博 士 学 位 論 文

内容の要旨

および

審査の結果の要旨

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はしがき

本号は学位規則（昭和28年4月1日文部省令第9号）第8条の規定による公表を目的として、平成２１年3月２０日に本学において博士の学位を授与した者の論文内容の要旨及び論文審査の結果の要旨を収録したものである。

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学位記の番号　　甲　第１２号（平成２１年３月２０日）

学位論文題目

Utilisation of mineral fibers with degradable resin-

Material design, properties and applications

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論文内容の要旨

Thesis abstract

The primary field of application for fibre-reinforced polymer composites is the aerospace industry. In recent years however, composite materials have been increasingly used for various technical tasks, where it is beneficial to apply lightweight construction materials which have high strength and stiffness characteristics. The favourable specific properties of fibre reinforced polymer composites are based on the low density of the matrix resins used and the high strength of the embedded fibres.

Regarding utilisation after service life, conventional fibre reinforced composites cause considerable problems, as the combination of miscellaneous, often highly durable components complicates the recycling process considerably and landfill disposal or incineration are becoming increasingly difficult due to environmental concerns and legal requirements. An additional problem, which accompanies the use of polymer resins as matrix material, is the depletion of finite natural petrochemical resources, accompanied by a rise in costs.

An answer to resolve these issues may be given by natural fibre reinforced polymers

based on renewable resources, 'green' composites. By embedding natural fibres into matrices based on biopolymers, e.g. derivates from cellulose, starch, shellac or plant oils, fibre reinforced polymers are obtained, which may be integrated back into the natural biodegradation cycles, e.g. by CO_2-neutral incineration, including recovery of energy, by classic recycling or by composting. Advantages of biodegradable composites therefore include: biological degradation, reduction in the volume of garbage, compostability, preservation of fossil-based raw materials, and protection of climate through reduction of carbon dioxide emission.

Although research on 'green' composites has been conducted worldwide since at least the mid-1990s, the practical applications of 'green' composites are still in its early stages. Most fibres that are currently used for manufacturing of green composites are cellulose-based plant fibres. Although cellulose-based fibres possess several advantages, they exhibit some severe drawbacks. The main aim of this thesis is to contribute to the enhancement of the application possibilities of 'green' composites by exploring natural mineral basalt fibres as reinforcement in biodegradable polymeric resins.

This study presents the development of a composite that uses environmentally degradable starch-based resin as matrix and basalt fibre plain fabric as reinforcement. Initially, the tensile strength of single basalt fibres was verified by means of single fibre tensile tests and statistically investigated by means of a Weibull analysis. Prepreg sheets were manufactured by means of a modified doctor blade system and a hot power press. The sheets were used to manufacture unidirectional-reinforced and bidirectional-reinforced specimens with fibre volume contents ranging from 33 % to 61 %. Specimens were tested for tensile and flexural strength, and exhibited values of up to 517 MPa and 157 MPa in case of UD-composites, and 373 MPa and 122 MPa in case of BD-composites, respectively. Through the analysis of the mechanical properties in relation to the volume fibre content, it was verified that tensile and flexural strength, as well as the according moduli, can be enhanced due to increasing fibre content. The enhancement of the properties was, however, not as continuous as desired, while the strain behaviour was largely independent of the fibre content. This behaviour indicated that the fibres are the predominant load-bearing component, especially at higher fibre fractions, and that the fibre-matrix bond still leaves room for improvement. A further sign of suboptimal fibre-matrix bond was the dominance of compressive fracture at the flexural tests. A relatively high void content

was identified as a further factor. The actual characteristics of the composites offered, therefore, the opportunity to improve interfacial bonding strength and other properties even further by the use of chemical treatments.

Subsequently, through application of silane coupling agents to the reinforcement fibres, the flexural composite properties of the bidirectional reinforced composites were improved by as much as 38 %. This showed that by application of silane coupling agents compatible with the resin system, flexural properties of the composite could be improved significantly

In order to enhance the fire retardancy and hence the applicability of the composite, fire retardants were applied to the resin, and their effectiveness was tested by means of flame rating (according to UL 94) and thermogravimetric analysis (TGA), respectively. The flammability behaviour of the base resin and of the composites with and without flame retardants, as well as the effect of flame retardant additives on the flexural properties of basalt fibre reinforced starch based composite was studied. It was shown that basalt fibres are a natural reinforcement material which significantly improves the flammability behaviour of the thermoplastic matrix. It was further shown that magnesium hydroxide and red phosphor can effectively reduce the flammability of the composite with contents as low as 4.5 wt%. The flexural properties of the retardant-filled composite exhibited a slight decrease compared to the composite without flame retardants, but at the same time showed better properties compared to the composites without treatment with silane coupling agent compatible to the polymer system.

Finally the biodegradability of the basalt fibre reinforced composite was evaluated and approved by means of soil burial tests and garbage-processing machine assisted composting, and the basalt fibres used as reinforcement for the starch based composite were successfully recovered and recycled, as short fibre reinforcement in thermoset unsaturated polyester resin.

Due to results of this study, it was shown that mineral basalt fibres are a promising alternative as reinforcing agents in 'green' composites due to their favourable mechanical properties and limited impact on the environment.

審査の結果の要旨

近年プラスチックの過剰使用に伴い、様々な環境問題が生じている。プラスチックの廃棄処理には、多くのエネルギーを消費するため、環境に対する負荷の小さい廃棄処理法、リサイクル方法の開発は目下の急務となっている。

無機人造繊維強化複合材料の代替材料を新規に創製することによりこれらの課題を解決する見地から、世界中でグリーンコンポジットについての研究と技術開発が盛んに行われている。

本研究は，従来の石油由来樹脂マトリックスに替え、生分解性樹脂（澱粉樹脂）をマトリックスとし、強化材に天然の鉱物繊維であるバサルと繊維を用いたグリーンコンポジットとしてのFRPの新規成形方法を開発し、そのFRPの機械的特性について探究したものである。はじめにドクターブレード成形装置を用い、繊維およびマトリックスともに天然素材の高強度一方向グリーンコンポジットおよび高強度二方向グリーンコンポジットの成形に成功した。本研究で成形したバサルト繊維強化プラスチックスは、最も汎用性の高いガラス繊維強化プラスチックスと同等の機械的特性を示すことが明らかにされた。また機械的特性を向上させる見地から、繊維/マトリックス間の界面特性およびぬれ性の改善の可能性についても論考した。さらに、バサルト繊維強化プラスチックスの耐炎性および生分解性とバサルト繊維のリサイクル性に対しても重要な知見を示した。

本研究の結果は，査読付論文として学会誌に 3 篇掲載されたのみならず、国際会議において7件の口頭発表を行った。

平成 20 年 9 月 16 日に提出された学位論文について 4 名の審査員による予備審査の結果、合格と判定されたことを受け本審査に入り、平成 21 年 2 月 13 日に論文公開発表会を開催した。口頭発表、質疑応答の内容及び本分野の本人の理解度は満足し得るものであった。生分解性材料としての将来の在り方に関する示唆に富む内容であり、博士の学位を与えるに相応しいものと判断された。