, and to methods of preparing these compounds. The invention also relates to the gels formed by these compounds, methods for making them and use of the gels in various applications.

Organogels have great potential for industrial applications due to their diversity in the structures and physical properties. Particular interest is in pharmaceutical industry for drug delivery, food and cosmetic industry, tissue engineering, electrooptic / photonic devices, as sensors or organic templates for the preparation of inorganic materials, etc. Gelation of commercial fuels and hazardous organic solvents and liquids can be useful for development of safety transport technology.

Background art

Investigation of gel formation by low molecular weight organogelators has attracted considerable interest in last 15 years. Some of the reviews are publicised (P. Terech *et al*, *Chem. Rev.*, **97**, 3133-3159 (1997), L. A. Estroff *et al*, *Chem. Rev.* 104,1201-1217 (2004), F. Fages *et al*, *Top. Curr. Chem.*, **256**, 77-131 (2005), M. Neralagatta *et al*, *Chem. Soc. Rev.*, **34**, 821-836 (2005), M. de Loos, *et al*, *Eur. J. Org. Chem.* (2005) 3615-3631).

First paper about selective oil gelation from oil / water mixtures with potential application in oil transport accident rehabilitations was reported by S Bhattacharya *et al*, *Chem. Commun.*, (2001) 185.

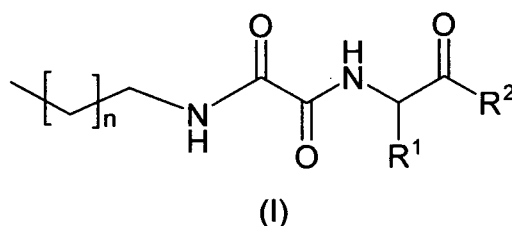
Some other publications describe investigation of commercial fuel gelation (D.R. Trivedi *et al*, *Chem. Mater.*, **18**, 1470-1478 (2006); D.R. Trivedi *et al*, *Chem. Eur. J.* 2004, 10, 5311 - 5322; D.R. Trivedi *et al*, *Crystal Growth & Design*, **6(9)**:2114-2121, 2006; M. Suzuki *et al*, *New J. Chem.*, **30**, 1184-1191, 2006.; D. Khatua *et al*, *Langmuir* **21**, 109-114, 2005.)

In our earlier publication (V. Čaplar *et al*, *Eur. J. Org. Chem.*, **2004(19)**, 4048-4059) we tested gelation properties of some alkylaminoundecanoic acid derivatives with oil (Diesel), water and appropriate organic solvents.

Detailed description of invention

The present invention relates to the new amphyphilic oxalamide derivatives which are capable of forming gels with commercial fuels. In addition, these compounds can form gels with water and some organic solvents.

The present invention relates to the compounds of general formula (I):



wherein:

R^1 is hydrogen, phenyl, C_{1-6} alkyl which can be further substituted with R^3

R^3 is a 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxy groups

R^2 is OR^4 , NHR^4

R^4 is hydrogen, C_{1-6} alkyl or benzyl

n is an integer from 1 to 12

or a salt thereof.

Organogelators of general formula (I) can form gels by dissolution of a compound in a tested solvent and cooling the resulting solution to the room temperature. This procedure causes gel formation, which is macroscopical manifestation of supramolecular self-assembly of certain organic molecules to the nanoscale gel network.

The compounds of general formula (I), especially amide derivatives, can form gels at very low concentrations.

In the present invention, the term “organogelator” refers to any low molecular weight compound (such as the compounds of general formula (I)) which is able to form gel in appropriate solvent. The compound is dissolved by heating and the gel is formed by cooling this solution.

In the present specification “ C_1 - C_6 alkyl” refers to a straight or branched hydrocarbon chains having one to six carbon atoms. Examples of alkyl moiety are methyl, ethyl, *n*-propyl, isopropyl, *t*-butyl, *n*-butyl and *n*-hexyl.

Salts of the compounds of general formula (I) include inorganic salts, ammonia salts and salts of substituted amines.

If a chiral centre is present in compounds of the present invention, all forms of such isomers are intended to be covered herein. Compounds of the invention containing a chiral centre may be used as enantiomers, racemic mixtures or enantiomerically enriched mixtures.

The term "aromatic moiety" in the context of the present specification refers to aromatic ring systems having 5 to 10 carbon atoms incorporated in one or two rings. Examples of aromatic moieties are benzene, naphthalene, biphenyl ring systems etc.

"Heteroaromatic" refers to aromatic ring systems as defined above but in which one or more carbon atoms is replaced by a nitrogen, oxygen or sulphur atom. Examples of heteroaromatic ring systems are indole, pyridine, imidazole, indazole, benzimidazole, quinoline, isoquinoline, benzthiazole, benzoxazole, etc.

In selected compounds of general formula (I) independently or in any combination:

R^2 is OH or NHR^4

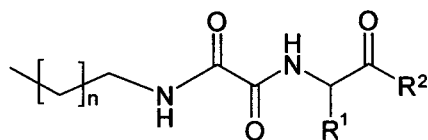
n is integer from 2 to 10

Selected salts of compounds of general formula (I) are Na^+ and K^+ .

In preferred compounds of general formula (I) n is 4 to 10 and R^2 is NH_2 .

Preferred salts of compounds of general formula (I) are Na^+ salts.

Particularly preferred compounds, according to general formula (I), include:



1 $n=2$, $R^1=CH_2CH(CH_3)_2$, $R^2=OMe$

2 $n=2$, $R^1=CH_2CH(CH_3)_2$, $R^2=OH$

3 $n=2$, $R^1=CH_2CH(CH_3)_2$, $R^2=O^-Na^+$

4 $n=2$, $R^1=CH_2CH(CH_3)_2$, $R^2=NH_2$

5 $n=4$, $R^1=CH_2CH(CH_3)_2$, $R^2=OMe$

6 $n=4$, $R^1=CH_2Ph$, $R^2=OMe$

7 $n=4$, $R^1=CH_2CH(CH_3)_2$, $R^2=OH$

8 $n=4$, $R^1=CH_2Ph$, $R^2=OH$

9 $n=4$, $R^1=CH_2CH(CH_3)_2$, $R^2=O^-Na^+$

10 $n=4$, $R^1=CH_2Ph$, $R^2=O^-Na^+$

11 $n=4$, $R^1=CH_2CH(CH_3)_2$, $R^2=NH_2$

12 $n=4$, $R^1=CH_2Ph$, $R^2=NH_2$

13 $n=10$, $R^1=CH_2CH(CH_3)_2$, $R^2=OMe$

14 $n=10$, $R^1=CH_2CH(CH_3)_2$, $R^2=OH$

15 $n=10$, $R^1=CH_2CH(CH_3)_2$, $R^2=O^-Na^+$

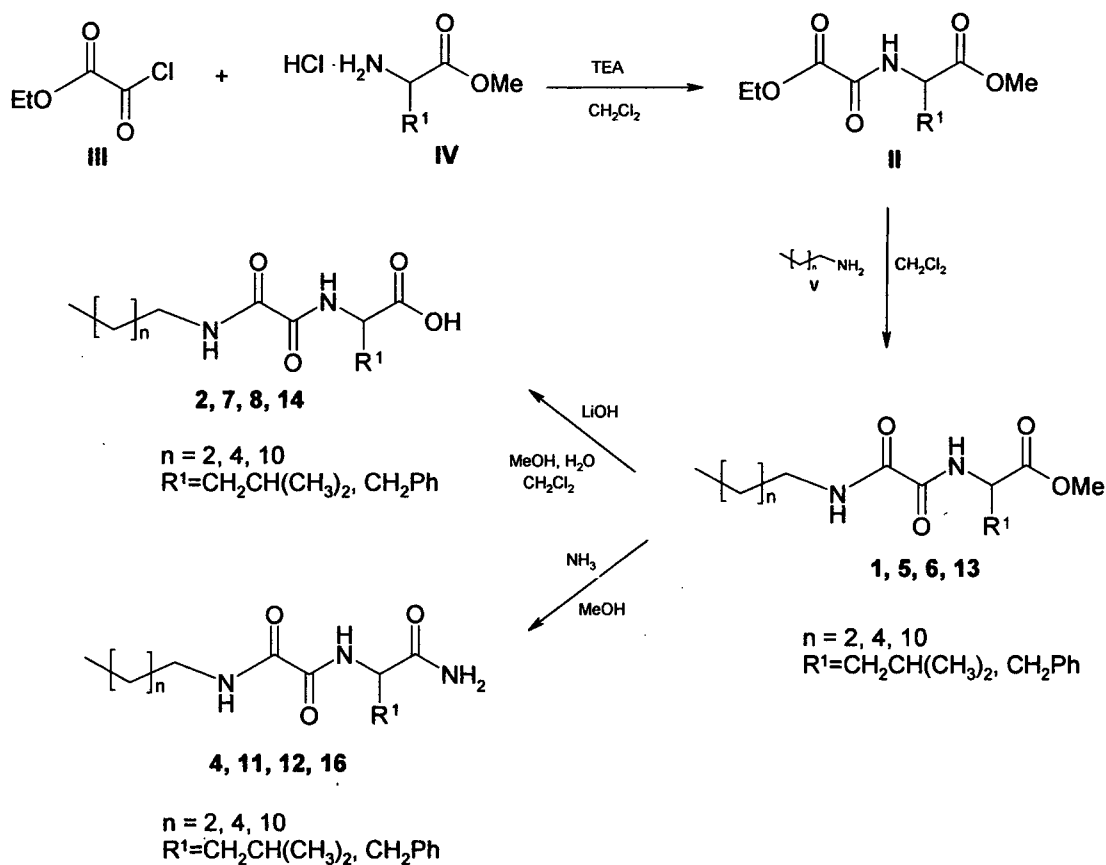
16 $n=10$, $R^1=CH_2CH(CH_3)_2$, $R^2=NH_2$

e.g. following compounds:

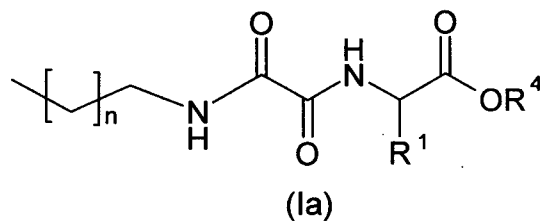
4

1. N-butyloxalamido-L-leucine methyl ester
2. N-butyloxalamido-L-leucine
3. N-butyloxalamido-L-leucine sodium salt
4. N-butyloxalamido-L-leucinamide
5. N-hexyloxalamido-L-leucine methyl ester
6. N-hexyloxalamido-L-phenylalanine methyl ester
7. N-hexyloxalamido-L-leucine
8. N-hexyloxalamido-L-phenylalanine
9. N-hexyloxalamido-L-leucine sodium salt
10. N-hexyloxalamido-L-phenylalanine sodium salt
11. N-hexyloxalamido-L-leucinamide
12. N-hexyloxalamido-L-phenylalaninamide
13. N-dodecyloxalamido-L-leucine methyl ester
14. N-dodecyloxalamido-L-leucine
15. N-dodecyloxalamido-L-leucine sodium salt
16. N-dodecyloxalamido-L-leucinamide

Preferred compounds of the invention are prepared according to the following scheme:



Compounds of general formula (I), where R^2 is OR^5 , can be presented as esters of general formula (Ia)



wherein:

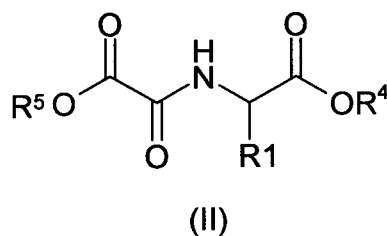
R^1 is hydrogen, phenyl, C_{1-6} alkyl which can be further substituted with R^3

R^3 is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups;

R^4 is C_{1-6} alkyl or benzyl

n is an integer from 1 to 12.

Compounds of general formula (Ia) can be prepared from the compounds with general formula (II)



wherein:

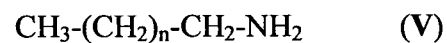
R^1 is hydrogen, phenyl, C_{1-6} alkyl which can be further substituted with R^3

R^3 is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups;

R^4 is C_{1-6} alkyl or benzyl

R^5 is C_{1-6} alkyl or benzyl

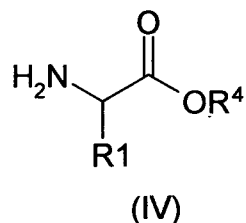
by an aminolysis with compounds of general formula (V):



wherein n is an integer from 1 to 12.

The reaction is performed by stirring the solution of compounds with general formula (II) and compounds with general formula (V) in an organic solvent at room temperature.

Compounds of general formula (II) can be prepared from compounds of general formula (IV):



wherein:

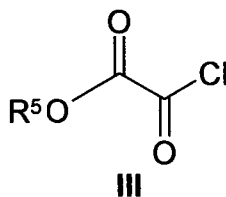
R¹ is hydrogen, phenyl, C₁₋₆ alkyl which can be further substituted with R³

R³ is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups;

R⁴ is C₁₋₆ alkyl or benzyl

or from a salt thereof, whereby salt can be hydrochloride or tosylate

by an acylation with compounds of general formula (III):



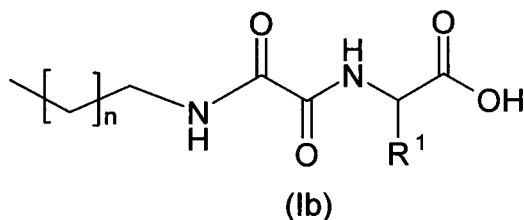
wherein:

R⁵ is alkyl or benzyl.

Acylation reaction can be carried out in organic solutions by dropwise addition of solution of compounds with general formula (III) to a solution of compounds with general formula (IV) and TEA at temperature -10–0 °C. After that the reaction mixture is allowed to warm up to room temperature and stirred overnight.

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Compounds of general formula (I), in which R^2 is OH, can be presented as compounds of general formula (Ib):



wherein:

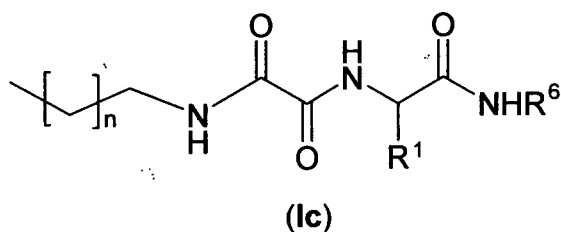
R^1 is hydrogen, phenyl, C_{1-6} alkyl which can be further substituted with R^3

R^3 is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups;

n is an integer from 1 to 12.

Compounds of general formula (Ib) can be prepared from the corresponding esters of general formula (Ia). The conversion of ester groups into carboxylic acids can be achieved by any known method, for example by hydrolysis of alkyl esters using lithium hydroxide solution or, alternatively, by hydrogenolysis of benzyl esters. In this reaction a solution of ester is hydrogenated over a suitable catalyst (typically palladium/carbon). Suitable protection and deprotection methodologies can be found, for example, in *Protecting Groups in Organic Synthesis*, by Theodor W. Greene and Peter G. M. Wuts, published by John Wiley & Sons Inc.

Compounds of general formula (I), in which R^2 is NHR^6 , can be presented as compounds of general formula (Ic):



wherein:

R^1 is hydrogen, phenyl, C_{1-6} alkyl which can be further substituted with R^3

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R^3 is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups;

R^6 is hydrogen, C_{1-6} alkyl or benzyl

n is an integer from 1 to 12.

Compounds of general formula (Ic) can be prepared from the corresponding esters of general formula (Ia) by the aminolysis reaction with the compounds of general formula (VI)

R^6-NH_2 (VI)

wherein R^6 is hydrogen, C_{1-6} alkyl or benzyl.

The conversion of ester group into carbamoyl group can be achieved by known methods, for example by reaction with saturated $NH_3/MeOH$ or with solution of substituted amines.

Compounds of general formula (I) are capable of forming gels when heated in appropriate solvent, followed by cooling to room temperature. Alternatively, gel can be formed by dissolving organogelator in one organic solvent, followed by mixing this solution with another solvent.

Therefore, this invention relates also to the gels formed by the compounds of formula (I) or a salt thereof mixed with solvents. It also relates to the methods for preparing such gels from compounds of formula (I) or a salt thereof and solvents as well as the use of these gels in various applications.

Solvents can be commercial fuels like petrol, diesel etc. Alternatively, solvents can be H_2O or organic solvents such as DMSO, DMF, EtOH, CH_3CN , THF, CH_2Cl_2 , acetone, toluene, *p*-xylene, tetraline, decaline or mixtures thereof.

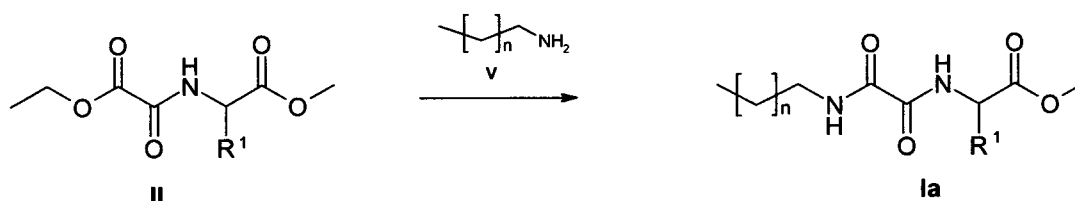
Chemical synthesis of the compounds with general formula (I)

General

Reagents were purchased from Aldrich, Fluka, Kemika, Merck and Sigma, and were used without further purification. All solvents were purified and dried according to standard procedures. The reactions were monitored by thin layer chromatography (TLC) on Merck

Kieselgel HF254 plastic sheets and spots were made visible using a UV lamp (254 nm) or I₂ vapours. Prepared compounds were purified chromatographically by preparative T.L.C. using silica gel *Merck HF₂₅₄* and by column chromatography using silica 0.063-0.2 mm (Merck). Reaction yields are not optimised. NMR spectra were recorded on a Bruker Avance spectrometer at 300/75 MHz with tetramethylsilane (TMS) as an internal standard. Chemical shifts (δ) were given in ppm, coupling constants (J) in Hz. Spin multiplicities; s (singlet), d (doublet), t (triplet), q (quadruplet), p (pentet) and m (multiplet). IR spectra were taken in KBr pellets on ABB Bomen MB 102 FTIR-spectrometer, wave numbers (ν) are reported in cm⁻¹. Optical rotations were measured on an Optical Activity AA-10 Automatic Polarimeter in a 1 dm cell at 589 nm; concentrations were given in g/100 ml.

1. General procedure for preparation of N-alkyl, N'-L-methyleucyl(phenylalanyl) oxalamides (Ia)

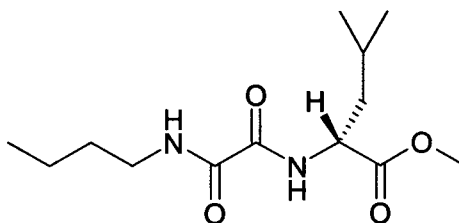


1. $n=2$, $R^1=\text{CH}_2\text{CH}(\text{CH}_3)_2$
5. $n=4$, $R^1=\text{CH}_2\text{CH}(\text{CH}_3)_2$
6. $n=4$, $R^1=\text{CH}_2\text{Ph}$
13. $n=10$, $R^1=\text{CH}_2\text{CH}(\text{CH}_3)_2$

- a) **Preparation of N-(ethoxyoxalyl)-L-leucine(phenylalanine)methyl ester (II);** To a cooled (0 °C) solution of hydrochloride of amino acid **IV** (1 mmol) and TEA (2 mmol) in dry CH₂Cl₂ (10 ml) a solution of oxalyl chloride **III** (1 mmol) in dry CH₂Cl₂ (5 ml) was added dropwise during the 15 minutes. The reaction mixture was stirred for 30 min at 0 °C and 20 hours at room temperature. Afterwards, CH₂Cl₂ (10 ml) was added and the mixture was washed successively with 5 % HCl, 5 % NaHCO₃ and water. The organic layer was dried over Na₂SO₄ and the solvent removed under reduced pressure. The ester **II** obtained in this way was used without additional purification.
- b) To the solution of N-(ethoxyoxalyl)-L-leucine(phenylalanine)methyl ester (**II**) in dry CH₂Cl₂ alkylamine **V** was added and the reaction mixture was stirred for 3 days at room

temperature. The product was purified by crystallisation (CH_2Cl_2 –light petroleum) or preparative TLC chromatography (EtOAc–light petroleum, 100:25).

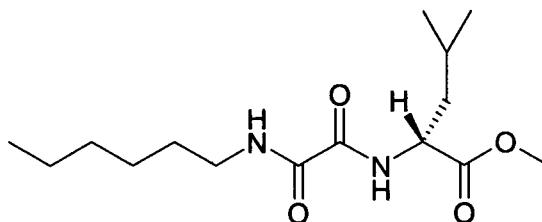
Example 1: N-butyloxalamido-L-leucine methyl ester (1)



Molecular Weight = 272,35
Molecular Formula = $\text{C}_{13}\text{H}_{24}\text{N}_2\text{O}_4$

To the solution of N-(ethoxyoxalyl)-L-leucine methyl ester (**II**) (0.270 g, 1.01 mmol) in dry CH_2Cl_2 (10 ml) *n*-butylamine (0.110 ml, 1.113 mmol) was added and the reaction mixture was stirred for 3 days at room temperature. The product was purified by crystallisation (CH_2Cl_2 –light petroleum) and by preparative TLC chromatography (EtOAc–light petroleum, 200:50). Yield: 78.7 %; $[\alpha]_{\text{D}} = -13$ (c 1, CH_2Cl_2); ^1H NMR (DMSO-d_6): δ 8.94 (1 H, d, $J = 8.3$, NH_{Leu}), 8.77 (1 H, t, $J = 5.7$, NH_{butyl}), 4.36 (1H, ddd, $J = 3.6$, $J = 8.3$, $J = 11.1$, $\text{CH}_{\alpha, \text{Leu}}$), 3.63 (3 H, s, OCH_3), 3.18 – 3.09 (2 H, m, CH_2N), 1.86 – 1.78, 1.59 – 1.49, 1.48 – 1.39, 1.31 – 1.19 (1H, 2H, 2H, 2H, 4 m, $\text{CH}_{\beta, \text{Leu}}$, $\text{CH}_{2, (\beta, \text{Leu})}$, $\text{CH}_{2, \text{butyl}}$), 0.89 – 0.83 (9 H, $\text{CH}_{3(\gamma, \text{Leu})}$ and $\text{CH}_3, \text{butyl}$), ^{13}C NMR (DMSO-d_6): δ 171.9 (COOMe), 160.3, 159.4 (CON), 52.0 (OCH_3), 50.4 ($\text{CH}_{\alpha, \text{Leu}}$), 38.9, 38.5 ($\text{CH}_2 (\beta, \text{Leu})$, $\text{CH}_2\text{N}_{\text{butyl}}$), 30.7, 19.5 ($\text{CH}_2, \text{butyl}$), 24.3, 22.8, 21.0 ($\text{CH}_{\gamma, \text{Leu}}$, $\text{CH}_3, \sigma, \text{Leu}$), 13.5 $\text{CH}_3, \text{butyl}$); IR: 3300, 1749, 1652, 1521.

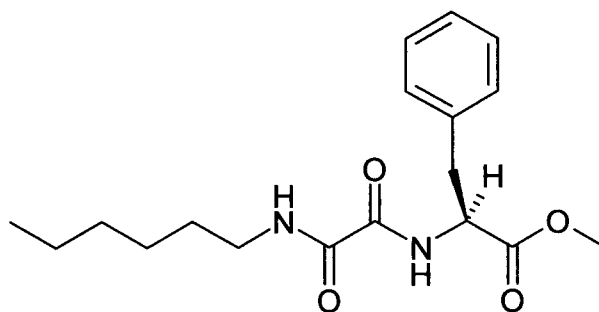
Example 2: N-hexyloxalamido-L-leucine methyl ester (5)



Molecular Weight = 300,40
Molecular Formula = $\text{C}_{15}\text{H}_{28}\text{N}_2\text{O}_4$

To the solution of N-(ethoxyoxalyl)-L-leucine methyl ester (**II**) (1.066 g, 4.346 mmol) in dry CH_2Cl_2 (40 ml) *n*-hexylamine (0.64 ml, 4.968 mmol) was added and the reaction mixture was stirred for 3 days at room temperature. The product was purified by crystallisation (CH_2Cl_2 -light petroleum) and by preparative TLC chromatography (EtOAc-light petroleum, 200:50). Yield: 70 %; $[\alpha]_{\text{D}} = -12$ (c 1, CH_2Cl_2); ^1H NMR ($\text{DMSO}-d_6$): δ 8.93 (1 H, d, $J = 8.4$, NH_{Leu}), 8.76 (1 H, t, $J = 5.8$, NH_{hexyl}), 4.36 (1H, ddd, $J = 3.5$, $J = 8.4$, $J = 10.6$, $\text{CH}_{\alpha, \text{Leu}}$), 3.63 (3 H, s, OCH_3), 3.16 – 3.08 (2 H, m, CH_2N), 1.86 – 1.77, 1.59 – 1.50, 1.49 – 1.41, 1.31- 1.18 (1 H, 2 H, 2 H, and 4 H, 4 m, $\text{CH}_{\beta, \text{Leu}}$, $\text{CH}_2 (\beta, \text{Leu})$, $\text{CH}_2, \text{hexyl}$), 0.88, 0.84 (6 H, 2 d, $J = 5.9$, $\text{CH}_3 (\gamma, \text{Leu})$), 0.86 (3 H, t, $J = 6.9$, $\text{CH}_3, \text{hexyl}$), ^{13}C NMR ($\text{DMSO}-d_6$): δ 172.4 (COOMe), 160.8, 159.8 (CON), 52.5 (OCH_3), 50.9 ($\text{CH}_{\alpha, \text{Leu}}$), 39.4, 39.3 ($\text{CH}_2 (\beta, \text{Leu})$, CH_2N), 31.4, 29.1, 26.4, 22.5 ($\text{CH}_2, \text{hexyl}$), 24.7, 23.3, 21.5 ($\text{CH}_{\gamma, \text{Leu}}$, $\text{CH}_3, \alpha, \text{Leu}$), 14.3 $\text{CH}_3, \text{hexyl}$); IR: 3294, 1745, 1687, 1653, 1525.

Example 3: N-hexyloxalamido-L-phenylalanine methyl ester (6)



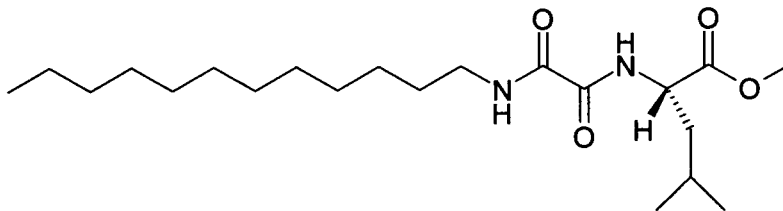
Molecular Weight = 334.42
Molecular Formula = $\text{C}_{18}\text{H}_{26}\text{N}_2\text{O}_4$

To the solution of N-(ethoxyoxalyl)-L-phenylalanine methyl ester (**II**) (4.900 g, 17.544. mmol) in dry CH_2Cl_2 (60 ml) *n*-hexylamine (2.32 ml, 17.562 mmol) was added and the reaction mixture was stirred for 3 days at room temperature. The product was purified by preparative TLC chromatography (EtOAc-light petroleum, 200:50). Yield: 75 %; ^1H NMR ($\text{DMSO}-d_6$): δ 8.92 (1 H, d, $J = 8.5$, NH_{Phe}), 8.71 (1 H, t, $J = 6.0$, NH_{hexyl}), 7.32-7.10 (5 H, m, CH_{arom}), 4.57 (1H, dt, $J = 5.9$, $J = 8.5$, $\text{CH}_{\alpha, \text{Phe}}$), 3.64 (3 H, s, OCH_3), 3.17 – 3.01 (4 H, m, CH_2N and $\text{CH}_2 (\beta, \text{Phe})$), 1.49 – 1.32, 1.32 – 1.08 (2 H and 4 H, 2m, $\text{CH}_2, \text{hexyl}$), 0.85 (3 H, t, $J = 6.8$, $\text{CH}_3, \text{hexyl}$), ^{13}C NMR (CDCl_3): δ 170.7 (COOMe), 159.6, 159.0 (CON), 136.4 (C_{arom}), 129.2, 128.8, 127.3

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(CH_{arom}), 53.7, 52.5 (CH_α, Phe and OCH₃), 39.7, 37.8 (CH₂ (β, Phe, and CH₂N), 31.4, 29.1, 26.5, 22.5 (CH₂, hexyl), 14.0 CH₃, hexyl).

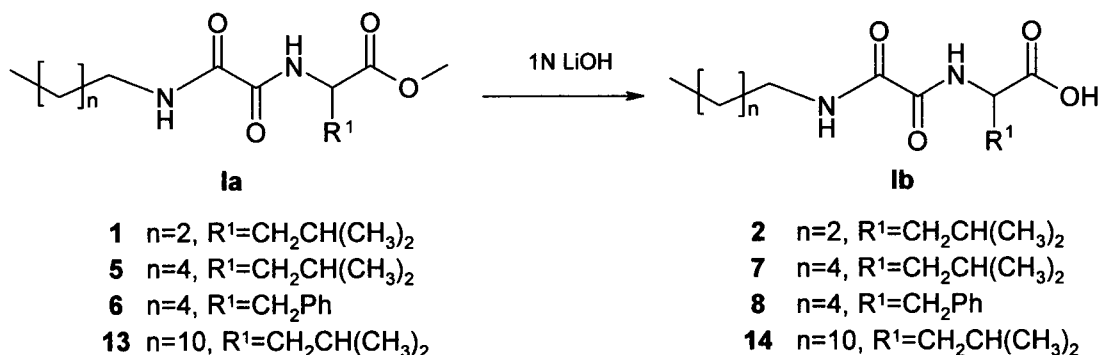
Example 4: N-dodecylloxalamido-L-leucine methyl ester (13)



Molecular Weight = 384.56
Molecular Formula = C₂₁H₄₀N₂O₄

To the solution of N-(ethoxyoxalyl)-L-leucine methyl ester (II) (0.372 ml, 1.517 mmol) in dry CH₂Cl₂ (20 ml), *n*-dodecylamine (0.35 ml, 1.522 mmol) was added and the reaction mixture was stirred for 3 days at room temperature. The product was purified by crystallisation (CH₂Cl₂-light petroleum) and by preparative TLC chromatography (EtOAc-light petroleum, 200:50). Yield: 70.3 %; [α]_D = - 9.5 (c 1, CH₂Cl₂); ¹H NMR (DMSO-d₆): δ 8.93 (1 H, d, *J* = 8.1, NH_{Leu}), 8.76 (1 H, t, *J* = 5.6, NH_{dodecyl}), 4.39 – 4.32 (1H, m, CH_α, Leu), 3.63 (3 H, s, OCH₃), 3.12 (2 H, q, *J* = 6.6, CH₂N), 1.89 – 1.75, 1.61 – 1.50, 1.49 – 1.39 (1 H, 2 H, 2 H, 18 H, 4 m, CH_γ, Leu, CH₂ (β, Leu), CH₂, dodecyl), 0.90 – 0.82 (9 H, m, CH₃(γ, Leu) and CH₃, dodecyl); ¹³C NMR (DMSO-d₆): δ 172.4 (COOMe), 160.8, 159.8 (CON), 52.5 (OCH₃), 50.9 (CH_α, Leu), 39.4, 39.3 (CH₂ (β, Leu), CH₂N_{dodecyl}), 31.8, 29.51, 29.47, 29.45, 29.42, 29.17, 29.14, 29.10, 26.8, 22.5 (CH₂, dodecyl), 24.8, 23.3, 21.5 (CH_γ, Leu), 14.4 CH₃, dodecyl); IR: 3300, 1750, 1656, 1524.

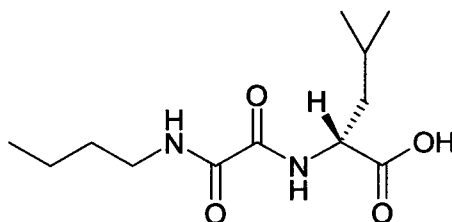
2. General procedure for preparation of N-alkyl, N'-L-leucyl-oxalamides (Ib)



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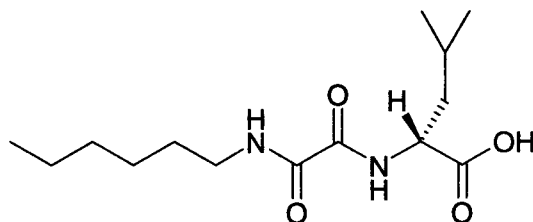
To the solution of ester **1a** in MeOH and CH₂Cl₂ 1N LiOH was added and the solution was stirred at room temperature overnight. The solvent was evaporated, the residue was dissolved in H₂O and the solution was acidified with 1N HCl. The product was extracted with EtOAc and organic layer was washed with H₂O and dried with Na₂SO₄. The solvent was evaporated at the reduced pressure to give solid product.

Example 5: N-butyloxalamido-L-leucine (2)



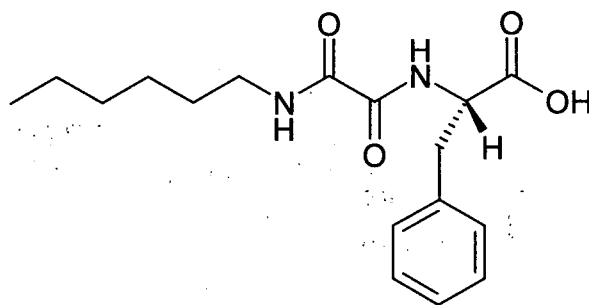
Molecular Weight = 258,32
Molecular Formula = C₁₂H₂₂N₂O₄

To the solution of N-butyloxalamido-L-leucine methyl ester (**1**) (0.247 g, 0.907 mmol) in MeOH (3 ml) and CH₂Cl₂ (1 ml) 1N LiOH (1.4 ml) was added and the solution was stirred at room temperature overnight. The solvent was evaporated, the residue was dissolved in H₂O and the solution was acidified with 1N HCl. The product was extracted with EtOAc, organic layer was washed with H₂O and dried with Na₂SO₄. The solvent was evaporated at the reduced pressure to give solid product. Yield: 92.2%; [α]_D = - 12 (c 1, CH₂Cl₂); ¹H NMR (DMSO-*d*₆): δ 12.83 (1 H, s br, COOH), 8.77 (1 H, t, *J* = 6.1, NH_{butyl}), 8.73 (1 H, d, *J* = 8.6, NH_{Leu}), 4.28 (1H, ddd, *J* = 10.5, *J* = 8.6, *J* = 3.4, CH_{α, Leu}), 3.14 (2 H, q, *J* = 6.7, CH₂N), 1.88 – 1.15 (7 H, m, CH_α, CH_{2(β, Leu)}, CH_{2, butyl}), 0.91 – 0.82 (9 H, m, CH_{3(σ, Leu)} and CH_{3, butyl}); ¹³C NMR (DMSO-*d*₆): δ 173.6 (COOH), 160.6, 160.0 (CON), 50.9 (CH_{α, Leu}), 39.6, 39.0 (CH_{2(β, Leu)}, CH_{2, butyl}), 31.3, 20.0 (CH_{2, butyl}), 24.9 (CH_γ), 23.4, 21.5 (CH_{3(σ, Leu)}), 14.1 (CH_{3, butyl}); IR: 3314, 1745, 1656, 1519.

Example 6: N-hexyloxalamido-L-leucine (7)

Molecular Weight = 286,37
Molecular Formula = C₁₄H₂₆N₂O₄

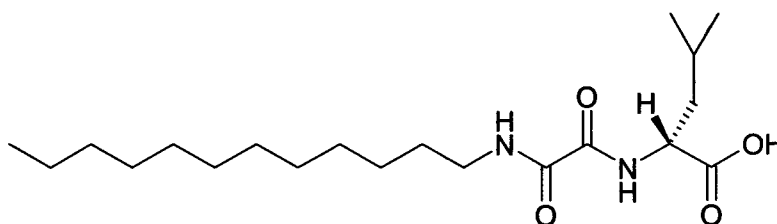
To the solution of N-hexyloxalamido-L-leucine methyl ester (5) (0.378 g, 1.258 mmol) in MeOH (2.5 ml) and CH₂Cl₂ (1 ml) 1N LiOH (1.9 ml) was added and the solution was stirred at room temperature overnight. The solvent was evaporated, the residue was dissolved in H₂O and the solution was acidified with 1N HCl. The product was extracted with EtOAc, organic layer was washed with H₂O and dried with Na₂SO₄. The solvent was evaporated at the reduced pressure to give solid product. Yield; 99 %: [α]_D = -14 (c 1, CH₂Cl₂); ¹H NMR (DMSO-*d*₆): δ 12.79 (1 H, s br, COOH), 8.78 (1 H, t, *J* = 6.1, NH_{hexyl}), 8.73 (1 H, d, *J* = 8.6 NH_{Leu}), 4.28 (1H, ddd, *J* = 10.4, *J* = 8.6, *J* = 3.3, CH_{α, Leu}), 3.17 – 3.07 (2 H, m, CH₂N), 1.88 – 1.12 (11 H, m, CH_{α, Leu}, CH_{2(β, Leu)}, CH_{2, hexyl}), 0.91 – 0.82 (9 H, m, CH_{3(σ, Leu)} and CH_{3, hexyl}); ¹³C NMR (DMSO-*d*₆): δ 173.6 (COOH), 160.6, 160.0 (CON), 50.9 (CH_α), 39.6, 39.3 (CH_{2(β, Leu)}, CH_{2, butyl}), 31.4, 29.1, 26.5, 22.5 (CH_{2, hexyl}), 24.9 (CH_γ), 23.4, 21.5 (CH_{3(σ, Leu)}), 14.3 (CH_{3, hexyl}); IR: 3297, 1725, 1658, 1523.

Example 7: N-hexyloxalamido-L-phenylalanine (8)

Molecular Weight = 320,39
Molecular Formula = C₁₇H₂₄N₂O₄

To the solution of N-hexyloxalamido-L-phenylalanine methyl ester (**6**) (0.043 g, 0.129. mmol) in MeOH (1 ml) and CH₂Cl₂ (0.2 ml) 1N LiOH (0.2 ml) was added and the solution was stirred at room temperature overnight. The solvent was evaporated, the residue was dissolved in H₂O and the solution was acidified with 1N HCl. The product was extracted with EtOAc, organic layer was washed with H₂O and dried with Na₂SO₄. The solvent was evaporated at the reduced pressure to give solid product. Yield: 60.6%; ¹H NMR (DMSO-d₆): δ 13.0 (1H, s br, COOH), 8.69 (1 H, t, *J* = 5.9, NH_{hexyl}), 8.66 (1 H, d, *J* = 8.6, NH_{Phe}), 7.82-7.17 (5 H, m, CH_{arom}), 4.57 (1H, dt, *J* = 4.7, *J* = 8.6, CH_α Phe), 3.19 – 3.03 (4 H, m, CH₂N and CH₂ (β, Phe)), 1.48 – 1.36, 1.30 – 1.14 (2 H and 6 H, 2m, CH₂, hexyl), 0.85 (3 H, t, *J* = 6.8, CH₃, hexyl), ¹³C NMR (DMSO-d₆): δ 172.0 (COOH), 159.8, 159.3 (CON), 137.5 (C_{arom}), 129.0, 128.2, 126.4 (CH_{arom}), 53.5 (CH_α Phe), 38.8, 35.7 (CH₂ (β, Phe) and CH₂N), 30.8, 28.5, 25.9, 22.0 (CH₂, hexyl), 13.8 CH₃, hexyl).

Example 8: N-dodecyloxalamido-L-leucine (15)



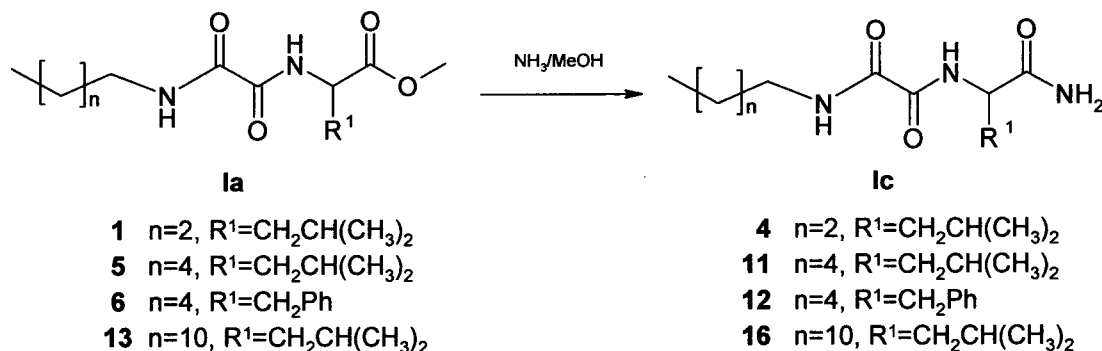
Molecular Weight =370.54
Molecular Formula =C₂₀H₃₈N₂O₄

To the solution of N-dodecyloxalamido-L-leucine methyl ester (**13**) (0.345 g, 0.897 mmol) in MeOH (2.5 ml) and CH₂Cl₂ (0.7 ml) 1N LiOH (1.4 ml) was added and the solution was stirred at room temperature overnight. The solvent was evaporated, the residue was dissolved in H₂O and the solution was acidified with 1N HCl. The product was extracted with EtOAc, organic layer was washed with H₂O and dried with Na₂SO₄. The solvent was evaporated at the reduced pressure to give solid product. Yield: 99%; [α]_D = -9 (c 1, CH₂Cl₂); ¹H NMR (DMSO-d₆): δ 12.79 (1 H, s br, COOH), 8.77 (1 H, t, *J* = 6.0, NH_{dodecyl}), 8.71 (1 H, d, *J* = 8.6, NH_{Leu}), 4.27 (1H, ddd, *J* = 10.0, *J* = 8.6, *J* = 3.5, CH_α Leu), 3.12 (2 H, q, *J* = 6.7, CH₂N), 1.87 – 1.12 (23 H, m, CH_α Leu, CH₂(β, Leu), CH₂, dodecyl), 0.90 – 0.82 (9 H, m, CH₃(γ, Leu) and CH₃, dodecyl); ¹³C NMR (DMSO-d₆): δ 173.6 (COOH), 160.6, 159.9 (CON), 50.9 (CH_α Leu), 39.6, 39.3 (CH₂(β, Leu), CH₂, dodecyl), 31.8, 29.51, 29.48, 29.46, 29.43, 29.18, 29.15, 29.12, 26.8, 22.6

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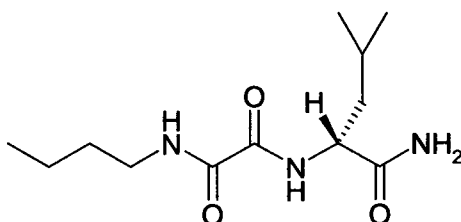
(CH₂, dodecyl), 24.9 (CH_α), 23.4, 21.5 (CH₃(γ, Leu)), 14.4 (CH₃, dodecyl); IR: 3313, 1745, 1656, 1519.

3. General procedure for preparation of *N* – alkyl, *N'* – *L* – leucylamide – oxalamides (Ic)



The solution of ester (**Ia**) in CH₂Cl₂ and conc. NH₃/MeOH was kept for 7 days at 4-8 °C. The precipitate was filtered off, washed with MeOH and dried under reduced pressure or the solvent was evaporated at reduced pressure.

Example 9: N-Butyloxalamido-L-leucylamide (4)



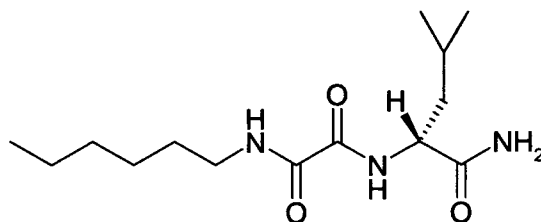
Molecular Weight = 272,35
Molecular Formula = C₁₃H₂₄N₂O₄

The solution of N-butyloxalamido-L-leucine methyl ester (**1**) (0.380 g, 1.395 mmol) in CH₂Cl₂ (1 ml) and conc. NH₃/MeOH (35 ml) was kept for 7 days at 4 -8 °C. The precipitate was filtered off, washed with MeOH. Yield: 89 %; ¹H NMR (DMSO-d₆): δ 8.78 (1 H, t, *J* = 6.0, NH_{butyl}), 8.36 (1 H, d, *J* = 9.0, NH_{Leu}), 7.48, 7.11 (2 x 1 H, 2 s, CONH₂), 4.28 (1H, dt, *J* = 9.3, *J* = 4.0, CH_{α, Leu}), 3.21 – 3.04 (2 H, m, CH₂N), 1.72 – 1.14 (7 H, m, CH_α, CH₂(β, Leu), CH₂, butyl), 0.96 – 0.71 (9 H, m, CH₃(γ, Leu) and CH₃, butyl); ¹³C NMR (DMSO-d₆): δ 173.6 (CONH₂),

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160.04, 160.01 (CON), 51.8 (CH_α), 41.4, 39.0 (CH_{2(β, Leu)}, CH_{2, butyl}), 31.2, 20.0 (CH_{2, butyl}), 24.8 (CH_{γ, Leu}), 23.5, 21.9 (CH_{3(δ, Leu)}), 14.1 (CH_{2, butyl}); IR: 3437, 3385, 3288, 3207, 1687, 1652, 1615, 1519.

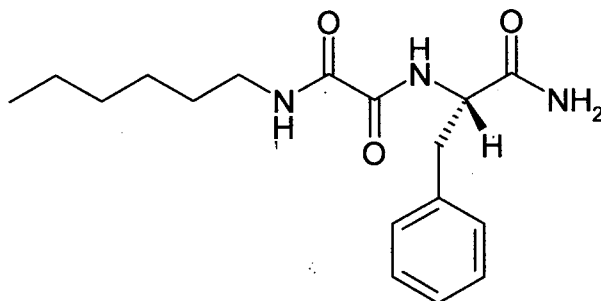
Example 10: N-hexyloxalamido-L-leucylamide (11)



Molecular Weight = 285,39
Molecular Formula = C₁₄H₂₇N₃O₃

The solution of N-hexyloxalamido-L-leucine methyl ester (**5**) (0.375 g, 1.248 mmol) in CH₂Cl₂ (1 ml) and conc. NH₃/MeOH (30 ml) was kept for 7 days at 4 -8 °C. The precipitate was filtered off, washed with MeOH. Yield: 86.2 %; ¹H NMR (DMSO-d₆): δ 8.78 (1 H, t, *J* = 6.0, NH_{hexyl}), 8.37 (1 H, d, *J* = 9.0, NH_{Leu}), 7.48, 7.11 (2 x 1 H, 2 s, CONH₂), 4.28 (1H, dt, *J* = 9.2, *J* = 3.9, CH_{α, Leu}), 3.17 – 3.07 (2 H, m, CH₂N), 1.72 – 1.14 (11 H, m, CH_α, CH_{2(β, Leu)}, CH_{2, hexyl}), 0.92 – 0.80 (9 H, m, CH_{3(γ, Leu)} and CH_{3, hexyl}); ¹³C NMR (DMSO-d₆): δ 173.6 (CONH₂), 160.05, 159.99 (CON), 51.8 (CH_{α, Leu}), 41.4, 39.4 (CH_{2(β, Leu)}, CH_{2, hexyl}), 31.4, 29.1, 26.4, 22.5 (CH_{2, hexyl}), 24.8 (CH_{γ, Leu}), 23.5, 21.9 (CH_{3(δ, Leu)}), 14.3 (CH_{2, hexyl}); IR: 3432, 3385, 3287, 3208, 1685, 1652, 1515.

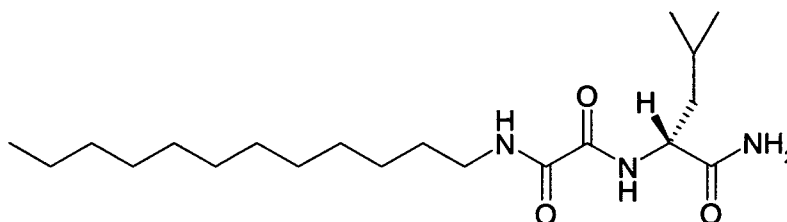
Example 11: N-hexyloxalamido-L-phenylalaninamide (12)



Molecular Weight = 319,41
Molecular Formula = C₁₇H₂₅N₃O₃

The solution of N-hexyloxalamido-L-phenylalanine methyl ester (**6**) (0.023 g, 0.0658 mmol) in CH_2Cl_2 (0.5 ml) and conc. NH_3/MeOH (5 ml) was kept for 7 days at 4 -8 °C. The precipitate was filtered off, washed with MeOH. Yield: 80.9 %; ^1H NMR (DMSO-d_6): δ 8.67 8.67 (1 H, t, $J = 5.9$, NH_{hexyl}), 8.37 (1 H, d, $J = 8.7$, NH_{Phe}), 7.58, 7.25 (2H, 2 s, CONH_2), 7.28-7.13 (5 H, m, CH_{arom}), 4.47 (1H, dt, $J = 4.5$, $J = 8.9$, $\text{CH}_{\alpha, \text{Phe}}$), 3.13 – 3.03 (4 H, m, CH_2N and $\text{CH}_{2\text{A}} (\beta, \text{Phe})$), 2.99 (1H, dd, $J = 9.2$, $J = 13.7$, $\text{CH}_{2\text{B}} (\beta, \text{Phe})$), 1.46 – 1.37, 1.29 – 1.15 (2 H and 6 H, 2m, CH_2 , hexyl), 0.85 (3 H, t, $J = 6.9$, CH_3 , hexyl), ^{13}C NMR (DMSO-d_6): δ 171.9 (CONH_2), 159.4, 159.3 (CON), 137.5 (C_{arom}), 129.1, 128.0, 126.3 (CH_{arom}), 54.0 ($\text{CH}_{\alpha, \text{Phe}}$), 38.8, 35.2 ($\text{CH}_2 (\beta, \text{Phe})$ and CH_2N), 30.8, 28.6, 25.9, 22.0 (CH_2 , hexyl), 13.8 CH_3 , hexyl).

Example 12: N-dodecyloxalamido-L-leucylamide (**16**)



Molecular Weight = 369,55
Molecular Formula = $\text{C}_{20}\text{H}_{39}\text{N}_3\text{O}_3$

The solution of N-dodecyloxalamido-L-leucine methyl ester (**13**) (0.375 g, 0.975 mmol) in CH_2Cl_2 (1 ml) and conc. NH_3/MeOH (25 ml) was kept for 7 days at 4 -8 °C. The precipitate was filtered off, washed with MeOH. Yield 81.3%; ^1H NMR (DMSO-d_6): δ 8.78 (1 H, t, $J = 5.9$, $\text{NH}_{\text{dodecyl}}$), 8.36 (1 H, d, $J = 9.1$, NH_{Leu}), 7.49, 7.11 (2 x 1 H, 2 s, CONH_2), 4.28 (1H, dt, $J = 9.1$, $J = 3.8$, $\text{CH}_{\alpha, \text{Leu}}$), 3.18 – 3.02 (2 H, m, CH_2N), 1.72 – 1.09 (23 H, m, CH_{α} , $\text{CH}_2 (\beta, \text{Leu})$, CH_2 , decyl), 0.91 – 0.81 (9 H, m, $\text{CH}_3 (\gamma, \text{Leu})$ and CH_3 , dodecyl); ^{13}C NMR (DMSO-d_6): δ 173.6 (CONH_2), 160.03, 159.99 (CON), 51.8 (CH_{α}), 41.4, 39.4 ($\text{CH}_2 (\beta, \text{Leu})$, CH_2 , dodecyl), 31.7, 29.50, 29.47, 29.45, 29.42, 29.17, 29.14, 29.11, 26.8, 22.6 (CH_2 , decyl), 24.8 ($\text{CH}_{\gamma, \text{Leu}}$), 23.4, 22.0 ($\text{CH}_3 (\delta, \text{Leu})$), 14.4 (CH_2 , dodecyl); IR: 3389, 3291, 3198, 1686, 1653, 1511.

Testing of gelation properties of organogelators

The gelation properties of the compounds of the invention were tested and minimal gelation concentrations (m.g.c.) were determined. The results are shown in the Table 1. and Table 2.

Procedure

The experiments were performed by dissolution of the weighted amount of organogelators (10 mg) in a measured volume of selected solvent. In the first case, when pure solvent was used, heating of the sample was necessary to produce solution. By cooling the solution to room temperature gel was obtained. The portions of solvent were repeatedly added until the gel disappeared.

In the second case, when mixtures of solvent were used, the sample (10 mg) was dissolved in first solvent and measured volume (0.05 - 0.25 ml) of second solvent was added. Gel was formed by heating of this mixture followed by cooling. The portions of second solvent were repeatedly added until the gel disappeared. For very efficient organogelators m.g.c. was determined on amount of gelators ca. 2 mg.

In order to compare the gelation properties and chemical structures of the compounds, we tested their gelation properties in several organic solvents of different polarity, water and commercial fuels.

Table 1. Maximum solvent volume (in μl) that can be gelated by the specific amount (10 mg) of organogelators (amides and salts of carboxylic acids)

	C4-NH ₂ 4	C6-NH ₂ 11	C6-Phe-NH ₂ 12	C12-NH ₂ 16	C4-O ⁻ Na ⁺ 3	C6-O ⁻ Na ⁺ 9	C12-O ⁻ Na ⁺ 15
petrol	45800	79200	7000	10300	NG	NG	NG
diesel	36100	67200	10000	42400	NG	NG	NG
H ₂ O	6 850	NG	NT	NG	NG	NG	NG
DMSO	NG	NG	NG	NG	NG	NG	NG
DMSO-H ₂ O	1500+750	2150+1650	7500+20500	17100+20700	NG	NG	NG
DMF-H ₂ O	600+500	1150+2550	3450+3450	10350+13300	NG	NG	NG
EtOH	250	NG	NG	1050	NG	NG	NG

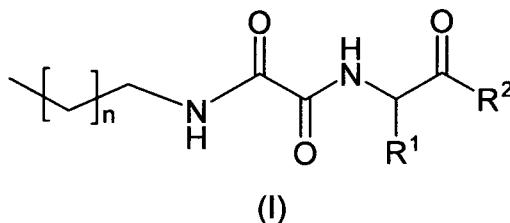
THF	100	NG	500	150	NG	NG	NG
EtOAc	200	NG	1000	NG	NG	NG	NG
acetone	NG	NG	NG	NG	NG	NG	NG
CH ₂ Cl ₂ CH ₂ Cl ₂ - DMSO	1050	750	NT 400+40	750	NG	NG	NG
CH ₃ CN	NG	NG	NG	1450	NG	NG	NG
toluene	24950	16200	4000	15200	NG	NG	NG
p-xylene	33600	68000	7000	5250	NG	NG	NG
decaline	37400	106250	26000	24100	NG	NG	NG
tetraline	500	150	500	300	NG	NG	NG

Table 2. Maximum solvent volume (in μ l) that can be gelled by the specific amount (10 mg) of organogelators (esters and carboxylic acids)

	C4- OMe 1	C6- OMe 5	C6-Phe- OMe 6	C12-OMe 13	C4- OH 2	C6-OH 7	C6-Phe- OH 8	C12- OH 14
petrol	NG	NG	400	NG	NG	NG	6500	NG
diesel	NG	NG	5750	NG	NG	NG	5750	NG
H ₂ O	NG	NG	NG	NG	NG	NG	10750	NG
DMSO	NG	NG	NG	NG	NG	NG	NG	550
DMSO-H ₂ O	NG	NG	9430+11750	2720+3050	NG	250+450	170+11750	NG
DMF-H ₂ O	NG	NG	5150+3350	14100+20700	NG	NG	550+7500	NG
EtOH	NG	NG	NG	NG	NG	NG	NG	NG
THF	NG	NG	NG	NG	NG	NG	NG	NG
EtOAc EtOAc+hexane	NG	NG	NG	NG	NG	NG	NG 200+1500	NG
acetone	NG	NG	NG	NG	NG	NG	NG	NG
CH ₂ Cl ₂ CH ₂ Cl ₂ +hexane	NG	NG	NG	NG	NG	NG	NG 2750+8750	NG
CH ₃ CN	NG	NG	NG	NG	NG	NG	NG	NG
toluene	NG	NG	NG	NG	NG	NG	2500	NG
p-xylene	NG	NG	100	NG	NG	NG	4250	NG
decaline	NG	NG	4300	NG	NG	NG	6950	NG
tetraline	NG	NG	NG	NG	NG	NG	NG	NG

Claims

1. Compounds of general formula (I):



characterised by:

R^1 is hydrogen, phenyl, C_{1-6} alkyl which can be further substituted with R^3

R^3 is 5- or 6- membered aromatic or heteroaromatic ring system which can be further substituted with benzyl or hydroxyl groups;

R^2 is OR^4 , NHR^4

R^4 is hydrogen, C_{1-6} alkyl or benzyl

n is an integer from 1 to 12

or the salts thereof.

2. Compounds according to the claim 1, characterised by that independently or in any combination:

R^1 is isobutyl

R^2 is OR^4 , OH, NHR^4

n is an integer from 1 to 10

or the salts thereof.

3. Compounds of general formula (I) according to the claims 1 to 2 characterised by that R^2 is NH_2 and n is 2 to 10.

4. Compounds according to the claim 2 characterised by that the group R^2 is OR^4 .

5. Compounds according to the claim 1 or claim 2 characterised by that the group R^2 is

OH.

6 Compounds according to the claim 1 or claim 2 characterised by that the group R^2 is NHR^3 .

7. Compounds according to any of the claims 1 to 6 characterised by that the group R^1 is isobutyl group.

8. Compounds according to any of the claims 1 to 7 characterised by they are sodium salts.

9. Compounds according to any of the claims 1 to 8 characterised by they are ammonium salts or alkyl (benzyl) amine salts.

10. Compounds according to any of the claims 1 to 9 characterised by they are selected from:

N-butyloxalamido-L-leucine methyl ester;

N-butyloxalamido-L-leucine

N-butyloxalamido-L-leucine sodium salt

N-butyloxalamido-L-leucinamide

N-hexyloxalamido-L-leucine methyl ester

N-hexyloxalamido-L-phenylalanine methyl ester

N-hexyloxalamido-L-leucine

N-hexyloxalamido-L-phenylalanine

N-hexyloxalamido-L-leucine sodium salt

N-hexyloxalamido-L-phenylalanine sodium salt

N-hexyloxalamido-L-leucinamide

N-hexyloxalamido-L-phenylalaninamide

N-dodecyloxalamido-L-leucine methyl ester

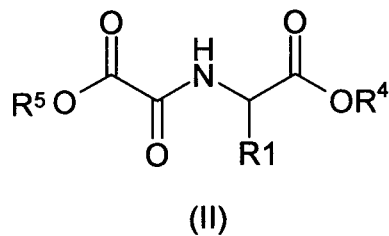
N-dodecyloxalamido-L-leucine

N-dodecyloxalamido-L-leucine sodium salt

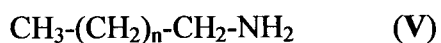
N-dodecyloxalamido-L-leucinamide

11. Method for the preparation of compounds of general formula (I) according to the claim 1 characterised by that comprises:

reaction of compounds with general formula (II)

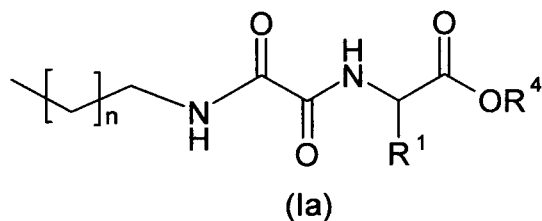


with alkylamines of general formula (V):

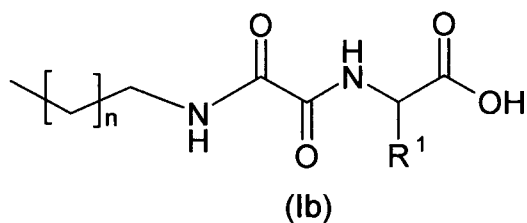


wherein R^1 , R^4 , R^5 and n are defined in the claim 1

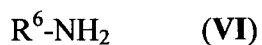
for the preparation of compounds of general formula (Ia),



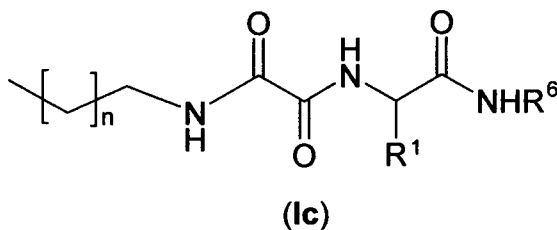
reaction of hydrolysis or hydrogenolysis of compounds of general formula (Ia) for the preparation of the compounds (Ib)



or reaction of the compounds (Ia) with the compounds (VI)



for the preparation of the compounds (Ic).



12. A gel characterised by that comprises any of the compounds defined in the claim 1.
13. A gel according to the claim 12 characterised by that it is prepared by dissolution of a compound in solvents.
14. Solvents according to the claim 13 characterised by that the solvents can be:
 - a) commercial fuels such as petrol or diesel;
 - b) organic solvents such as DMSO, DMF, EtOH, CH_3CN , THF, CH_2Cl_2 , acetone, toluene, *p*-xylene, tetraline, decaline;
 - b) water and mixtures of water with organic solvents such as DMSO or DMF.
15. Use of the compounds according to the claim 1 for the preparation of gels.
16. Use according to the claim 16 for the gelation of commercial fuels.