

Method for producing a multi-phase polymer system

The present invention relates to the technical field of polymerisation methods, which are used in a wide variety of forms, both in terms of the apparatus employed and the method conditions, for the targeted production of plastics with specific property profiles. More specifically, the invention proposes a method for producing a multi-phase polymer system comprising two polymerisation stages, which initially leads to a first polymerisate, in the presence of which the polymerisation is then continued within a closed casing. The most important application background for the multi-phase polymer systems produced in this way is pressure-sensitive adhesives for adhesive tapes, which are used in various technical fields to create both temporary and permanent material bonds.

For industrial adhesive tape applications, polymers based on the polymerisation of monomers with functionalised vinyl groups are frequently used. A well-known example of such polymers are poly(meth)acrylates. Poly(meth)acrylates offer various advantages over other plastics used as a basis for adhesive applications. For example, they are highly resistant to high-energy radiation and oxidising substances. In contrast to poly(meth)acrylates, synthetic rubbers and natural rubbers often contain C-C double bonds, which make these polymers - and consequently the adhesive components based on them - vulnerable to, for example, UV radiation, oxygen and ozone. Furthermore, poly(meth)acrylate-based plastic formulations can be transparent and are suitable for use across a wide temperature range.

In the past, polymers based on monomers with functionalised vinyl groups were regularly produced from solution by means of free-radical polymerisation. The polymers are coated onto a substrate from solution using a coating bar or a similar tool and then dried. To increase cohesion, the polymers are usually cross-linked, which is typically achieved either thermally, by UV radiation or by electron beam irradiation. The solvent process is, on the whole, economically rather inefficient and environmentally harmful, as the solvents are either not recovered and thus pollute the environment, or must be recovered at high energy cost.

Furthermore, quality issues regularly arise. For instance, adhesive tapes with a higher coating weight based on the polymers described can often scarcely be produced without bubbles.

To mitigate these drawbacks, attempts have been made to apply the polymers from the melt to the substrate in a so-called hot-melt process. However, this technique also has its

limitations. As the polymers are still largely produced from solution, the solvent must be removed from the melt prior to processing. This is done, for example, in a dry extruder. The drying process involves high temperatures and shear forces acting on the polymers, meaning that higher-molecular-weight polymers in particular suffer relatively severe damage during processing. For example, gelation may occur, with the result that the polymers can no longer be coated at all, or only with great difficulty. Molecular weight degradation can lead to a significant enrichment of the low-molecular-weight fractions; these then act as lubricants and cause undesirable properties such as a lack of shear strength. In the prior art, various approaches to processing poly(meth)acrylates into pressure-sensitive adhesives by means of hot-melt processes are described in particular.

For example, WO 00/06637 A1 discloses a method for producing an article, wherein the method comprises:

- a) melt-mixing a polymer composition and a plurality of microspheres, at least one of which is an expandable polymeric microsphere, under process conditions comprising temperature and shear rate and selected such that an expandable, extrudable composition is formed;
- b) extruding the expandable, extrudable composition through a die to form a foam having a smooth surface with a Ra value of less than 75 μm ;
- c) at least partially expanding one or more of the expandable polymeric microspheres before the polymer composition leaves the die.

WO 2010/112346 A1 relates to a method for producing a foamed composition containing thermally sensitive substances, in which the composition is foamed in a first step at a first temperature and the thermally sensitive substances, which may for example be thermal cross-linkers, are added to the composition in a subsequent step at a second, lower temperature than the first temperature.

WO 2008/122489 A1 describes a method for the thermal cross-linking of polyacrylates with functional groups capable of undergoing cross-linking reactions with epoxy groups, wherein a cross-linker-accelerator system comprising epoxy-group-containing substances (cross-linker) and at least one substance (accelerator) which acts as an accelerator for the cross-linking reaction at a temperature below the melting temperature of the polyacrylate are used.

In the prior art, attempts have also been made to combine the polymerisation and processing of acrylate polymers in a continuous process. WO 2004/101627 A1 describes a method for the continuous polymerisation of acrylic monomers to form polyacrylates in the presence of polymerisation-controller substances, wherein at least one polymerisation step is carried out within at least one reaction extruder. The disadvantages of such methods have been found to include, amongst other things, low throughput, high capital costs and limited flexibility when changing formulations.

Another factor that influences the properties of polymers based on monomers with functionalised vinyl groups is the polymer architecture. In the distant past, work was mostly carried out with random copolymers, in which the monomers are randomly distributed along the polymer chain. In recent decades, the focus has shifted to polymers with controlled structures, such as block copolymers or copolymers with a monomer distribution gradient. The aim is therefore to specifically control the property profile of the polymers and the resulting plastic formulations via the distribution or arrangement of the monomers. In this way, it is possible to achieve properties that are inherently contradictory using one and the same polymer base. In this context, methods of controlled (meth)acrylate polymerisation have been described in particular in the prior art.

WO 2004/101627 A1 describes a method for the continuous polymerisation of acrylic monomers to polyacrylates in the presence of polymerisation-controller substances, wherein at least one polymerisation step is carried out within at least one reaction extruder.

WO 2013/072120 A1 relates to a method for producing acrylate-based polymers, in which a controlled radical polymerisation reaction of a reaction mixture comprising at least one acrylate-based monomer is first carried out in the presence of at least one controller substance having at least one functional group $S-C=X$, where $X = S, O$ or N , wherein the controller substance is selected from the group comprising dithioesters, dithiocarbonates, dithiocarbamates, trithiocarbonates, imido-dithio-carbonates and xanthates ('RAFT controllers'),

wherein the majority of the controller substance molecules are incorporated into the resulting polymer chains, whereupon the respective polymer chain exhibits the functional group of the incorporated controller,

characterised in that

at least one chemical compound having at least two conjugated double bonds (hereinafter referred to as a “conjugated diene”) is brought into contact with the polymers thus obtained, so that hetero-Diels-Alder reactions are induced between the C=X double bonds of the functional groups S-C=X incorporated into the polymer chains and the conjugated double bonds of the conjugated diene.

EP 2 607 394 A1 discloses a method for producing pressure-sensitive adhesives, in which a controlled radical polymerisation reaction of a reaction mixture comprising one or more monomers is carried out in the presence of at least one radical initiator to produce at least one polymer, characterised in that

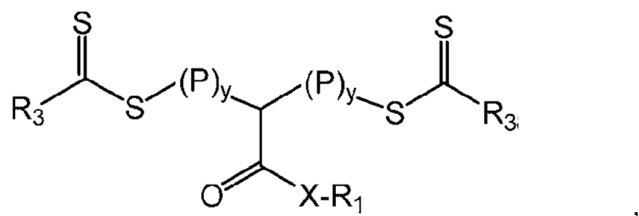
at least one cyclic 1,4-diradical is used as the radical initiator, which is generated by means of a cyclisation reaction from a compound having at least two unsaturated C-C bonds (“diradical-forming compound”),

wherein the polymerisation reaction is carried out in the presence of at least one controller substance (“polymerisation controller”).

WO 2013/055978 A1 describes acrylic copolymers which involve the controlled placement of specific functional groups within the polymer structure. The copolymers contain at least two reactive segments and are produced via a controlled radical polymerisation method. The copolymers are suitable for use in the production of adhesives and elastomers.

WO 2018/118905 A1 describes a cross-linkable composition comprising

- a) a polymeric material of the formula



wherein

R₁ is hydrogen, alkyl, fluorinated alkyl, aryl, aralkyl or substituted aralkyl;

X is oxygen or $-NR_2$, wherein R_2 is hydrogen, alkyl, fluorinated alkyl, aryl, aralkyl or substituted aryl;

each R_3 is an alkoxy, fluorinated alkoxy or $-N(R_4)_2$, wherein each R_4 is alkyl or fluorinated alkyl or, together with the nitrogen to which they are both attached, forms a heterocyclic ring;

each P is a polymeric block comprising a polymerised product of a first monomer composition comprising at least one monomer having a single ethylenically unsaturated group;

y is an integer from 1 to 5; and

- b) a second monomer composition which is miscible with the polymeric material of the formula and comprises a cross-linking monomer having at least two ethylenically unsaturated groups.

WO 2018/178829 A1 describes controlled radical initiators, reaction mixtures containing these and various ethylenically unsaturated monomers; polymeric materials formed from these reaction mixtures; cross-linkable compositions containing the polymeric materials; and cross-linked compositions formed therefrom. The controlled radical initiators are bis-dithiocarbamate and bis-dithiocarbonate compounds having a single carbon atom between the two dithiocarbamate or dithiocarbonate groups.

There is a continuing need for methods for producing polymers based on monomers with functionalised vinyl groups, which can be carried out efficiently from both an economic and environmental perspective and result in polymers with a controlled architecture that are suitable for straightforward further processing. However, the problem remains that polymerisation is easiest to manage in solution, but the solvent must be removed for the further processing of the polymers. This is regularly associated with high temperatures, for example in the feed preheating unit of concentration extruders. In polymers produced by controlled polymerisation in particular, this causes thermal degradation of the controller substances used, which on the one hand often results in a strong unpleasant odour and, on the other hand, depending on the position of the controller substances in the polymer, may also lead to polymer degradation and changes in the mechanical properties of the polymer systems.

One object of the invention was to avoid the problems known from the prior art and to provide a method for producing polymers based on monomers with functionalised vinyl groups, by

means of which polymers with a controlled architecture can be produced efficiently and without the need for subsequent thermal treatment, such that the resulting polymer architecture and the mechanical properties of the polymers based thereon are retained during their further processing.

A further object of the invention was to provide such a method that offers a high degree of flexibility for changes to the polymer formulation and the desired polymer architecture.

A supplementary object of the invention was to make the resulting polymers available in an efficient manner such that they can be easily stored and transported whilst still being suitable for efficient further processing both from the melt and from solution.

A supplementary object of the invention was to provide polymers producible in this manner with good cohesion and good processability from the melt.

A further supplementary object of the invention was to provide polymers based on monomers with functionalised vinyl groups, which are particularly well suited for adhesive tape applications.

A first and general subject of the invention, by which these objects are achieved, is a method for producing a polymer system present in at least two phases, comprising the following steps:

- a) polymerising a monomer composition I, comprising in total at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers with at least one polymerisable vinyl group, in order to form a polymerisate A;
- b) polymerising a monomer composition II, comprising in total at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers with at least one polymerisable vinyl group, wherein at least 50 wt.% of the monomers in monomer composition II differ from those in monomer composition I, in the presence of polymerisate A in order to form polymerisate B, thereby obtaining a polymer system which is present in at least two phases and comprises polymerisates A and B;

characterised in that step b) is carried out in a closed casing.

Details and embodiments of the invention are described below. Such embodiments which are hereinafter designated in any form as preferred are combined in particularly preferred embodiments with features of other embodiments designated in any form as preferred.

Combinations of two or more of the embodiments described below as 'particularly preferred' in any form are therefore particularly preferred. Also preferred are embodiments in which a feature of an embodiment described as 'preferred' to any extent is combined with one or more further features of other embodiments described as 'preferred' to any extent.

Insofar as both specific quantities or proportions of an element and preferred embodiments of the element are disclosed below, the specific quantities or proportions of the preferred embodiments are also disclosed in particular. Furthermore, it is disclosed that, for the corresponding specific total quantities or total proportions of the elements, at least some of the elements may be in a preferred configuration and, in particular, that elements in a preferred configuration may in turn be present within the specific total quantities or total proportions.

The term 'polymer system' as used in the invention refers to both a single polymer and a mixture of two or more different polymers. In accordance with the understanding of those skilled in the art, a "polymer" or "a single polymer" is understood to mean not only a single macromolecule, but also a plurality of macromolecules originating from one and the same polymerisation process and exhibiting a specific molecular weight distribution amongst themselves.

The polymer system resulting from the method according to the invention exists in at least two phases. The person skilled in the art understands this to mean that one phase is rich in one component of the polymer system, for example polymerisate A, or consists substantially of that component, and the other phase is rich in another component, for example polymerisate B, or consists substantially of that component. The presence of small amounts of one component in the other, which does not prevent the formation of the multi-phase system, is regarded as negligible. If the polymer system resulting from the method according to the invention comprises more than two phases, the foregoing applies accordingly to all of these phases.

The phase separation is particularly preferably carried out in such a way that discrete regions ('domains'), which are rich in polymerisate A or polymerisate B – i.e. are substantially formed from polymerisate A or polymerisate B – are present in a continuous matrix which is rich in the respective other polymer – i.e. is substantially formed from the respective other polymerisate. A polymer system resulting from the method according to the invention is regarded as being present in at least two phases in particular if at least one of the criteria a) – c) listed below is met:

- a) Phase boundaries are discernible in an elevation profile analysis or an analysis of the Young's modulus of an *atomic force* microscopy (AFM) image of the polymer system.
- b) At least two independent glass transition temperatures are obtained from a differential scanning calorimetry (DSC) measurement performed on the polymer system.
- c) At least two $\tan \delta$ maxima are obtained from a dynamic mechanical analysis (DMA) performed on the polymer system.

According to the invention, the polymer system, which is present in at least two phases, also comprises a microphase-separated polymer system, i.e. a polymer system in which the discontinuous phase is present in a microscopically fine distribution.

The method according to the invention comprises a step a) in which the polymerisation of a monomer composition I, comprising in total at least 80 wt.%, based on the total weight of the monomer composition I, of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group, to form a polymerisate A.

Preferably, the monomer composition I comprises one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group in an amount of at least 85 wt.%, more preferably at least 90 wt.%, particularly preferably at least 95 wt.%, and in particular at least 98 wt.%. Most particularly preferably, the monomer composition I comprises exclusively one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group.

The polymerisation of step a) may in principle be carried out in any manner. Preferably, it is carried out in a reactor, in particular in a reactor designed for the processing of highly viscous masses, or in a closed casing; more preferably, it is carried out in a planetary mixer or in a closed casing.

It is also preferred that the polymerisation of step a) takes place in the absence of solvent. Solvent concentrations in minute amounts, e.g. resulting from production residues or lying in the range of background concentrations, are regarded as negligible in this context.

The polymerisate A resulting from the polymerisation of step a) is preferably a polymer block from which, or to which, at least one further, distinguishable polymer block can be polymerised.

The method according to the invention comprises a step b) in which, in the presence of polymerisate A, the polymerisation of a monomer composition II is carried out, which comprises, in total, at least 80 wt.%, based on the total weight of monomer composition I, of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group, wherein at least 50 wt.% of the monomers in monomer composition II differ from those in monomer composition I, to form a polymerisate B, yielding a polymer system present in at least two phases comprising polymerisates A and B.

Preferably, monomer composition II comprises one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group in an amount of at least 85 wt.%, more preferably at least 90 wt.%, particularly preferably at least 95 wt.%, and in particular at least 98 wt.%. Most particularly preferably, monomer composition II comprises exclusively one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group.

According to the invention, the polymerisation of step b) takes place in a closed casing. This is understood to mean any system whatsoever which, in its design, does not impede the course of the polymerisation and which spatially confines and completely surrounds the composition to be polymerised or undergoing polymerisation, and which at least limits, preferably prevents, the passage of at least air, water and the enclosed monomers. Preferably, the closed casing is elastic.

The material of the closed casing is preferably selected from the group consisting of ethylene-vinyl acetate copolymers, ethylene-acrylic acid copolymers, polyethylene, polypropylene, polybutadiene, ionomer films and mixtures thereof; more preferably, it is selected from polyethylene, ethylene-vinyl acetate copolymers and mixtures thereof. The thickness of the material of the closed casing is preferably 30 to 100 μm , more preferably 35 to 70 μm , and in particular 40 to 60 μm . The thickness of a closed sleeve filled with the materials to be polymerised is preferably 2 to 22 mm, more preferably 5 to 21 mm, and in particular 7 to 20 mm. The materials mentioned are not critical for the initiation of polymerisation by means of UV radiation.

The closed sleeve is produced, for example, by placing two film webs on top of one another and heat-sealing them along the long sides and the bottom edge. The monomer mixture is then filled in using a liquid moulding, filling and sealing machine. Subsequently, the film is sealed in sections through the liquid level, resulting in several filled closed casings, which are then separated from one another.

Preferably, the polymerisation of step b) takes place in the absence of solvent. Solvent concentrations in minute amounts, e.g. resulting from production residues or lying in the range of background concentrations, are regarded as negligible. It is particularly preferred that the method according to the invention is carried out entirely in the absence of solvents.

In a preferred embodiment of the method according to the invention, in step b) the polymerisate A is dissolved in the monomer composition II. In principle, this can be done in any manner known to the skilled person, for example by stirring polymerisate A into monomer composition II. Provided that the polymerisation of step a) has taken place in a closed casing and polymerisate A is thus present in the closed casing, the casing material

- is destroyed – preferably mechanically – at least to the extent that polymerisate A can be removed and introduced into monomer composition II, and
- subsequently separated from the polymerisate A or the mixture of polymerisate A and monomer composition II.

It goes without saying that, in this context, monomer composition II is not to be understood as a solvent in the above sense and therefore does not fall under the feature 'in the absence of solvents'.

It is preferred, provided that the polymerisation of step a) has taken place in a closed casing and polymerisate A is thus present in the closed casing, that

- the closed casing, together with the polymerisate A contained therein, is introduced into a mixing apparatus, more preferably into a kneader; wherein the mixing apparatus already contains the monomer composition II or the monomer composition II is subsequently introduced into the mixing apparatus;
- polymerisate A is brought into contact with monomer composition II whilst breaking down the closed casing, or, more preferably, dissolved in monomer composition II;
- the casing material is separated from the mixture of polymerisate A and monomer composition II by means of filtration; and

- the mixture of polymerisate A and monomer composition II is introduced into a further closed casing.

It is preferred that at least the polymerisation in step b) of the method according to the invention is a controlled radical polymerisation. A "controlled polymerisation" is understood to mean a polymerisation which allows at least control of the lengths of the resulting polymers or polymer blocks and of the polymer architecture. Examples of controlled radical polymerisations include atom transfer radical polymerisation (ATRP), nitroxide/TEMPO-controlled polymerisation and the RAFT process.

In one embodiment of the invention, at least the polymerisation in step b) of the method according to the invention is a RAFT polymerisation. 'RAFT polymerisation' stands for 'reversible addition-fragmentation chain transfer polymerisation'. This refers to a polymerisation in which the reaction is controlled by reversible chain transfer reactions. In this process, an active, growing radical chain adds to a specific controller substance, the so-called RAFT agent, which is already linked to another chain and is thus present as a higher-molecular-weight RAFT agent (macro- e RAFT agent). The addition of the active radical chain produces an intermediate which, due to its structure, has the potential to fragment in various directions. This process again yields a macro-RAFT agent and an active radical chain available for propagation, wherein the latter need not correspond to the previous active radical chain. In this way, the probability of propagation is distributed equally across all chains, resulting in a tight molecular weight distribution.

In this embodiment, the polymerisation in step a) is preferably a controlled radical polymerisation, and more preferably a RAFT polymerisation.

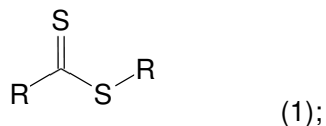
Irrespective of the specific embodiment, the polymerisation in step a) is preferably a radical polymerisation, i.e. it is initiated by a radical-forming substance and then proceeds via a radical propagating by monomer addition. In step a), radical formation and thus polymerisation is preferably initiated by radiation, particularly preferably by UV radiation. In step b) too, radical formation and thus polymerisation is preferably initiated by radiation, particularly preferably by UV radiation.

In accordance with the foregoing, the polymerisation in step b) is preferably carried out in the presence of at least one controller substance containing at least one sequence $-S-C(=X)-$, wherein X stands for S, O or NR' , wherein R' stands for an organic group. It is even more

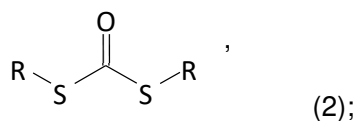
preferred that the polymerisation in step a) is also carried out in the presence of such a controller substance.

The controller substance containing at least one sequence -S-C(=X)- is preferably selected from the group consisting of

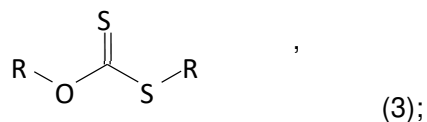
dithioesters, i.e. compounds of the general structure (1)



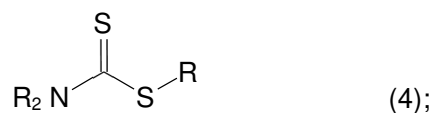
dithiocarbonates, i.e. compounds of the general structure (2)



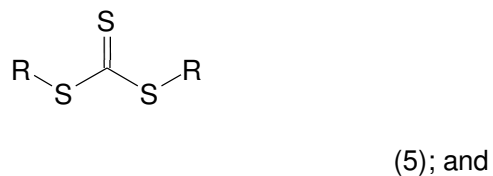
xanthates, i.e. compounds of the general structure (3)



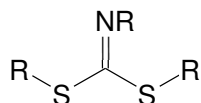
dithiocarbamates, i.e. compounds of the general structure (4)



trithiocarbonates, i.e. compounds of the general structure (5)



Imido-dithio-carbonates, i.e. compounds of the general structure (6)



(6),

wherein, in the general structures (1) to (6), the substituents R each represent, independently of one another, an organic or inorganic group, preferably an organic group. In particular, at least one substituent R in the general structures (1) to (6) comprises a polymer chain formed during the polymerisation in the relevant step.

Particularly preferred is the controller substance containing at least one sequence -S-C(=X)- selected from trithiocarbonates and xanthates; in the above sense, therefore, preferably from compounds according to general structures (3) and (5).

It is particularly preferred that the controller substance contains at least one sequence -S-C(X)- in its original state, i.e. not yet incorporating growing polymer chains, selected from the group consisting of dibenzyltrithiocarbonate, O-ethyl-S-(1-methyloxycarbonyl)ethyl xanthate, 1,4-phenylene bis(methylene) didodecyl tricarbonotrithioate, 2,2'-[carbonothioylbis(thio)]bis[2-methylpropanoic acid] and 4-cyano-4-(((dodecylthio)carbonothioyl)thio)pentanoic acid.

In one embodiment of the method according to the invention, at least the polymerisation in step (b) is carried out in the presence of at least one controller substance containing at least one sequence -S-C(=X)-, wherein X represents S, O or NR', where R' represents an organic group, and is initiated by at least one radical-forming photoinitiator present alongside the at least one controller substance. The term "photoinitiator" refers to a substance which, under the influence of light of specific wavelengths, usually at least under the action of UV radiation, and optionally also of UV radiation in the wavelength range of visible light (approx. 300–500 nm), forms radical species. The at least one radical-forming photoinitiator is preferably selected from the group consisting of 1-hydroxycyclohexylphenylketone and 2,2-dimethoxy-2-acetophenone.

It is also preferred that the polymerisation in step a) is carried out in the presence of at least one controller substance containing at least one sequence -S-C(=X)-, wherein X stands for S, O or NR', wherein R' stands for an organic group, and is initiated by at least one radical-forming photoinitiator present alongside the at least one controller substance.

However, within the scope of the method according to the invention, it is also possible for the polymerisation in one or both of steps a) and b) to be carried out in the presence of at least one controller substance containing at least one sequence $-S-C(=X)-$, wherein X stands for S, O or NR' , and R' stands for an organic group, and for the controller substance itself to act as a polymerisation initiator. In this case, the polymerisation can be triggered without a further initiator and carried out entirely without the use of a further polymerisation initiator. Such controller substances are also known to those skilled in the art under the designation 'Iniferter', as they combine the functions of an initiator, transfer agent and terminator. In this respect, the controller substance is preferably a trithiocarbonate.

A preferred subject of the invention is a method for producing a polymer system present in at least two phases, comprising the following steps:

- a) UV-initiated, radical polymerisation of a monomer composition I, comprising in total at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group, to form a polymerisate A;
- b) UV-initiated, radical polymerisation of a monomer composition II, comprising in total at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group, wherein at least 50 wt.% of the monomers in monomer composition II differ from those in monomer composition I, in the presence of polymerisate A to form polymerisate B, thereby obtaining a polymer system comprising polymerisates A and B present in at least two phases;

characterised in that step b) takes place in a closed casing.

Even more preferred is a method for producing a polymer system present in at least two phases, comprising the following steps:

- a) UV-initiated RAFT polymerisation of a monomer composition I comprising, in total, at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one polymerisable vinyl group, to form a polymerisate A;
- b) UV-initiated, radical polymerisation of a monomer composition II comprising, in total, at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers having at least one

polymerisable vinyl group, wherein at least 50 wt.% of the monomers in monomer composition II differ from those in monomer composition I, in the presence of polymerisate A to form polymerisate B, thereby obtaining a polymer system present in at least two phases comprising polymerisates A and B;

characterised in that step b) is carried out in a closed casing.

The method according to the invention is intended to result in a polymer system present in at least two phases. Furthermore, it was an objective of the invention to provide the two- or multi-phase polymer system with the highest possible cohesion. It therefore appeared important, when selecting the monomers used in the two polymerisation steps, to take these and, where applicable, further properties of the final polymer system into account.

Against this background, in one embodiment of the invention, one of the monomer compositions I and II comprises in total at least 50 wt.%, more preferably in total at least 60 wt.%, in particular in total at least 75 wt.%, of one or more monomers having a glass transition temperature of the respective homopolymer of $\leq 0^{\circ}\text{C}$, more preferably of $\leq -10^{\circ}\text{C}$, in particular of $\leq -20^{\circ}\text{C}$; and the other of the monomer compositions I and II comprises in total at least 60 wt.%, more preferably in total at least 70 wt.%, in particular in total at least 80 wt.%, of one or more monomers having a glass transition temperature of the respective homopolymer of $\geq 50^{\circ}\text{C}$, more preferably of $\geq 75^{\circ}\text{C}$, in particular of $\geq 100^{\circ}\text{C}$.

In a development of this embodiment, one of the monomer compositions I and II preferably comprises, in total, at least 50 wt.%, more preferably at least 60 wt.%, and in particular at least 75 wt.%, of one or more monomers selected from the group consisting of ethyl acrylate, n-butyl acrylate, iso-butyl acrylate, iso-amyl acrylate, n-hexyl acrylate, 2-heptyl acrylate, n-heptyl acrylate, 2-ethylhexyl acrylate, 2-octyl acrylate, n-octyl acrylate, iso-octyl acrylate, n-nonyl acrylate, iso-nonyl acrylate, n-decyl acrylate, iso-decyl acrylate, 2-[[[(butylamino)carbonyl]oxy]ethyl acrylate, 2-cyanoethyl acrylate, 2-(2-ethoxyethoxy)ethyl acrylate, phenoxyethyl acrylate, iso-stearyl acrylate, docosyl acrylate, and 2-[2-(2-methoxyethoxy)ethoxy]ethyl acrylate; and

the other of the monomer compositions I and II comprises, in total, at least 60 wt.%, more preferably at least 70 wt.%, and in particular at least 80 wt.%, of one or more monomers selected from the group consisting of isobornyl acrylate, norbornyl acrylate, benzyl acrylate, 3,3,5-trimethylcyclohexyl acrylate, cyclohexyl acrylate, 4-[(6-acryloyloxy)hexyloxy]4'-cyanobiphenyl, N-succinimidyl acrylate, 1-ethylcyclopentyl acrylate, N-tert-octyl acrylamide, N-

tert-butyl acrylamide, dimethyl acrylamide, diethylacrylamide, N-isopropylacrylamide, acrylamide, N-[3-(dimethylamino)propyl]acrylamide, diacetoneacrylamide, N-(butoxymethyl)acrylamide, N-phenylacrylamide, N-[2-(dimethylamino)ethyl]acrylamide, N-[2-(diethylamino)ethyl]acrylamide, methyl methacrylate, ethyl methacrylate, cyclohexyl methacrylate, benzyl methacrylate, isobornyl methacrylate, glycerol formal methacrylate, 2-dimethylaminoethyl methacrylate, phenoxyethyl methacrylate, 9-anthrylmethyl methacrylate, 2-ethyl-2-adamantyl methacrylate, 2-(acetoacetyloxy)ethyl methacrylate, 2-isopropyl-2-methacryloyloxyadamantane, isopropyl methacrylate, isobutyl methacrylate, tert-butyl methacrylate, furfuryl methacrylate, 2-methacryloyloxy-2-methyladamantane, 2-morpholinoethyl methacrylate, phenyl methacrylate, N-succinimidyl methacrylate, 2-(tert-butylamino)ethyl methacrylate, 2-cyclohexylpropan-2-yl methacrylate, 1-adamantyl methacrylate, 1-methylcyclopentyl methacrylate, 3-dimethylaminopropyl methacrylamide, N-tert-butyl methacrylamide, N-(methoxymethyl)methacrylamide, N,N-dimethylmethacrylamide, methacrylamide, N-phenylmethacrylamide, N,N-dimethylmethacrylamide, N-isopropylmethacrylamide, N-vinylformamide, N-vinylpyrrolidone, N-vinylcaprolactam, N-vinylcarbazole, N-vinylimidazole, vinylmethyloxazolidinone and N-vinyl-N-methylacetamide.

More preferably, one of the monomer compositions I and II comprises in total at least 50 wt.%, more preferably at least 60 wt.%, and in particular at least 75 wt.%, of one or more monomers selected from the group consisting of n-butyl acrylate, isoamyl acrylate, n-heptyl acrylate, 2-ethylhexyl acrylate, 2-octyl acrylate, iso-octyl acrylate, n-octyl acrylate, n-decyl acrylate, iso-decyl acrylate and lauryl acrylate; and

the other of the monomer compositions I and II comprises in total at least 60 wt.%, more preferably at least 70 wt.%, and in particular at least 80 wt.%, one or more monomers selected from the group consisting of isobornyl acrylate, norbornyl acrylate, tert-butyl methacrylate, benzyl acrylate, N-isopropyl acrylamide, N-tert-octyl acrylamide, N-tert-butyl acrylamide and dimethyl acrylamide.

In a further development of this embodiment, one of the monomer compositions I and II preferably comprises in total at least 50 wt.%, more preferably in total at least 60 wt.%, and in particular in total at least 75 wt.%, of one or more monomers selected from the group consisting of ethyl acrylate, n-butyl acrylate, iso-butyl acrylate, iso-amyl acrylate, n-hexyl acrylate, n-heptyl acrylate, 2-heptyl acrylate, 2-ethylhexyl acrylate, 2-octyl acrylate, n-octyl acrylate, iso-octyl acrylate, n-nonyl acrylate, iso-nonyl acrylate, n-decyl acrylate, iso-decyl acrylate, 2-[[[(butylamino)carbonyl]oxy]ethyl acrylate, 2-cyanoethyl acrylate, 2-(2-ethoxyethoxy)ethyl

acrylate, phenoxyethyl acrylate, iso-stearyl acrylate, docosyl acrylate, and 2-[2-(2-methoxyethoxy)ethoxy]ethyl acrylate;

up to a total of at most 35 wt.%, more preferably up to a total of at most 25 wt.%, and in particular up to a total of at most 20 wt.%, of one or more monomers selected from the group consisting of isobornyl acrylate, norbornyl acrylate, methyl methacrylate, dimethylacrylamide, diethylacrylamide, 4-tert-butylcyclohexyl acrylate, tert-butyl acrylate, tert-butyl methacrylate, cyclohexyl methacrylate and cyclohexyl acrylate;

up to a total of at most 10 wt.% of one or more functionalised monomers; and

the other of the monomer compositions I and II comprises in total at least 60 wt.%, more preferably in total at least 70 wt.%, and in particular in total at least 80 wt.%, of one or more monomers selected from the group consisting of isobornyl acrylate, norbornyl acrylate, benzyl acrylate, 3,3,5-trimethylcyclohexyl acrylate, cyclohexyl acrylate, 4-[(6-acryloyloxy)hexyloxy]4'-cyanobiphenyl, N-succinimidyl acrylate, 1-ethylcyclopentyl acrylate, N-tert-octyl acrylamide, N-tert-butyl acrylamide, dimethyl acrylamide, diethyl acrylamide, N-isopropyl acrylamide,, N-[3-(dimethylamino)propyl]acrylamide, diacetoneacrylamide, N-(butoxymethyl)acrylamide, N-phenylacrylamide, N-[2-(dimethylamino)ethyl]acrylamide, N-[2-(diethylamino)ethyl]acrylamide, methyl methacrylate, ethyl methacrylate, cyclohexyl methacrylate, benzyl methacrylate, isobornyl methacrylate, glycerol formal methacrylate, 2-dimethylaminoethyl methacrylate, phenoxyethyl methacrylate, 9-anthrylmethyl methacrylate, 2-ethyl-2-adamantyl methacrylate, 2-(acetoacetyloxy)ethyl methacrylate, 2-isopropyl-2-methacryloyloxyadamantane, isopropyl methacrylate, isobutyl methacrylate, tert-butyl methacrylate, furfuryl methacrylate, 2-methacryloyloxy-2-methyladamantane, 2-morpholinoethyl methacrylate, phenyl methacrylate, N-succinimidyl methacrylate, 2-(tert-butylamino)ethyl methacrylate, 2-cyclohexylpropan-2-yl methacrylate, 1-adamantyl methacrylate, 1-methylcyclopentyl methacrylate, 3-dimethylaminopropyl methacrylamide, N-tert-butyl methacrylamide, N-(methoxymethyl)methacrylamide, N,N-dimethylmethacrylamide, N-phenylmethacrylamide, N,N-dimethylmethacrylamide, N-isopropylmethacrylamide, N-vinylformamide, N-vinylpyrrolidone, N-vinylcaprolactam, N-vinylcarbazole, N-vinylimidazole, vinylmethyloxazolidinone and N-vinyl-N-methylacetamide; as well as

up to a total of at most 10 wt.% of one or more functionalised monomers,

wherein the functionalised monomers are selected from the group consisting of 1-acryloyloxy-3-hydroxyadamantane, 2-hydroxyethyl acrylate, 2-hydroxyethyl methacrylate, hydroxypropyl acrylate, 4-hydroxybutyl acrylate, 2-hydroxy-3-phenoxypropyl acrylate, 2-hydroxyethyl methacrylate, hydroxypropyl methacrylate, 4-hydroxybutyl methacrylate, 2-hydroxy-3-phenoxypropyl methacrylate, 4-hydroxyethylacrylamide, acrylic acid, methacrylic acid, 2-

acryloyloxyethylsuccinate, methacryloyloxyethylsuccinate, sulfoethyl methacrylate, acrylamide, methacrylamide, glycidyl acrylate, glycidyl methacrylate, 3,4-epoxycyclohexyl methyl acrylate, 3,4-epoxycyclohexyl methyl methacrylate, 4-hydroxybutyl acrylate glycidyl ether, 4-hydroxybutyl methacrylate glycidyl ether, isocyanatoethyl acrylate, isocyanatoethyl methacrylate, 2-[2-(methacryloyloxy)ethyloxy]ethyl isocyanate, 2-[2-(acryloyloxy)ethyloxy]ethyl isocyanate and α,α -dimethyl-m-isopropenylbenzyl isocyanate.

In particular, one of the monomer compositions I and II comprises in total at least 50 wt.%, more preferably in total at least 60 wt.%, and in particular in total at least 75 wt.%, of one or more monomers selected from the group consisting of n-butyl acrylate, isoamyl acrylate, n-heptyl acrylate, 2-ethylhexyl acrylate, 2-octyl acrylate, iso-octyl acrylate, n-octyl acrylate, n-decyl acrylate and iso-decyl acrylate;

up to a total of at most 35 wt.%, more preferably up to a total of at most 25 wt.%, and in particular up to a total of at most 20 wt.%, of one or more monomers selected from the group consisting of isobornyl acrylate, norbornyl acrylate, methyl methacrylate, dimethylacrylamide, and

up to a total of at most 10 wt.% of one or more monomers selected from the group consisting of acrylic acid, methacrylic acid, 2-hydroxyethyl acrylate, 2-hydroxyethyl methacrylate, hydroxypropyl acrylate, hydroxypropyl methacrylate, 4-hydroxybutyl acrylate, 4-hydroxybutyl methacrylate, 3,4-epoxycyclohexyl methyl acrylate, 3,4-epoxycyclohexyl methyl methacrylate, glycidyl acrylate and glycidyl methacrylate; and

the other of the monomer compositions I and II comprises in total at least 60 wt.%, more preferably in total at least 70 wt.%, and in particular in total at least 80 wt.%, of one or more monomers selected from the group consisting of isobornyl acrylate, norbornyl acrylate, tert-butyl methacrylate, benzyl acrylate, N-isopropylacrylamide, N-tert-octylacrylamide, N-tert-butylacrylamide and dimethylacrylamide; as well as

up to a total of at most 10 wt.% of one or more monomers selected from the group consisting of acrylic acid, methacrylic acid, 2-hydroxyethyl acrylate, 2-hydroxyethyl methacrylate, hydroxypropyl acrylate, hydroxypropyl methacrylate, 4-hydroxybutyl acrylate, 4-hydroxybutyl methacrylate, 3,4-epoxycyclohexyl methyl acrylate, 3,4-epoxycyclohexyl methyl methacrylate, glycidyl acrylate and glycidyl methacrylate.

In a specific embodiment of the method according to the invention, at least the monomer composition II is free from methyl methacrylate; more preferably, it is free from methacrylic acid esters and methacrylamides; in particular, it is free from any methacrylic compounds.

It is also preferred that monomer composition I is free of methyl methacrylate; more preferably, it is free of methacrylic acid esters and methacrylamides; in particular, it is free of any methacrylic compounds.

It has been shown that the exclusion of the aforementioned compounds has a beneficial effect on the polymerisation rate.

A further subject of the invention is a polymer system present in at least two phases and within a closed casing, which can be obtained by a method according to the invention.

Preferably, the polymer system resulting from the method according to the invention is a block copolymer. The polymer blocks of this block copolymer are preferably formed by the polymerisates A and B. It is more preferred that the polymer system resulting from the method according to the invention is a triblock copolymer, in particular a triblock copolymer of the form ABA or BAB, wherein A and B denote the polymerisates A and B.

It is also preferred that the polymer system resulting from the method according to the invention is an elastomer.

A further subject of the invention is the use of a polymer system according to the invention as at least one polymer component in an extrusion method for producing an adhesive composition, in particular a pressure-sensitive adhesive.

According to technical understanding, a pressure-sensitive adhesive is an adhesive that possesses pressure-sensitive properties, i.e. the ability to form a permanent bond with a substrate even under relatively light pressure. Such adhesives or pressure-sensitive adhesive tapes are generally permanently self-adhesive even at room temperature, which means that they exhibit a certain viscosity and tackiness, enabling them to wet the surface of a substrate even under light pressure. Without wishing to be bound by this theory, it is often assumed that a pressure-sensitive adhesive can be regarded as an extremely high-viscosity liquid with an elastic component, which consequently exhibits characteristic viscoelastic properties leading to the permanent self-adhesiveness and pressure-sensitive adhesive properties described above. It is assumed that, in pressure-sensitive adhesives, mechanical deformation gives rise to both viscous flow processes and the build-up of elastic restoring forces. The viscous flow

component serves to achieve adhesion, whilst the elastic restoring forces are necessary, in particular, to achieve cohesion. The relationships between rheology and pressure-sensitive adhesion are known in the prior art and are described, for example, in 'Satas, Handbook of Pressure Sensitive Adhesives Technology', Third Edition, (1999), pages 153 to 203. To characterise the extent of the elastic and viscous components, the storage modulus (G') and the loss modulus (G'') are usually employed; these can be determined by means of dynamic mechanical analysis (DMA), for example using a rheometer. In the context of the present invention, an adhesive composition is preferably understood to be pressure-sensitive and thus a pressure-sensitive adhesive if, at a temperature of 23°C in the deformation frequency range from 10^0 to 10^1 rad/sec, G' and G'' each lie at least partly in the range from 10^3 to 10^7 Pa.

To produce an adhesive composition, in particular a pressure-sensitive adhesive, by an extrusion method, the polymer system according to the invention, present within the closed casing, is heated in the extruder to a temperature at which the casing material melts and can thus be homogeneously incorporated into the polymer system. It has been shown that the influence of the casing material on the properties of the adhesive compositions produced with the polymer system of the invention is below the threshold of technical relevance. The polymer system is heated to such an extent that it becomes deformable, and in particular flowable. As has been established, the specific polymer systems of the present invention frequently exhibit lower viscosities and thus better deformability and flowability than conventional polymers used in the production of pressure-sensitive adhesives in particular. The polymer system of the invention can thus be processed in an extruder under the influence of heat and shear, and if necessary with the incorporation of further components, to form an adhesive compound and finally be moulded. The temperatures typically used in this process do not lead to the decomposition of the controller substances incorporated into the polymer frameworks and thus do not result in polymer degradation. A key advantage of the invention is that no removal of solvents is required prior to processing the polymer systems in the extruder, a step which regularly leads to the degradation processes just described.

Further process steps, such as mixing with additives, filtration or degassing, may also take place in the extruder. The moulding of the adhesive composition thus obtained, in particular the pressure-sensitive adhesive composition, into a desired layer form may be carried out, for example, by means of a calender onto a substrate or a release liner.

During the processing of the polymer system into an adhesive composition, in particular a pressure-sensitive adhesive, the polymer system may be mixed with further components.

These further components may be selected from the group consisting of further polymers; tack-enhancing resins; fillers, for example electrically conductive fillers, thermally conductive fillers and the like; flame retardants, for example ammonium polyphosphate and its derivatives; foaming agents; anti-ageing agents; light stabilisers; plasticisers and compounding particles.

A further subject of the invention is a composition for producing a pressure-sensitive adhesive, comprising a polymer system according to the invention, wherein the polymer system is no longer present in a closed casing and the composition is free from cross-linking agents. A key advantage of the present invention is that polymer systems produced according to the invention frequently exhibit such inherent cohesion that they can serve as a basis for pressure-sensitive adhesives without the need for further cross-linking. They thus differ from conventional polymer systems used in the manufacture of pressure-sensitive adhesives, which are based on monomers with functionalised vinyl groups and are usually cross-linked by means of high-energy radiation or thermally to achieve sufficient cohesion.

Examples

Measurement and test methods:

Method 1 – Determination of the glass transition temperature of polymers

The static glass transition temperature of the polymers was determined using dynamic scanning calorimetry (DSC). To do this, 5 mg of an untreated sample of the polymer in question was weighed into an aluminium crucible (volume 25 μ l) and sealed with a perforated lid. A DSC 204 F1 from Netzsch was used for the measurement. The experiment was carried out under nitrogen for inerting. The sample was first cooled to -150°C , then heated at a rate of 10 K/min to $+150^{\circ}\text{C}$ and cooled again to -150°C . The subsequent second heating curve was again run at 10 K/min and the change in heat capacity recorded. Glass transitions are identified as steps in the thermogram (heat flow-temperature diagram, see Figure 1).

The glass transition temperature T_g was obtained as follows (see Figure 1):

The respective linear sections of the measurement curve before and after the step are extended in the direction of rising (section before the step) or falling (section after the step) temperatures (extended lines ① and ②). In the region of the step, a fitting line ⑤ is drawn parallel to the y-axis so that it intersects the two extended lines, in such a way that two areas ③ and ④ (between the respective extended line, the fitting line and the measurement curve) of equal area are formed. The point of intersection of the fitting line positioned in this way with the measurement curve gives the glass transition temperature.

Method 2 – Determination of molar masses

The values for the weight-average molar mass M_w given in this paper refer to the well-established determination by gel permeation chromatography (GPC). The determination is carried out on 100 μ l of a clear-filtered sample (sample concentration 3 g/l). Tetrahydrofuran is used as the eluent. The measurement is carried out at 25°C.

A PSS-SDV column, 5 μ m, 10^3 Å, 8.0 mm $\times$ 50 mm (specifications here and below in the order: type, particle size, internal diameter $\times$ length; 1 Å = 10^{-10} m) is used as a precolumn. For separation, a combination of PSS SDV columns, 5 μ m, 10^3 Å, 10^5 Å and 10^6 Å, each 8.0 mm $\times$ 300 mm, is used (columns from Agilent; detection via PSS SECcurity² differential refractometer). The flow rate is 1.0 ml per minute. Calibration is carried out using the commercially available ReadyCal-Kit Poly(styrene) high from Agilent. This is universally converted to polymethyl methacrylate (PMMA) using the Mark-Houwink parameters K and alpha, so that the data is reported in PMMA mass equivalents.

The polydispersity index (PDI) is determined, as is known to those skilled in the art, as the ratio of the weight-average to the number-average molecular weight.

Method 3 – Dynamic Mechanical Analysis (DMA)

G' and G'' are determined using a rheometer. The material under investigation is subjected to a sinusoidally oscillating shear stress in a plate-plate configuration. In shear stress-controlled instruments, the deformation is measured as a function of time, along with the time lag of this deformation relative to the application of the shear stress. This time lag is referred to as the phase angle δ .

The storage modulus G' is defined as follows: $G' = (\tau/\gamma) \cdot \cos(\delta)$ (τ = shear stress, γ = strain, δ = phase angle = phase shift between the shear stress vector and the strain vector). The definition of the loss modulus G'' is: $G'' = (\tau/\gamma) \cdot \sin(\delta)$ (τ = shear stress, γ = strain, δ = phase angle = phase shift between the shear stress and strain vectors).

$$\tan \delta = G''/G'.$$

Instrument: strain-controlled rheometer (ARES), plate-plate, $\varnothing$ 25 mm

Deformation: 1 %

Frequency: 10 rad/s.

Method 4 – Determination of static shear strength (shear life)

Shear strength is a measure of the internal strength of the adhesive and was tested in the so-called static shear test as follows:

The test was carried out under standard conditions (23°C, 50 % relative humidity) using a weight of 1 kg. A 1.3 cm wide strip of the sample (50 μ m polymer layer on 36 μ m etched PET film) was bonded to a polished steel plate over a length of 2 cm by rolling over it with a 2 kg roller (twice back and forth). The plates were equilibrated for 30 minutes under test conditions but without a load. The test weight (1 kg) was then attached so that a shear stress parallel to the bonded surface was created, and the time until the bond failed was measured. The measurement result is given in minutes. The median of three individual measurements is reported.

Preparation of the casing material:

Two sheets of ethylene-vinyl acetate copolymer film (5 % VA) were laid on top of one another and heat-sealed along three of the four side edges to form a rectangular pouch 10 cm wide and 20 cm long.

Example 1, step a) – Polymerisate 1A

To prepare polymerisate 1A, 120 g of a reaction mixture consisting of 99.2 wt.% IBOA (isobornyl acrylate), 0.5 wt.% DBTTC (dibenzyl trithiocarbonate) and 0.3 wt.% Irgacure® 184 (1-hydroxycyclohexylphenyl ketone) was filled into the bag prepared as described above using a liquid forming, filling and sealing machine. The reaction mixture was inerted by passing nitrogen through it for 30 minutes. The bag was then sealed at the top in a transverse direction. The thickness of the filled bag was 8 mm.

The bag was placed in a water bath, which was maintained at 20°C, so that it was completely surrounded by water. A Heraeus Semray UV2000 LED 365 nm UV lamp was positioned 23 cm above and below the bag. It was irradiated for 10 minutes at an intensity of 10 mW/cm² resulting from the described arrangement.

The weight-average molecular weight (M_w) and the polydispersity index (PDI) of the prepared polymerisate 1A are given in Table 1.

Example 1, step b) – Polymerisate 1B

Polymerisate 1A from step a) was mechanically separated from the bag material. Subsequently, 17 wt.% of polymerisate 1A was dissolved in 83 wt.% n-butyl acrylate. The resulting reaction mixture was filled into another bag prepared as described above and inerted for 30 minutes by passing nitrogen through it. The bag was sealed at the top end with the monomer.

The contents of the bag were irradiated in the same manner as in step a).

The weight-average molecular weight (M_w) and the polydispersity index (PDI) of the resulting polymerisate 1B are given in Table 1.

Example 2, step a) – Polymerisate 2A

For the preparation of polymerisate 2A, 120 g of a reaction mixture consisting of 99.2 wt.% nBA (n-butyl acrylate), 0.5 wt.% BM1812®; (phenylene bis(methylene) didodecyl dicarbonotrithioate) and 0.3 wt.% Irgacure® 184 (1-hydroxycyclohexylphenyl ketone) were placed into a bag prepared as described above. The reaction mixture was inerted by passing nitrogen through it for 30 minutes. The bag was then sealed at the top in a transverse direction.

The contents of the bag were irradiated in the same manner as in Example 1, step a). The molecular weight (M_w) and polydispersity (PDI) of the resulting polymerisate 2A are given in Table 1.

Example 2, step b) – Polymerisate 2B

Polymerisate 2A was mechanically separated from the bag material. Subsequently, 83 wt.% of polymerisate 2A was dissolved in 17 wt.% of IBOA (iso-bornyl acrylate). The resulting reaction mixture was transferred into another bag prepared as described above and inerted for 30 minutes by passing nitrogen through it. The bag was sealed at the top.

The contents of the bag were irradiated in the same manner as in Example 1, step a). The weight-average molecular weight (M_w) and the polydispersity index (PDI) of the resulting polymerisate 2B are given in Table 1.

Example 3, step b) – Polymerisate 3B

The polymerisate A from Example 1, step a) was mechanically separated from the bag material. Subsequently, 25 wt.% of this polymer was dissolved in 75 wt.% LA1214F® (a mixture of lauryl acrylate and myristyl acrylate). The reaction mixture thus obtained was filled into another bag prepared as described above and inerted for 30 minutes by passing nitrogen through it. The bag was sealed at the upper end.

The contents of the bag were irradiated in the same manner as in Example 1, step a). The weight-average molecular weight (M_w) and the polydispersity index (PDI) of the resulting polymer 3B are listed in Table 1.

Example 4 (comparative example)

To prepare polymerisate 4, 120 g of a reaction mixture consisting of 82.2 wt.% nBA (n-butyl acrylate), 17 wt.% iBOA (iso-bornyl acrylate), 0.11 wt.% DBTTC (dibenzyl trithiocarbonate) and 0.3 wt.% Irgacure® 184 (1-hydroxycyclohexylphenyl ketone) was placed into a bag prepared

as described above. The reaction mixture was inerted by passing nitrogen through it for 30 minutes. The bag was then sealed at the top in a transverse direction.

The contents of the bag were irradiated in the same manner as in Example 1, step a).

The molecular weight (M_w) and polydispersity (PDI) of the resulting polymer 4 are given in Table 1.

Table1 : Weight-average molecular weight, polydispersity and shear life of the polymerisates

Polymer	M_w [g/mol]	PDI	Shear life [min]
1A	52,700	2.4	
1B	299,000	2.5	> 10,000
2A	146,000	1.7	
2B	271,000	2.7	> 10,000
3B	594,000	5.6	
4	271,000	1.6	< 5

Claims

1. A method for producing a polymer system present in at least two phases, comprising the following steps:
 - a) polymerising a monomer composition I, comprising in total at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers with at least one polymerisable vinyl group, in order to form a polymerisate A; and
 - b) polymerising a monomer composition II, comprising in total at least 80 wt.% of one or more monomers selected from the group consisting of methacrylic acid esters, methacrylamides and monomers with at least one polymerisable vinyl group, wherein at least 50 wt.% of the monomers in monomer composition II differ from those in monomer composition I, in the presence of polymerisate A in order to form polymerisate B, thereby obtaining a polymer system which is present in at least two phases and comprises polymerisates A and B;characterised in that step b) is carried out in a closed casing.
2. The method according to claim 1, characterised in that at least the polymerisation in step b) is a controlled radical polymerisation.
3. The method according to any one of claims 1 and 2, characterised in that at least the polymerisation in step b) is carried out in the presence of at least one controller substance containing at least one sequence $-S-C(=X)-$, wherein X stands for S, O or NR' , wherein R' stands for an organic group.
4. The method according to claim 3, characterised in that the controller substance is selected from the group consisting of trithiocarbonates and xanthates.
5. The method according to any one of the preceding claims, characterised in that the material of the closed casing is selected from the group consisting of ethylene-vinyl acetate copolymers, ethylene-acrylic acid copolymers, polyethylene, polypropylene, polybutadiene and ionomer films.

6. The method according to any one of the preceding claims, characterised in that, in step b), polymerisate A is dissolved in monomer composition II.
7. The method according to any one of the preceding claims, characterised in that the polymerisation of step b) takes place in the absence of solvent.
8. A polymer system present in at least two phases and in a closed casing, obtainable by a method according to any one of the preceding claims.
9. The polymer system according to claim 8, characterised in that the polymer system is a block copolymer.
10. The polymer system according to any one of claims 8 and 9, characterised in that the polymer system is an elastomer.
11. Use of a polymer system according to any one of claims 8 to 10 as at least one polymer component in an extrusion method for producing an adhesive composition.
12. A composition for producing a pressure-sensitive adhesive comprising a polymer system according to any one of claims 8 to 10, wherein the polymer system is no longer present in the closed casing and the composition is free from cross-linking agents.