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(Article begins on next page)

1 Development of disposable filtering mask recycled materials: impact
2 of blending with recycled mixed polyolefin and their ageing stability

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8

9 **Abstract**

10 A survey on Covid-19 protecting masks habits carried out on the Italian population at the end
11 of 2020 revealed that disposable face masks are the most used, thus resulting in a considerable
12 quantity of waste. Therefore, a separate collection of these devices based on local platforms
13 such as schools, offices and factories is proposed. This would limit collecting costs, ensure the
14 origin and quantity and simplify the sanitizing treatment of the masks, in order to allow their
15 introduction into the industrial recycling stream of plastic materials.

16 In this scenario, an urban separate waste collection material (namely mixed polyolefin) was
17 selected and melt blended in several ratios with the disposable filtering masks. Two recycling
18 solutions have been envisaged: the use of the filtering part alone or the joint introduction of the
19 ear loops.

20 Compared to the mixed polyolefin, the resulting recycled materials from filtering face masks
21 have lower viscosity but comparable toughness and superior stiffness and strength. The
22 presence of ear loops in the recycled improves the rigidity but slightly decrease the viscosity
23 and worsen both the strength and toughness. Furthermore, conceiving an outdoor application,
24 the stability to photo oxidation was investigated through an accelerated ageing process. The

25 elaborated degradation rate of the masks material is similar to a commercial polypropylene,
26 while for the mixed polyolefin is greatly reduced. The mixed compounds have intermediate
27 degradation rates.

28 **Key words**

29 Face mask; mechanical recycling; recycled mixed polyolefin; mechanical properties; photo
30 degradation; waste.

31

32 1. Introduction

33 An updated published research on recycling of plastic packaging, reported that in Europe only
34 32% of polymers were collected for recycling, while 25% were sent to landfill and 43% were
35 incinerated (Schyns and Shaver, 2020). It is therefore clear the need to develop and promote
36 methods that increase the recycling fraction compared to the other two solutions recognized as
37 less sustainable (Bora et al., 2020; Gu et al., 2017; Sharma et al., 2020). Due to the currently
38 pandemic situation, one of the main challenges is represented by disposable face masks, which
39 mostly ended up in landfills or incineration (Armentano et al., 2021; Cesaro and Pirozzi, 2020;
40 Rhee, 2020; Selvaranjan et al., 2021). The main constituent material was found to be
41 polypropylene (PP) (Battezzore et al., 2020). According to a study conducted in Japan by
42 Narita et al. (Narita et al., 2002), the CO₂ emissions from the production of 1 kg of PP would
43 be 1.4 kg. The main body of the surgical mask is made up of just some grams, however, must
44 be multiplied by the huge number of people who use them globally and by the fact that are
45 single use devices.

46 The most sustainable scenario in this field is to mechanically recycle the waste to obtain a
47 regenerated material that can be used in place of a first use material and possibly recycle it over
48 and over again (Bora et al., 2020).

49 In a recent article it was shown that the materials with which most disposable masks are made,
50 are potentially recyclable between 78 % and 91 % of the total weight (Battezzore et al., 2020).
51 However, the same article also highlighted the variability of the materials used by many
52 producers on the market and the difficulty in mechanically recycling the different components
53 all together. In addition, other types of masks, such as FFP2, can be collected and recycled but
54 have a further heterogeneity of materials (Crespo et al., 2021). Therefore, the properties of the
55 recycled materials obtained require an adequate development to fulfil different industrial
56 application fields. Lastly, it has to be taken into account that the accurate amount of material
57 potentially available nowadays is still unknown (Prata et al., 2020; Singh et al., 2020).

58 Starting from this last aspect, forecasts in the early stages of the pandemic estimated a large use
59 of these devices (1 billion face masks each month in Italy - on March 2020 as reported by the
60 Politecnico di Torino (Torino, 2020). In fact, it is extremely difficult to quantify the actual use
61 of disposable masks as there is no data on sales or quantities disposed. Due to the lack of
62 information, the first part of the article focuses on the results of a survey that directly
63 investigated the citizens habits. This was carried out in December 2020 in Italy and received
64 over 1000 answers. The main result is that the majority of respondents still use disposable
65 “surgical” masks and are willing to throw them into a separate collection system. Although no
66 economic assessments are presented, a dedicated recycling system spread at local level must be
67 provided. Its affordability will be directly connected to the proximity of the collection site.
68 Moreover, the elaborated data showed that the amount of waste generated by the disposed
69 masks is not marginal compared to the other municipal waste and therefore is worth being
70 industrially taken into consideration. This first important result made meaningful the further
71 development of the research.

72 In the study, a new recycle perspective for disposable masks is presented, together with other
73 urban waste plastics. A mixed polyolefin (MPO) waste fraction separated using flotation sorting
74 of plastic wastes was selected as dilution source since it requires minimal sorting and is much
75 cheaper than neat PP and PE (Schyns and Shaver, 2020).

76 To get rid of the possible influence of the heterogeneity of mask material (Armentano et al.,
77 2021), in the present article only the “surgical” masks are considered and two approaches were
78 deepen: use only the filtering part of the masks (M) or the filtering part together with the ear
79 loops (ME). From the first approach, filtering part consists of about 78% of the total weight and
80 is made of polypropylene (PP), like all the masks previously analyzed (Battezzore et al.,
81 2020). This material can be separated by floatation in recycling plants for plastics. In the second
82 option, all the plastic parts were considered together, reaching over 90% of the total weight but
83 having a heterogeneous system. In this latter, the separation is not necessary except for the

84 metal nose clip. In both cases, the recycling study was carried out using only new disposable
85 face masks.

86 The materials obtained with different mixing ratio of MPO and M or ME were characterized
87 for their rheological and mechanical properties and correlated to their morphological
88 peculiarity. Finally, designing possible outdoor applications, the photo-oxidation ageing was
89 investigated pointing out the differences that adding M or ME has on the MPO.

90 2. Materials and Methods

91 2.1. Materials

92 The disposable face masks were purchased from Xiantao Wenjun non-woven co., Ltd. (China)
93 Standard GB/T 32610-2016.

94 The neat filtering face mask is used and coded as M in the paper while the filtering face mask
95 together with the ear loop parts are coded as ME (Characterization added in the SI and Figure
96 S1).

97 The recycled mix of PP and PE (MPO) obtained from municipal wastes is a commercial grade
98 Bretene 003GR160 from Breplast S.p.a., made of 70 wt.% of PP and 30 wt.% of PE from the
99 supplier datasheet and coded as rPP in the text.

100 2.2. Survey

101 The online survey was created with “Google Forms” and consisted of 15 multiple choice
102 questions in Italian language, 9 of them were compulsory while the others requested only if
103 relevant. All the question have been translated and listed in the Supporting Information.

104 The survey was promoted via local newspaper, social networks and word-of-mouth, in order to
105 achieve the larger spreading possible. Data collection lasted for one month before Christmas
106 holiday when in Italy were witnessing the second wave of the pandemic. During this period the
107 on-site working was allowed, even though remote working was encouraged. The activities of

108 the catering were allowed only until 6.00 pm and the school were open with alternation of on-
109 site and distance learning.

110 2.3. Face mask mechanical recycling process

111 To have a uniform and homogeneous starting material, a primary recycling step was performed
112 using an internal mixer Brabender. A quantity of about 50 g of the mask filtering part were
113 separated manually from the other parts and cut in squares of about 10 mm, then processed at
114 190°C and 30 rpm for 10 minutes. The second recycling phase involved the mixing of the
115 material obtained in the first step with a co-rotating twin screw micro extruder DSM Xplore 15
116 ml microcompounder. The screw speed was maintained at 50 rpm for the feeding time and
117 increased up to 100 rpm for the residence time, fixed for all runs at 2 min. The heating
118 temperature was selected at 190°C.

119 The mixing of different concentrations of M or ME in the rPP was investigated: the first number
120 in the material code is the weight ratio of rPP and the last part of the name is the weight ratio
121 and type of material from the recycled mask (e.g. 25rPP75M is the formulation with 25 wt.%
122 of rPP and 75wt.% of M).

123 The extruded materials were manually pelletized and placed inside a mold made of a 0.1 mm
124 thickness aluminum foil with a 100x100 mm² hole inside two metal plates. Using a hot
125 compression molding press Collin P200T at 190°C for 2 min under a pressure of 5 MPa the
126 thin film was obtained. The same conditions were adopted for the discs for rheology tests, with
127 a diameter of 25 mm and a thickness of 1 mm.

128 2.4. Ageing

129 UV ageing of films has been carried out by irradiations in air in a SEPAP 12/24 unit (Atlas
130 Material Testing Technology LLC) at a wavelength > 300 nm. The apparatus was equipped
131 with four medium-pressure mercury lamps with borosilicate envelope able to filter wavelengths
132 below 300 nm. It was designed for accelerated artificial UV ageing tests in conditions

133 comparable to natural outdoor weathering. Samples were homogeneously exposed on a rotating
134 support in the center of the chamber. The surface temperature of the samples was accurately
135 controlled and maintained at $60^{\circ}\text{C} \pm 1^{\circ}\text{C}$ through a thermocouple placed behind a reference
136 film with similar chemical composition as the exposed samples.

137 2.5. Analyses

138 The thermal properties have been evaluated by Differential Scanning Calorimetry (DSC) and
139 Thermogravimetric analyses (TGA). DSC measurements have been performed by a DSC TA
140 Q20, using 8 ± 1 mg of sample and the chamber has been purged by nitrogen. Each sample has
141 been heated from -50°C to 300°C at $10^{\circ}\text{C min}^{-1}$. The melting temperature, as well as the melting
142 enthalpy have been obtained from the peak maximum and as the integral of the area under the
143 heat flow curve, respectively.

144 TGA were carried out in air, from 50 to 700°C with a heating rate of 10°C/min . The used
145 equipment is a Discovery thermo balance (TA Instruments) (experimental error: ± 0.5 wt.%, \pm
146 1°C) with samples of approximately 10 mg placed in open alumina pans and fluxed with air at
147 25 mL/min.

148 The morphologies of the film cross sections of rPP and compounded materials were examined
149 after mechanical tests and gold-metallization using a EVO 15 Scanning Electron Microscope
150 (SEM) from Zeiss (beam voltage: 20 kV working distance: 8.5 mm). Elemental analysis was
151 carried out by EDS (Energy Dispersive X-ray spectroscopy) using an X-ray probe (Oxford
152 Ultim Max, model 40).

153 Attenuated Total Reflection (ATR) was used to investigate chemical composition of the rPP
154 and ME using a Frontier FT-IR spectrophotometer (16 scans and 4 cm^{-1} resolution, Perkin
155 Elmer) equipped with a universal ATR sampling accessory and a diamond crystal.

156 The rheological properties of the melt blended materials were analyzed using an ARES
157 rheometer fitted with a 25 mm parallel plate geometry. The gap between the plates was set to 1

158 mm. Dynamic strain sweep tests were carried out to confirm the linearity of the viscoelastic
159 region up to 10% strain at 100 rad/s frequency. Frequency sweeps were carried out to determine
160 the complex viscosity (η^*) over a frequency range of 0.1–100 rad/s at 10% strain. Tests were
161 performed under a nitrogen atmosphere to avoid any degradation.

162 The complex viscosity curves of materials have been fitted using a modified Carreau model
163 (Filippone et al., 2015):

$$164 \quad \eta^*(\omega) = \frac{\eta_0}{[1 + (\lambda\omega)]^{(1-n)}} + \frac{\sigma_0}{\omega}$$

165 Where σ_0 is the melt yield stress, η_0 is the zero shear viscosity, λ is the relaxation time and n is
166 the dimensionless power law index.

167 Tensile tests were performed at room temperature using a loading cell of 50 N (error <0.25%),
168 a strain rate of 1 mm/min and a gauge length of 20 mm with an Instron 5966 model machine
169 equipped with 250 N rubber face pneumatic grips. The specimens for the stress–strain analyses
170 were 40x10x0.1 mm³ obtained by cutting the compression-molded films with scissors. Three
171 specimens were used for each formulation and the average values and corresponding standard
172 deviations of the tensile modulus (E), elongation at break (ϵ) and maximum tensile strength
173 (σ_M) were calculated and reported.

174 The samples for the mechanical tests were conditioned at 23°C and 50% of relative humidity
175 before analyses.

176 The compression molded films were subjected to accelerate ageing using a SEPAP 12/24 unit
177 and monitored with a Frontier FT-IR spectrophotometer (16 scans and 4 cm⁻¹ resolution, Perkin
178 Elmer) equipment. The photo-oxidation has been followed by monitoring the intensity of the
179 maximum absorbance in the 1800-1690 cm⁻¹ range (C=O vibration stretching band range) as a
180 function of exposure time. In order to avoid differences due to film thickness absorption, the
181 degradation peak has been normalized to the absorption peak at 2723 cm⁻¹ (C-H vibration

182 stretching band of PP). The Oxidation Induction Time (OIT) has been calculated as the time at
183 which the C=O peak starts to increase linearly with time and the slope of the line after the OIT
184 has been indicated as the degradation rate.

185

186 3. Main results and implications of the survey on the use of 187 disposable face masks

188 The survey was published online in December 2020 via Google's web platform and consisted
189 of several questions with multiple choice answers. The main cross-section of respondents
190 focused on young people (more than 60% are under 40 and the 35 % is in the 20-30 age group
191 - Figure 1a) from the North of Italy (84%).

192 According to the 2020 Italian population census (ISTAT), the amount of citizens between 15
193 and 20 years old was the 4.8% of the population, while the age group 20 - 40 represented the
194 22%, the range 40-60 years was the 31% and the over 60 reached approximately the 30 % of
195 the overall population. Lastly, the under 15 years old group accounted for the 13% but does not
196 have entirely the legal obligation to wear masks and has not been greatly reached by the
197 presented survey.

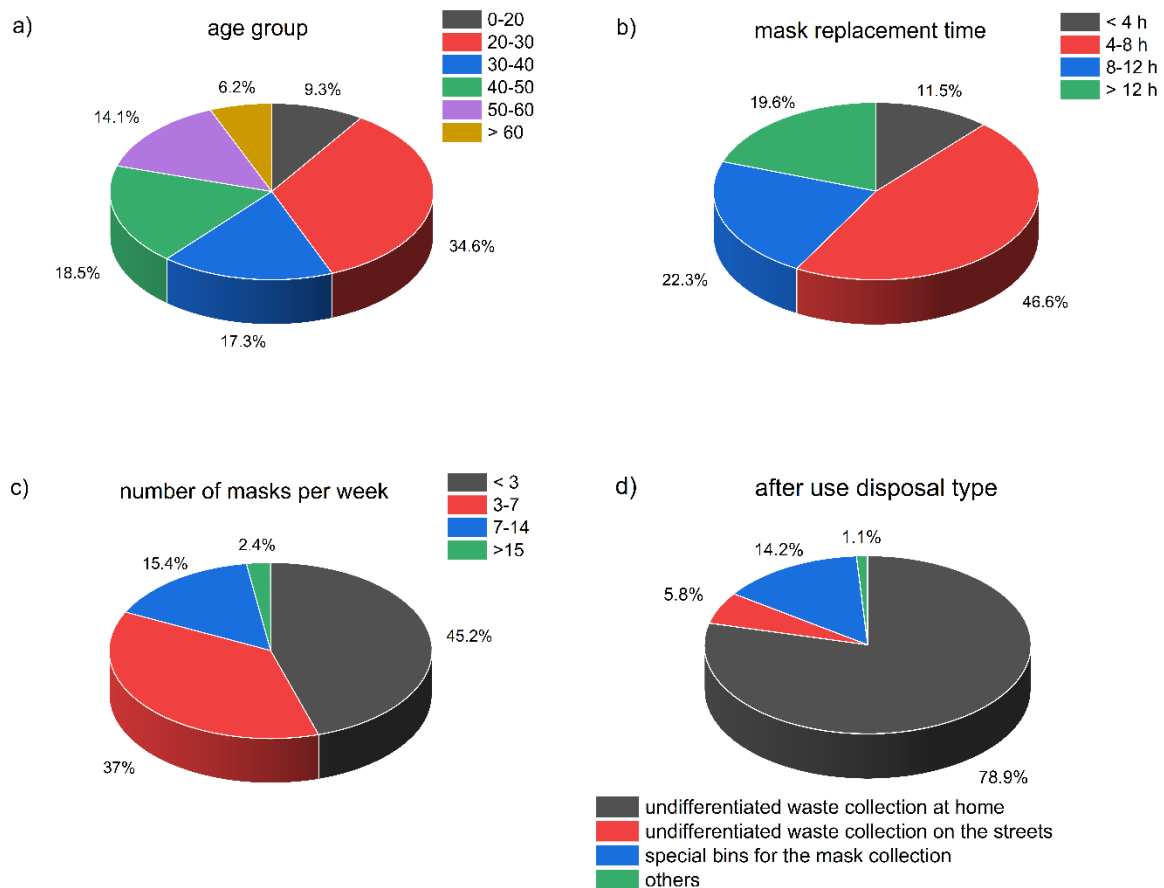
198 Considering that more than 80 % of respondents are in the age group 20-60 and that on the
199 overall Italian population the same range accounts for over 50% (about 30 million people), the
200 survey on the use of masks could be taken as particularly reliable of this subset of population.

201 Also, in this type of interview it is important to consider if a statistically significant sample is
202 reached. The general rule is that the larger the sample size, the greater its validity. The
203 confidence interval, also called margin of error, is the plus-or-minus figure usually reported in
204 opinion poll results. The confidence level, expressed as a percentage, represents the value of
205 the total population that is within the confidence interval. For a sample size of 800 people (the
206 interviewed in this survey in the 20-60 age group), a population of 30 million and a standard
207 confidence level of 95%, the confidence interval is from 1 to 3%. This means that all the data
208 subsequently reported can be considered with a plus-or-minus value of 1-3%.

209 Considering all the interviewed, more than 55% declared that they are working or studying at
210 the workplace where, about half of them, receive the personal protective equipment.

211 Referring to the masks, 78% of respondents declared to use disposable surgical masks and 47%
212 of those questioned replaced it after 4-8 hours of use. Moreover, 45% of the interviewed used
213 less than 3 masks per week, but 37% said they use between 3 and 7 masks and 15% between 7
214 and 14 (main data are reported in Figure 1, more data in the SI Figure S2). By making a rough
215 statistical calculation in which the frequency found is multiplied for each sampling class, an
216 average of 4.5 masks per week can be estimated. Multiplying this number by 0.78 (the
217 percentage that declared to use this kind of masks) and assuming the survey representative of
218 about half of the Italian population (30 million of people), it is possible to estimate the use of
219 105 million masks per week and therefore about 5 billion per year. This estimated value is still
220 relevant, even though is less than half of that estimated in March 2020 by the Politecnico di
221 Torino and published in a document used by the Italian Ministry as reference for the device
222 supply (Torino, 2020).

223 The last part of the survey referred to the habit of masks dispose after use. 79% declared to
224 dispose them in the unsorted waste collection at home, while 6% directly in the unsorted bins
225 on the streets and 14% in special bins in the workplace or schools, 1% in another way (Figure
226 1d). Finally, the last question in the survey, probably the most important related to the theme
227 of this scientific article, stated: "If there would be the possibility to collect the after use masks
228 in a dedicated bin in your city, would you do it?" 93% answered yes. This important propensity
229 makes a strategic recycling program more likely to be designed.



230

231 **Figure 1. Online survey results regarding the use of masks: age groups of**
 232 **participants (a), time declared for “surgical” mask replacement (b), number of**
 233 **disposable “surgical” masks used per week individually (c), habit of mask disposing**
 234 **after use (d).**

235

236 Based on the previous data and considering only the weight of the filtering part, with an average
 237 use of 4.5 masks per week, a quantity of 600 g/year per capita is generated. This value has then
 238 been compared to the current waste situation in Italy.

239 From the data on separate collection it can be deduced that in 2019 the percentage of municipal
 240 waste per capita was 500 kg/year (Frittelloni et al., 2019) and around 20 kg/per capita of these
 241 are plastics sent to recycling plants (2020). In this regard, the previously calculated quantity of
 242 possibly recycled masks material (0.6 kg/per capita) would be only 0.1% of the all municipal
 243 solid waste, but about 3% of the total quantity of recyclable plastic material. In addition, taking
 244 into account that PP is only a fraction of all recycled plastic (Ragaert et al., 2017), this
 245 percentage would still be much higher.

246 In addition to this evaluation, it is also necessary to consider how to carry out the collection.
247 Due to the need to sanitize the masks (Armentano et al., 2021; Rubio-Romero et al., 2020;
248 Schwartz et al., 2020), it is evident that they must be collected separately from other wastes.
249 Thus the specific mask collection can be easily arranged only in densely populated centers e.g.
250 in a city with 50000 inhabitants or in factories and schools where such devices are distributed,
251 in order to have a predictable quantity and homogeneity over time. Only after sanitization they
252 could be delivered to recycling plants and treated as the polyolefin fraction found in urban
253 separate waste collection.

254 As an example, the numbers referring to students in public schools will be discussed. According
255 to the data published by the Italian Ministry of Education (MIUR, 2020) in 2020-21, the number
256 of students in primary and secondary schools is 6.6 million people. Because of the fact that on
257 both scholastic levels is mandatory to wear masks, these students are provided with a disposable
258 face mask every day. Considering about 200 days of attendance per year, this results in over
259 1.3 billion masks disposed, corresponding to 4000 tons per year produced throughout Italy.
260 Even considering a scholar year as 2020-21 where the days in attendance were about half of the
261 total (Marcello, 2021), the quantity remains high. This material, not only would be relatively
262 constant and easy to recover, but is actually already collected in each school and thrown into
263 the unsorted wastes. The before discussed separate collection at local level, is even more likely
264 to be designed if considering the regional numbers of wasted masks in school. Considering only
265 a region like Piedmont, there are approximately half a million students generating about 1 ton
266 per day of used masks (300 tons per year). Such quantity could power a regional recycling plant
267 which usually would processes over 100 kg per hour. The amount would be grater if also
268 universities and factories would adopt the same collection strategy.

269 4. rPP characterization

270 A physical-chemical characterization of the recycled MPO was obtained through DSC, TGA,
271 FTIR and SEM investigations. Through these analyses it was possible to determine that, in
272 addition to the two main materials declared in the technical data sheet, other plastic materials
273 and fillers are present. As can be seen from the DSC analysis (Figure 2a), in addition to the two
274 peaks related to the crystalline part of PE (127°C) and PP (166°C), there are two other peaks
275 centered at 209 and 249°C that can be hypothesized as PVC or PA and PET. By deep evaluating
276 the proportions of the enthalpy areas, the crystalline quantity of the latter appears to be much
277 smaller than the peaks assigned to PP and PE. Therefore, even if this evaluation is limited to
278 the crystalline part of the materials, a presence of around 6% of these can be estimated.

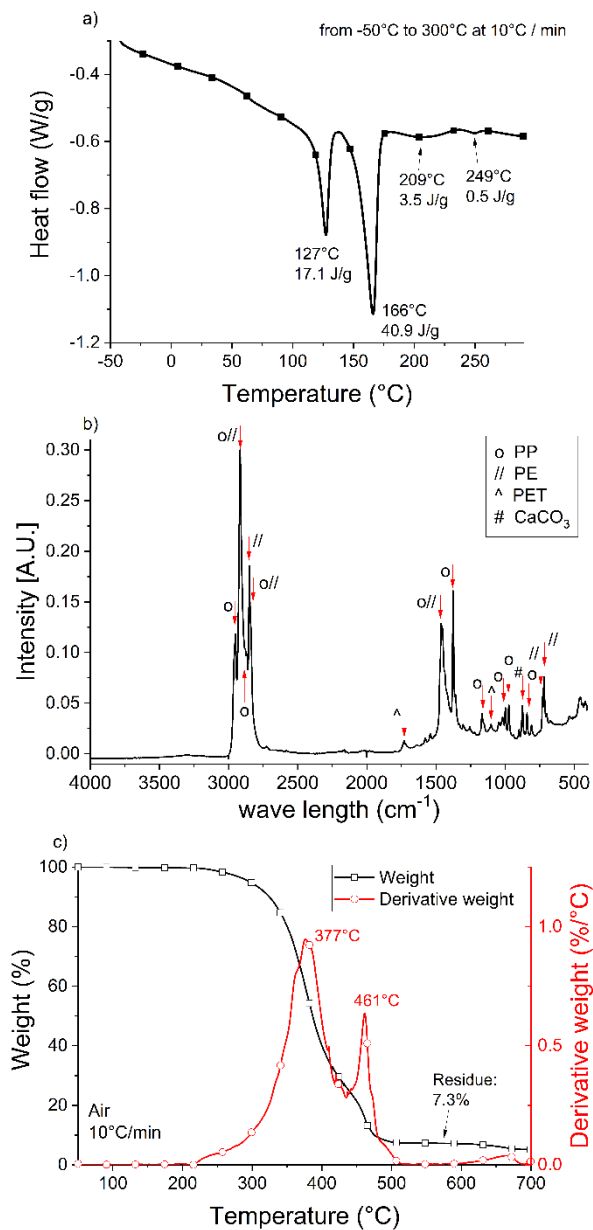
279 The FTIR spectroscopy shows the presence of the typical peaks of PE and PP in addition to the
280 ones distinctive of PET and calcium carbonate (Figure 2b). The attributions are detailed in the
281 SI Table S1 (Battegazzore et al., 2020 Gopanna et al., 2019 Socrates, 2004 Kanelli et al., 2015).

282 From the TGA analysis in air (Figure 2c), it can be seen as the thermal degradation of the
283 material follows two main degradation phases centered at the temperature of 377 and 461°C.
284 The residue at 550°C is 7.3 % and can be attributed to the inorganic filler present.

285 In the morphological and elementary analyses carried out on a section of the material (Figure
286 3), a multiphase structure has been observed. The presence of the inorganic filler (e.g. point 1
287 in Figure 3) was confirmed thanks to the backscattered electrons images and the elementary
288 analyses, of which the results are: C (~38% atomic), O (~44% atomic) and Ca (~16% atomic).
289 Moreover, an XRD analysis was also carried out on the residues after thermo-oxidation at
290 600°C (reported in the SI Figure S3) to confirm the identification of the filler as calcium
291 carbonate. In addition to the inorganic filler, the morphology presents a second phase in
292 spheroid shape within the matrix (Figure 3 point 2): C (~99% atomic) and Ca (<1% atomic).
293 Probably, the second phase is the PE as already reported in the literature for blend of PP/PE

294 (Lin et al., 2015) and the presence of Ca is due to the penetration of the EDS into the bulk of
295 the material where calcium carbonate particles may be present.

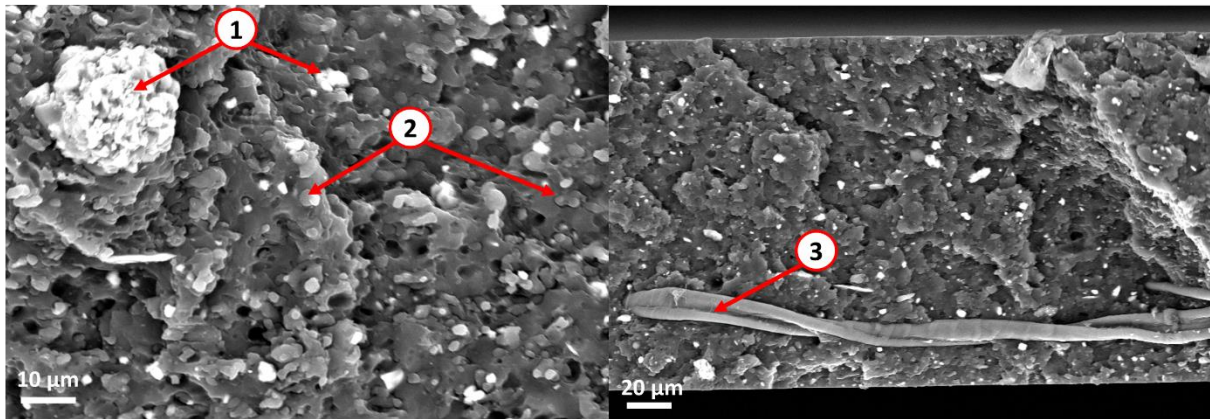
296 An accurate observation of the fracture portions of the material revealed the presence of other
297 phases different from those already reported within the matrix. Such morphologies resemble
298 fibers or result of crushing of packaging. The elemental analysis detected the presence of C (~
299 75% atomic) and O (~ 25% atomic) (Figure 3 point 3) which, considering the results reported
300 by the DSC and ATR analyses (Figure 2a and b), can be assimilated to PET.



301

302 **Figure 2. rPP characterization with DSC from -50°C to 300°C at 10°C min⁻¹ (a); ATR**
303 **from 4000 to 400 cm⁻¹, where the peaks associated to PP (Gopanna et al., 2019) are**

304 high lined with (o), PE (Socrates, 2004) with (/), PET (Kanelli et al., 2015) with (^),
305 calcium carbonate (Battezzore et al., 2020) with (#) (b) and TGA from 50 to 700°C
306 at 10°C/min in air (c).
307



308

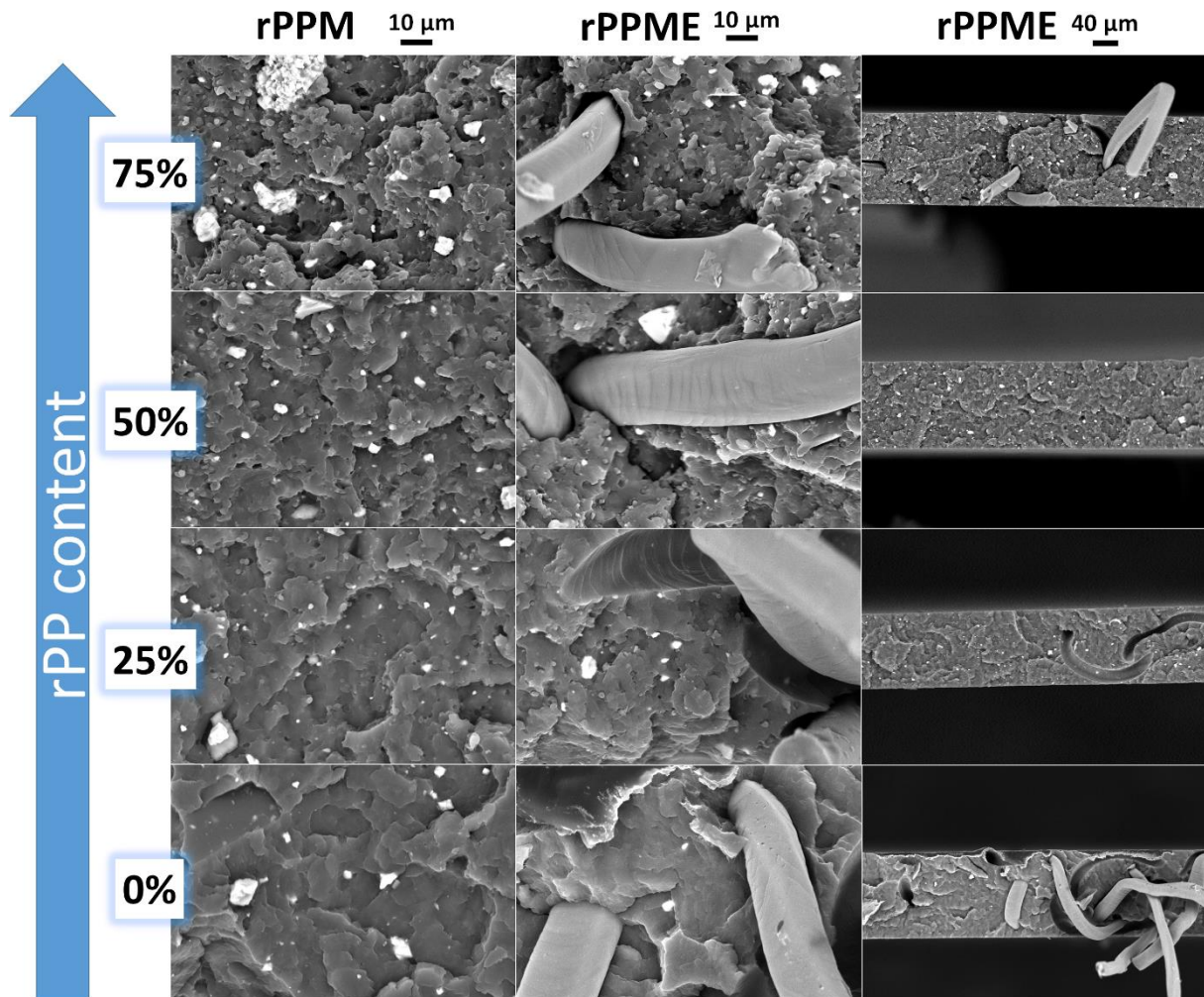
309 **Figure 3. SEM magnifications of rPP cross-section and three characteristic points**
310 **where EDS analyses were carried out.**

311 5. Properties of the compounded materials (rPPM and rPPME 312 systems)

313 In a previous article (Battezzore et al., 2020), the recycling of the disposable mask materials
314 separated from other recycled plastics was assumed. Conversely, in this new investigation, the
315 influence on the main properties of a commercial material obtained from the recycling of urban
316 waste with the masks was assessed.

317 5.1. Morphology

318 The first analysis carried out concerns the morphology of all the formulations. The purpose of
319 this analysis is to verify the distribution of the filler and the ear loops in the compounded
320 materials. The rPPM series shows both the gradual dilution of calcium carbonate and the
321 progressive disappearance of the second spherical polymer phase (PE) with the increasing in
322 the mask content. On the other hand, the rPPME series clearly shows the fibers of the ear loops
323 when not uniformly distributed in the matrix. In fact, as the presence of material deriving from
324 ME is diluted, the fibers are less frequent but always concentrated in some areas. Moreover,
325 can be appreciated that the adhesion between the fibers and the matrix is poor, indeed, there are
326 gaps between the fibers and the matrix as well as imprint left by fibers pulled out or detached.



328

329 **Figure 4. SEM magnifications of rPPM and rPPME formulations. From bottom to**
 330 **top, the figures show an increasing amount of rPP as indicated by the band on the left**
 331 **side. The first column reports the rPPM formulations, while the second and third**
 332 **ones the rPPME formulations at two different magnifications.**

333

334 5.2. Rheology

335 The graphs of the complex viscosity of formulations based on rPPM as a function of the
 336 frequency are shown in the SI Figure S4.

337 M sample shows a typical PP-like behavior with a Newtonian plateau in the low frequency
 338 range and a shear thinning behavior as the frequency increases. As already reported in the
 339 literature (Battezzore et al., 2020), the viscosity of this material is extremely low. Conversely,
 340 the rPP material has higher viscosity than M and present a yield stress behavior in the low

341 frequency region. This is mainly attributable to the largest amount of filler present in this
 342 material if compared to M.

343 These two aspects can be numerically evaluated thanks to the fitting with the Carreau-like
 344 equation, in which η_0 and σ_0 parameters represent the apparent viscosity at zero shear and the
 345 yield stress, respectively.

346 Observing the values listed for all the formulations in Table 1, it can be noted that at 190 °C the
 347 η_0 progressively increases from 171 Pa s of M to 3397 Pa s of rPP, while σ_0 changes from 0 Pa
 348 to 234 Pa. A similar behavior was observed also at the highest temperature studied (230°C) as
 349 reported in the SI Figure S4 and Table 1.

350 This trend is due to the uniform distribution of the filler in the matrix of all the formulations
 351 investigated, which allows to have a progressive and predictable behavior.

352 Thanks to this first analysis campaign, it was found that mixing M with rPP can improve the
 353 potential low viscosity problem of M. Indeed, the rheological properties are progressively
 354 shifted towards those of the rPP and therefore a formulation can be designed to have certain
 355 tailored rheological characteristics.

356 **Table 1. Data from Carreau like fitting of rPPM system.**

Name	190°C				230°C			
	η_0 [Pa.s]	λ [1/s]	n	σ_0 [Pa]	η_0 [Pa.s]	λ [1/s]	n	σ_0 [Pa]
rPP	3397	0.78	0.53	234	1810	0.74	0.62	148
75rPP25M	1810	0.50	0.57	81	992	0.25	0.60	52
50rPP50M	788	0.28	0.63	12	435	0.22	0.69	5
25rPP75M	346	0.13	0.67	3	184	0.09	0.73	1
M	171	0.05	0.66	0	88	0.02	0.67	0

357

358 **Table 2. Data from Carreau like fitting of rPPME system.**

Name	190°C				230°C			
	η_0 [Pa.s]	λ [1/s]	n	σ_0 [Pa]	η_0 [Pa.s]	λ [1/s]	n	σ_0 [Pa]
rPP	3397	0.78	0.53	234	1810	0.74	0.62	148
75rPP25ME	1471	0.71	0.53	156	1042	0.72	0.59	125
50rPP50ME	1341	0.84	0.54	181	886	0.78	0.60	161
25rPP75ME	1736	1.15	0.53	396	1545	2.14	0.58	183
ME	1852	2.59	0.55	299	1377	2.47	0.54	136

359

360 Considering the ME materials, the most impressive rheological characteristic to be highlighted
361 is the higher viscosity compared to M samples and, consequently, the recycled system rPPME
362 has a higher viscosity compared to the rPPM counterpart (see η_0 in Table 2 versus Table 1).
363 This is due to the ear loops that remain intact without melting at these temperatures. In fact,
364 differently to what was studied in the previous article (Battezzore et al., 2020), the ear loops
365 are based on PET fabric and this increases the viscosity of the recycled material. In addition, a
366 yield stress behavior (σ_0) is already present in the neat ME material as was observed in the rPP
367 (Figure S4 and Figure S5).

368 However, it has to be noted that the properties of the intermediate formulations are not strictly
369 proportional to that of the two boundaries (ME and rPP). Indeed, they have both viscosity and
370 yield stress lower than expected.

371 These variations could be due to the heterogeneity of the materials as highlighted in the SEM
372 observations. A greater local concentration of ear loops fibers would actually increase or lower
373 both the viscosity and the yield stress.

374 Despite these limitations in the accurate determination of rheological properties of the rPPME
375 system, it is evident that both the viscosity and the yield stress of all materials are in the same
376 order of magnitude as neat rPP (SI Figure S5).

377 Furthermore, the "crossover point", which defines the boundary between mainly viscous and
378 mainly elastic behavior, for rPP is at $\omega=51.2$ rad/s with a value of $G'=G''$ of $2.1 \cdot 10^4$ Pa at 190°C
379 (details in the SI Figure S6).

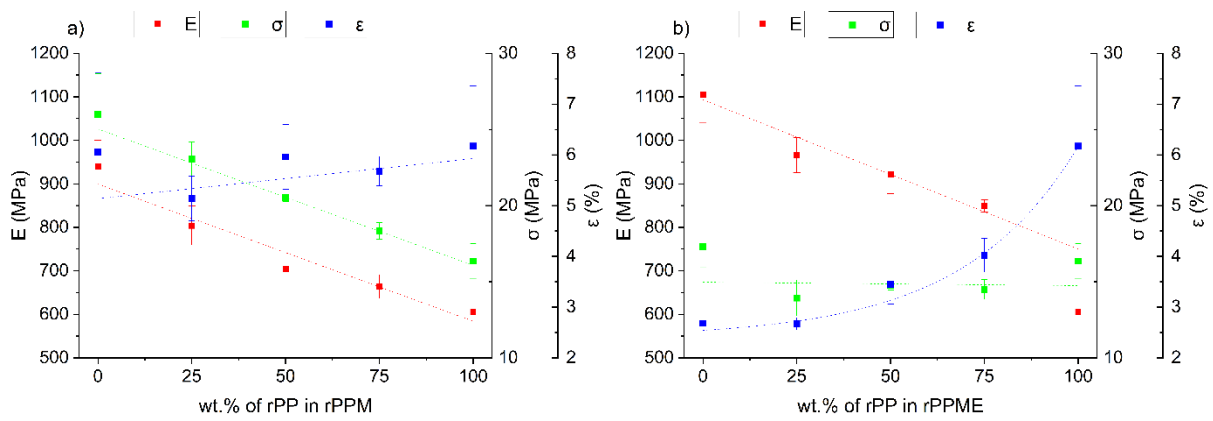
380 By mixing the material with the masks, the crossover point is shifted to higher frequencies due
381 to the lower molecular weight of the latter polymer.

382 This shifting is less intense in the rPPME formulation than in the rPPM, as can be seen for the
383 25% of mask content formulations. In the first case, it is still possible to find the crossover point
384 (75rPP25ME 65.4 rad/s $1.1 \cdot 10^4$ Pa @ 190°C Figure S6) while in the second case no crossover
385 is detected.

386 Summing up, both the solutions adopted are therefore suitable to improve limitations due to the
387 low viscosity of the recycled mask material alone, and the rPPME system represents the better
388 solution.

389 5.3. Mechanical properties

390 In Figure 5 the tensile tests data on films for both the rPPM and rPPME based formulations are
391 summarized.



392

393 **Figure 5. Mechanical main properties of rPPM (a) and rPPME (b) formulations with**
 394 **fitting curves.**

395

396 The mechanical properties of rPP does not differ much from the ones of M or ME. In particular,
 397 M has a higher modulus (940 ± 60 MPa) and strength (26.0 ± 2.7 MPa) than rPP (605 ± 57 and
 398 16.4 ± 1.1 MPa) while the elongation at break is comparable (6.1 ± 1.5 %). On the other hand,
 399 ME has the highest modulus (1105 ± 65 MPa) and a strength similar to rPP (17.3 ± 1.4 MPa) but
 400 lower than M, as well as the worst elongation at break (2.7 ± 0.3 %). The overall lower properties
 401 of the ME compared to the M formulation were predictable from the SEM observations (Figure
 402 4), in which the poor adhesion between the matrix and the fibers has been observed. This
 403 generates stress concentration points in the matrix which, inevitably, decrease the strength and
 404 the elongation at break of the specimen. In fact, the Figure S7 in the SI shows the plastic
 405 deformation areas of the matrix near the fibers, after the mechanical tests. Moreover, the fibers
 406 result not evidently deformed but detached from the matrix. In addition, the separation surface
 407 between matrix and fiber is smooth, indicating poor adhesion. All these facts confirm the
 408 supposed fracture mechanism in which the load is sustained only by the matrix.

409 Comparing the results with the ones obtained by Crespo et al. (Crespo et al., 2021), who studied
 410 the FFP2 face masks material, it can be noted that the overall mechanical properties of the
 411 recycled material M are similar to the ones from the FFP2 masks (modulus 1.4 ± 0.3 GPa;
 412 strength 23.6 ± 0.2 MPa; elongation at break 7.2 ± 0.4 %).

413 Regarding the compounded materials, the dilution of M or ME in rPP produces intermediate
414 behaviors. Again, thanks to the homogeneity, rPPM formulations are much more regular so that
415 trend lines can be obtained both for the elastic modulus and stress, basically following the rule
416 of mixtures between the two components (dotted lines in Figure 5a). The toughness of the
417 system seems not to be greatly influenced by the diluting of M in rPP, the trend line is indeed
418 quite horizontal. The result is predictable also in this case because both M and rPP have a similar
419 elongation at break.

420 For rPPME formulations the values are so fluctuating that only the modulus shows a clear trend
421 with the variation of the dilution ratios (red dotted line in Figure 5b). The strength seems
422 essentially indifferent to the composition due to the presence of defects in the sample originated
423 by not melted inclusions. These could be ear loop fibers for ME or impurities of the rPP material
424 as reported in the SEM magnifications (Figure 3 and Figure 4). The defects are statistically
425 present in the tested specimens and differs in size, causing variance in the results.

426 On the other hand, the toughness is definitely dependent on the quantity of ME introduced. It
427 indeed seems to follow an exponential proportionality as presented by the dotted blue curve in
428 Figure 5, where the ME increasing amount results in a great detrimental effect.

429 This result is also highlighted by the comparison of the SEM images after the mechanical tests
430 reported in Figure S7 of SI. An evident extensive deformation of the matrix is observed for neat
431 rPP sample while a reduced one for neat ME sample. In the intermediate formulations,
432 therefore, the ME fracture mechanism dominates.

433 In conclusion, M has mechanical properties as good as or better than the rPP. For this reasons,
434 it can be used as rPP replacement for the same applications. Furthermore, the mixing with rPP
435 does not substantially affect its characteristics in none of the percentage ratio. On the other
436 hand, ME has lower characteristics. In order not to bring important variations in the mechanical
437 properties, ME has therefore to be introduced in small percentages only (lower than 25 wt.%).

438 5.4. Ageing

439 One of the possible exploitation of the recycled PP is in outdoor applications. In order to verify
440 whether the materials can be used directly or requires the addition of stabilizing additives, a
441 light accelerated ageing study was performed. Moreover, the masks have also been compared
442 to a commercial PP analyzed in a previous study in the same conditions (Battezzore et al.,
443 2014).

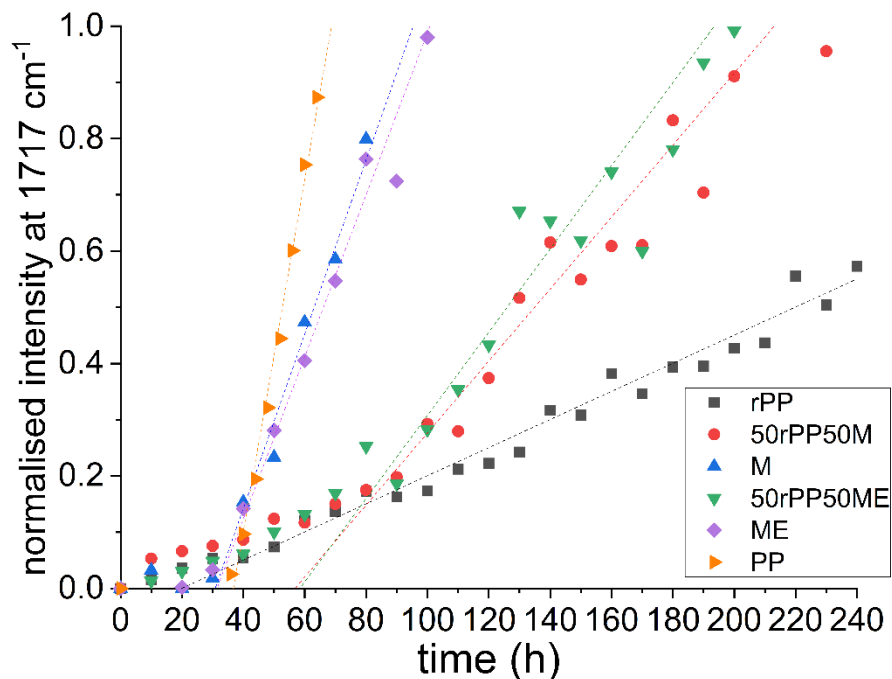
444 The FTIR analysis on samples have been carried out every 10 hours of exposure and in Figure
445 6 the variation of the intensity at 1717 cm^{-1} normalized to the peak at 2723 cm^{-1} is shown.
446 According to the literature, the normalized value of the absorbance in this area is related to the
447 formation of photo-degradation products, namely carbonyl groups (Battezzore et al., 2014;
448 Cerruti et al., 2009; Nanni et al., 2019; Philippart et al., 1999).

449 Generally, PP is characterized by an Oxidation Induction Time (OIT) during which no oxidation
450 of the polymer is observed. This value is calculated by superimposing the two trend lines of the
451 data obtained before and after the OIT. Their intersection defines the value of the OIT. In
452 particular, the already mentioned stabilized commercial PP (Moplen HP500N grade) proved
453 to have the OIT around 35h of exposure (Battezzore et al., 2014). Similarly, the OIT
454 calculated for the M and ME formulations was around 30h. It is thus clear that the neat materials
455 recycled from the masks keeps the stabilizing effect of the additives generally introduced in
456 industrial formulations.

457 In addition, the photo-oxidation rates were calculated as the slope of the interpolating curves
458 (Table S2 in SI). Considering the experimental data fluctuation, it remains almost comparable
459 for the M and ME samples (1.55×10^{-2} vs. 1.44×10^{-2}). Moreover, the rates are lower than that
460 of commercial PP (3.13×10^{-2}). It has to be noted that probably also the typical light blue color
461 of the external filtering part may have contributed to lower the photo-oxidation rates.

462 On the other hand, the photo degradation behavior of the rPP differs from the ones of the
463 materials just described. In particular, it presents a progressive degradation with a limited slope
464 variation between the first and second part. The OIT value is therefore not easy to define. At
465 the same time, the degradation rate is significantly lower than that of the previous materials
466 (0.25×10^{-2}). The behavior may be due to the presence of fillers such as calcium carbonate or,
467 above all, to the colorant. In fact, these formulations turns out to be black and this characteristic
468 may have been obtained with carbon black as coloring agent. As known, it is also active as UV
469 shield and therefore able to decrease the degradation rate (Allen et al., 1998; Horrocks et al.,
470 1999; Pena et al., 2001).

471 For the evaluation of the diluted systems one intermediate formulation was studied. Like for
472 the starting mask materials, also the ageing behaviors of 50rPP50M and 50 rPP50ME are quite
473 similar to each other, and intermediate between the neat M or ME formulations and the rPP. In
474 particular, the trend before the 80 h of exposure resemble the rPP ones, while the degradation
475 rate of the second part is intermediate between that of M or ME and that of rPP ($0.64-0.74 \times 10^{-2}$)
476 ²) and is highlighted by dotted straight line in the graph in Figure 6.



477

478 **Figure 6. Ageing of M, ME, rPP, 50rPP50M, 50rPP50ME and PP (Battezzore et**
 479 **al., 2014).**

480

481 6. Conclusions

482 From the data obtained from the survey, a quantity of 0.6 kg/year per capita of waste derived
 483 from the use of disposable masks was calculated. If completely recycled, this amount would be
 484 equivalent to 3% of the actual recycled plastic materials in Italy. Even reducing the collecting
 485 area to only local realities such as regional schools, recycling plant could be envisaged.

486 A promising strategy would appear to mix this waste stream after sanitization with municipal
 487 recycled plastic waste. For this purpose, the physical and mechanical properties of the MPO
 488 and of the compounds containing mask materials have been compared. The joint recycling with
 489 the filtering part of the mask results in the progressive decreasing of the zero shear viscosity, in
 490 comparable toughness and superior stiffness (10-33%) and strength (12-41%). On the other
 491 hand, the additional presence of the ear loops in the recycling system gives a compounded
 492 material having low viscosity but steady for all formulations at about 50% of rPP value. The

493 elongation and strength are lowered with respect to rPP in the range of 35-56% and 6-15%,
494 respectively.

495 All the above considered, if the mechanical properties are important in the final application any
496 concentration of the filtering part can be introduced, but only limited amounts of ear loops are
497 accepted.

498 Considering the results obtained with FFP2 masks that gave mechanical results similar to those
499 obtained in the present research, the properties reported in this study could be extended also to
500 this type of masks or the mixture of the two.

501 Referring to the photo degradation ageing of the recycled filtering mask material, it is
502 comparable to a commercial PP. Also, the presence of ear loops does not affect its behavior.

503 Considering the compounded material instead, the photo degradation ageing is proportional to
504 the concentration of mask and/or ear loop introduced.

505 The main reason is probably the lower ratio of carbon black in the material if compared to the
506 mixed polyolefin alone. The same shielding effect of rPP would have probably reached adding
507 carbon black to the formulations.

508 In conclusion, neither the waste collection and processing nor the final properties revealed any
509 objection to the recycling of the disposable masks together with other urban waste plastic
510 materials.

511

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