Project No: 50766

Project Acronym: EXTRU CO_2

Development of a low-temperature processing method for the production of natural long-fibre filled polypropylene sheet for automotive applications

PUBLISHABLE FINAL ACTIVITY REPORT

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Confidential Technical Report
46571

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Date: 9th August 2006

PROJECT NO: JL0003

EXTRU CO_2: DEVELOPMENT OF A LOW-TEMPERATURE PROCESSING METHOD FOR THE PRODUCTION OF NATURAL LONG-FIBRE FILLED POLYPROPYLENE SHEET FOR AUTOMOTIVE APPLICATIONS

PUBLISHABLE FINAL ACTIVITY REPORT

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PUBLISHABLE FINAL ACTIVITY REPORT

SECTION 1: PROJECT EXECUTION

1.1 Project Background

Natural fibres, such as hemp kenaf, flax and jute find an increasing role in providing reinforcement for polymer matrices used in the automotive sector. Currently most of the consumption is geared towards thermoset chemistry, typified by low shear and temperature processes such as resin transfer moulding. Their use in thermoplastic applications is limited to compression or other press moulding operations, principally because the heat and shear required by conventional processes such as compounding and extrusion can damage the natural fibres. Polypropylene, the main automotive polymer resin has a processing temperature of around 200-220ºC and at these temperatures, particularly in the presence of shear, natural fibres can suffer thermal degradation leading to loss of mechanical performance, discolouration and odour problems.

This issue has stunted the growth of such materials in automotive applications, which is unfortunate since they offer significant benefits over traditional reinforcements such as glass fibre, not least because of their environmental credentials. Thermoplastic materials containing natural fibre reinforcement are recyclable and also offer a move towards sustainable products and processes. Couple this to low weight and the design freedom offered by thermoplastics and the benefits become very.

It is well known that super critical carbon dioxide ($scCO_2$) has a unique set of properties that make it a very powerful solvent. The critical point is reached at a temperature of 31.1ºC and 73.8 bar and at these temperatures it assumes the solvating power of a liquid coupled to the diffusive capacity of a gas. Moreover the critical point is reached under conditions that are achievable under typical thermoplastic polymer processing conditions such as extrusion. The literature indicates that when $scCO_2$ is injected into a polymer melt, its solvating power has a plasticizing effect on the melt that can reduce the viscosity of the system. Unlike traditional plasticization systems, CO_2 can be considered a fugitive plasticizer since its removal is facilitated by diffusion at the end of the process.

Reduced viscosity allows for melt flow at reduced processing temperatures. It is possible that the $scCO_2$ induced viscosity reduction can lead to lower processing temperatures, thus preserving the integrity of the melt phase. This is the key challenge for the Extru CO_2 project.

1.2 Project Objectives

The stated aims of the project are to develop the necessary extrusion and mixing technologies to allow for low-temperature processing of natural fibre filled polypropylene using $scCO_2$ as a processing aid to produce natural fibre polypropylene compounds suitable for thermoforming.

This overall aim is crystallised in the following objectives.

- To develop a pre-competitive industrial process to produce solid, natural fibre filled thermoplastic sheet.

- To produce a pre-competitive solid, natural fibre filled thermoplastic sheet.

- To reduce the environmental impact of both the developed process and the sheet materials produced.

- To reduce production costs.

- To increase SME competitiveness.

1.3 Project consortium

The project consortium consist of a vertically integrated group of SME contractors covering all aspect of the supply chain from natural fibre producers, compound manufacturers, polymer converters- both sheet extrusion and thermoforming- as well as end users with close links to the automotive sector.

The consortium is complemented by two RTD Performers.

SME Contractors

Hemcore Ltd.	Natural Fibre Supplier	United Kingdom
Kefi S.p.A	Natural Fibre Supplier	Italy
Aspin Engineering Ltd.	Precision Engineers	United Kingdom
Whittaker Technical Plastics Ltd.	Technical Compounder	United Kingdom
Candea Industrials Plasticas S.A	Sheet Extruder	Spain
Plasticos Flome S.L.	Thermoformer	Spain
ESP Plastics Ltd.	Thermoformer	United Kingdom
Carcoustics Espana. S.A	Converter	Spain
Sivel Ltd	Converter	Bulgaria

RTD Performers

Asociación de Investigación de Materiales Plásticos (Aimplas)	RTD Performer	Spain
Rapra Technology	RTD Performer	United Kingdom

1.4 Overview of Work Performed

The two year project has been divided up into structured work packages which were integrated together to achieve the objectives of the project.

Materials and Specifications

The first three months of the project concentrated on reviewing the state of the art with regard to thermoplastic natural fibre composites, defining target material applications and arriving at a specification which could be used to bench mark success for the automotive industry (Table 1). The targeted applications are lightly loaded internal automotive body panels such as door and trunk linings, head lining and acoustic insulation panels.

Table 1: Typical Automotive Property Specification for Natural Fibre Thin Sheet Composites

Property	Standard	Range in Automotive sector
Density	DIN 53479	1.00-1.38g/cm^3
Ball Hardness	ISO 2039	80-125MPa
Flexural Strength	ISO 178	23ºC: 20 – 30MPa
		-40ºC: 20 – 90MPa
		80ºC: 9 – 12MPa
Tensile strength	ISO 527	> 35MPa
Strain at Break		20-250%
Heat Behaviour	DIN 53497	90-110ºC
Vicat Softening	ISO 306	110-130ºC
Ball Impact		No break
Charpy (notched)	ISO 179	9-20kJ/m^2
Flammability	According to TL1010	
Odour	PV3900	Maximum mark: 3
Fogging	DIN 75201	Fog mass (procedure A): 1.0mg
Carbon emission	PV 3341	Maximum 50µgC/g
Formaldehyde emission	PV 3925	Maximum 10mg/kg

With the specification identified two parallel actives progressed to look at characterising the effect of $scCO_2$ and developing the natural fibre compounding process.

Compound Formulation and Preparation

Compound formulations have been developed using various grades of polypropylene containing hemp and kenaf natural fibre reinforcement in a range from 10-30%w/w. The role of fibre compatibilisers have been explored and optimum formulations developed to balance the requirements of processability, notably the elastic properties required for thermoforming, and the ultimate property characteristics demanded by the application.

A pleasing aspect of the work was that it proved possible to use conventional bast fibre with no special treatment beyond a natural retting process to separate the fibres from the woody plant core. Again trials were undertaken to optimise the retting process such that a balance could be struck between the conflicting demands of fibre strength and odour.

Natural fibre compound manufacture presented its own set of challenges, principally in resolving how best to incorporate the low bulk density fibre into the twin screw compounding process at relatively high loadings. The problem was resolved by developing a fibre pre-pelletisation process which allowed fibre length to be preserved whilst at the same densifying the fibre to achieve a bulk density similar to the resin carrier. This allowed the fibre to be introduced into the process on conventional feeding equipment. Figure 1 shows an image of the densified kenaf fibre.

Figure 1: Densified Kenaf Fibre Pellet- whole and unravelled

The fibre pre-pelletisation process is an elegant solution to a difficult problem. Whilst it does add an additional step and some cost, it increases the applicability of the technology and provides an avenue for the SME fibre supplier to increase their product range.

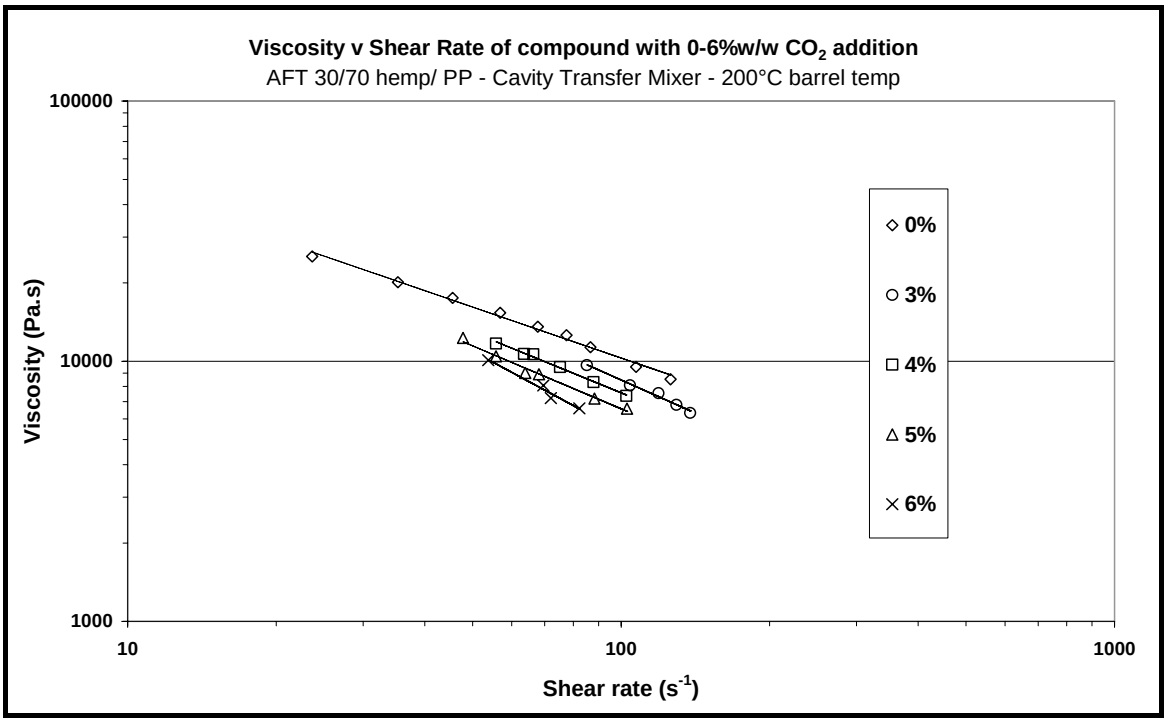
The natural fibre compounds that were optimised from these activities subsequently provided the feedstock for $scCO_2$ development trials as well as the sheet extrusion trials, both pilot and industrial scale.

Supercritical CO_2 Investigation

The effect of the $scCO_2$ process was investigated by performing in-line capillary rheometry measurements. A slit die rheometer was specially constructed and attached to a 38mm single screw extruder which was modified for barrel injection of $scCO_2$. Much effort was devoted to develop and optimise a gas handling and pumping system to control the accuracy and consistency of $scCO_2$ injection. As part of the process several speciality in-line mixers were manufactured to investigate the distribution of the supercritical fluid within the polymer matrix. The work has shown that incorporation of such mixers are beneficial to process stability.

The work has clearly demonstrated the plasticization effect of $scCO_2$ on the natural fibre compounds (Figure 3).

Figure 3: Plasticisation Effect of $scCO_2$ in the range of 0-6%w/w

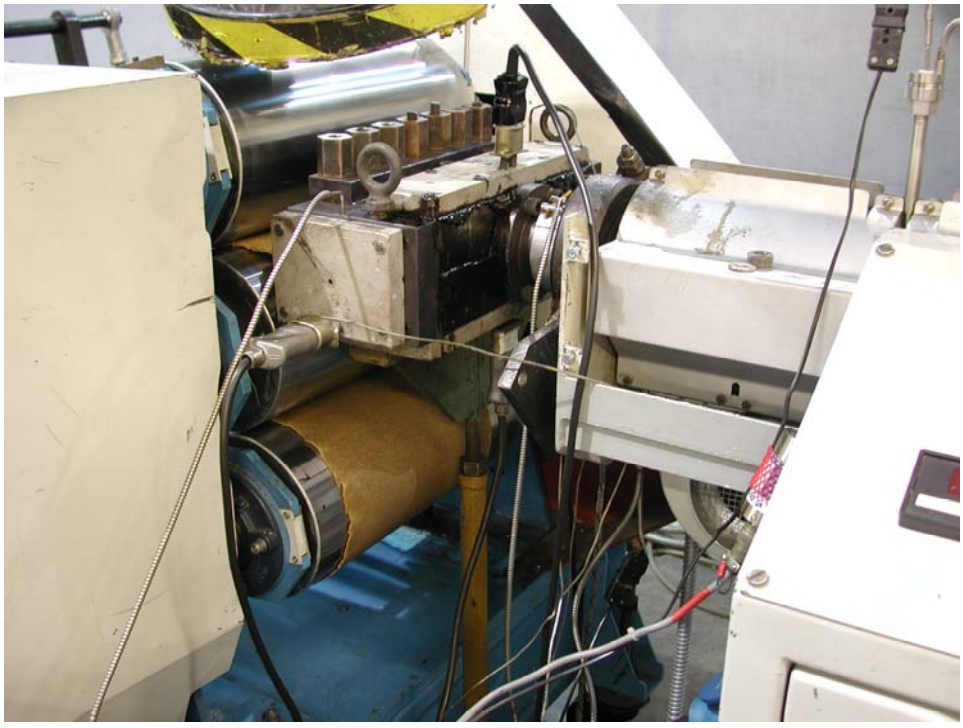


Furthermore it has proved possible to use this reduction in viscosity to progressively lower the processing temperature of the extrusion process post the gas injection system. In this manner a 20ºC reduction in melt temperature depresses the melt temperature of polypropylene extrudate below the 200ºC mark. The approach is possible because the $scCO_2$ plasticisation effect ensure that the viscosity of the system is still sufficiently mobile for the melt to remain processable.

Pilot Sheet Extrusion Trials

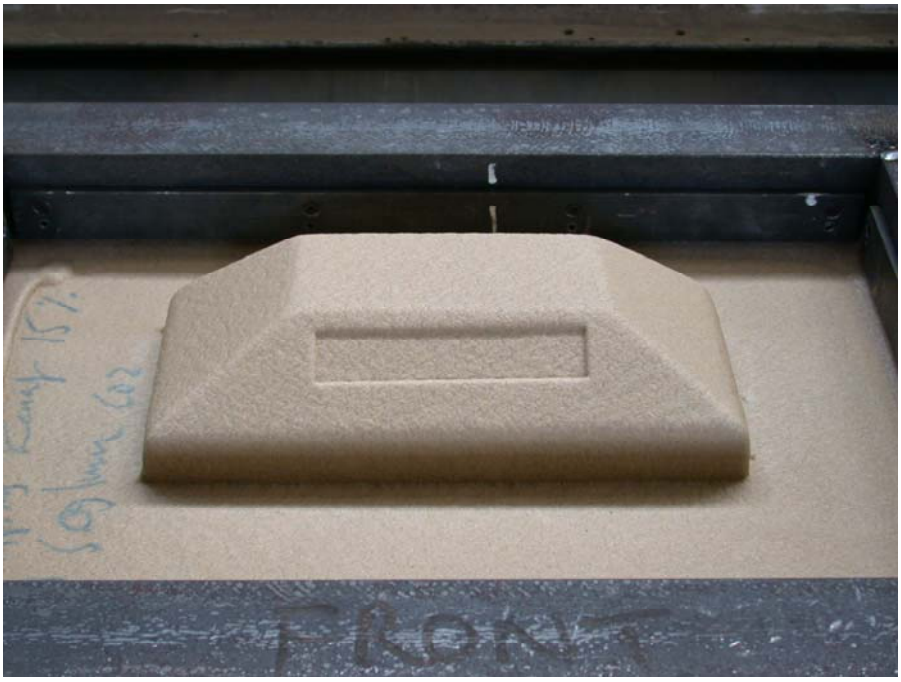
Demonstration of the $scCO_2$ plasticisation effect enabled sheet extrusion trials to be performed on a pilot scale. Sheet 300mm wide sheet and up to 3mm thick was manufactured in combination with $scCO_2$ induced melt temperature reduction (Figure 4).

Figure 4: Pilot Natural Fibre Sheet Production



The sheets produced in this manner were subsequently thermoformed by the consortium partners and accepting for certain limitations such as excessively deep draw angles and high fibre content, the material processed well, producing well defined crisp mouldings. (Figure 5)

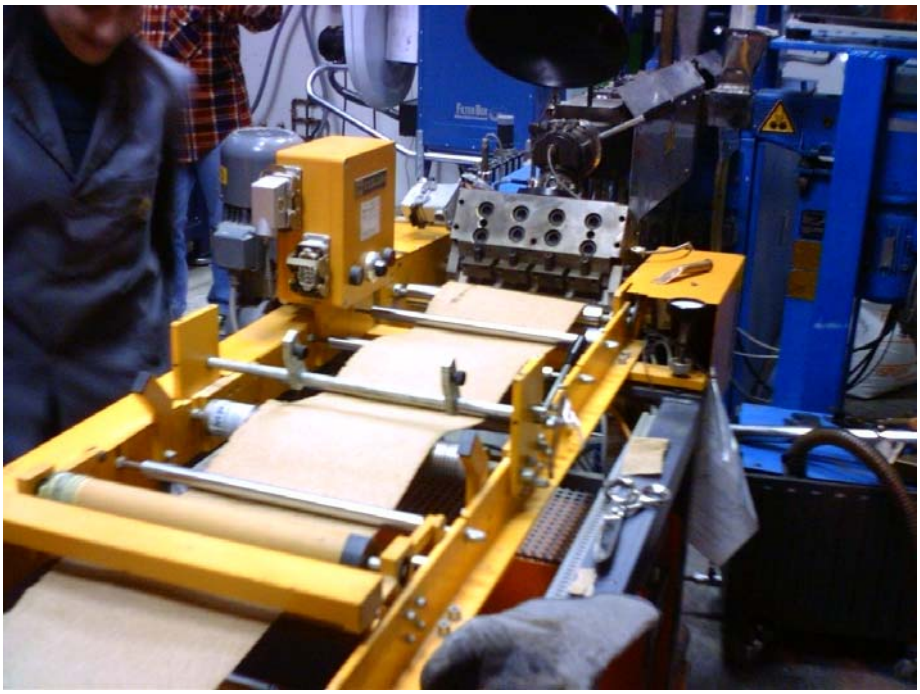
Figure 5: Typical Thermoformed Component



One Sep Process- Direct Inline Compounding

It was appreciated by the consortium that process economics will have a significant bearing on uptake of the technology, particularly so when the target audience is automotive. To address some of these issue the consortium developed a combined compounding and extrusion process capable of mixing the raw ingredients whilst extruding the sheet profile simultaneously (Figure 6). This development has been one of the key success features of the project.

Figure 6: Direct In Line Compounding of Natural Fibre Sheet



1.5 Results and Achievements

The project has demonstrated conclusively the plasticization effect of $scCO_2$ which has in turn has allowed a sheet extrusion process to be developed to manufacture polypropylene natural fibre composites at temperatures below 200ºC. In this respect both the prototype pre-competitive process and product have been realised: these provide additional opportunities to expand the remit of natural fibre products beyond the techniques that currently exist - mat technology etc.

The mechanical properties of the composites produced by the route are inline with the requirements of the automotive industry. Charpy impact properties greater than $25kJ/m^2$ have been obtained and tensile modulus values greater than other types of organic fillers have also been achieved.

With respect to competitor material currently available as pre-compounded feedstocks the properties of the products developed are superior and the cost comparison quite promising.

The environmental credentials of the product have been reviewed by undertaking recycling studies. Again the data is promising with results indicating that up to 50% w/w of recyclate can be re-incorporated without deterioration in mechanical properties. This feature will again help support the process economics.

The principle area for improvement is in maximising mechanical property. So far complete solid profiles have not been attained and it is likely that future effort should be devoted to this are to better understand the complex interaction between the materials and the supercritical fluid.

Further Information

Further Information is available on the Extru CO_2 web site http://www.rapra.net/projects/overview.asp), or by contacting the Project Coordinator Emyr Peregrine (eperegrine@rapra.net)

SECTION 2: DISSEMINATION AND USE

Find below a brief and publishable description of the main characteristics of each of the exploitable results. At the time of writing (June 06) there is no collaboration sought or offered. An Industrial Secret is applied for all the four results. The results will be commercialized according to:

a) The content of the Plan for Using and Disseminating the Knowledge approved by the all the co-owners of the results and

b) The conditions and terms of the corresponding Confidentiality Agreement signed among the co-owner/s interested in the sale (former project partner/s) and the potential client/s.

The four results have common contact details: Project Co-coordinators.

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1. New compound formulations

- **Result description:** Specific compounding formulations made of polypropylene and natural fibres (kenaf or hemp) in different ratios not higher than 30% in weight with good degree of mixing, dispersion and fibre length. To have entire natural fibres (length around 3 cm) in the polymeric matrix is what differentiates these compounds from the commercial ones that use natural fibres but reduced to dust. The presence of entire natural fibre (not dust particles) confers the outstanding properties on the new compounds.

- **Possible market applications/Further research:** The first field addressed is automotive sector since the compound formulations meet its standards in terms of mechanical properties (being manufactured by extrusion process). However, other types of plastic transformation processes where applying this compound are considered to be researched in the mid/long term.

- **Stage of development:** Industrial Scale-up step already done (Compound obtained at industrial scale to test the process viability). The new compound formulations are ready to be used as a new industrial product.

2. Sheets obtained from the new compound formulations

- **Result description:** Thermoplastic extruded sheet made of polypropylene and natural fibres (hemp or kenaf in different ratios not higher than 30% in weight). This new sheet presents advantages over the traditional sheet materials manufactured for internal automotive applications. The new sheet shows:

  a) Reduced fibre breakage.
  b) Enhanced thermal properties.
  c) Enhanced mechanical properties (high impact strength and tensile modulus).
  d) Reduced odour.

  Furthermore, its finish ('varnished wood' aspect) and environmental characteristics make this sheet more attractive to be commercialized.

- **Possible market applications/Further research:** Any thermoformed product manufacturers interested in benefiting from the properties of the new sheet listed above. The new thermoplastic extruded sheet is applicable mainly for automotive sector where its compliance has been proved, but it can be useful in any other potential application where the new sheet turns out to be an innovation (furniture, toys, gardening tooling, etc).

- **Stage of development:** Industrial Scale-up step already done (Extruded sheet obtained at industrial scale to test the process viability). The new thermoplastic extruded sheet is ready to be used as a new industrial product.

3. One-step compounding-extrusion process

- **Result description:** Specific coupling between a compounding machine and a part of an extruder machine which enables to produce a sheet in just one step. This new design of the two machines into one shows savings in the raw material and energy during the one-step process, and consequently, this reduces the total process cost and then, the products cost without losing the products quality.

- **Possible market applications/Further research:** New design to be used by those compounds and extruded sheet manufacturers who can be interested in the advantages of the new one-step process. In principle, applicable for all type of materials.

- **Stage of development:** Pilot-plant prototype. It is necessary to carry out the industrial scale-up study to analyze all the adjustments and modifications required.

4. Pelletization process

- **Result description:** A complete know-how about the work in the pelletization process, the adjustment of critical parameters and auxiliary processes necessary to avoid any loss in the final properties of the fibres. As a result of this knowledge, natural fibre pellets can feed the compounding machine as standard polymeric pellets do it.

- **Possible market applications/Further research:** This knowledge can be useful for those natural fibre suppliers who are interested in offering a new product (natural fibres in pellets) and compounding sectors interested in adding this previous step to their compounding process.

- **Stage of development:** Industrial Scale-up step already done (Natural fibre pellets obtained at industrial scale to test the process viability). Ready to be used as a new industrial product.

5. Best practices for the use and recycling of the new composite and the new products

- **Result description:** A Best Practice Guide written for the production of new compound formulations, extruded sheet and thermoformed parts from natural fibre (up to 30% in weight) and polypropylene. It covers materials advice, processing conditions, and health and safety product design device.

- **Possible market applications/Further research:**
  Recycling/Compounding, Sheet Extrusion and Thermoforming sectors.

- **Stage of development:** Ready to be published.

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CONDITIONS OF BUSINESS – 11-04-2006

FORMATION OF CONTRACT

1.1 All quotations are made and all orders are accepted subject to the following conditions. All other terms, conditions or warranties whatsoever are excluded from any contract between the parties unless expressly accepted by Rapra Technology ("Rapra") in writing.

1.2 Quotations shall be available for acceptance for a maximum period of 30 days from the dates thereof and may be withdrawn by Rapra within such period at any time by written or oral notice. "Work" shall mean the work and services that Rapra agree to provide in the quotation.

1.3 If any statement or representation has been made to the Client by Rapra, or its employees upon which the Client relies (other than in the documents enclosed with Rapra's quotation) then the Client must set out that statement or representation in a document to be attached to the return copy of the quotation and in any such case Rapra may accept or reject the same and/or submit a new quotation.

1.4 The supply of materials, products or information by the Client pursuant to the quotation shall constitute acceptance of these conditions where acceptance has not previously been communicated by the Client to Rapra.

PRICES

2.1 All prices are, unless otherwise stated, quoted net exclusive of VAT.

2.2 All requests for variations or addition to the Work must be made by the Client in writing. In the event of any variation or addition being so requested and agreed to by Rapra, Rapra shall be entitled to make an adjustment to the contract price fairly reflecting such variation or addition.

PAYMENT

3.1 Unless otherwise agreed by Rapra in writing the terms of payment shall be 30 days from receipt of invoice by the Client, which shall be deemed to be two working days after posting. Rapra may submit interim invoices in respect of each stage of Work completed for the Client.

3.2 Rapra reserves the right to vary the payment terms of this contract in the event that it considers payment in advance (in part or whole) is necessary.

3.3 No disputes arising under the contract nor delays beyond the reasonable control of Rapra shall interfere with prompt payment in full by the Client.

3.4 In the event of default in payment by the Client Rapra shall be entitled at its option to treat the whole contract as repudiated by the Client or to suspend all further work on any contract or contracts between Rapra and the Client without notice and to charge interest on any amount outstanding at the rate of 2% per annum above the Base Rate of National Westminster Bank plc in force at the time when payment was due.

COMPLETION

4.1 Time for completion of Work is given as accurately as possible but is not guaranteed. The Client shall have no right to damages or to cancel the order for failure for any cause to meet any time stated for completion of Work.

4.2 Any estimate of the date of completion of Work shall in every case be dependent upon prompt receipt of all necessary information, instructions or approvals from the Client. Variations or additions to the Work requested by the Client may result in delay in completion.

CANCELLATION

5. Either party may cancel the contract on 30 days written notice to the other on condition that all costs and expenses incurred by Rapra up to the time of cancellation and, where cancellation is at the insistence of the Client, all loss of profits and other loss or damage resulting to Rapra by reason of such cancellation, will be paid forthwith by the Client to Rapra.

LIABILITY

6.1 Rapra undertakes that it will indemnify and keep the Client indemnified against all liabilities, costs and expenses in respect of claims in relation to death or injury to persons or damage to tangible property to the extent that such death, injury, loss or damage is attributable to the negligent acts or omissions of Rapra, its officers, employees, agents or sub-contractors.

6.2 Save where Rapra is shown to have failed to exercise reasonable care in the performance of the Work and such failure results in death or personal injury, Rapra shall not be liable in respect of claims arising by reason of death or personal injury. Further, under no circumstances whatsoever shall Rapra be liable for consequential loss, loss of profits, damage to property or wasted expenditure.

6.3 Rapra's liability, whether in respect of one claim or the aggregate of various claims other than claims for death or personal injury due to negligence on the part of Rapra shall not exceed £500,000 in any Year and the Client agrees to insure adequately to cover claims in excess of such amount.

CONFIDENTIAL INFORMATION AND INDUSTRIAL PROPERTY RIGHTS

7.1 All data, information and reports are produced for the benefit of the addressee only. Rapra accepts no liability arising from unauthorised use of such information or reports by a third party.

7.2 The Client shall not reproduce or abstract for the purpose of advertising or otherwise any report or other information on the Work or use the name of Rapra either expressly or by implication in any of its advertising or sales promotional material without the prior written consent of the Company Secretary of Rapra.

7.3 All drawings, documents, confidential records, computer software and other information supplied by Rapra are supplied on the express understanding that copyright is reserved to Rapra and that the Client will not, without the written consent of Rapra, either give away, loan, exhibit or sell any such drawings, documents, records, software or other information or extracts therefrom or copies thereof or use them in any way except in connection with the Work in respect of which they are issued.

7.4 All intellectual Property Rights belonging to or otherwise in the control of either party prior to entering into the contract shall remain the property of the party owning such Intellectual Property Rights.

7.5 All title and ownership of, or relating to, any intellectual property, including, but not limited to ideas, inventions, discoveries, creations, improvements or any other property subject to patent protection or intellectual property rights as developed or resulting from work under this agreement, shall directly or indirectly be solely owned by Rapra Technology unless otherwise agreed to in writing by all participating parties.

7.6 In the event that Rapra Technology does not wish to apply for or maintain patent protection for any invention owned by it in accordance with clause 7 herein, it will on request assign its rights in respect of that patent to the client but in any event Rapra Technology shall be granted a royalty free, irrevocable, non-exclusive, world-wide right to use such intellectual Property Rights assigned under this condition 7.6.

7.7 Rapra Technology will on request grant rights to the client for exploitation or patenting of the ideas, inventions, discoveries, creations, improvements arising from the work, in the client's traditional or defined new areas of business. In all other areas, rights remain vested with Rapra Technology.

SAMPLES

8. Rapra retains the right to return or dispose of the samples at the customers cost after a period of 6 months unless otherwise agreed with the client. Storage of the samples beyond the initial 6 month period will be charged for, invoiced in advance for the agreed period (minimum additional 6 months).

CUSTOMER'S INFORMATION

9.1 The Client shall be solely responsible for ensuring that all drawings, information, advice and recommendations given to Rapra, either directly or indirectly by the Client or by the Client's agents, servants, consultants or advisers, are accurate and sufficient for completion of the Work. Examination or consideration by Rapra of such drawings, information, advice or recommendations shall in no way limit the Client's responsibility hereunder unless Rapra specifically agrees in writing to accept responsibility.

9.2 Rapra shall not disclose to any third party any knowledge or information relating to the Work which is, on receipt by Rapra, marked 'confidential' by the Client unless and until such information becomes public knowledge.

INSOLVENCY

10. If either party shall become bankrupt or under the provisions of Section 123 of the Insolvency Act 1986 is deemed to be unable to pay its debts or compounds with creditors or in the event of a resolution being passed or proceedings commenced for its administration or liquidation (other than for a voluntary winding up for the purposes of reconstruction or amalgamation) or if a Receiver or Manager is appointed of all or any part of its assets or undertaking, the other party shall be entitled to cancel the contract in whole or in part by notice in writing without prejudice to any other right or remedy accrued or accruing to that party.

FORCE MAJEURE

11. In the event of the performance of any obligation accepted by Rapra being prevented, delayed, or in any way interfered with by direction of government, war, industrial dispute, strike, breakdown of machinery or plant, accident, fire or by any other cause beyond Rapra's control Rapra may at its option suspend performance or cancel its obligations under the contract without liability for any damage or consequential loss resulting therefrom, such suspension or cancellation being without prejudice to Rapra's right to recover all sums owing to it in respect of works performed and costs incurred prior to the date of suspension or cancellation.

ASSIGNMENT

12. This Contract is personal to the parties and may not be assigned or transferred without the prior written consent of the other party.

LEGAL

13. The contract shall be governed and interpreted exclusively according to the Law of England and shall be subject to the jurisdiction of the English Courts only.