In This Issue:

Welcome to the Year of Recycling Policy

Effect of Mechanical Recycling on Fiber Reinforced Thermoplastic Composites
Arrows vs. Triangles: Good Intentions Gone Awry

By Conor Carlin, SPE Vice President, Marketing & Communications

Increased public awareness of plastics and the environment has led to a search for more information about how best to manage these materials at the end of their useful lives. Plastics, polymers, and chemistry are complex, with many branches of knowledge spreading out in different directions. Most people who are not scientists tend to interact with plastics in a utilitarian way – the materials are ubiquitous and useful. In our consumption-driven world, however, plastics can pose problems when they are no longer useful to us. This post is not meant to address waste management, sustainable materials management, or plastics recycling. The goal is to highlight a single, small element to address a larger point: the confusion created by the numbers found on most plastic packaging.

Contrary to popular (though perhaps waning) belief, these numbers are not recycling codes. They are resin identification codes. They do not imply recyclability, though many people assume that they do, causing a chain reaction of problems. In 1988, the plastics industry trade association (then known as SPI, or Society of the Plastics Industry), introduced resin identification codes (RIC) to help materials recovery facilities (MRFs) and recycling facilities sort different types of plastic resin. The “chasing arrows” logo was created with numbers to identify these resins. Prior to 1988, plastic items were not marked or stamped with any identification. In response to rising costs associated with tipping fees at landfills, the plastics industry attempted to create a system that would allow waste management groups to segregate potentially useful, valuable materials. In the US, 39 states adopted the SPI RIC system in some form and created legislation mandating the use of the codes, though slight differences existed among states, e.g. some mandated that all items over 16 ounces required coding, while others started coding at 8 ounce items.

In 2008, the American Society for Testing and Materials (ASTM) took over management of the RIC system. This group issued new guidelines in 2010, including changing the logo from chasing arrows to a solid triangle. The numbers did not change. Because the new standard acknowledges prior regulation, it makes clear that “…existing statutes or regulation will take precedence…” over the new one. In addition, the new regulation only applies to new molds or tooling, though no enforcement mechanism is evident in the text, and modification to older items is not required. This explains why we still see chasing arrow symbols today, despite the authors’ attempts to decouple the RIC system from recycling messages.

Plastic material derived from non-fossil resources such as polylactide (or PLA, which was not fully commercialized in 1998) is still classified as 7 which means it gets lumped in with such diverse materials as acrylonitrile-butadiene-styrene (or ABS, used for Lego bricks) and polycarbonate (or PC, used in many optical applications), and multilayer materials. Paradoxically, the increase in lightweight, multilayer, flexible packaging reduces overall carbon accounting, but poses new and thorny issues for end-of-life management. Some industry participants point out that today’s materials are not compatible with recycling and waste management infrastructure that was mostly developed in the 1990s. Declining municipal budgets and a relative dearth of private investment in the sector have stalled greater technological innovation and improvement. In short, consumption habits – and associated waste streams are – changing faster than infrastructure systems’ ability to manage that waste. Can we ask if the RIC is still relevant? What would a new system look like? Is one even required given advances in high-speed, near-infrared sorting technologies (link from 1993!!) and digital watermark systems?

We don’t all get updates from ASTM in our email inboxes, so it requires some effort to stay current with a topic that overlaps industry, technology, and politics (you can find a good summary here). Yet we must acknowledge that convenience can lead to thoughtlessness, and bureaucratic approaches to fast-moving societal issues are sub-optimal. Much changes with time; we are required to adapt to new rules, regulations, and realities. Little signs on the bottom of empty bottles might not seem worthy of our attention, but sometimes a small change can make a big difference.

Editor’s Note: This article was originally posted to LinkedIn. SPE Recycling Division members are encouraged to join the discussion on social media platforms to ensure a balanced conversation on the topic of plastics and the environment.
A Procurement-Driven Sustainability Initiative

By Mercedes Landazuri, Techmer PM

With growing interest in environmental issues, both industry and professional associations are hosting more sustainability-minded events to learn best practices and discover new success stories. On December 10, 2019, the SPE Chicago Section hosted a sustainability dinner in Elk Grove Village with two featured speakers, George Staniulis, past-Chair of the SPE Sustainability Division, and Tina Burnet, Director of Procurement for Freudenberg Household Products. This article will provide a summary of the talk given by Tina Burnet.

Freudenberg Household Products (FHP) is a global technology group operating in 40 market segments with several manufacturing sites across the US. The household division employs 350 people at their Aurora, IL facility where they do injection molding and extrusion. This location is also a warehouse and distribution center.

FHP started taking on sustainability seriously in 2015 when customer expectations for sustainable initiatives were growing. The catalyst for FHP’s recycled polypropylene program, however, came from an unlikely source.

Procurement teams live by data, and the team at FHP is no different. In 2015, Ms. Burnet noticed that graphs of polypropylene prices were very erratic. As a result, she initially looked at the option of utilizing recycled polypropylene to stabilize this cost.

In starting this project, Burnet stressed the importance of aligning goals within the company.

“When I look at the production performance goals for the plant, I’m a nuisance to them,” she said. “The first time a company starts using recycled material there are problems that impact production performance. Processing, flash, tiger striping, etc. I know all the terms now. I understood that for this to work, people had to have skin in the game, so I had to get leadership buy-in and make the use of recycled material an overall company goal. To implement culture change you need to define success and celebrate your wins.”

“It took a few years to successfully implement the change to recycled resin, but now FHP is able to boast that in 2019, 85% of their total resin consumption was recycled material. In addition, they have the #1 selling mop in the United States, with 3.5 million units produced.

With corrections in the polypropylene market, and virgin prices dropping, Burnet was asked if FHP is considering going back to virgin material.

“We’re temped to take advantage of some of the prices of the virgin resins. I gave key players the opportunity, but the same people I had to beg to get on board with the recycled program said no. The plant is fully skilled on using recycled material and we never want to go back to operators not knowing what to do in the middle of the night, and now our customers are asking us for the recycled material.”

“These days in B2B you’re competing for business from a younger generation that doesn’t have brand loyalty in certain market segments. You need to have something that makes your company important to them. You want them to be able to state that their supplier scored above all others in their sustainability study.”

“We did it for money,” Burnet said candidly. “That’s the truth. But we’re in a really good situation right now. We can work on continuous improvement for sustainability.”

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Welcome to the Year of Recycling Policy

By Dan Leif, Editor, Resource Recycling

With the new year now underway, government leaders areretu...
Notable developments are also taking place in Oregon. There, a steering committee brought together by the state’s Department of Environmental Quality has been working to identify and outline a suite of policies and tools to help “modernize” Oregon’s recycling system. This multi-stakeholder process could result in a framework that brings producer funding into the system in an innovative format.

Then there is California.

In 2019, Assembly Bill 1080 and its companion, Senate Bill 54, brought national industry attention (and plenty of lobbying dollars) to Sacramento. The legislation, which sought to reduce waste from single-use packaging by 75%, died on the final day of the legislative session. But its traction made clear the political will exists in the Golden State to push forward significant changes.

Meanwhile, Assembly Bill 792, which established recycled-content minimums for containers in the state’s redemption system, passed both legislative chambers before getting vetoed by Gov. Gavin Newsom. The governor’s qualm was that the bill didn’t do enough to force action on the part of manufacturers.

Backers of those pieces of California legislation have vowed to put forward their proposals again this year, and a ballot initiative taking aim at single-use plastics could also be in front of state voters this November.

In short, the momentum around materials management is alive and well in America’s largest state. The wider repercussions of that fact could be pronounced.

**Tied to Image Crisis**

Those who have been around the waste and recycling sectors for some time know that policy is always part of the equation. Bottle bill debates have been happening at the state level for decades, companies and trade groups have long needed to understand how regulatory structures would impact their operations, and decisions at city halls nationwide have determined the specifics of local programs for as long as those programs have existed.

But at the outset of 2020, the industry finds itself entwined with policy in a bigger, more visible way. That’s because recycling is tied to the life cycle of packaging, and packaging is in the midst of an image crisis.

The calls for EPR and other systemic shifts in materials recovery at both the state and federal levels have their root in the public’s recent determination that current end-of-life options for plastic and other packaging materials just aren’t working.

It might be true that criticisms from the masses (and mainstream media) fail to take into account important nuances of material use and management, but it’s also important for recycling stakeholders to understand that this environment is the new normal – and that they’re going to have to navigate these waters whether they want to or not.

A year from now, at least in some regions, the frameworks underlying materials recovery may be undergoing significant shifts due to the decisions of lawmakers. For some stakeholders, this will be a very good thing. For others, it will be full of costs and frustrations.

But no one should be saying they didn’t see it coming.

*Dan Leif is the managing editor of Resource Recycling and can be contacted at dan@resource-recycling.com.*
Abstract

Recyclability of natural fiber and glass fiber reinforced polypropylene composites and glass fiber reinforced nylon composites have been studied through injection molding and mechanical grinding. Mechanical properties of virgin and recycled composites were assessed through flexural, tensile and impact tests. No significant degradation in mechanical properties of natural fiber composites was observed after several rounds of recycling. However, severe degradation in mechanical properties was observed for glass fiber composites. For instance, after five cycles of recycling, only 59% of flexural strength and 64% of flexural modulus was retained for glass fiber reinforced nylon composite, which is mainly due to attrition in the length of glass fibers after subsequent recycling. Water absorption tests conducted at room temperature and subsequent environmental conditionings showed no effect on any of the natural or glass fiber reinforced polypropylene composite. However, nylon composites absorbed about 8% water before reaching saturation point after 45 days of immersion.

1. Introduction

Extensive use of thermoplastics and their composites in our society is causing growing concerns due to its adverse effects on our environment [1]. There are several ways to reutilize polymers for the sustainable model such as reuse, chemical recycling and mechanical recycling. Of these, the latter two are the most widely practiced methods of recycling [2]. This study focuses on mechanical recycling of polymer composites with various mineral and natural fillers. In general, mechanical recycling includes regrinding and reprocessing the polymer composite to produce a new component. The main problem with this type of recycling includes degradation in mechanical properties due to shortening of fibers, damage on the surface of the fiber, delamination on the wall of natural fiber, fiber matrix debonding, thermomechanical degradation on coupling agent, and reduction in molecular weight [3]. Poor retention of mechanical properties leads to recycled material generally being downgraded for next cycle of application. Although there has been attempts to use additives to upgrade the mechanically recycled polymer composites [4].

In the automotive industry, the use of polymer composites continues to grow due to its contribution to light weighting, which directly impacts fuel economy. Metal components in automotive vehicles are replaced by plastics or composites resulting in lightweight vehicle. A reduction in 10% weight leads to 3% to 7% improvement in fuel efficiency [5]. High density fillers such glass fibers or minerals (talc, calcium carbonate) are generally used to reinforce polymer matrix to improve the mechanical properties of the composite without sacrificing the overall cost of the component. However, mechanical recycling has an adverse effect on the recyclability of these composites. The length of the fiber, which directly relates to the improvement in the mechanical properties, is greatly reduced due to the brittle nature of glass fibers. Fiber attrition has also been observed in injection molded components where the fibers must navigate through thin and convoluted sections. Besides, the reduction in molecular weight of the polymer matrix due to thermomechanical degradation could further exacerbate the issue with recycling. In contrast to glass fiber reinforcement, composites reinforced with natural fiber exhibit very little to no changes in mechanical properties after recycling [6]. This is because natural fibers can withstand external mechanical forces and are less likely to break due to their complex internal compositions [6] [7]. Injection molding is the widely utilized manufacturing method for polymer composites based automotive components. The presence of resin rich layer at the surface...
of injection molded components protects the hydrophilic natural fillers from moisture intrusion. In this study, various natural fiber and glass fiber composites are investigated for degradation in mechanical properties due to grinding and reprocessing, and the effect of recycling on water absorption behavior. Tensile tests, flexural tests and impact tests were conducted to assess the mechanical properties of dry (control) and saturated specimens subjected to various hygrothermal conditions.

2. Materials

Table 1 shows the list of composite materials considered in this study. There are three natural fiber reinforced composites, one hybrid composite and four composites with mineral fillers (talc and glass fibers).

<table>
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<tr>
<th>Notation</th>
<th>Matrix</th>
<th>Filler</th>
<th>Filler type</th>
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<td>WF</td>
<td>PP Copolymer</td>
<td>20% Wood pine fiber</td>
<td>Natural fillers</td>
</tr>
<tr>
<td>RH</td>
<td>Recycled polyolefin (PP and PE)</td>
<td>10% Rice hull</td>
<td></td>
</tr>
<tr>
<td>C20</td>
<td>PP</td>
<td>20% Cellulose</td>
<td></td>
</tr>
<tr>
<td>CGF</td>
<td>PP</td>
<td>10% Cellulose 15 Long glass fiber</td>
<td></td>
</tr>
<tr>
<td>T40</td>
<td>PP Homopolymer</td>
<td>40% Talc</td>
<td></td>
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<tr>
<td>GB</td>
<td>PP</td>
<td>29% Talc 7% Glass bubbles 5% Short glass fiber</td>
<td></td>
</tr>
<tr>
<td>GFC</td>
<td>Chemically coupled PP</td>
<td>30% Short glass fiber</td>
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<tr>
<td>LGF</td>
<td>PP</td>
<td>30% Long glass fiber</td>
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</tr>
<tr>
<td>NGF</td>
<td>Nylon 6</td>
<td>15% Short glass fiber</td>
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</table>

Table 1. Thermoplastic composites considered in the study.

3. Experimental Work

3.1. Sample preparation and recycling

Figure 1 shows the mechanical recycling process adopted in this study. During the first cycle, virgin pellets are fed into the injection molding machine to fabricate test specimens. A set number of specimens are collected for mechanical and physical characterization. The rest of the specimens are ground and then oven dried at 105°C for four hours. In the second cycle, the oven dried pellets are injection molded and the process is repeated up to five cycles. All specimens were prepared via injection molding using the parameters presented in Table 2.

3.2. Water absorption tests

Prior to starting the water absorption tests, specimens were conditioned at 23°C and 50% relative humidity for at least seven days before taking the initial dry weight measurement. Water absorption tests were carried out by immersing the specimens in tap water in accordance to ASTM D570. While immersing the specimens in water, spacers were kept between the flat surface of the specimens to allow water absorption. Absorption tests were performed on three sets of specimens for each type of composite at 23°C. Each set of specimen consists of seven specimens.
To determine mass loss due to water absorption, specimens saturated at 23°C were re-dried in an oven at 50°C. Specimens were periodically weighed until the average change in weight was less than 5 mg.

### 3.4. Re-drying

Table 2. Processing parameters for injection molding.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>PP composite</th>
<th>Nylon composite</th>
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<tr>
<td>Barrel temperature (from hopper to nozzle):</td>
<td>182, 188, 191, 193, 193°C</td>
<td>238, 243, 246, 249, 249°C</td>
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<tr>
<td>Back pressure</td>
<td>689.5 kPa</td>
<td></td>
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<tr>
<td>Hold pressure</td>
<td>1999.5 kPa</td>
<td></td>
</tr>
<tr>
<td>Screw speed</td>
<td>70 rpm</td>
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<tr>
<td>Injection time</td>
<td></td>
<td></td>
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<tr>
<td>Hold time</td>
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<tr>
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<td>Mold temperature:</td>
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<tr>
<td>PP Composites</td>
<td>29.4°C</td>
<td></td>
</tr>
<tr>
<td>Nylon composite</td>
<td>60°C</td>
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</table>

Weight measurements were taken after 1, 2, 3, 7, 14 and every two weeks thereafter, until the specimens saturated. The procedure for taking readings for specimens immersed at 23°C are as follows: the specimens were taken out from the water one at a time, placed vertically to allow water to drain for 30 s, then surface moisture was wiped off, weight measurement taken and placed back in water. Water absorption percentage was calculated by using the following equation:

\[
%M = \frac{M_{\text{wet}} - M_{\text{dry}}}{M_{\text{dry}}} \times 100
\]

Where, %M is the percentage water absorbed, \(M_{\text{wet}}\) is the mass of the specimen (g) subjected to water absorption for a certain period of time, \(M_{\text{dry}}\) is the initial dry mass (g).

### 3.5. Mechanical tests

Intron Dual Column Universal Testing System (Model 3366) was used for both tensile and flexural tests. Tensile tests were conducted on Type V specimens (ASTM D638) at a loading rate of 5 mm/min until failure or up to a maximum engineering strain of 10%, whichever occurred earlier. A 5 kN load cell was employed to monitor the force in the specimen and an extensometer was used to directly monitor the strain in the gage length. A 500 N load cell was used for quasi-static three-point flexural tests. Specimens were loaded at a strain rate of 5% per min until failure or up to a maximum strain of 5%, whichever occurred earlier. Calculation of strain was based on the deflection at the middle of the specimen monitored from the crosshead displacement of the machine in accordance to ASTM D790.

All the test specimens were conditioned at 23°C for at least seven days. For instance, the specimens subjected to freeze-thaw cycling were conditioned by immersing them in water at 23°C for at least seven days before testing. While performing tests on wet specimens, the specimens were taken out of the water one at a time and weight measurement taken following the procedure outlined earlier and placed in the Intron and tested.

Notched Izod impact testing was also conducted on control specimen using a pendulum arm impact tester (Model 43-02-03) in accordance to ASTM D256. Specimen preparation included creating a standard type V notch using a notch cutter (Model TMI 22-05) on rectangular bars. At least ten specimens for each composition were tested where each specimen was clamped with a constant force.

### 3.3. Freeze thaw exposure

Since these materials are to be used for exterior application, composite materials were subjected to freeze-thaw exposure. In accordance to ASTM D7032, one set of specimens saturated at 23°C were subjected to -29°C for 24 h, followed by thawing at 23°C for 24 h. This completes one freeze-thaw cycle. Specimens were subjected to five such cycles. These specimens were subsequently conditioned at 23°C for at least seven days while still being immersed in water for the assessment of mechanical properties through tensile and flexural tests.
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Figure 2. Tensile properties of control samples for thermoplastic composites.

Figure 3. Thermomechanical degradation of tensile properties after mechanical recycling (a) tensile strength, (b) tensile modulus, (c) tensile strain at maximum force, (d) legend.
4. Experimental Results

4.1. Mechanical properties

4.1.1. Tensile properties

Tensile strength, tensile modulus and strain to failure for control samples are presented in Figure 2. As expected, natural fiber reinforced composites have the lowest tensile properties. Both wood fiber and rice hull reinforced composites did not fail within 5% strain, while all other composites failed below a tensile strain of 5%. GB composite with talc, glass bubbles and glass fiber was found to be the most brittle one with a strain to failure of only 1.67%. Long glass fiber PP composite has the best overall tensile properties.

After recycling, there is significant decrement in the tensile strength of glass fiber reinforced composites (Figure 3). Whereas, marginal effect was found on natural fiber reinforced composites or talc composite due to recycling. Effect of recycling on modulus is not as pronounced as strength. In general, strain to failure increases slightly with recycling. For nylon glass fiber composite, strain to failure after fifth cycle is very high (~8%) compared to 2 – 2.5% for earlier cycles (Figure 3). This could be attributed to the moisture present in the nylon matrix in the later cycles, which act as plasticizer. Yield stress decreases with water absorption due to an increase in chain mobility in the presence of water [8]. It is worth noting that no appreciable changes in the appearance was observed after recycling. Least retention of properties for glass fiber reinforced composites was observed after five batches of recycling. Natural fiber reinforced composites and talc reinforced composites were least affected by recycling with retention of more than 90% tensile strength property. Whereas, glass fiber reinforced composites have about 60-70% modulus property retention after five batches of recycling.

4.1.2. Flexural properties

Results on flexural properties of control samples are similar to the tensile properties. Long glass fiber composites have overall best flexural properties (Figure 4). Whereas, natural fiber reinforced PP composites have the lowest flexural properties. Trends of decrement in flexural properties after recycling are also similar to tensile properties. Significant decrement was observed in flexural strength of glass fiber reinforced composites, especially the ones with long glass fibers. Decrement in modulus is not as significant as flexural strength. Recycling has marginal effect on natural fiber reinforced composites or talc composite.

Similar to tensile properties, least retention of properties was observed for glass fiber reinforced composites after five batches of recycling with retention of only 60-70% strength properties after recycling (Figure 5). More than 90% tensile strength property retention was observed for natural fiber reinforced composites. Talc reinforced composites seem to be unaffected by recycling.
4.1.3. Impact properties

As expected, wood fiber and rice hull reinforced composites have the highest impact resistance and are least affected by recycling (Figure 6). Impact resistance of glass fiber reinforced composites is severely affected by recycling. For long glass fiber reinforced polypropylene composite, after five recycles, impact resistance is only 39% of the first cycle.

Figure 5. Thermomechanical degradation of flexural properties after mechanical recycling (a) flexural strength, (b) flexural modulus, (c) flexural strain at maximum force, (d) legend.

Figure 6. Degradation in impact properties after mechanical recycling of the composites.

Figure 6. Degradation in impact properties after mechanical recycling of the composites.
4.1.3. Impact properties
As expected, wood fiber and rice hull reinforced composites have the highest impact resistance and are least affected by recycling (Figure 6). Impact resistance of glass fiber reinforced composites is severely affected by recycling. For long glass fiber reinforced polypropylene composite, after five recycles, impact resistance is only 39% of the first cycle.

4.2. Water absorption tests
Only glass fiber reinforced nylon composites showed water absorption at 23°C with maximum intake being 6.5% to 7.7% (by wt.) at saturation. Percentage water absorption at saturation decreased slightly with recycling. No dimensional instability (warping) or changes in physical appearance was observed for nylon composites after immersion in water. However, thickness swelling of about 6.5% was observed. All other composites did not absorb water ever after 45 days of immersion.

4.3. Hygrothermal effects on mechanical properties
Strength and modulus properties decreased significantly after subjecting the saturated nylon composites to hygrothermal conditions. Subjecting saturated specimens to five cycles of freeze thaw did not have any significant effect on the mechanical properties of the nylon composites. Water absorption had irreversible effects on the mechanical properties since they could not be completely regained after re-drying the saturated specimens. Loss in mechanical properties is attributed to an increase in chain mobility in the presence of water. Under wet condition, cellulose PP composites are better than glass fiber reinforced nylon composites (Figure 8).

5. Conclusions
Following are the conclusions drawn from this study:

- Effect of mechanical recycling (regrinding) on tensile, flexural, impact properties were studied on nine types of composites.
- Mechanical properties of natural fiber composites were not significantly affected by recycling retaining more than 90% of the original properties.
- Tensile and flexural strength properties of glass fiber reinforced composites were significantly affected by recycling retaining only 60-70% of the original values.
- Talc filled composites were unaffected by mechanical recycling.
- Impact properties of long glass fiber composite was also significantly affected, retaining only 39% of original value after five batches of recycling.
- Only nylon composite was found to absorb water with 6.5% to 7.7% absorption at saturation.

Figure 7. Water uptake (by wt.) for glass fiber reinforced nylon composites.

Cellulose composites were at par or better than nylon composites under wet condition.
6. References


Figure 8. Comparison between hygrothermal effects on mechanical properties of glass fiber reinforced nylon composites and cellulose reinforced polypropylene composites.
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