



Modification of epoxy resins with functional silanes, polysiloxanes, silsesquioxanes, silica and silicates



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ABSTRACT

Epoxy resins are very important and widely used thermosetting polymers that find many practical applications. Very often their properties can be effectively modified by an addition of reactive silanes, polysiloxanes, silsesquioxanes, silica, montmorillonite, and other fillers. This review considers the literature concerning: (a) synthesis of carbofunctional silanes (CFS), polysiloxanes (CFPS) and polyhedral silsesquioxanes (POSS); (b) properties of neat epoxy resins and their composites and nanocomposites, obtained by modifications with reactive silanes, silicon containing monomers and polymers, and silica based fillers, enabling improvement of their mechanical properties, thermal and flame resistance as well as providing corrosion and antimicrobial protection.

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Abbreviations: PCL-TESi, alkoxy silane-functionalized polycaprolactone; AEPE, allyl 2,3-epoxy propyl ether; AMS, aminommodified silica nanoparticles; APTMS, aminopropyl(trimethoxy)silane; APTES, 3-amino-propyltriethoxysilane; AMDES, (3-aminopropyl)methyldiethoxysilane; H₂N(CH₂)₃MeSi(OEt)₂; ACAF, anisotropic conductive adhesive film; BDMA, benzyl(*N,N*-dimethyl)amine; APDS, bis(4-amino-phenoxy)dimethylsilane; BAMPO, bis(3-aminophenyl)methyl phosphine oxide; DS, 1,3-bis(3-aminopropyl)-1,1,3,3-tetramethyldisiloxane; BISE, 1,3-bis[3-(4,5-epoxy-1,2,3,6-tetrahydrophthalimido)-propyl]tetramethyldisiloxane; BGDMSB, 1,4-bis-(glycidylloxymethylsilyl)benzene; BGTMS, 1,3-bis-(glycidylloxypopyl)-1,1,3,3-tetramethyldisiloxane; HEPA, 2,2'-bis(4-β-hydroxyethoxy)phenyl-propane; BT, bis-maleidetriazine resin; BEPDS, 1,3-bis[2-(3-(7-oxabicyclo[4.1.0]-heptyl)ethyl)tetra-methyldisiloxane; BTMSTS, bis[3-(trimethoxysilyl)-1-phenylpropyl]tetrasulfide; *t*-BOC, *t*-butoxy-carbonyl; CNFs, carbon nanofibers; CNTs, carbon nanotubes; CFS, carbofunctional silanes; CFPS, carbofunctional polysiloxanes; XNBR, carboxylated acrylonitrile butadiene rubber; CTBN, carboxyl-terminated poly(butadiene-co-acrylonitrile) rubbers; *m*-CPBA, *m*-chloroperoxybenzoic acid; CTE, coefficient of thermal expansion; *o*-CFER, *o*-cresol formaldehyde epoxy resin; CE/E-Si, cyanate ester resin CECE-polysiloxane resin; CER, cycloaliphatic epoxy resins; MD₂M, decamethyl-tetrasiloxane (D = Me₂SiO; M = Me₃SiO_{0.5}); DICY, dicyanodiamide; DDM, 4,4'-diaminodiphenyl-methane; DDS, 4,4'-diaminodiphenyl sulfone; DGMPs, diglycidylloxymethylphenylsilane; DEP, diethylphosphite; DMAP, 4-(dimethylamino)pyridine; DDSQ, double-decker silsesquioxane; DMA, dynamic mechanical analysis; DOPO, 9,10-dihydro-9-oxa-10-phosphaphenanthrene-10-oxide; DSC, differential scanning calorimetry; Gly-HPO, (2,5-dihydroxyphenyl)diphenyl phosphine oxide; DGEBAs, diglycidyl ether of bis-phenol A; ES, epoxyfunctional silanes and siloxanes; EMCs, epoxy molding compounds; EP, epoxy resin; GPTMS, (3-glycidioxypropyl)trimethoxysilane; FTIR, Fourier transformation infrared spectroscopy; FS, fumed nanosilica; GF, glass fiber; PDMS-DGE, α,ω-(glycidioxypropyl)poly(dimethylsiloxane); glycidyl-*iso*-butyl-POSS, 1-(3-glycidyl)propoxy-3,5,7,9,11,13,15-isobutylpenta-cyclo[9.5.1.1(3,9).1(5,15).1(7,13)]octasiloxane; D₃D^H, heptamethylcyclotetrasiloxane; MM, hexamethyldisiloxane Me₃SiOSiMe₃; HTHPB, hydroxyl-terminated hydrogenated polybutadiene; HPSiE, hyperbranched polysiloxane; IPDT, integral procedural decomposition temperature; IPN, interpenetrating polymer network; IPTES, isocyanatopropyl(triethoxysilane); ICPTMS, (3-isocyanatopropyl)triethoxysilane; PPSQ, ladderlike

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Contents

1. Introduction	68
2. Reactive silanes and siloxanes in silicon chemistry	68
2.1. Carbofunctional silanes (CFS)	69
2.2. Carbofunctional polysiloxanes (CFPS)	69
2.2.1. Syntheses of CFPS	69
2.3. Polyhedral functional silsesquioxanes (POSS)	73
2.4. Synthesis of epoxyfunctional silanes and siloxanes	75
3. Properties and applications of epoxy resins	81
4. Epoxy resins modified with reactive silanes and siloxanes	84
4.1. Modification of epoxy resins with epoxyfunctional silanes and siloxanes	85
4.2. Preparation, properties and applications of nanocomposites from epoxy resins, silica, silicates and other nanofillers ..	85
4.3. Nanocomposites of epoxy resins with POSS	95
4.4. Flammability of neat and modified epoxy resins	103
4.5. Composites of epoxides modified with polyurethanes and silanes	105
4.6. Epoxy resins in anticorrosion protection	105
4.7. Epoxy resins modified with silanes and silicones in combination with other polymers	109
4.8. Antimicrobial coatings from silane modified epoxy resins	113
5. Summary and conclusions	114
Acknowledgements	114
References	114

1. Introduction

Considerable interest has developed in applications of reactive functional silanes and polysiloxanes, mostly involving compounds with carbofunctional groups. They are used in many fields of science, and especially in materials science and a chemical technology. This interest widens the range of their numerous applications [1]. Moreover, silica, in situ generated nanosilica (by a sol–gel process), silane modified silicas, other fillers and nanofillers have often been used for modifications of different polymers and polymeric materials.

2. Reactive silanes and siloxanes in silicon chemistry

In organosilicon chemistry most important are following functional groups: $\equiv\text{Si}-\text{Cl}$, $\equiv\text{Si}-\text{OR}$, $\equiv\text{SiOCOR}$ (R—usually alkyl group), $\equiv\text{Si}-\text{H}$, and $\equiv\text{Si}-\text{CH}=\text{CH}_2$. Functional organic silanes include: chlorosilanes, alkoxy silanes, and acetoxysilanes, hydrosilanes, and different functional and carbofunctional silanes [2–5]. These silyl functional groups are present both in organosilicon monomers (e.g., dimethyldichlorosilane, methyltriethoxysilane, methyltriacetoxysilane, etc.) and polymers, and can be grafted on

polyphenylsilsesquioxane; LED, light emitting diodes; LOI, limiting oxygen index; LCD, liquid crystal display; LAH, lithium aluminum hydride; MPOSS, mercaptopropyl polyhedral oligomeric silsesquioxane; MAPTMS, (methacryloxy-propyl)trimethoxysilane; MDEA, 4,4'-methylenebis(2,6-diethylaniline); MTHPA, methyl tetrahydro-phthalic anhydride; m-silica, modified silica; POSS-NH₂, monoaminopropyl functionalized POSS; MMT, montmorillonite; MMT, montmorillonite; NCS, nanocomposites; NAD, nonaqueous dispersion; NMR, nuclear magnetic resonance; Me₂SiO₄(D₄), octamethylcyclotetrasiloxane; OE-CH-POSS, octaepoxy-cyclohexyldimethylsilyl-POSS; (HSiO_{1.5})₈(T^H)₈, octahydroxysilsesquioxane; (HSiMe₂O)Si₈O₁₂(Q₈M^H)₈, octakis-(dimethylsiloxy)octasilsesquioxane; OAPS, octa(aminophenyl)silsesquioxane; OC-POSS, octakis(ethyltrimethylsilyloxycyclohexenyl epoxide); OG, octakis(glycidyltrimethylsiloxy)-octasilsesquioxane [(glycidyl-Me₂SiO_{1.5})₈]; OHPS, octakis(3-hydroxypropyldimethylsiloxy)octa-silsesquioxane; OGDMS, octa(dimethylsilylglycidylloxypropyl) POSS; MPOSS, octa(3-mercaptopropyl)silsesquioxane; OMPS, octakis(3-methacryloxypropyldimethylsiloxy)octasilsesquioxane; OE-POSS, octa(propylglycidyl ether) polyhedral oligomeric silsesquioxane; OMMT, organomodified montmorillonite; OTR, oxygen transmission rate; POTS, 1H,1H,2H,2H-perfluorooctyl triethoxysilane; MPDA, *m*-phenylene diamine; BSA, 4,4'-(1,3-phenylenediisopropylidene)bis-aniline; Karstedt's catalyst, platinum divinyltetramethyldisiloxane [Pt₂(dvs)₃]; PAPS, poly(aminophenyl)silsesquioxane; PAMS, poly(3-aminopropyl-methylsiloxane); PDMS, polydimethylsiloxane; PMHS, polymethylhydro-siloxanes; PCL, polycaprolactone; PEG, poly(ethylene glycol); POSS, polyhedral oligomeric silsesquioxane; AEAP-POSS, polyhedral oligomeric silsesquioxane containing eight *N*-aminoethyl-3-aminopropyl groups; PHE-BA, poly(hydroxyether of bisphenol A); BSA, 4,4'-(1,3-phenylenediisopropylidene) bis-aniline; PBT, poly(butylene terephthalate); PCNCs, polymer-clay nanocomposite; PMPSQ, poly(methylphenyl)-silsesquioxane; POPDA, poly(oxypropylenediamine); PTU, polythiourethane; PU, polyurethane; SEM, poly(urethaneimide) PUI scanning electron microscopy; PVAc, poly(vinyl acetate); PCB, printed circuit boards; QAS, quaternary ammonium salt; SCA, silane coupling agents; SM-IATE, silane-modified isocyanuric acid triglycidyl ester; SiC, silicon carbide; POSS, silsesquioxanes; SEC, size exclusion chromatography; SAXS, small angle X-ray scattering; SSP, spherical silica powders; TES-POSS, tetraepoxidized octa-2-styryloctasilsesquioxane [(PhCHCHO)₄(Si₈O₁₂)(CH—CHPh)₄]; TEOS, tetraethoxysilane Si(OEt)₄; TEPA, tetraethylene-pentamine; TMOS, tetramethoxysilane Si(OMe)₄; TMES, tetrakis(methacryloyloxy)ethoxy silane; TPTES, 3-thiopropyltriethoxysilane; ASHP, triaryl sulfonate hexafluorophosphate; PCL-PDMS-PCL, triblock copolymer polycaprolactone-polydimethylsiloxane-polycaprolactone; TGA, thermo-gravimetric analysis; PUI-Si, triethoxysilane-functionalized poly(urethaneimide); TEM, transmission electron microscopy; TBT, tributyltin; TETA, triethylenetetraamine; TMSHU, trimethoxysilane terminated HEPA urethane; TMS-Ph, tris(trimethylsilyl)phosphate; TMPTGE, trimethylpropane triglycidyl ether; TPIP, two photon induced polymerization; VAc, vinyl acetate; vinyl-PDMS, vinyl-terminated PDMS; VOC, volatile organic compounds; YI, yellow index.

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