PRIMUS

WP3. Production of quality recyclates

Tasks 3.1-3.4

DEMO 1: r-PC/ABS for Automotive interior

DEMO 2: r-TPV for Automotive cooling circuit and its elements

DEMO 3: r-HIPS from refrigerators to refrigerators demonstrating food contact

DEMO 4: r-EPDM for washing machine door seal

Deliverable 3.5

PRIMUS demo case studies report

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DISCLAIMER

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DEFINITIONS/GLOSSARY

Recyclate: recycled polymer, either from post-consumer, or post-industrial origin that has undergone necessary pre-treatments steps, such as sorting, washing and melt filtration, etc. allowing the use in new plastic products. Recyclate can be used as such, as one component in a polymer compound, or polymer blend (*i.e.,* final formulation).

Recycled content: Percentage of recycled polymer in the final plastic formulation.

Compatibilizer: usually polymeric modifier & additive that increased the miscibility of incompatible polymer phases in a blend (such as PC/ABS blend)

Impact modifier: usually a rubbery additive, for example, a derivative of polybutadiene or acrylic rubber, that is applied in polymer compounding to improve impact strength or notched impact strength of the base polymer.

Statistical variation of process data: mechanically recycled feedstocks typically have greater variability than virgin plastics in properties. Variation can be *within or between* collected and processed material batches. Variability is often manifested in the measured melt viscosities (Melt flow index, MFI) and other physical properties, such as impact strength and elastic moduli, as compared to virgin plastics. This is due to the facts that recycled polymers always contain impurities, such as mixed non-targeted polymers (e.g., polystyrene in ABS), and the recycled polymers have usually undergone thermo-oxidative and/or UV-degradation. Moreover, recycled feedstocks usually show further degradation of properties in the mechanical recycling and second life. This variability of properties can be compensated during the compounding of formulations, for example by combining strategies of (i) controlling the composition inline during production, (ii) applying reactive additives and stabilizers, (iii) removing non-melting impurities by melt filtration, or (iv) adaptive process control.

ABBREVIATIONS

ABS: Acrylonitrile butadiene styrene copolymer

PC: polycarbonate (of bisphenol A)

PP: polypropylene

EPDM: Ethylene propylene diene rubber

TPV: Thermoplastic vulcanizate. Dynamically vulcanized PP/EDPM thermoplastic elastomer blend

EXECUTIVE SUMMARY

Objective of WP3 was to deliver high-performance recyclates and raw material formulation with recycled content for the PRIMUS demos. Moreover, the consistency and quality variations of the recycled materials streams were assessed at VTT. Sourcing of recycled polymers, compound testing and formulation, and lab scale to bench scale PC/ABS and HIPS compounding test were done at VTT for the Demo 1 (automotive PC/ABS) and Demo 3 (refrigerator HIPS). PP and EPDM were investigated at Cikatek for the Demo 2 (automotive cooling circuit PP/EPDM TPV) and Demo 4 (washing machine gasket, EPDM rubber). Coolrec was the supplier the recycled HIPS, ABS and EPDM test materials. Maier was conducting injection moulding tests, surface coating tests, and 2D validation tests of the developed r-PC/ABS blends.

Broadly summarized, material formulations for each four demo cases have been successful:

1) New r-PC/ABS formulations with high (>80%) recycled content and very consistent properties were generated. Moreover, the material properties enable the demo part made fully of the r-PC/ABS and with aesthetic painted surface.
2) The novel dynamically vulcanized r-PP/EPDM (r-TPV) samples with up to 40% recycled content passed the material thermal aging and chemical exposure tests required for cooling circuit materials. Mechanical properties, performance after aging in the lab and chemical stability of the vulcanizate (r-TPV) were retained well. Due to the unfavourable melt viscosity for tube extrusion and thermoforming, recycled content of the r-TPV was limited to maximum of approximately 25%. However, higher recycled content was demonstrated for injection moulded r-TPV supporting component of the cooling circuitry.
3) Promising results were achieved from the upgrading of r-HIPS flakes from EoL refrigerators. Mechanical properties and melt flow characteristics for extrusion-thermoforming were upgraded to a level comparable to the primary HIPS material. Recycled content up to 70% was demonstrated. Mechanical and rheological testing, and NIAS screening tests showed that closed loop recycling of HIPS in refrigerator liners is technically feasible.
4) Mechanically re-grinded EDPM rubber particles from EoL washing machine gaskets can be used as filler in EDPM compound intended for the washing machine gasket application. Mechanical properties, abrasion resistance, thermal aging behaviour and chemical resistance of the r-EDPM compounds with up to 20% regrind content are very similar compared to the primary EPDM.

On behalf of Authors

Jani Pelto, VTT

PRIMUS PROJECT

PRIMUS project is dedicated to significantly contribute to the goals of the European Strategy for Plastics and enhance the amount of quality and safe recycled plastics that enter the European markets. PRIMUS is a project funded by the Horizon Europe in the following call: HORIZON-CL4-2021-RESILIENCE-01-10: Paving the way to an increased share of recycled plastics in added value products (RIA). PRIMUS is a 3-year project with a total budget of 7 M€. PRIMUS has 10 partners, and 2 affiliated entities.

PRIMUS will actively engage with the plastics value chain stakeholders and innovatively develop novel methods and technologies to significantly increase the circularity, and production and use of sustainable, safe and quality recyclates in added value products. The main technological focuses are on advanced mechanical recycling coupled with broad analytics and novel pretreatment methods for removal of hazardous substances and counteracting degradation. PRIMUS will produce 4 demonstrations where new added value products will be made from recycled and upgraded non- or underutilized plastic waste streams from waste electronics and electrical equipment (WEEE) and end-of-life vehicles (ELV). The four demo products will be automotive interior parts, automotive cooling circuits and its elements, a food contact application refrigerator, and a closed-loop demonstration of washing machine seals.

The project aims at establishing EU widely accepted and transparent procedures to control quality and safety of recyclates, especially for the waste streams containing hazardous substances like brominated flame retardants. The framework related work will include broad engagement of the European plastics sector and recyclers, but also the society, citizens and communities as well as consumers. Safety and trackability back to origin, traceability, are consistent and overlapping themes in PRIMUS. PRIMUS will not only technically and industrially support the uptake of recyclates in products but will also address and support the concerns of the society and enhance the uptake of products that have recycled content.

1 INTRODUCTION

1.1 Scope

This report describes the status and progression of the WP3 material development tasks for each four PRIMUS demo cases. The necessary validation steps towards the full scale industrially relevant demonstrations in the WP4 are explained.

1.2 Audience

The target audience is the PRIMUS consortium and the public.

1.3 Contributions of partners

The following **Error! Reference source not found.** depicts the main contributions from participant partners in the development of this deliverable.

Table 1. Partners´ contributions

Participant short name	Contributions
VTT	Overall content to sections 1,2, 3.1, 3.3., 4.1, 4.3, 5
CIK	Overall content to section 3.2, 3.4, 3.4 and 4.3
MAI	Trials and critical tests for DEMO 1, section 3.1
COR	Sourcing of EPDM, collection, washing, separation and sorting HIPS and ABS recyclates

1.4 Relation to other activities in the project

The following **Error! Reference source not found.** depicts the main relationship of this deliverable to other activities (or deliverables) developed within the PRIMUS project and that should be considered along with this document for further understanding of its contents.

Table 2. relation to other activities in the project

Deliverable Number	Contributions
D3.1	This deliverable provides the overall description of the recycled feedstock sourcing for all four demo cases, quality variations of recycled feedstock, analyses of the melt processing data variability from r-PC, r-ABS and r-HIPS plastics before and after upgrading. Initial technical property datasheets of the demo material formulations were generated.

D3.2	Automotive interior r-PC/ABS part demo materials developed were tested at Maier. Results of the critical tests on 2D sample geometry were given with recommendation for further improvement of surface properties allowing the r-PC/ABS to be used directly used in the visible aesthetic surface on automotive dashboard fascia.
D3.3	Formulation of PP/EPDM thermoplastics vulcanizates with recycled PP and recycled EPDM rubber recovered from aramid reinforced flexible hoses was described. Thermomechanical reclaiming process for peroxide crosslinked EPDM was used. Suitability of the developed formulations with recycled content for tube extrusion and injection moulding, and the mechanical properties of TPVs were initially assessed.
D3.4	Three different thermomechanical grinding routes for reclaiming EPDM were investigated. Mechanical properties of moulded EPDM compounds with recycled content were quantified before after thermal and harsh chemical stress simulating the end application (washing machine gasket).

1.5 Structure

- **Section 1:** Contains an overview of this document, providing its Scope, Audience, and Structure
- **Section 2:** Contains the objectives and expected impacts of the project.
- **Section 3:** contains the final status of the demo material development in PRIMUS WP3.

2 OBJECTIVES AND EXPECTED IMPACT

2.1 Objective

Work package (WP) 3 was started in the beginning of the PRIMUS project with the goal to provide high quality recyclates and formulated materials to the four selected demonstrators:

1) DEMO1 PC/ABS for automotive interior,
2) DEMO2 TPV for electric vehicle cooling circuit,
3) DEMO3 high impact PS for food contact refrigerator liner, and
4) DEMO4 EPDM washing machine gasket.

Development work or the demonstrators started from sourcing of polymer feedstocks, and studying the consistency of the materials streams, and the variation of their initial material properties. In all cases the recycled feedstocks were used as components in material formulations (blends, compounds, and mixtures) and designed for specific technical application. The objective of the WP is to provide realistic and tangible data on how recycled polymer can be upgraded into high performance products, allowing also accurate techno-economic calculations, and, moreover, pave way for better acceptance by the industries and the public.

2.2 Expected Impact

PRIMUS WP3 will likely open new business opportunities and generate new plastics products with recycled content. In broader perspective the WP3 attempts to demonstrate replacement of virgin polymer resins by recycled feedstocks in high-value technical applications. The demonstrations will likely boost the acceptance of recyclates in domestic appliance and on automotive sectors, which as heavily standardized, and where safety is often the primary driver in material selection. Therefore, for good reason these industries have been traditionally rather conservative in taking recycled plastic resins into their products.

3 STATUS OF DEMO MATERIAL DEVELOPMENT

3.1 DEMO #1 PC/ABS Automotive interior part

3.1.1 Functional specifications

The DEMO #1 Fascia developed by MAIER as functional prototype is a component that includes different injected plastic parts, joined to the cockpit with standard snap fits molded on back of the parts. The Electric +Electronic devices (audio, video, GPS, etc.) are fixed with a metallic frame to the fascia and cockpit by standard fasteners (steel screws and nuts). The design of the DEMO #1 Fascia is based on commercial versions of similar applications currently on the market produced by MAIER. This prototype fascia has been re-designed to be injected by a two-material (2K) molding process. The front side of the part is made of a conventional plastic material to obtain the high-quality aesthetic functionality. The back side includes the same structural functionalities than the commercial version.

Figure 1. Fascia-Central Console. Commercial version made by MAIER

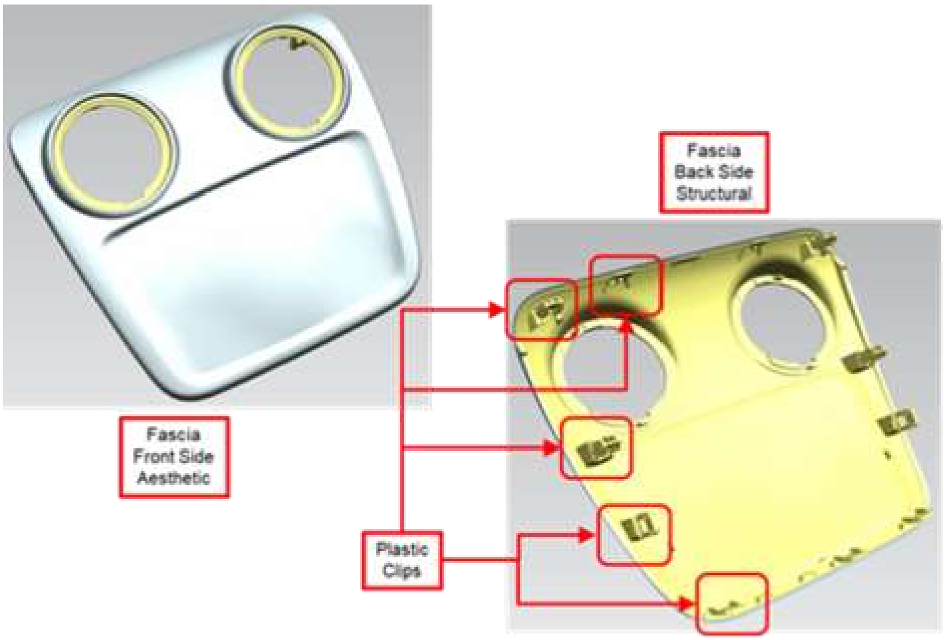


Figure 2. DEMO # 1 Fascia-Central Console. Prototype version.

Validation tests according to the technical specification of European car makers must be done on the 2K dashboard fascia sample parts, to verify the general behavior under normal conditions of use. The tests will be carried out on the different versions of injected and painted parts. The following table (Table 3) summarizes the main functional specifications for an automotive interior application like a dashboard fascia (from Renault and Stellantis standards for definition of functional specifications for automotive parts made of thermoplastics).

Standard tests listed in Table 3 include different methods for the evaluation of mechanical performance, thermal ageing, chemical resistance, visual aesthetic evaluation, assembly/dismantling efforts, part drop, odor, weight, and dimensional control.

Table 3. DEMO #1 Functional Specification.

N°	Test description	Method	Specification
2.2	Stiffness (5daN / 5 s / Ø14mm)	CAR03 0009	Deflection < 2 mm
3.1	Heat ageing (22h/100°C)	D45 1234	No change
4.1	Impact resistance (B/1000/50/23°C)	D42 1235	No break
4.1	Impact resistance (B/1000/50/-30°C)		
5.3	VOC	D45 1601	Report
5.4	Flammability	D45 1333	<100 mm/min
5.5	Fogging (1h / 110°C)	D45 1727	85±5%
	Fogging (24 h / 110°C)		
5.6	Odour	D10 5517	Frequency of descriptor's presence <6/10
5.7	Light resistance (WOM) (240 h)	D47 1431	≥4
5.8	Colour fastness to rubbing (10 Cycles)	D45 1010	

	Dry		≥4/5
	Soapy water		≥4/5
	Technical Heptane		≥4/5
	Essence F		≥4/5
	Acid Perspiration		≥4/5
	Basic Perspiration		≥4/5
	Distilled water		≥4/5
5.9.1	Wear by rubbing	D14 1055	≥4/5
5.9.2	Dry abrasion	D42 1775	≥4/5
5.10	Resistance to micro-organism in humid atmosphere (7days/40°C/95%HR)	D47 1217	No Odour No Micro-organism
	7 days		
	24 h		
5.11.1	Accelerated ageing (72h/40°C/>95%HR)	D47 1165 B15 5050	
5.11.2	Finished parts	B15 5050	
5.11.4	Colour fasteness to water, sea water and perspiration	D47 1020 B15 5050	
	Acid perspiration		5
	Basic perspiration		5
5.12	Chemical resistance (hand cream)	B62 0400	
	Nivea		≥4/5
	La Roche-Posay		≥4/5
5.13	Paint coatings and similar products staining	D27 5144	Colour change: 0 Adhesion: a/b No change
	Initial adhesion	D27 1327	
	Adhesion after water inmersion (72h/40°C)	B62 0400	
6.3	Part fall test (1m)		No break

Commercial versions of the Fascia-Central Console are made of thermoplastic materials like ABS, PC/ABS blend and PC. The choice between them depends on the quality level specified by each OEM. In general, due to the level of functional and aesthetic specifications sought in the final component, the materials used are commercial approved primary grades, with a painted surface finish. The following table (Table 4) summarizes the main technical characteristics of these slightly different type of ABS, PC/ABS and PC materials (values as in Technical Data Sheets from the material suppliers).

Table 4. DEMO #1 Material Specification for Primary materials.

Code				PLA00005	PLA00200	PLA00072	PLA00083	PLA00090	PLA00244
Type				ABS	ABS	PC/ABS	PC/ABS	PC	PC
Melt volume-flow rate		ISO 1133	cm3/10min	25	14	18	20	16	34
Temperature			°C	220	220	260	260	300	300
Load			Kg	10	10	5	5	1,2	1,2
Moulding shrinkage	60x60x2 mm	ISO 294	%	0.4 - 0.6	0.4 - 0.7	0.5 - 0.7	0.5 - 0.7		0.5 - 0.7
Flexural modulus		ISO 178	MPa	2,400	2,350	2,350	2,200	2,450	2,350
Flexural Stress		ISO 178	MPa	72	73	84	82	96	97
Tensile modulus		ISO 527	MPa	2,400	2,300	2,350	2,200	2,300	2,400
Yield stress	50 mm/min	ISO 527	MPa	47	49	54	54	67	65
Yield strain		ISO 527	%	2.5		4.4	4.5	6.0	6.0
Stress at break		ISO 527	MPa			47	56	70	60
Strain at break	50 mm/min	ISO 527	%	>15		>50	100,0	>100	>50
Charpy impact strength	23°C	ISO 179	kJ/m2	90.0			NB	NB	NB
Charpy impact strength	-30°C	ISO 179	kJ/m2	80.0			NB		NB
Charpy impact strength	notched/ 23°C	ISO 179	kJ/m2	17.0	25.0		60.0	30.0	55.0
Charpy impact strength	notched/ -30°C	ISO 179	kJ/m2	7.0	9.0		45.0	15.0	12.0
Izod impact strength	23°C	ISO 180	kJ/m2			NB	NB	NB	
Izod impact strength	-30°C	ISO 180	kJ/m2			NB	NB		
Izod impact strength	notched/ 23°C	ISO 180	kJ/m2	17.0	23.0	48.0	55.0	25.0	55.0
Izod impact strength	notched/ -30°C	ISO 180	kJ/m2	7.0		35.0	40.0	10.0	12.0
HDT A (Unannealed)	1.80 MPa	ISO 75	°C	83	85	102	106	117	124
HDT B (Unannealed)	0.45 MPa	ISO 75	°C	95	92	122	126		137
Vicat softening temp	50°C/h 50 N	ISO 306	°C	101	101	118		136	145
Density		ISO 1183	kg/m3	1,050	1,050	1,130	1,140	1,200	1,190

3.1.2 PRIMUS r-PC/ABS blends for testing

In total sixteen different formulations (DM1-001 to DM1-016) has been generated, having recycled content between 62% and 95%. The nomenclature of test samples in the Maier application test laboratory, and names and compositions of the PRIMUS samples are listed in the Tables 5 and 6, respectively. The first four formulations are with recycled ABS flakes batch (30009) (2021), and from DM1-005 onwards with a new r-ABS flake batch (30012).

DM1-003 and DM2-010 are otherwise the same formulations and generated with the same compounding condition but with different recycled ABS batch (ABS 30009/30012 from Coolrec).

DM1-011 is the blend without added compatibilizer, 11.5% impact modifier, and with high (88%) recycled content.

The five samples DM1-012-016 have similar composition to the DM1-008, but they are with 0.25-0.5% four different surface modifiers which were added to enhance the surface wetting or r-PC/ABS for decorative painting at the part manufacturer.

Table 5. Internal coding of PC/ABS test materials a Maier, including two primary PC/ABS blends, six commercial PC/ABS blend with recycled content and the thirteen of the prototype formulations developed in PRIMUS.

Source	Source	r-Cont (%)	Code MAIER	Code VTT
Recycled	PIR	100	MTC317	
Recycled	PCR	50	MTC344	
Recycled	PCR	50	MTC345	
Recycled	PCR	50	MTC346	
Recycled	PIR	100	MTC398	
Recycled	PIR	75	MTC401	
Primary		0	PLA00072	
Primary		0	PLA00074	
Recycled	PIR+PCR	62	MTC538	DM1-005
Recycled	PIR+PCR	75	MTC559	DM1-006
Recycled	PIR+PCR	80	MTC540	DM1-007
Recycled	PIR+PCR	88	MTC541	DM1-008
Recycled	PIR+PCR	87	MTC542	DM1-009
Recycled	PIR+PCR	86	MTC543	DM1-010
Recycled	PIR+PCR	84	MTC544	DM1-011
Recycled	PIR+PCR	86.8	MTC611	DM1-012
Recycled	PIR+PCR	86.5	MTC612	DM1-013
Recycled	PIR+PCR	86.5	MTC613	DM1-014
Recycled	PIR+PCR	86.5	MTC614	DM1-015
Recycled	PIR+PCR	86.5	MTC615	DM1-016

Table 6. r-PC/ABS formulations developed at VTT. All formulations except DM1-011 contain also 4.0 wt-% compatibilizer and 0.5 wt-% thermal stabilizers.

Formulation number	r-PC (%)	virgin ABS (%)	impact modifier (%)	r-ABS (%)	recycled content (%)
DM1-001	62	33.4	0	0	62
DM1-002	62	0	0	33.4	95.4
DM1-003	62	0	11.5	22	84
DM1-004	62	0	6.7	26.7	88.7
				new batch of r-ABS (COR 30012) in use ↓	
DM1-005	62	20.5	0	13	75
DM1-006	62	15.5	0	18	80
DM1-007	62	0	7.5	26	88
DM1-008	62	0	8.5	25	87
DM1-009	62	0	9.5	24	86
DM1-010 (control for 003)	62	0	11.5	22	84
DM1-011 (no compatibilizer)	62	0	11.5	26	88
DM1-012 surf. mod. 1_conc.1	62	0	8.5	24.8	86.8
DM1-013 surf. mod. 1_ conc. 2	62	0	8.5	24.5	86.5
DM1-014 surf. mod. 2_c1	62	0	8.5	24.5	86.5
DM1-015 surf. mod. 3_c1	62	0	8.5	24.5	86.5
DM1-016 surf. mod. 4_c1	62	0	8.5	24.5	86.5

3.1.3 Mechanical properties and melt flow properties of PRIMUS r-PC/ABS blends versus targeted performance in automotive interior application

Table 7 shows technical performance of few PC/ABS blends that have been previously studied as potential candidates for the dashboard fascia material. Table 8 presents the tensile properties, Charpy Notched impact and MFI of the PRIMUS r-PC/ABS formulations developed at VTT. The targeted numbers received from the part manufacturer are presented in green on the first row. It can be deduced from the tabulated data that all the sample formulations except for the sample DM1-002 with the maximum of 95.5% recycled content and without impact modifier can meet the targets of good mechanical properties. The mostly varying parameter between the formulations is the (low) MFI, which indeed can be a limiting factor in the production of thin-walled injection moulded parts.

It is to be noted that the r-PC/ABS blends from DM1-005 onwards are based on new r-ABS batch (30012). Composition of DM1-007-011 was systematically varied in the same range as DM1-003-004. Recipes DM1-005-006 do not contain impact modifier but have virgin ABS content between 15-20% instead. Adding virgin ABS (high MW grade) improves further the tensile strength and Young's modulus, but with the expense of lowering the recycled content of the blend.

Composition of DM1-012 to DM1-016 blends with added (0.25-0.5%) surface are similar in composition with DM1-008. Mechanical properties of those samples are within the expected range.

Table 7. Technical performance of PC/ABS blends previously validated for dashboard fascia at Maier test center.

Code				MTC317	MTC345	MTC346	MTC347	MTC398	MTC401	PLA00072	PLA00074
Melt volume rate, MVR		ISO 1133	cm3/10 min	30	20	18	22	22	22	18	19
Temperature			°C	260	260	260	260	260	260	260	260
Load			Kg	5	5	5	5	5	5	5	5
Moulding shrinkage	60x60x2 mm	ISO 294	%	0.70			0.4 - 0.6			0.5 - 0.7	0.5 - 0.7
Tensile modulus		ISO 527	MPa	2,350	2,260	2,270	2,300	2,051	2,200	2,350	2,300
Yield stress	5 mm/min	ISO 527	MPa								54
Yield stress	50 mm/min	ISO 527	MPa	50				39	45	54	
Yield strain		ISO 527	%						5.0	4.4	4.7
Stress at break		ISO 527	MPa		51.0	53.0	52.0			47.0	50.0
Strain at break	5 mm/min	ISO 527	%								>50
Strain at break	50 mm/min	ISO 527	%	>50	>90	>90		61.0		>50	
Charpy impact strength	23°C	ISO 179	kJ/m2	NB			NB	NB	NB	NB	NB
Charpy impact strength	-30°C	ISO 179	kJ/m2							NB	NB
Charpy impact strength	notched/23 °C	ISO 179	kJ/m2	40.0	50.0	55.0	45.0	42.0	45.0	50.0	50.0
Charpy impact strength	notched/-30°C	ISO 179	kJ/m2	30.0	38.0	44.0				35.0	37.0
Heat Deflection Temp	1.80 MPa	ISO 75	°C	100	99	106	100		105	102	107
Vicat softening temp	50°C/h 50 N	ISO 306	°C	116	121		121	115		118	128
CLTE		ISO 11359	E-4/°C							0.80	0.75

Water absorption	23°C/sat	ISO 62	%	0.70					0.20	0.70	
Moisture absorption	23°C/50%rh	ISO 62	%	0.20						0.20	
Density		ISO 1183	kg/m3	1,110	1,130	1,150	1,120	1,120	1,150	1,130	1,140

Table 8. Summary of mechanical properties, melt flow index and recycled content of the four initial formulations DM1-001-004 based on r-ABS batch (30009), and seven other formulations (presented in the previous table).

#sample	Recycled content (%)	Charpy notched impact @RT. (kJ/m2)	Young's modulus (MPa)	Tensile Stress at max force (MPa)	MFI +260°C /5 kg (g/10 min)
Target	**80**	**30<**	**2000<**	**45< (yield)**	**15<**
DM1-001	62.1	43.0 ± 1.9	2190 ± 50	55.6 ± 0.4	7.5 ± 0.5
DM1-002	95.5	10.0 ± 0.9	2410 ± 20	60.7 ± 0.2	26.9 ± 0.8
DM1-003	84.0	32.8 ± 1.0	2000 ± 30	48.5 ± 0.2	9.2 ± 0.5
DM1-004	88.9	26.3 ± 1.4	2240 ± 30	52.7 ± 0.3	16.4 ± 1.1
DM1-005	62.0	37.8 ± 1.9	2260 ± 10	55.4 ± 0.2	12.9 ± 0.5
DM1-006	75.0	32.6 ± 1.9	2300 ± 30	56.1 ± 0.4	16.0 ± 0.5
DM1-007	80.0	37.2 ± 0.7	2150 ± 40	51.4 ± 0.3	14.6 ± 0.5
DM1-008	88.0	38.2 ± 0.7	2110 ± 50	50.6 ± 0.3	11.0 ± 0.9
DM1-009	87.0	39.7 ± 0.6	2090 ± 50	49.7 ± 0.4	11.3 ± 0.7
DM1-010	86.0	40.9 ± 0.9	1990 ± 30	48.4 ± 0.3	8.6 ± 0.4
DM1-011	84.0	23.5 ± 0.5	2010 ± 30	47.7 ± 0.2	>90 (not stabile)
0.25-0.5% surface modifiers added					
DM1-012 surf. mod. 1_conc.1	86.8	31.2 ± 0.8	2100 ± 40	47.2 ± 0.1	n.a.
DM1-013 surf. mod. 1_ conc. 2	86.5	30.3 ± 1.1	2030 ± 30	45.9 ± 0.3	n.a.
DM1-014 surf. mod. 2_c1	86.5	31.4 ± 1.2	2050 ± 40	46.8 ± 0.3	n.a.
DM1-015 surf. mod. 3_c1	86.5	--	--	--	n.a.
DM1-016 surf. mod. 4_c1	86.5	--	--	--	n.a.

3.1.4 2D Validation test of aesthetics properties and chemical compatibility

Injection moulding

The results from the characterization tests show that the pre-selected commercial recycled PC/ABS materials have a general behavior that seems to be adequate for the considered Demo 1 Fascia application. In any case, further technical activities are needed to confirm that these recycled materials are compatible with the injection molding and post-processing (painting) of the proposed final component. The following processing tests have been carried out under controlled conditions at MAIER pilot plant facilities:

- Injection molding of 2D sample parts (120x100x2.0 mm3) in a prototype mold (Figure 4), to define the optimized processing window (melt temperature, mold temperature, injection speed) for each material under evaluation.

- Aerographic painting of 2D sample parts, by using different paint formulations (One-Layer Paint, Tri-Layer Paint).
- Preliminary functional validation tests (visual aesthetic evaluation, color, gloss, initial adhesion, adhesion after immersion in water, high-pressure washing).

Both the commercial recycled PC/ABS materials and the prototype formulations developed by VTT have been injection molded to obtain flat sample parts of 120x100 mm2 and 2.0 mm in thickness (Figure 3). The injection tests have been carried out on a molding machine DEMAG 80-370 with 800 kN of clamping force. The following table (Table 9) summarizes the actual recorded values set to obtain the 2D sample parts with optimized quality, for each of the materials under evaluation. No relevant differences were detected during the injection moulding of the recycled materials compared to the prime materials, considered as the base line for the evaluation of the processability. All the critical factors for the preliminary evaluation of the quality of the samples (weight, dimensional control, visual effect) are of the same level as the primary reference samples.

A prototype mould (Figure 3), with interchangeable inserts on both fixed and moving half, has been used for the injection of the 2D sample parts. The ejection system is on the moving half, while the cavity, the sprue and the injection gate are on the fixed half.

The overall surface appearance of the selected injection molded 2D parts of the commercial (r-)PC+ABS grades, and the prototype PRIMUS r-PC+ABS grades are shown in Figures 4 and 5, respectively.

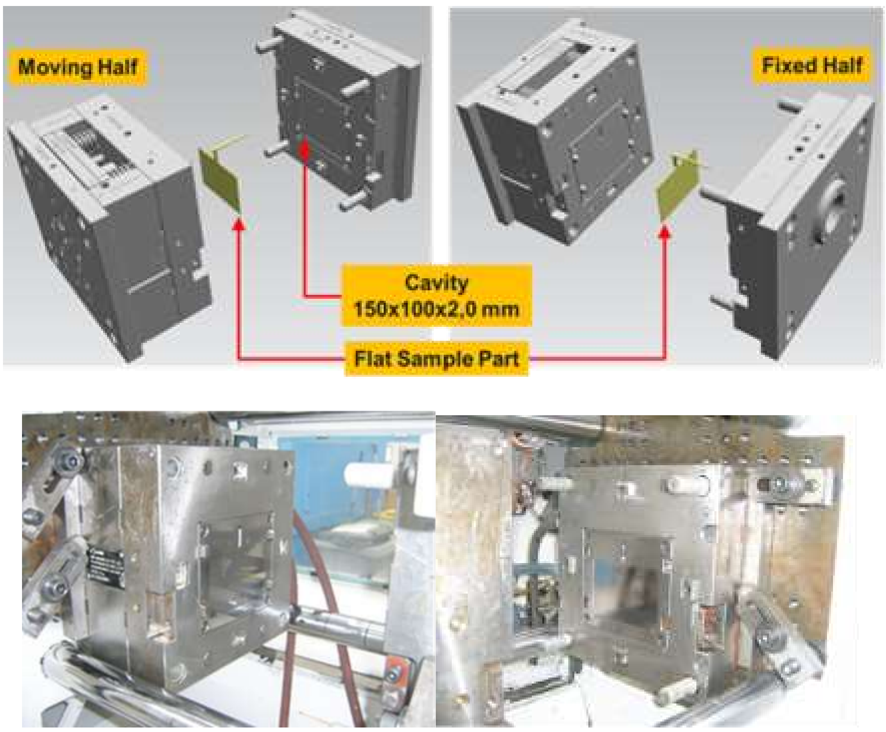


Figure 3. Injection mould. Flat Sample Part.

Table 9. Injection Moulding Processing

Material	Code	MTC317	MTC344	MTC345	MTC346	PLA00072
Drying	(h)	2	2	2	2	2
	(°C)	80	80	80	80	80
Injection Filling Time	(s)	2.1	2.1	2.1	2.1	2.1
Dossing Time	(s)	5.4	5.4	5.4	5.4	5.4
Injection Pressure	(bar)	938	996	1,068	1,242	1,097
Injection Speed	(cm^3/s)	17.2	17.2	17.2	17.2	17.2
Holding Pressure Time	(s)	8.0	8.0	8.0	8.0	8.0
Holding Pressure	(bar)	578	578	578	578	578
Cooling Time	(s)	30.0	30.0	30.0	30.0	30.0
Screw Rotation	(rpm)	300	300	300	300	300
Backpressure	(bar)	3	3	3	3	3
Nozzle	(°C)	230	230	230	230	230
Front	(°C)	220	220	220	220	220
Center	(°C)	210	210	210	210	210
Rear	(°C)	200	200	200	200	200
Mold Temperature	(°C)	70	70	70	70	70

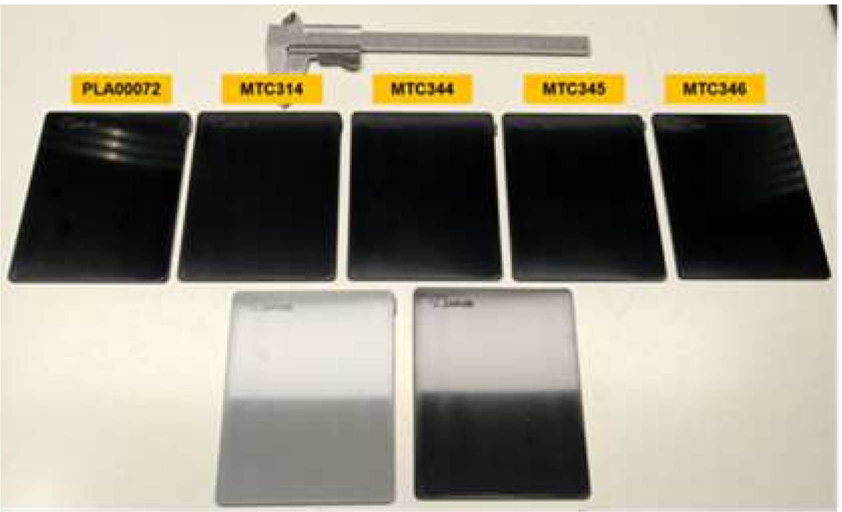


Figure 4. Injected Sample Parts. PC+ABS Recycled. Commercial grades.

MTC538 (DM1-005)

MTC539 (DM1-006)

MTC540 (DM1-007)

MTC541 (DM1-008)

Figure 5. Injected Sample Parts. Recycled PC+ABS. Prototype formulations DM-005 to DM-008 developed in EU-PRIMUS.

Post-Processing (Aerographic painting)

The injected 2D sample parts have been surface coated by aerographic painting, at MAIER pilot plant facilities, to reproduce the same processing conditions used at industrial scale in a robotic facility, for the manufacture of painted automotive components. The processing conditions of the painting process are the same as in serial production:

- Pre-Drying: 15 min / 25°C
- Oven Drying: 30 min / 80°C

To evaluate properly the behavior of the recycled PC/ABS materials, compared to the primary grades, two different paint formulations have been used. Mixing ratios for each component of the formulations listed below (Table 10) are the same as in serial production of automotive applications with aesthetic finishes.

The painting process is depicted in Figure 6. Examples of painted parts made of commercial materials and of PRIMUS prototype materials are shown in Figure 7 and Figure 8, respectively.

Table 10. A) Tri-Layer Paint (Satin Chrome), B) One-Layer Paint (Deep Black).

A	Code	Type	(%)
Primer	P00278	Primer	47.6
	P00350	Thinner	47.6
	P7421	Hardener	4.8
	Total		100.0
Colour		Paint	83.2
	P00052	Thinner	16.6
	P7421	Hardener	0.2
	Total		100.0
Coat		Coat	66.6
	P00350	Thinner	33.3
	P7421	Hardener	0.1
	Total		100.0

B	Code	Type	(%)
Paint One-Layer Paint (Deep Black)	P7433	Paint	71.4
	P7421	Hardener	14.3
	P00054	Thinner	14.3
	Total		100.0

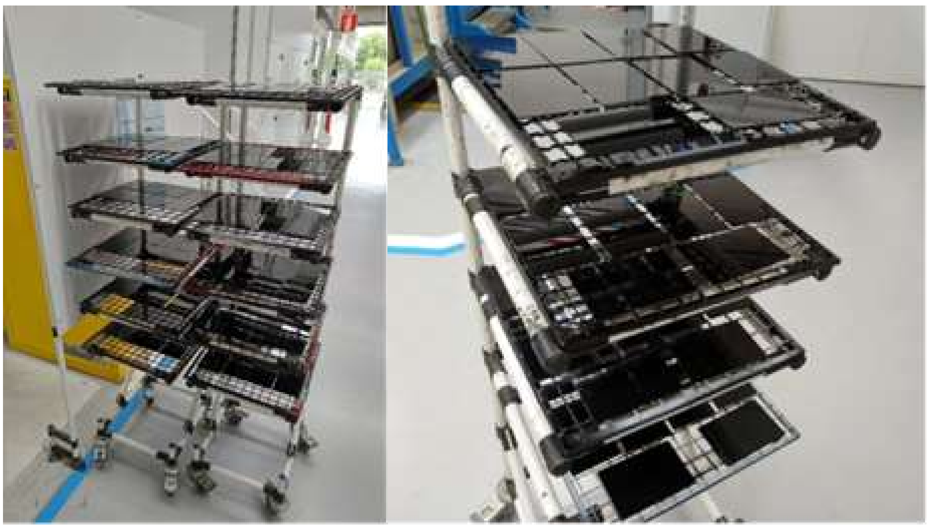


Figure 6. 2D samples after the painting process.

Figure 7. Painted sample parts. Tri-Layer Paint (left) and On-Layer Paint (right)

MTC538 DEM1-005 MTC539 DEM1-006

Figure 8. Painted sample parts. On-Layer Paint. r-PC/ABS prototype formulations.

The 2D painted sample parts have been evaluated according to the most relevant technical specifications, to check the actual performance of the recycled PC/ABS materials, the paint formulations and processing conditions, compared to the quality criteria of a conventional automotive application with a painted aesthetic finish produced at industrial scale in serial production conditions.

The following Table 11 and Table 12 and the selected graphical presentations shown in Figures 9 and 10 summarize the results obtained for the commercial grade samples and the PRIMUS prototype materials, respectively.

To make easier the discussion of the results, a quantitative comparative scale has been proposed, where a value of Assessment-Overall Rating = 100% represents a painted part that fulfills all the functional specifications proposed for the DEMO #1 Fascia demonstrator.

Briefly summarized, of the commercial (recycled or virgin grade) materials only one, namely MTC 398 (100% post-industrial recycled material) had 100% rating in the tests. In other cases, the samples did not pass the Resistance to immersion in water (Ford Tank) test and/or failed in the "resistance to washing with high pressure cleaners" test. This is underlining the highly destructive nature of these tests (Table 11, Figure 9). The corresponding data for the DM1-005-DM1-011 prototype samples (Table 12, Figure 10) shows more limited success in the water immersion test and the visual appearance of the painted parts. Therefore, it is anticipated that the main difference comparing the commercial versus prototype materials is the paint adhesion, which is is most likely due to the differences in the surface wetting (surface energy) of the materials.

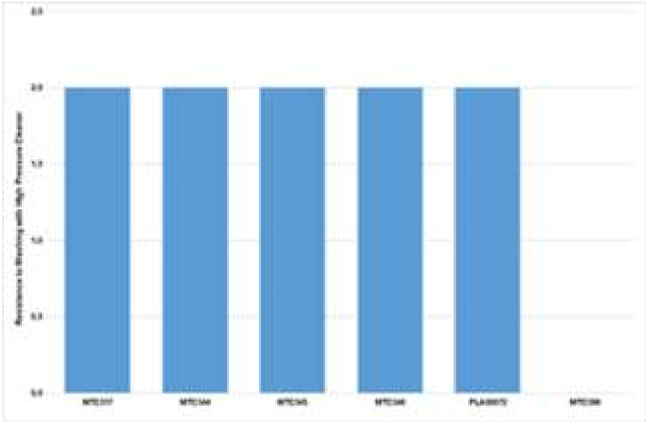
Validation tests results (Commercial Recycled materials)

The following Tables 11-12 and Figures 9-10 summarize the validation test results of the commercial materials with recycled content.

Table 11. Validation Tests Results. Commercial Recycled Materials.

			Paint	Tri-Layer					One-Layer
Test	Standard		Code	MTC317	MTC344	MTC345	MTC346	PLA00072	MTC398
Colour	D25 5479 / ASTM D2244		L	82.60	82.08	81.65	79.53	81.83	24.36
			a	-1.03	-1.01	-0.96	-0.87	-0.99	-0.06
			b	-0.05	0.05	0.04	-0.07	-0.03	-0.25
			G	117.57	114.90	116.67	121.30	120.30	92.33
Initial Adhesion	B72 0200 / D25 1075		a-b	OK	OK	OK	OK	OK	OK
Aesthetic (Visual)	Injection		0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Painted		0.0	1.0	0.0	1.0	1.0	0.0	0.0
	Total		0.0	1.0	0.0	1.0	1.0	0.0	0.0
				NOK	OK	NOK	NOK	OK	OK
Resistance to immersion in water (Ford Tank)	B72 0200 / D27 1327	Blistering	0	2	1	2	2	0	0
				NOK	NOK	NOK	NOK	OK	OK
		Adhesion	a-b	d	a	a	a	a	a
				NOK	OK	OK	OK	OK	OK
Resistance to washing with high pressure cleaners	B72 0200 / D25 5376		0.0	2.0	2.0	2.0	2.0	2.0	0.0
				NOK	NOK	NOK	NOK	NOK	OK
Assessment - Overall Rating			%	20	60	60	40	80	100

A



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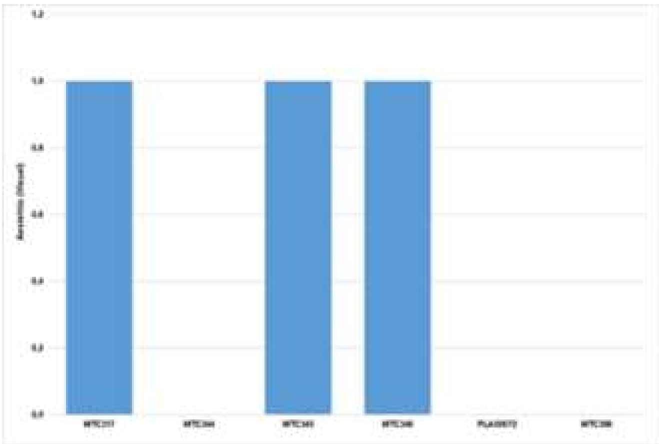


C

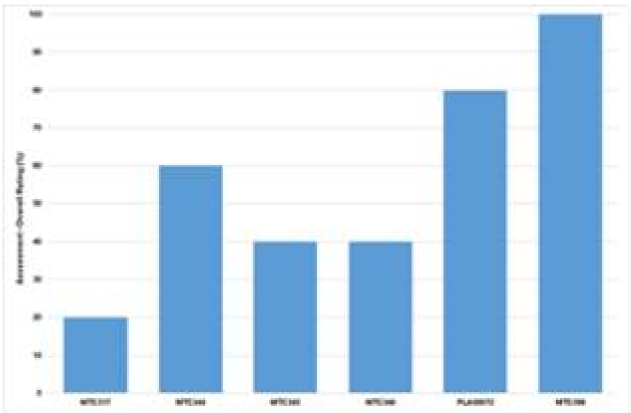
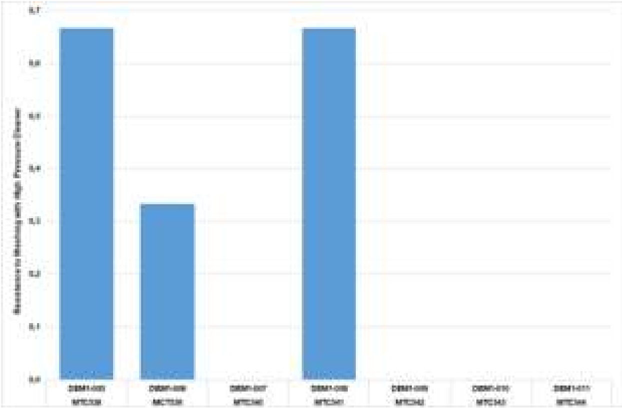


Figure 9. Validation test data for commercial materials with recycled content. A) Resistance to Washing with High Pressure Cleaner, B) Aesthetic (Visual) C) Assessment – Overall Rating.

Table 12. Overall Rating from validation tests of PRIMUS r-PC/ABS samples.

			Paint	One-Layer (Deep Black)						
Test	Standard		Code	DEM1-005	DEM1-006	DEM1-007	DEM1-008	DEM1-009	DEM1-010	DEM1-011
Colour	D25 5479 / ASTM D2244		L	23.80	23.80	23.81	23.78	23.79	23.77	23.76
			a	-0.11	-0.10	-0.10	-0.11	-0.13	-0.11	-0.12
			b	-0.58	-0.59	-0.57	-0.60	-0.59	-0.60	-0.60
			G	89.97	90.27	89.87	90.40	90.60	90.37	90.30
Initial Adhesion	B72 0200 / D25 1075		a-b	OK	OK	OK	OK	OK	OK	OK
Aesthetic (Visual)	Injection		0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
	Painted		0.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
	Total		0.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0
				NOK	NOK	NOK	NOK	NOK	NOK	NOK
Resistance to immersion in water (Ford Tank)	B72 0200 / D27 1327	Blistering	0							
				NOK	NOK	NOK	NOK	NOK	NOK	NOK
		Adhesion	a-b	a	a	a	a	a	a	a
				OK	OK	OK	OK	OK	OK	OK
Resistance to washing with high pressure cleaners	B72 0200 / D25 5376		0.0	0.7	0.3	0.0	0.7	0.0	0.0	0.0
				NOK	NOK	OK	NOK	OK	OK	OK
Assessment - Overall Rating			%	40	40	60	40	60	60	60

A



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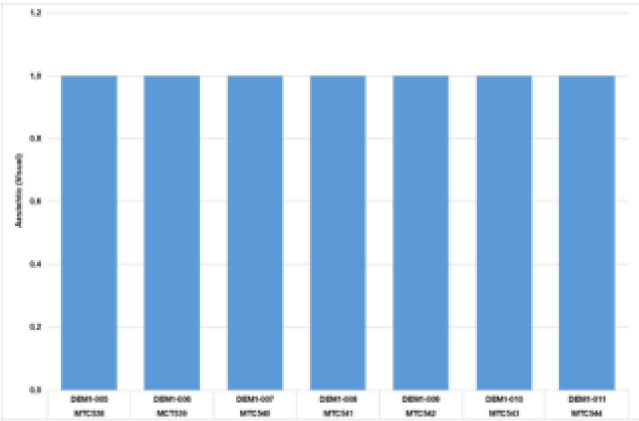

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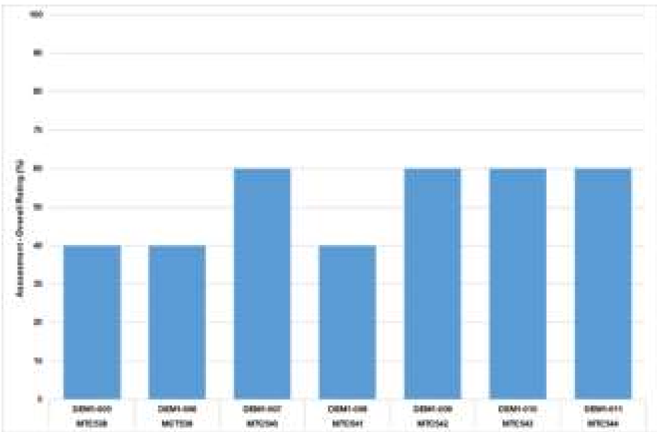

Figure 10. Validation test data for PRIMUS demo materials. A) Resistance to Washing with High Pressure Cleaner, B) Aesthetic (Visual) C) Assessment – Overall Rating.

3.2 r-TPV (rPP/EPDM) for automotive cooling circuit

3.2.1 Components of the cooling circuit

Demonstrator 2 involves the cooling circuit, tubes and supporting parts for automotives (Figure 11). The cooling circuit is necessary to eliminate the excess of heat that is generated when the car is operating. The cooling system is composed by several elements, like thermostat, radiator, heater, fan, air condenser, coolant reservoir. Basically, all the individual parts are connected by thermoformed hoses.

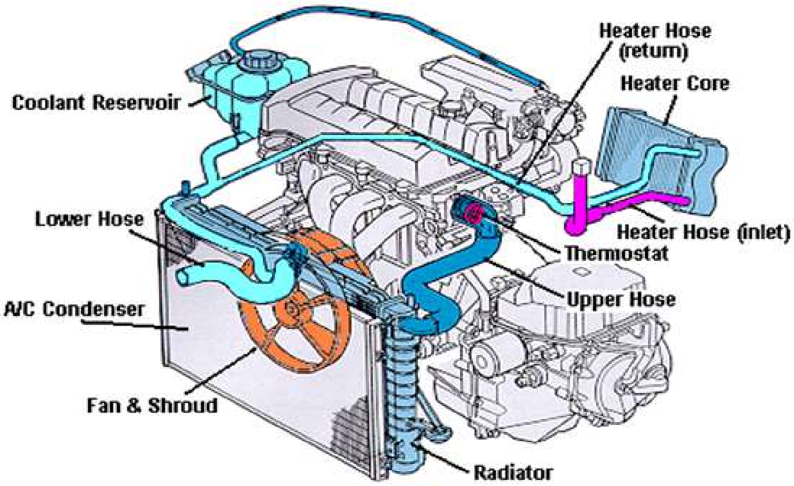

Figure 11. Cooling circuit of the combustion engine

Nowadays, EPDM hoses are the ones mostly used for this application in combustion engine type cars and multilayer hoses (with a textile reinforcement) (Figure 12) or monolayer (Figure 13) can be used depending on the working pressure that the hose needs to withstand.

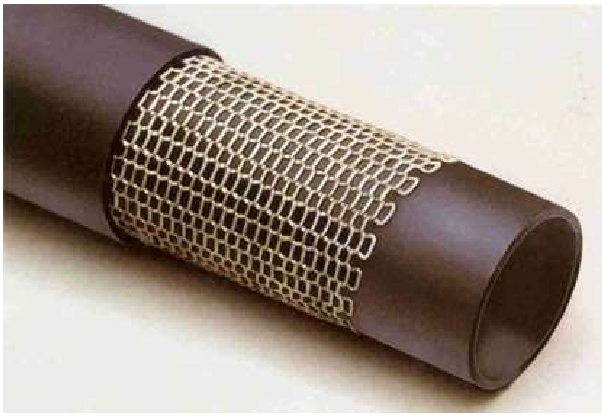

Figure 12. Multilayer EPDM hose with textile reinforcement

In electric vehicles the working temperatures and pressure ranges are lower than in combustion engine type cars and other alternative materials can be used for the cooling hoses. Thermoplastic vulcanizates (TPV) are good candidates for this type of hoses and in this project polypropylene (PP) and ethylene-propylene-diene (EPDM) rubber-based TPVs have been developed. Being thermoplastic, TPV hoses provide many advantages in comparison to the conventional ones:

- Recyclability
- Reduction in weight
- Shorter production times

Figure 13. Monolayer TPV pipe

The selected materials need to be resistant against the permanent contact to coolant fluids. Mixtures of 50% coolant and 50% water are used to withstand the wide temperature range that is reached near the cooling system, guaranteeing a low freezing point and a high boiling point.

3.2.2 Sourcing of r-PP, r-EPDM and r-PP/EPDM

The r-PP has been sourced from a local recycled material supplier and has been introduced in the formulation together with the virgin material. The r-PP is collected from post industrial waste of alimentary food cans and grinded to obtain the r-PP that is shown in the following Figure 14.

Figure 14. r-PP sourced from a local supplier

The recycling of the EPDM is more complex due to the chemical bonds that are in the structure of the vulcanized rubber. To obtain the recycled EPDM a devulcanization is needed and it has been performed in an external company using a co-rotating twin screw extruder.

The post-industrial EPDM hoses have been collected from CIKAUTXO factory and grinded before the reclaiming process. The obtained r-EPDM is shown in the Figure 15 and has been introduced in TPV formulations together with the primary EPDM.

Figure 15. Reclaimed EPDM.

Another way to introduce recycled material could be to regrind the extruded TPV pipes and use this material again to manufacture pipes. Limiting amount of the regrinded TPV in extruded pipe is to be analysed.

3.2.3 Processing of dynamically vulcanized r-PP/EPDM thermoplastic elastomer

The TPV mixtures have been manufactured in the co-rotating twin screw extruder that is in the mixing room of Cikautxo factory. The EPDM that is part of these formulations is vulcanized dynamically during melt mixing at high shear and elevated temperatures and the product obtained is in form of pellets as is shown in the Figure 16.

Figure 16. TPV pellets manufactured in CIKAUTXO.

3.2.4 Technical preformance vs. specifications for the TPV hoses in the cooling systems

In the first period of the project, 8 TPV formulations have been developed with different recycled content and the properties of these materials are shown in the following Table 13.

Table 13. The initial PRIMUS TPV formulation DM2-000-007.

Formulation no.	Recycled content (%)	Tensile Strength (MPa)	Elongation at break (%)	MFI (g/10min)
Requirement		> 7	> 400	1-9
DM2-000	0	15.0	430	0.7
DM2-001	10	15.5	436	1.9
DM2-002	21	16.0	461	7.1
DM2-003	12	19.1	421	0.7
DM2-004	24	17.4	425	1.5
DM2-005	49	11.5	154	40
DM2-006	70	11.9	180	100
DM2-007	36	13.1	500	24

The formulations from DM2-000 to DM2-004 exhibit good flow properties for extrusion process but the latest formulations from DM2-005 to DM2-007 showed too high MFI values that means that these materials have very low consistency for extrusion.

Although the first 5 formulations (DM2-000 to DM2-004) have more suitable MFI, these formulations have too high hardness (ShD 50) which is out of range from the desired hardness (ShD40) and were therefore not the selected for the extrusion tests.

Five more formulations were investigated with the adequate hardness (ShD40) and with recycled content up to 36%. The properties of these formulations are shown in the following Table 14.

Table 14. The final PRIMUS TPV formulation DM2-008 (primary reference) to DM2-012.

Formulation no.	Recycled content (%)	Tensile Strength (MPa)	Elongation at break (%)	MFI (g/10min)
Requirement		> 7	> 400	1-9
DM2-008	0	13.7	440	2.4
DM2-009	36	13.3	510	30
DM2-010	30	12.9	467	21
DM2-011	23	14.0	422	14
DM2-012	18	14.7	496	10

The formulations with the highest recycled content showed the highest MFI and due to that, will not be suitable for the extrusion.

The latest formulations DM2-011 and DM2-012 were selected to the extrusion of the TPV pipe samples. The mechanical test results and flow behaviour of these formulations is to be compared with the reference primary TPV DM2-008.

3.2.5 Chemical compatibility with cooling liquids and aging in air

Table 15 summarizes the chemical compatibility and aging test results. To perform these material tests, 2D material sheets with dimensions 90x90x2 mm3 have been injected in Cikautxo's facilities and samples have been taken out to obtain dumb-bell shaped samples Overall, aging in coolant and the exposure to hot air (+110°C) for up to 1000h does not significantly affect the mechanical properties of the primary TPV reference nor the r-TPV samples having 18% and 23% recycled contents.

At initial state, the TPV materials containing recycled material showed higher tensile strength, although these materials are 3 points softer in the Shore D scale. The change in hardness after the ageings is lower in the formulations containing recycled material and for tensile/elongation at break changes there is not significant difference between different formulations.

Table 15. Data from the chemical compatibility and aging tests of r-TPVs.

	Formulation no.	DM2-008	DM2-012	DM2-011
	Recycled content (%)	0	18	23
TEST	Requirement			
Initial				
Hardness (ShD)	40 ± 5	41	38	38
Tensile Strength (MPa)	> 7	13.7	14.7	14.0
Elongation at break (%)	> 400	440	496	422
Air Ageing 110°C 168h				
Hardness (ShD)		43	38	39
ΔShD	-5 to 5	+2	0	+1
Tensile Strength (MPa)		14.5	14.8	14.5
ΔTS, %	-30 max	6	1	4
Elongation at break (%)	> 300	431	442	462
ΔEB, %	-40 max	-2	-11	9
Air Ageing 110°C 504h				
Hardness (ShD)		45	40	39
ΔShD	-5 to 5	+4	+2	+1
Tensile Strength (MPa)		14.2	15.2	15.1
ΔTS, %	-30 max	4	3	8
Elongation at break (%)	> 300	414	431	460
ΔEB, %	-40 max	-6	-13	9
Air Ageing 110° 1008h				
Hardness (ShD)		46	41	40
ΔShD	-5 to 5	+5	+3	+2
Tensile Strength (MPa)		13.9	15.5	15.1
ΔTS, %	-30 max	1	5	8
Elongation at break (%)	> 300	396	438	471
ΔEB, %	-40 max	-10	-12	12
Coolant Ageing 110° 168h				
Hardness (ShD)		40	37	37
ΔShD	0 to 10	-1	-1	-1
Tensile Strength (MPa)		13.8	14.3	14.7
ΔTS, %	-20 max	1	-3	5
Elongation at break (%)	> 300	392	402	455
ΔEB, %	-30 max	-11	-19	8
Coolant Ageing 110° 504h				
Hardness (ShD)		44	40	39
ΔShD	0 to 10	3	+2	+1
Tensile Strength (MPa)		13.7	14.5	15.1
ΔTS, %	-20 max	0	-2	8
Elongation at break (%)	> 300	405	409	456
ΔEB, %	-30 max	-8	-18	8

Coolant Ageing 110° 1008h				
Hardness (ShD)		40	37	38
ΔShD	0 to 10	-1	-1	0
Tensile Strength (MPa)		14.3	15.1	15.6
ΔTS, %	-20 max	4	+3	+11
Elongation at break (%)	> 300	400	440	461
ΔEB, %	-30 max	-9	-11	+9

3.3 Recycled HIPS (r-HIPS) for refrigerator liner

3.3.1 Background and motivation

Refrigerators most commonly have their inner liners made of high impact strength polystyrene plastic (HIPS) (Figure 17). The stringent requirement for the HIPS in this application is the food contact approval. This is due to potential contact with food inside the refrigerator and requires special HIPS grades (such as the Ineos Styrolution HIPS resin used as reference in this project) that have passed specific migration tests and are known for their enhanced environmental stress cracking resistance. For this reason, it was considered the already once food contact approved HIPS grade collected from end-of-life refrigerators could be potentially good source for recycled (r-HIPS) feedstock. Moreover, upgrading the processing properties for extrusion-thermoforming of such recycled feedstock should be viable.

Figure 17. Refrigerator with thermoformed HIPS inner liners visible.

3.3.2 Sourcing of r-HIPS

The r-HIPS flakes were received from Coolrec B.V., Waalwijk, Netherlands by grade name PS30025, which is collected from the post-consumer fridge inner liners (Figure 18). The r-HIPS was delivered form 13 different production time points in the form of flakes. To evaluate the variation in the content and quality of the r-HIPS, 3 samples from the different locations in the container of each production point was taken. The 39 samples were grinded and flushed with distilled water before the processing and analyses for the r-HIPS to get rid of Sodium chloride from density separation at Coolrec B.V. and other water-soluble contaminants. For a reference, virgin HIPS, Styrolution PS ESCRimo, was received from INEOS Styrolution Group GmbH.

Figure 18. Recycled (r-HIPS) flakes sourced from Coolrec B.V.

3.3.3 Technical performance requirements for r-HIPS

The criteria for the acceptable performance of r-HIPS are based on the properties of primary HIPS (Table 16). Importantly, r-HIPS should have the required food contact approval. Second, the processibility with extrusion-thermoforming must be similar compared to the primary resin. Moreover, elastic properties and notched impact strength of the material should be in the same range as in the primary resin.

Table 16. Requirement and analysis results for reference primary HIPS.

	Primary HIPS (Ineos Styrolution)	Requirement	Units
Charpy impact strength	15	>10	kJ/m^2
Melt flow rate / index (MFI)	3.4	3-7	g/10 min
Storage modulus (E') DMA	1700	>1700	MPa at 20°C

3.3.4 Screening of the properties of r-HIPS between different processing points and different batches at the recycler

The grinded and washed recycled HIPS were dried and processed into test samples of 4x10x80mm3 using twin screw micro compounder (DSM Xplore, 15 cm3 mixing volume). The drying and processing of virgin HIPS was made with the same parameters but using the supplied pellets without grinding or washing. Test samples were analysed using impact strength test and from the impact strength results 3 highest and 3 lowest impact strength samples were also analysed for melt flow rate (MFR), solid state viscoelastic Storage and Loss moduli by dynamical mechanical analysis (DMA) and chemical composition by Fourier transform infrared spectroscopy (FT-IR). The results for the analyses are summarized in Figures 19-22.

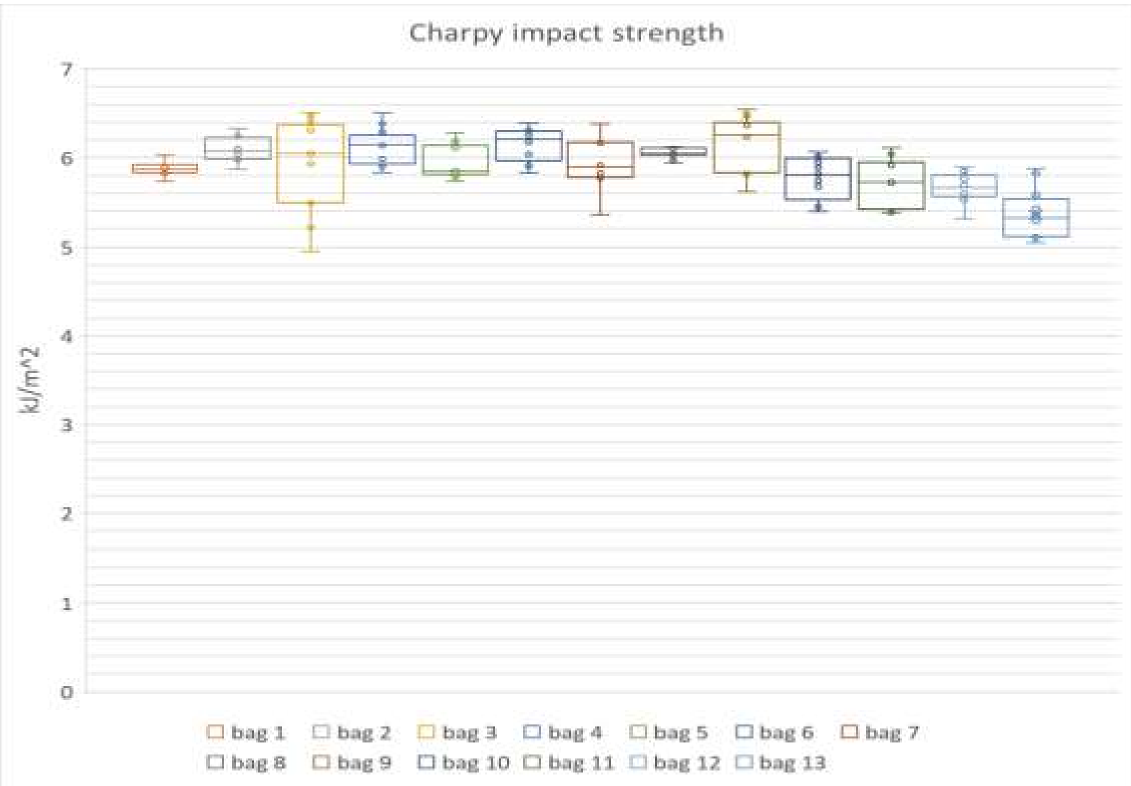

Figure 19. Charpy impact strength results for injection moulded r-HIPS samples at each of the 13 production time points (n=12).

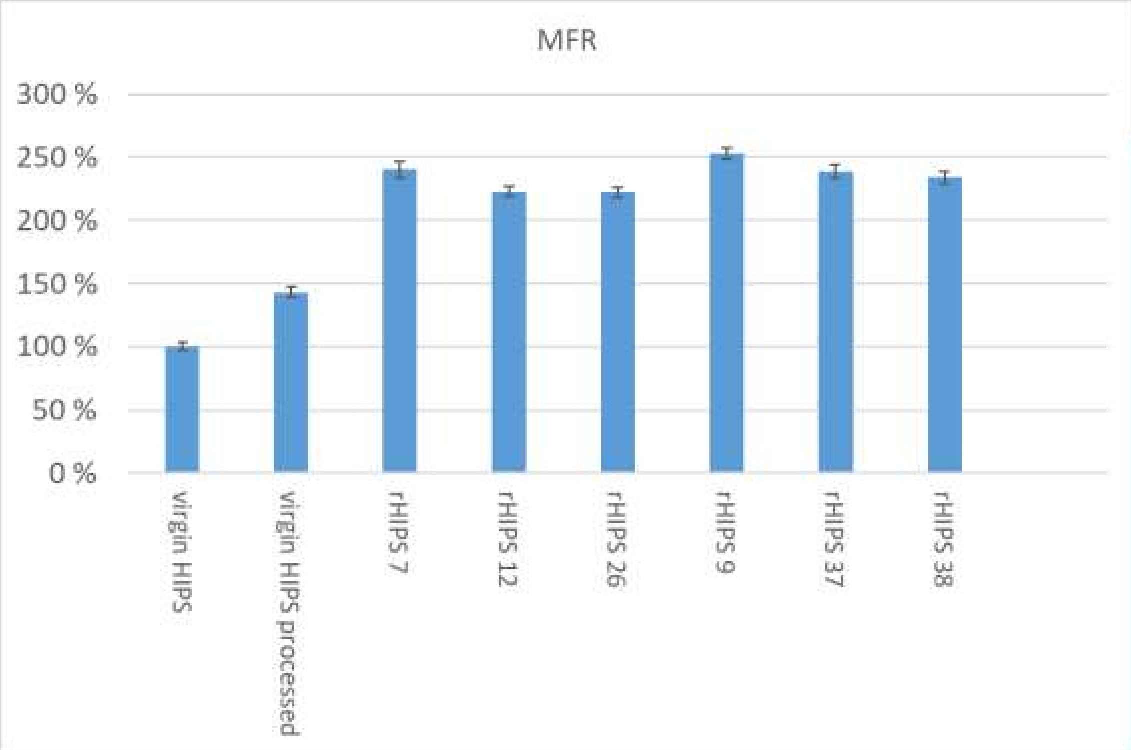

Figure 20. MRF analysis results (200°C, 5 kg). The error bars represent the standard deviation. The results are normalized and compared to primary HIPS result.

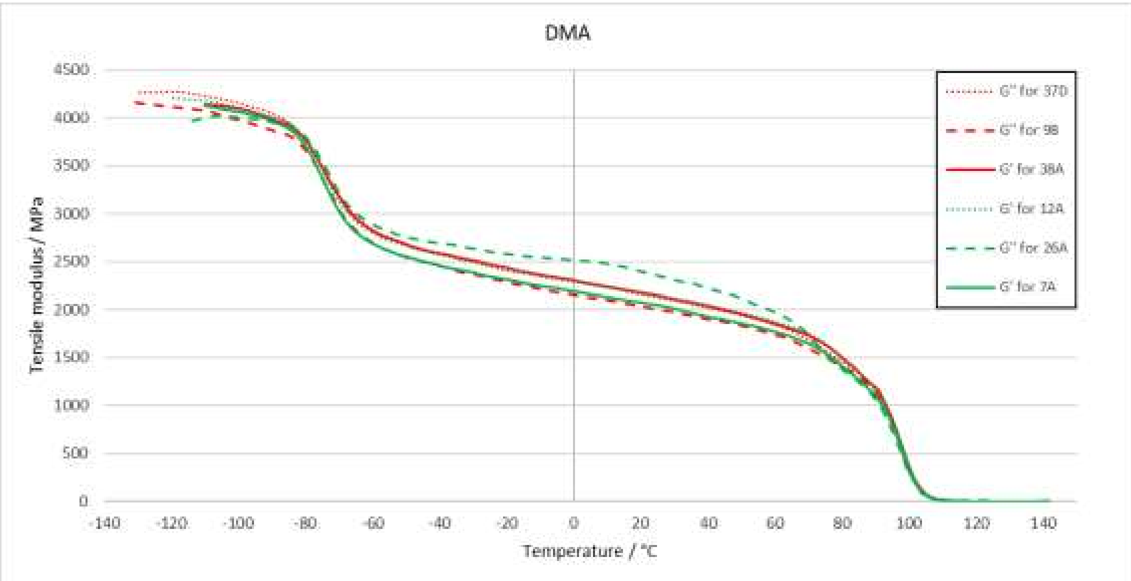

Figure 21. DMA analyses of the different r-HIPS samples.

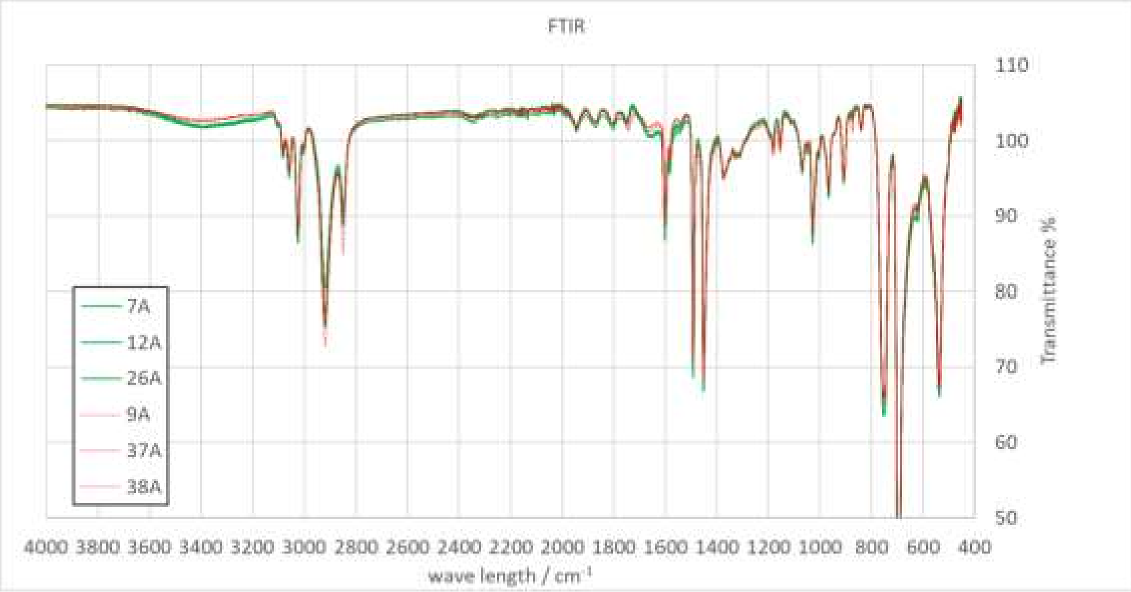

Figure 22. FTIR analyses of the different r-HIPS samples.

Conclusion on the broad analyses of the r-HIPS feedstock was that there was great variation in the impact strength between the samples. However, the largest variation was between the samples *within* each production point, and less variation between the parallel samples from each production point. In all cases impact modification was necessary to return the required impact strength. There was clear no correlation found between the impact strength and other parameter such as MFR, solid viscoelastic properties (DMA) of composition (FT-IR). For example, referring to the MFR data (figure 22) the samples 7, 12 and 26 had the three highest impact strength and samples 9, 37 and 38 had the lowest impact strength. Similarly, the high impact strength was not associated with thermal relaxations or storage modulus (G'') in DMA nor any features seen in FT-IR spectra. In addition, it is noticeable that FT-IR spectra the of the r-HIPS samples was indicating only HIPS and no significant amount any other polymers in the mixture, nor signs of aging or degradation.

3.3.5 Upgrading of r-HIPS

From the feedstock analysis it was obvious that upgrading of the r-HIPS was required. Lab scale screening of the best impact modifiers and other additives and their composition was made, and the best candidates were selected for pilot scale compounding with ZSE 27mm 60D Twin screw extruder (VTT/VAREX). The formulas are their properties are described in Table 17.

Table 17. Upgraded r-HIPS formulas and their properties.

Formulation no.	Recycled content (%)	Charpy impact strength, kJ/m^2	Flexural modulus, GPa	MFI (g/10min, 200°C, 5kg)
Requirement		> 10	>1.5	3-7
DM3-004	70	10.0	1.7	5.6
DM3-005	70	11.1	1.6	6.3
DM3-007	50	10.7	1.6	4.9

During the upgrading of r-HIPS, also the decontamination of the r-HIPS was conducted using melt filtration and melt degassing with the same compounding line (ZSE 27mm 60D Twin screw extruder (VTT/VAREX). Samples for the NIAS analysis were taken after the compounding process but without additives or addition of primary HIPS. Migration test was one for the washed r-HIPS before and after the other studied decontamination steps. Based on the initial results, further processing steps and the testing conditions were re-evaluated. New migration tests of different upgraded HIPS bathes are on-going. For the different batches, additional decontamination procedures such as vacuum oven degassing, milder conditions in the compounding process, extra washing step with alcoholic solvents, and/or alternative migration test conditions were considered. Initial assessment shows that all the studied improvement in the processing and pre-treatment alone indeed reduced the amount or dimers and trimers in HIPS.

3.4 **r-EPDM for washing machine gasket**

3.4.1 Sourcing of suitable EPDM for washing machine gasket application

Cikautxo collaborated with Coolrec in getting access to EPDM gaskets. Samples were manually collected from dismantled end-of-life washing machines at Coolrec facility (Figure 23).

Figure 23. End-of-life washing machine door seals.

The target of the demonstrator was to produce dark grey rubber mixture for producing new washing machine door gaskets, hence dark grey color gaskets were selected for the available source. Light colored (Figure 24) were discarded from the demo.

Figure 24. Example of a door gasket that has been discarded to produce dark grey gaskets due to its colour.

After the collection the gaskets were cleaned with a 1% soap solution and subsequently rinsed to remove dust and dirt. Any visible contamination or objects that remained in the parts, such as plastic inserts and metal brackets, were also removed from the rubber part.

3.4.2 Optimized thermomechanical processing route to recycled o reclaimed EPDM

Broadly described, mechanical recycling of cured EPDM rubber process has two important steps:

STEP 1: Chunking the parts into 4-5mm size granulates by a cutting machine with rotating blades and particle filters for the generated fines to get the desired particle sizes fraction. In the following Figure 25, such cutting machine mechanism is depicted.

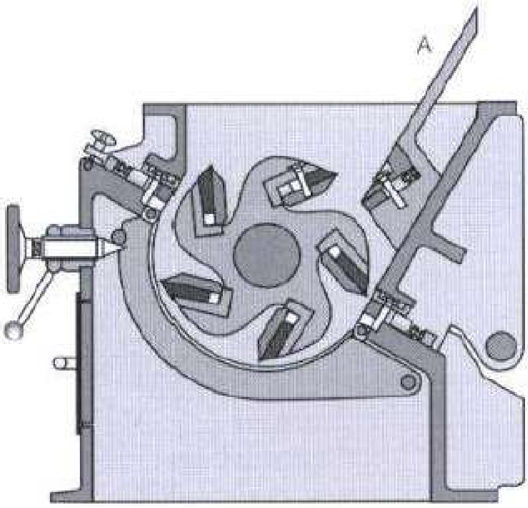

Figure 25. Scheme of the cutting machine for EPDM granulates.

The maximum rotor speed maximum is 450 rpm and the rotor diameter 500mm. After the first step the chunked door gaskets are in the form of 4-5mm granulates which is are suitable to the subsequent micronizing step (STEP 2) (Figure 6).

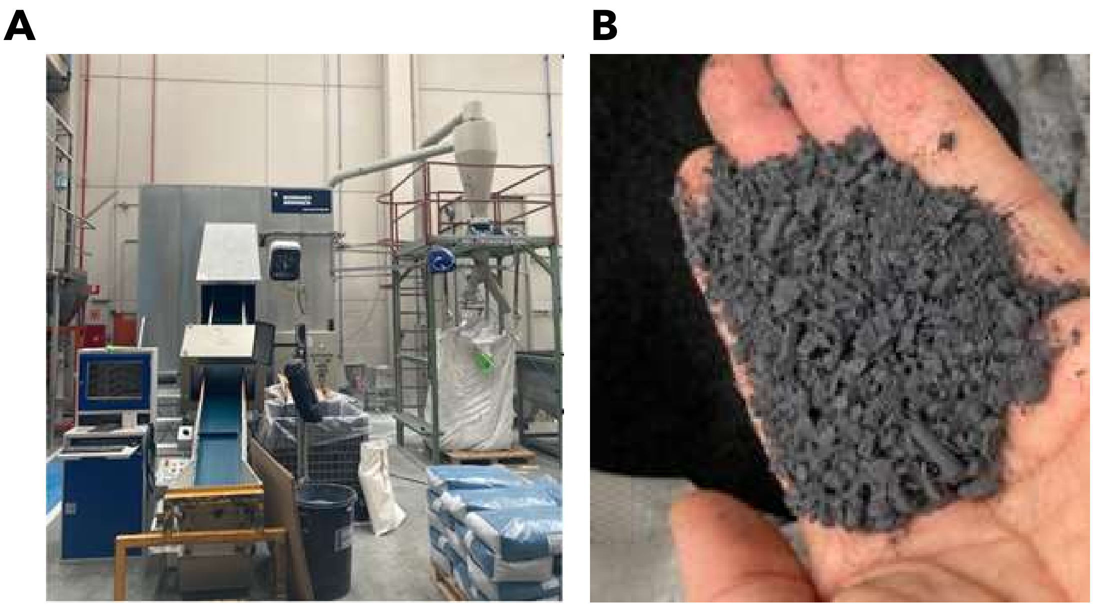

Figure 26. A) The cutting machine installation, B) ground material after the cutting process

STEP 2: The second step is the mechanical micronisation process of the EPDM granulates. For this purpose, special compressing and cutting equipment is used.

With this type of cutting technology a relatively coarse particle size is achieved (< 0.25mm), but, beneficially large specific surface area is generated due to the irregular cut surface of the micronized powder. This helps in achieving good adhesion to the rubber EPDM matrix and results in good mechanical properties of the final EPDM compound. Noteworthy, In this case cut powder is used as solely as particulate filler. The cutting installation is the following Figure 27.

Figure 27. Micronizing installation

The micronized EPDM powder produced with the cutting is shown in the following Figure 28.

Figure 28. Micronised EPDM powder obtained by cutting end-of-life gaskets.

3.4.3 Production of EPDM rubber compound with r-EPDM content

The micronized EDPM powder was used to produce mixtures in an industrial scale. For the mixing, a 150kg internal mixer and a mill installation were used (Figure 29).

Figure 29. 150kg internal mixer and a mill installation for the preparation of PRIMUS EPDM compounds

3.4.4 PRIMUS EPDM compounds

For the PRIMUS demo, the following 3 compounds were generated:

- EPDM compound for washing machine with 0% recyclate
- EPDM compound for washing machine with 10% recyclate
- EPDM compound for washing machine with 20% recyclate

The raw EPDM rubber strips of the 3 compounds are shown in the Figure 30.

Figure 30. rubber compounds with 0%, 10% and 20% of recyclates.

3.4.5 Chemical compatibility of r-EPDM compounds with detergents in elevated temperatures

A comparative study of mechanical properties, thermal and chemical resistance was done for the 3 studied compounds. The results are summarized in the following Table 18.

The following conclusions on the influence of added recyclate to the properties of EPDM rubber compoundcan be drawn from the tests data :

- Tensile strength and elongation at break were both gradually reduced when the amount of recyclate EPDM was increased from zero to 20%. However, up to 20% the material performance remained in the desired range.
- Compression set is suffering from the introduction of the recyclates. Again, at 20% recycled content the compression set was still suitable for the gasket application.
- Abrasion resistance was reduced by the addition of the recyclate, but still inside the required range at 20% loading.
- The heat ageing was slightly worse with the introduction of recyclates. The behaviour of the materials with recyclates up to 20% met the requirements.
- Chemical resistance to powder detergent solution, bleach solution and softener solution was reduced slightly with the introduction of recyclates. The increase of sample volume (uptake of fluid and swelling) was higher with recycIate powder present.
- In general, the introduction of recyclate coming from end-of-life gaskets reduced the mechanical properties, heat ageing resistance and detergent and bleach resistance. However, up to a 20% of recycled content the material still met the specification.

Table 18. Mechanical properties, thermal and chemical resistance of PRIMUS r-EPDM compounds

Compound			DM4-0013 (ref production)	DM4-0010	DM4-0012	
Recyclate content, %			**0**	**10**	**20**	
TEST	Standard	Requirement	DM4-0013 (ref production)	DM4-0010	DM4-0012	Evaluation
Hardness, ShA	ISO 48-4	38 ± 3	37	37	38	OK
Tensile Strength, MPa	ISO 37	>8	10,3	9,4	8,2	OK
Elongation at break, %	ISO 37	>550	835	754	705	OK
Compression set, 24h at 100°C	ISO 815, type B, method A	<50	35	38	48	OK
Abrasion resistance, mm³	ISO 4649, method A	<500	421	481	490	OK
Heat ageing, 72h at 100°C	ISO 188					
ΔShA	ISO 48-4	± 5	4	4	5	OK
ΔTS, %	ISO 37	± 30	2,1	0,9	0,2	OK
ΔEB, %	ISO 37	± 30	-6,2	-12,2	-15,5	OK
Dixan Powder detergent 1,5%, 168h	ISO 1817					
ΔShA	ISO 48-4	± 10	-3	-5	-7	OK
ΔTS, %	ISO 37	± 30	-10	-13	-19	OK
ΔEB, %	ISO 37	± 30	-14	-18	-24	OK
ΔVolume, %	ISO 1817	± 10	7	8,2	9,2	OK
ACE Bleach 2%, 168h 95°C	ISO 1817					
ΔShA	ISO 48-4	± 5	-2	-3	-4	OK
ΔTS, %	ISO 37	± 30	1.9	1.1	0,2	OK
ΔEB, %	ISO 37	± 30	-2,8	-5,9	-9,7	OK
ΔVolume, %	ISO 1817	± 10	3.2	4,6	5,8	OK
Softener 0,2%, 168h 95°C	ISO 1817					
ΔShA	ISO 48-4	± 5	-1	-1	-2	OK
ΔTS, %	ISO 37	± 30	-6,6	-7,6	-11,2	OK
ΔEB, %	ISO 37	± 30	-6,2	-7,9	-12,3	OK
ΔVolume, %	ISO 1817	± 10	7.0	7.6	8,8	OK

4 NEXT STEPS FOR UPSCALING AND PRODUCTION OF FULL DEMO PARTS

4.1 DEMO1: r-PC/ABS for automotive interior

4.1.1 Remaining 2D tests (Q2-Q3/24)

Surface energy measurements of the DM-012-016 at Maier are to be completed before making the final selection of the most suitable formulation for the full-scale demonstrator.

4.1.2 Upscaling the compounding process (Q3/24)

VTT will produce a larger batch of the r-PC/ABS and send this to Maier for 3D -2K injection moulding tests. Tests will be done using the real production moulds.

4.1.3 Injection moulding of the final demo part (Q3-Q4/24)

Maier will manufacture single material demo parts using the 2K-injection mould. The parts will be decoratively painted on the top surface.

4.1.4 Further recycling of the injected 3D parts (Q3-4/24)

Maier will send to VTT few pristine and few painted demo parts. The parts will be re-grinded, injection moulded into standard dogbone mechanical test samples, and tested at VTT for their mechanical properties, aesthetic properties and VOCs after the reprocessing.

4.2 DEMO 2: r-TPV for automotive cooling circuit

4.2.1 Tuning of the melt viscosity of TPV for tube extrusion

TPV pipes are processed as conventional thermoplastic tube extrusion technology. This is possible as the rubber phase is dynamically vulcanized during the compounding of the material, there is no need for any vulcanization or post-annealing process after extrusion of the pipes.

Although the material developed formulations are already suitable for the final demonstrator. The possibility to increase the recycled content by compensating for the non-optimally low viscosity (or high MFI) of the current r-TPV formulations will be investigated further. VTT will carry out rheological measurements and examine the possibilities to modify the melt viscosity of the r-PP-part of the r-TPV formulation. If this proves successfully, then it will be possible to increase the recycled content of the TPV without compromising the flow behaviour in the tube extrusion. Moreover, high viscosity will likely enable easier thermoforming of the extruded pipe by steam, hot air or autoclave (Figure 31).

Figure 31. Extrusion of TPV pipes.

4.2.2 Recyclability of r-TPV

Re-grinding and re-processing of the extruded TPV and r-TPV tubes will be investigated further at Cikatek (Q3-4/24).

4.3 **DEMO 3: r-HIPS compound for extrusion-thermoforming**

4.3.1 Routes to HIPS for reduced level of NIASes (Q3/24)

New migration test results are available in August/24 from UEF. These results will enable comparison between the various optional decontamination procedures for HIPS and r-HIPS, such as vacuum oven degassing, milder conditions in the compounding process, extra washing step with alcoholic solvents, and/or migration test conditions adjusted to get more realistic data relevant for the application. The optimum pre-treatment procedure and material formulation will be then used in the final demonstrator. Results of the initial NIAS screening at UEF will be repeated with more samples, and in the end, also validated by accredited NIAS tests in external laboratory in WP4.

4.3.2 Final demonstrator 3

The size and the form of the final thermoformed HIPS parts has not been decided yet. Possibilities to manufacture full size refrigerator (TLR 6) in EU are known, but availability of materials at production scale (approximately one ton) and the production machinery for thick (>2mm) and wide cast sheet preforms are currently unavailable. Smaller size refrigerator demo has been initially discussed, but no decision has been made thus far. The final demo will be subject of WP4. However, sheet extrusion and thermoformability of the current materials with high recycled content 50-70% has been already validated in small scale (TLR 4).

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4.4 DEMO 4: r-EPDM compound for sulfur-crosslinked washing machine gasket (CIK)

4.4.1 DEMO4 and the final formulation of the EPDM compound

The final formulation will be likely fine-tuned during the continuation of the project in WP4. Reassessment may be needed after full evaluation of the production process of the gaskets with 10% and 20% of recyclates. For example, curing characteristics of the rubber in the production mould are critically important and cannot be adjusted solely based on laboratory testing. First production of the demo washing machine gasket has been scheduled to the week 25/2024, which is after the submission of D3.5.

5 CONCLUSIONS AND DISCUSSION

5.1 Summary of achievements

Broadly summarized, material formulations for each four demo cases have been successful:

r-PC/ABS formulations with high (>80%) recycled content has been generated, which is higher than in known r-PC/ABS commercial grades, that are typically limited into approximately 50-60%. Moreover, the material properties enable the demo part made fully of the r-PC/ABS and with aesthetic painted surface. Single recycled material demo which was not planned or expected originally. Hence, the PC/ABS demo is fully successful and paving the way for the wider industrial application of r-PC/ABS in automotive sector.

Recycled thermoplastic vulcanizate (r-TPV) made of recycled PP thermoplastic and thermo-mechanically reclaimed EPDM rubber has been previously very little investigated. According to the authors knowledge, usage of recycled feedstocks has been demonstrated for the first time in PRIMUS. The generated r-TPV samples have passed all the material thermal aging and chemical exposure tests for cooling circuit materials. The biggest challenge has been low viscosity of the recycled feedstocks, which is essential for maintaining processibility of the r-TPV in tube extrusion and the subsequent thermoforming of the tube. In that sense the recycled content of r-TPV has been limited to maximum of 23%. Much higher recycled content has been demonstrated for injection moulded r-TPV parts which are supporting structures for the cooling circuit. Note: TPV contains 30% inorganic filler, which is naturally not contributing at all to the recycled content.

Promising results have been achieved from the upgrading of r-HIPS flakes from EoL refrigerators. Mechanical properties and melt flow characteristics for extrusion-thermoforming have been upgraded to a level comparable to the primary HIPS material. Recycled content up to 70% has been demonstrated. Mechanical and rheological testing, and NIAS screening has shown that closed loop recycling of HIPS in refrigerator liners is technically feasible.

At least up to 20% of mechanically re-grinded EDPM rubber particles from EoL washing machine gaskets can be used as filler in EDPM compound intended for the washing machine gasket application. Of the three studied grinding technologies, the cutting with blades is the most techno-economically feasible method. Mechanical properties, abrasion resistance, thermal aging behaviour and chemical resistance of the r-EDPM compounds with up to 20% regrind content are very similar compared to the primary EPDM, but the curing kinetics of the EPDM compound gradually change when more regrind is added. Therefore, in the demonstrator the recycled content is limited to 20%.

5.2 Relation to continued developments

In the short term, the next steps of the project work will include the utilisation of material developed in WP3 in the production of the final PRIMUS demonstrations in WP4.

In broader sense, together with the work done on PRIMUS WP1 and WP2, this WP3 supports the development of EU wide standards for reliable assessment of potentially hazardous substances, and the sampling protocols in styrenic plastics (HIPS and ABS) recyclates, aiming to boost their safe use and to enable their high value applications in domestic appliances and automotive sector, for example.

PRIMUS has made notable advances in the aim to demonstrate the viability and profitability of recycling business models through its four demonstrative cases. These cases have showcased the potential for high-value applications using high recyclate content.

The success of the PRIMUS demo cases has reinforced confidence in the using recyclates among the OEM and manufacturing industry, encouraging adaptation and implementation of innovation and in the long-run, investments. The successful demonstration of high performing recyclate recipes for all the demo cases showcases the possibility for businesses to incorporate recycled materials into their products, creating new business for the plastic recycling industry and driving forward the circular economy.

The next steps in the project focus additionally on addressing the business viability of the solutions to prove that recyclate-based business models can be profitable. This in turns supports the overall investments into the field and leads to creation of more jobs within the recycling industry and its associated value chain. The demonstration cases can serve as concrete evidence that economies can thrive on circular economy principles. They provide inspiration for similar sectors to achieve sustainable growth while reducing reliance on virgin fossil resources. The use of demo products as a basis for further recyclate and process development has established a feedback loop for continuous improvement, fostering a culture of research and innovation. In addition, the sustainability of the solutions will be evaluated next.

In conclusion, the PRIMUS project's demonstration cases’ recipes have been successfully developed to be further integrated into scaled up production. The development work has been done in collaboration with the research organisations and universities, OEMs and manufacturers as well as other experts in the field. The project's outcomes are a testament to the power of collaborative effort in driving systemic change towards a more sustainable future.

5.3 Deviations to the plan

The deliverable report D3.5 was submitted two months later than originally planned due to missing essential parts of experimental data at the time of intended submission.