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Review article

Rheological stability as the missing criterion in HDPE circularity: A critical review and a new decision framework

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Abstract. The greatest obstacle to recycling post-consumer high-density polyethylene (PCR-HDPE) is typically the degradation of properties caused by impurities and heterogeneity. However, a critical analysis of the literature reveals that the real bottleneck is not the material composition, but rather rheological stability, which simultaneously determines the degradation history of the waste stream, melt behavior, and processability at the cycle level. This review proposes a new perspective: the decision among mechanical, chemical, and energetic recycling is better made based on a unified rheological stability index (*RSI*), which integrates carbonyl index, viscosity change after multiple instances of melting, melt flow index (MFI) instability, in-mold pressure fluctuation, and the degree of polymer incompatibility. *RSI* enables the prediction of the processability of PCR-HDPE and identifies which recycling path a fraction is most suitable for. The study demonstrates how an *RSI*-based approach can reduce quality risk, improve cycle stability, and support circular decision-making in an industrial environment.

Keywords: *recycling, polymer processing technologies, material testing and properties, mechanical properties, circular economy, polymer blends, rheology*

1. Introduction

High-density polyethylene (HDPE) and polypropylene (PP) play a key role in European plastics supply [1–7]. However, the industrial recycling of post-consumer fractions continues to result in low and highly variable quality. Current interpretations attribute quality degradation primarily to heterogeneity [7–12], contaminants [13–17], incompatible polymers [18–20], or inadequate sorting [21–28]. These factors indeed impair process stability, but the main bottleneck in post-consumer HDPE streams is not caused by these factors, but rather by their hidden degradation history: long-term polymer instability associated with chain scission, oxidation, crosslinking, and processing-induced structural changes [10, 29–34]. This previously underestimated instability is reflected in the viscosity drop experienced during processing [10–12, 17, 29, 31–37], in-mold pressure fluctuations [38, 39], and in the variance of energy demand [38–44]. Traditional quality parameters, such as MFI [14, 45–47], point-like Fourier-transform infrared spectroscopy (FTIR) peaks [32, 33, 37, 48, 49], or single-component differential scanning calorimetry (DSC) profiles [32, 47, 50, 51], are unable to predict this background degradation. The industrial processability of post-consumer HDPE (PCR-HDPE is therefore determined primarily by its rheological stability, rather than its composition.

This approach provides a new interpretative framework for comparing mechanical [8, 11, 12, 14, 16,

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17, 19, 52, 53], chemical [5, 13, 52, 54–61] and energetic recycling pathways [30, 62, 63–66], as degradation history and the rheological behavior of the melt influences the output of the process more than CO_2 emission [8, 30, 53, 61, 62, 67–69], energy consumption [38–44, 70] or cleaning options [15, 71–73].

This review includes:

(1) a comprehensive critical synthesis of the root causes of the rheological instability of PCR-HDPE and the limitations of currently used quality indicators [9, 11–14, 17, 45–47, 50, 51],

(2) a new decision support framework, the rheological stability index (*RSI*), which combines degradation history [10, 30–36], incompatibility [11, 12, 17–20, 31], and processability [10–12, 15, 17, 30, 38–40, 74, 75] into a single metric.

2. Why current quality metrics fail for PCR-HDPE

The assessment of the quality of post-consumer HDPE (PCR-HDPE) relies on routine tests, such as the melt flow index (MFI) or melt flow rate (MFR), FTIR spectroscopy, and DSC- and NIR-based (near-infrared spectroscopy) identification, worldwide. These methods all provide important information, but they measure separate phenomena, and none of them can comprehensively describe the entire degradation history of the polymer or its dynamic behavior during processing. Research has shown that the most critical properties of PCR-HDPE are not reflected in point-level structural or flow data, but in the stability of viscosity over time, the progression of chain scission, and the response of the melt to injection molding conditions [9, 29, 38, 39, 46, 47, 50, 51, 76].

The MFI is a widely used reference value; however, it refers to a single state point and does not reflect either the molecular weight distribution or the actual progress of degradation [14, 32, 50, 76]. Thermo-oxidative chain breaks during repeated melting can lead to unpredictable changes in the MFI [29], which are often not consistent with the stability of the melt viscosity profile during injection molding [38]. For this reason, the MFI is not suitable for predicting processability.

The carbonyl stretching vibration is a sensitive indicator of thermo-oxidative damage [32–34]. Recent studies have shown that spectral changes can be further refined using machine learning methods [48, 49, 77–79]. However, the increase in the carbonyl index is not closely related to melt stability, in-mold pressure changes, or cycle time fluctuations [38, 39]. FTIR is therefore a structural diagnostic method, but not a rheological predictor.

DSC is used to determine crystallinity, melting point, and enthalpy [32], thus indicating the basic structural state of the polyolefin. However, these parameters do not reflect the behavior of the melt under shear or the instability that develops during repeated cycles [9, 13, 50, 51, 76]. DSC is therefore not suitable for predicting processing risk.

NIR and hyperspectral technologies are excellent at sorting PE–PP fractions and reducing heterogeneity [21–23, 48]. Extended databases and ML-based (machine learning) classifiers further improve identification [24–28, 48, 49, 72, 73, 79], but spectral features do not reflect degradation history or rheological stability. The material purity achieved during sorting therefore, does not guarantee the stability of viscosity and pressure during industrial processing [9, 45].

A common limitation of these four routine tests is that they measure static or point-like properties, while dynamic, cycle-level phenomena largely determine the properties of PCR-HDPE. Although the low material demand of FTIR, DSC, MFI, and NIR measurements is generally advantageous, in the case of heterogeneous plastic waste streams, it may compromise the representativeness and reproducibility of sampling. Chain scission, oxidation, crosslinking, and melt instability occurring during degradation cannot be detected from a single MFI value, carbonyl peak, DSC profile, or NIR spectrum. These methods, therefore, capture discrete signals but cannot be used for an integrated interpretation of the entire degradation history of the polymer and its processing consequences. The limitations of these measurement methods are summarized in Table 1.

Table 1 shows that the common flaw of the four methods is that they measure static conditions, whereas the most important property of PCR-HDPE is its dynamic nature: the stability of viscosity under shear, heat, and repeated cycles. Therefore, all of the above metrics systematically miss the most important property of PCR-HDPE: rheological stability. These tools are suitable for gathering structural or identification information, but systematically ignore the fact that processability is determined by degradation history, the collapse of viscosity over time, and cycle-level instability. This justifies the need for an integrated,

Table 1. Common analytical metrics for PCR-HDPE and their diagnostic limitations.

Method	What does it measure?	What is the critical limit? (Why it fails)	References
MFI or MFR	Mass flow rate for a single state point is an indirect indicator of viscosity.	It does not reflect molecular weight distribution, degradation, or changes in viscosity per cycle. Thermo-oxidative chain scission and crosslinking cause unpredictable deviations. It does not correlate with stability during the injection molding process.	[14, 29, 32, 38, 46, 47, 50, 76]
FTIR (carbonyl stretching vibration)	Oxidation, formation of carbonyl groups, and chemical structural changes.	Although sensitive to degradation, it is not a good predictor of melt strength, in-mold stability, and cycle time variation. The carbonyl index does not follow rheological collapse. A small sample size may reduce representativeness in the case of inhomogeneous waste materials.	[32–34, 38, 39, 48, 49, 77–79]
DSC	Crystallinity, melting point, melting enthalpy, and morphological state.	Structural parameters do not reflect the melt's resistance to shear. It does not predict cycle-level instability or the rheological response of the melt. A small sample size may reduce representativeness in the case of inhomogeneous waste materials.	[9, 13, 32, 50, 51, 76]
NIR / HSI	Spectral identification of polymer type, additives, and surface properties.	Excellent for polymer identification, but the spectrum does not carry information about chain scission, oxidation, or the rheological stability of the melt. Better sorting does not guarantee better processability.	[21–28, 49, 72, 73, 79]
Sampling and material changes (additional)	Effects of cross-contamination, fraction mixing, grinding, and reprocessing.	Sampling errors and mixing (including PP and additives) significantly distort laboratory parameters. The thermal history remains hidden behind a single measurement.	[46, 47, 51, 76]

multivariate stability index that can quantitatively describe the entire history and process stability of the polymer.

3. Why rheological stability dominates stand recyclability factors

Most of the problems in post-consumer HDPE recycling (heterogeneity, impurities, incompatibility, and sorting errors) indeed degrade material quality, but the literature consistently shows that these factors only partially explain the unstable behavior of the polymer during injection molding [7, 9, 11–14, 17, 37, 80]. The dominant factor is the latent rheological instability created by the combination of degradation, incompatible mixing, and viscosity changes during processing. This affects not only mechanical performance but also energy requirements and technoeconomic viability, as noted in several technoeconomic and circular analyses [19, 81].

The history of PCR-HDPE is determined by thermal, UV, and mechanical stresses during use. These accelerate chain scission, oxidation, and molecular weight loss – the degradation profile that determines all subsequent process stability [10, 30, 32–36, 76, 82]. Repeated melting and extrusion accelerate this process further: chain length decreases, MFI increases, and the viscosity changes over time become unpredictable [11, 12, 17, 31, 35, 36]. FTIR and DSC analyses indicate that oxidation is not only manifested in the formation of carbonyl groups but also in subtle shifts in crystallinity and melting point [31–34, 37, 76, 82]. This results in HDPE not having fixed rheological properties, but rather having a dynamically changing melt behavior that can vary from cycle to cycle [38, 39]. The industrial context of cycle-dependent changes is also reinforced by good practices and processing guidelines, especially in an injection molding environment [83].

In addition to degradation, polymer incompatibility is the second main source of instability. PE–PP blending causes phase separation, morphological inhomogeneity, and local viscosity fluctuations; this has been systematically demonstrated in several experimental studies [19, 34]. The different deformation behavior of PP domains, additive residues, and small impurities during injection molding creates local pressure peaks, filling differences, and deviations in quality [10, 11, 15, 17, 37]. Incompatibility can also increase depending on the size distribution and grinding parameters, as confirmed by coarse grinding and particle size modeling for mixed fractions [84]. Sorting technologies (*e.g.*, NIR) undoubtedly improve fractionation [21–23, 48], but cannot eliminate rheological instability due to either degradation or incompatibility [31, 80, 85].

During injection molding, the most sensitive indicator is the pressure profile, which directly indicates the stability of the melt over time. Experiments show that if the material is rheologically unstable, the cycle-by-cycle pressure and viscosity pattern breaks down, which directly leads to a deterioration of mechanical properties [10, 30, 38]. This has not only qualitative but also energetic consequences: an unstable melt means higher energy requirements and slower cycle stabilization [10, 30, 36, 39]. In terms of energy requirements, rheological instability occurs not only in injection molding, but also in grinding and preprocessing steps; for example, the grinding of film residues and contaminants significantly increases power requirements [86]. For this reason, when a recycling route is chosen for PCR-HDPE, it is not the nominal MFI or impurity ratio, but rheological instability that determines whether a given fraction can be processed mechanically or chemical recycling is more appropriate [11, 12, 17, 30, 31, 80, 85].

Literature evidence suggests that the recycling performance of PCR-HDPE is not determined by its macroscopic composition, but rather by its rheological stability. Degradation, incompatibility, and process-instability are mutually reinforcing, multidimensional phenomena (Table 2). Evaluation and techno-economic analyses of circular systems also support this finding: the performance of the HDPE–PP system is not a function of the starting composition, but of its stability [19, 81]. Therefore, processability can only be described reliably if these are presented in a unified, integrated stability framework. This need is met by the Rheological Stability Index, which can become a decision support parameter for the recycling of PCR-HDPE.

Table 2 shows that the nominal characteristics of material composition do not determine mechanical recyclability, but rather the rheological stability of the material, which is determined by the three mechanisms. Since these effects reinforce each other, PCR-HDPE does not have a single stable, predictable melt behavior. Therefore, we need an integrated metric that can interpret the entire degradation and compatibility history in rheological space (*RSI*).

4. Comparative recycling evaluation routes based on rheological stability

The comparison of HDPE recycling routes has traditionally been organized around three main aspects: environmental performance, energy requirements, and material quality. Although these are all determining factors, literature evidence shows that the processability of PCR-HDPE is fundamentally determined by rheological stability. The decision among mechanical, chemical, and energetic recycling strategies can therefore only be well-founded if rheological stability is given a central role in the assessment, rather than an additional one.

Mechanical recycling is the most widely used industrial route, due to its favorable energy requirements and low carbon footprint [7, 67]. However, the variable viscosity of PCR-HDPE fractions, the degree of oxidation, and the degradation of polymer chains can cause significant fluctuations during the injection molding cycle [29, 39, 87]. Such instabilities often

Table 2. Key mechanisms controlling rheological stability in PCR-HDPE.

Mechanism	Main phenomenon/cause	Rheological consequence	Industrial (process) effect	References
Degradation	Chain scission, oxidation, molecular weight loss during the use cycle, and repeated processing.	Viscosity loss and temporal fluctuation. Narrowing of molecular weight distribution. Nonlinear melt response per cycle.	Fluctuation of filling. Pressure profile destabilization. Deterioration of mechanical properties.	[10–12, 17, 30–37, 76]
Incompatibility	PE–PP phase separation, additive residues, heterogeneous domains; purification and separation are incomplete.	Local viscosity contrast. Morphological instability. Different deformation of domains.	Different filling per cycle. Pressure peaks and filling errors. Increase in quality variance.	[10, 11, 15, 17, 19, 21–23, 34, 37, 48]
Process instability	The interaction of degradation and incompatibility during injection molding: sensitive viscosity and pressure changes.	Unstable melt strength. Different rheological responses per cycle.	Pressure profile collapse. Increase in energy demand. Difficult cycle stabilization.	[10–12, 17, 30, 31, 36, 38, 39, 80]

have a stronger impact on the process than material heterogeneity or even the presence of contaminants. It is also well documented that multiple extrusion cycles increase MFI fluctuations and reduce macromolecular integrity, which directly affects the stability of processing [32, 87]. The technological advantages of the mechanical route therefore, prevail until the rheological instability of the PCR fractions reaches a level where in-cycle behavior becomes unpredictable.

Chemical recycling addresses the degradation history problem by breaking down polymer chains and yields monomers or oil fractions from which new polymers can be produced [13, 52, 54]. Metathesis is one possible approach for monomer recovery from pyrolysis oil, with several relevant contributions reported [55–57]. In this sense, metathesis can be regarded as an intermediate route between mechanical and chemical recycling, as monomers are recovered and subsequently converted into new polymers using equipment typical of conventional polymer processing. According to the literature, pyrolysis and depolymerization are only more favorable from an energetic and environmental point of view than the mechanical route in certain cases [58]. Chemical technologies mostly allow rheologically unstable fractions to be returned to the cycle, but due to the great energy and infrastructure requirements, they are mostly relevant for waste streams that can no longer provide stable melt behavior. Consequently, the decision criterion for the chemical route is not contamination or complexity, but the point at which rheological instability increases the quality and economic risk of mechanical processing too high.

Energy recovery is more favorable than landfilling from an life cycle analysis (LCA) perspective in many cases, but it means the permanent loss of polymer chains and cannot be considered a circular solution [87]. In addition, full oxidation involves high emissions, so this strategy can only be justified for fractions that are effectively out of the cycle or that are rheologically and materially irretrievable. The comparison of the three routes, therefore, requires a framework that can treat rheological performance on an equal footing with environmental and energetic indicators.

Table 3 presents a comparison of virgin HDPE (vHDPE), recycled HDPE (rHDPE), and blended v/r systems, highlighting that rheological stability is crucial in all scenarios, as evident from the literature. Virgin HDPE exhibits homogeneous and stable behavior; however, it is also associated with the highest energy and carbon footprint [9, 97]. In the case of purely recycled HDPE, the processing window is narrowed due to the increased risk of rheological instability, which is a significant source of cycle fluctuations and quality deviations [29, 38, 39]. Blended systems, on the other hand, can stabilize the MFI, reduce cycle variance, and decrease the sensitivity of machine settings [18]. This stabilizing effect can be explained by an overall improvement in rheological stability, as the molecular weight distribution of the virgin material partially compensates for fluctuations introduced by the degraded fraction, including competing chain scission and crosslinking reactions.

Table 3 fully supports the claim that the key to industrial optimization of mechanical recycling is not only the control of heterogeneity or the removal of

Table 3. The comparison of three industrial scenarios for the HDPE-based material cycle: virgin (vHDPE), recycled (rHDPE), and blended (v/rHDPE) systems in terms of energy requirements, environmental performance, and processability.

Features	Virgin HDPE, (vHDPE)	100% recycled HDPE, (rHDPE)	Mixture, (v/rHDPE)
Advantages	Homogeneous properties, stable mechanical performance, excellent processability	40–85% energy savings, 25–75% CO_2 savings, lower material costs, fossil fuel replacement	Stabilized properties, better processability, less scrap, less virgin material requirement, cost-optimized
Disadvantages	High energy requirement (~10–12 MJ/kg), fossil-based, high CO_2 emissions, expensive	Property fluctuations, chain scission, oxidation, high cleaning, sorting requirements, and sensitivity to contamination	Quality depends on the ratio, validation required for some applications, and risk of incompatibility
Environmental performance	Highest environmental impact, no material cycle, fossil input	According to LCA, the most favorable: 0.28–0.53 kg CO_2/kg; primary energy <1 kWh/kg	32–80% CO_2 reduction based on LCA, intermediate energy demand
Processability	Stable MFI, no degradation, predictable extrusion and injection molding behavior	MFI increase, thermo-oxidative degradation in extrusion, narrow processing window	More stable viscosity, consistent MFI, fewer machine adjustments, lower specific energy
Studies	[9, 97]	[29, 39, 68, 71]	[7, 18, 38, 89]

contaminants, but also the ability to obtain a melt with stable rheological behavior from the PCR fraction. Pressure-controlled injection molding (PCIM), for example, is a technological approach that specifically improves the processability of rHDPE by reducing pressure and viscosity fluctuations [38]. Such solutions provide direct evidence that improving rheological stability not only provides processing advantages but also reduces energy requirements and quality rejects.

The comparison of the three recycling routes shows that rheological stability should play a decisive role in the decision. The mechanical route is a mature and environmentally friendly solution as long as the viscosity profile of the material remains predictable from cycle to cycle. When degradation is such that this can no longer be ensured, the chemical route becomes a rational choice. Rheological stability is therefore not a secondary but a primary criterion that determines the effectiveness of any recycling strategy. The evidence is clear that the industrial applicability of PCR-HDPE can only be reliably described if recycling decisions are based on the rheological performance of the material, rather than relying solely on traditional indicators of composition or purity. This justifies the introduction of an integrated indicator that can uniformly describe the degradation history, viscosity change, and processing stability of PCR-HDPE.

5. The rheological stability index (*RSI*): definition, structure, and justification

The greatest uncertainty in choosing the recycling route for PCR-HDPE is that the available material characteristics (MFI, FTIR, DSC, NIR identification) only partially describe the behavior of the melt during processing. The choice among mechanical, chemical, or energetic routes, therefore, requires a decision framework that can interpret degradation history, the rheological response as a function of time, and the effect of incompatible components in a uniform manner. The rheological stability index (*RSI*) introduced in this study meets this requirement. It organizes the most important factors determining the processability of PCR-HDPE into a single index normalized between 0 and 1.

The *RSI* is conceptually composed of four sub-indices as presented in Figure 1, each of which is a stability score scaled between 0 and 1: S_{chem} is polymer chemical stability (oxidation, chain scission), S_{rheo} is rheological viscosity stability during multiple instances of melting, S_{comp} is incompatibility and morphological instability, and S_{proc} is injection molding process stability (pressure and cycle variance). The

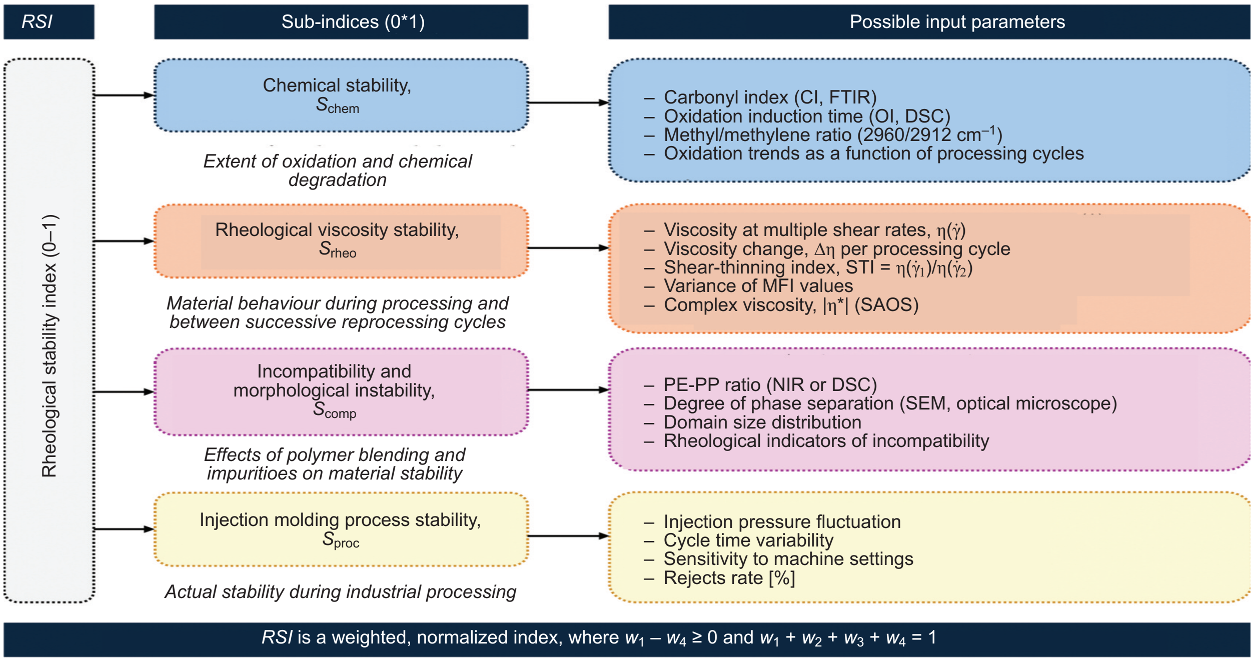

Figure 1. Conceptual structure of the rheological stability index (*RSI*) and its four sub-indices. The *RSI* is composed of normalized stability scores describing chemical stability (S_{chem}) [32, 33, 45, 48], rheological viscosity stability (S_{rheo}) [29, 33, 37], incompatibility- and morphology-related instability (S_{comp}) [16, 18, 20, 21–23], and injection molding process stability (S_{proc}) [38–44, 83].

RSI is a weighted, normalized index as expressed in Equation (1):

$$RSI = w_1 \cdot S_{\mathrm{chem}} + w_2 \cdot S_{\mathrm{rheo}} + w_3 \cdot S_{\mathrm{comp}} + w_4 \cdot S_{\mathrm{proc}} \quad (1)$$

where $w_1 - w_4 \geq 0$ and $w_1 + w_2 + w_3 + w_4 = 1$. The weights can be chosen depending on the specific industrial application (*e.g.* in food packaging S_{chem} and S_{comp} are given more weight, while in engineering components, S_{rheo} and S_{proc} dominate). This formalized index definition enables *RSI* to be more than just a theoretical concept, but a quantifiable decision-support tool.

The first pillar of *RSI* is polymer chemical stability, which is mainly characterized by oxidation indices provided by FTIR spectroscopy. The intensity of the carbonyl stretching band around 1720 cm^{-1} is a sensitive indicator of thermo-oxidative degradation in HDPE, while chain scission cannot be directly inferred from this signal alone. Instead, changes associated with chain scission are more accurately reflected by variations in the stretching vibrations of methyl and methylene groups in the 2960–2912 cm^{-1} region. Several studies have shown that the carbonyl index is closely related to oxidation-induced structural changes arising from repeated processing [32–34]. This progressive evolution of the carbonyl band and the corresponding increase in the carbonyl index, as a function of degradation severity and processing history, is schematically illustrated in Figure 2. Recent FTIR databases, such as the FTIR-Plastics dataset [48], further strengthen the reproducibility of oxidation indices and enable rapid, standardizable pre-rating of PCR-HDPE. For all these reasons, the FTIR-based carbonyl index is a critical input to the *RSI*, even if it is not suitable for an accurate prediction of injection molding behavior on its own, thus S_{chem} is essentially a normalized function of the oxidation state.

The second pillar is rheological behavior, especially the time-dependent change in viscosity during repeated melting. Many studies have shown that the viscosity stability of PCR-HDPE is a far more sensitive indicator of reprocessability than the single-point MFI or MFR value [29, 32]. The varying MFI due to chain scission and the different pattern of shear-thinning behavior from cycle to cycle cause uncertainty that directly affects the filling and post-pressing stages of injection molding [38, 39]. As a result of competing degradation mechanisms, repeated processing of HDPE leads to cycle-dependent viscosity drift and progressively increasing rheological instability [29, 32, 33]. This behavior is schematically illustrated in Figure 3.

While virgin materials exhibit stable viscosity behavior, post-consumer HDPE shows cycle-dependent and increasingly variable rheological responses due to competing degradation mechanisms, ultimately limiting safe mechanical recyclability. To describe this complex behavior, the *RSI* regards the change in

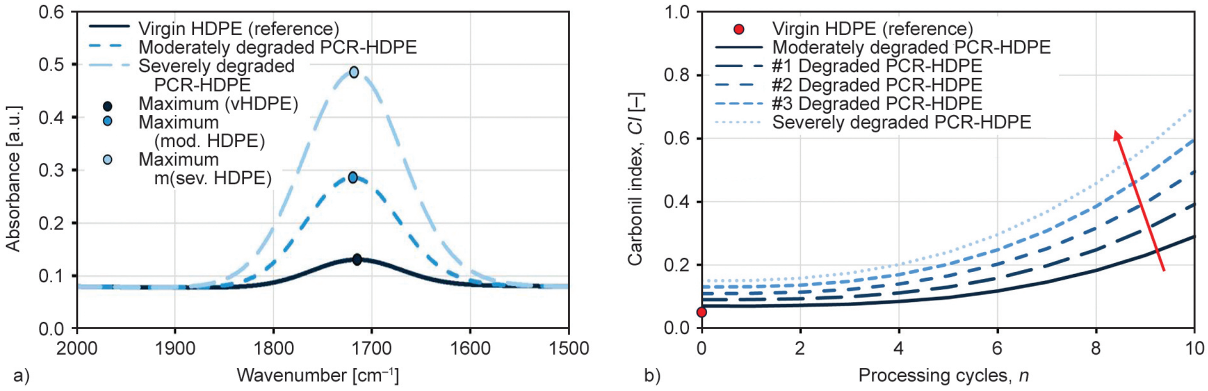

Figure 2. FTIR-based conceptual representation of thermo-oxidative degradation in HDPE and post-consumer recycled HDPE, based on literature reports. The figure is a schematic illustration derived from reported FTIR and carbonyl index trends and does not correspond to a single experimental dataset [17, 32, 33, 48]. a) Schematic FTIR spectra in the carbonyl region (1650–1800 cm^{-1}) illustrating the qualitative increase in oxidation severity from virgin HDPE to moderately and severely degraded PCR-HDPE, reflected by the growth and broadening of the carbonyl absorption band, consistent with typical literature assignments. b) Conceptual evolution of the carbonyl index (*CI*) with increasing processing cycles, showing cycle-dependent growth and divergence of *CI* trends under oxidation-dominated reprocessing conditions. Virgin HDPE is shown as a reference state, while the separation between curves reflects increasing variability associated with progressive thermo-oxidative degradation.

viscosity measured over multiple instances of melting as a key parameter; therefore, a high S_{rheo} indicates a small variance of viscosity and pressure between cycles, and a low value indicates high rheological instability.

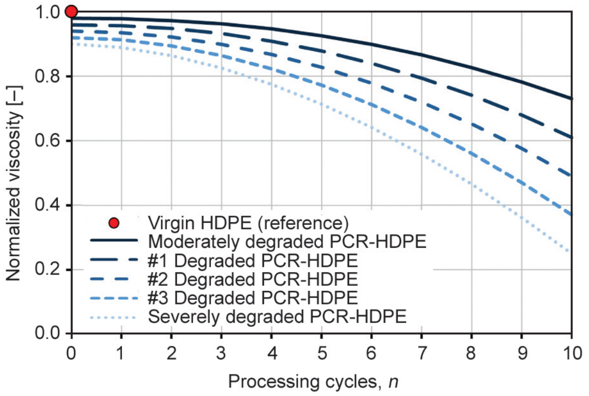

Figure 3. Conceptual illustration of literature-based rheological degradation trends of HDPE during repeated processing cycles. The schematic shows the qualitative evolution of apparent viscosity for virgin HDPE as a reference state and for post-consumer recycled HDPE at different degradation levels. The set of curves represents cycle-dependent viscosity drift and increasing rheological instability during repeated melting, where the progressive separation between curves reflects growing cycle-to-cycle variability and processing uncertainty not captured by single-point MFI or MFR measurements [12, 17, 29, 32, 33].

The third pillar is the rheological instability resulting from the incompatibility of polymer blends. In the case of HDPE–PP blends, differences in phase separation, domain shape and local flow conditions cause significant heterogeneity in the melt, which causes pressure fluctuations, cycle variance, and quality degradation during injection molding [19, 20]. This effect is also evident in diagnostic systems: although rapid NIR- and FTIR-based sorting significantly reduces heterogeneity [21–23], it cannot eliminate rheological instability resulting from incompatibility (2–5 wt% impurities).

Therefore, the third component of the *RSI* specifically includes the weighted effect of the PE–PP ratio, domain distribution, and compatibility factor, so S_{comp} shows morphological homogeneity and compatibilization. Table 4 clearly shows that instabilities resulting from incompatibility often have the same impact on processability as degradation itself.

The fourth pillar is the process stability characteristics measured during injection molding. The pressure profile of the injection phase and the viscosity change under holding pressure are direct indicators of the ability of the PCR-HDPE melt to maintain stable flow. The more stable pressure curves and smaller cycle fluctuations achieved by using PCIM technology [38, 39] are clear evidence that rheological

Table 4. Main advantages and limitations of the mechanical recycling of HDPE and PP, and comparison of related literature examples.

Technological and material-related aspects			
Aspects	**Benefits/Results**	**Limitations/problems**	**References**
Technological maturity	Most established and industrially proven (high-TRL) recycling route.	–	[14, 52]
Polymer chain stability	PP and HDPE can retain structural integrity over multiple controlled processing cycles.	Degradation-induced rheological instability increases downcycling risk.	[19, 29, 90]
Selection and the effect of impurities	NIR-based sorting enables HDPE-rich fractions (≈95–98% HDPE).	Residual impurities (additives, pigments, PVC, PET) impair processability and quality.	[7, 9, 13]
Compatibility and miscibility	When mixed with virgin material, properties can be well stabilized.	The incompatibility of HDPE–PP causes phase separation and requires compatibilization.	[16, 19]
Crosslinked HDPE (xHDPE)	Can be reused as a mechanically stable fraction via grinding and compression molding.	Not melt-processable; requires dedicated treatment routes.	[35]
Environmental and economic aspects			
Energy demand and CO_2 footprint	Lower energy demand and CO_2 footprint than chemical or thermal recycling.	Strongly dependent on extrusion and injection molding process optimization.	[38, 39, 67]
Life cycle impacts	The mechanical route has the most favorable environmental impact (CO_2, energy, eutrophication).	–	[68, 89]
Economy (CAPEX, OPEX)	Lowest CAPEX (capital expenditure) and OPEX (operating expenditure) of all recycling technologies.	Economic performance depends strongly on input material purity and homogeneity.	[3, 7]
Handling of special industrial fractions	Certain contaminated industrial fractions can still be mechanically recycled.	Multi-step cleaning and decontamination are required.	[71]

stability directly improves the predictability of the processing window. In addition, energy efficiency analyses of mechanical recycling also show that stable melt flow is associated with reduced cycle time and lower energy requirements [74, 75]. Accordingly, S_{proc} can be interpreted as a normalized function of pressure and cycle variance.

The final structure of the *RSI* is therefore a four-component model in which polymer chemical stability (S_{chem}), rheological viscosity change (S_{rheo}), incompatibility factors (S_{comp}) and injection molding cycle stability (S_{proc}) are displayed on a single scale normalized between 0 and 1. This approach allows different recycling routes to be compared. This comparison is presented in Table 5.

It is particularly important for *RSI* that mechanical, chemical, and energetic pathways are not mutually exclusive alternatives, but complementary options, the optimal choice between which is determined by the rheological performance of the material. If the viscosity stability of the PCR fraction is sufficient, the mechanical pathway has the most favorable environmental impact and energy requirements [7, 67]. If the degree of degradation or incompatibility is too high, the chemical pathway may be a solution [13, 52, 54]. The current practice of mechanical recycling is typically based on LCA, TEA (techno-economic analysis), and traditional quality parameters [67]. However, several comprehensive frameworks suggest that the real basis for decision-making should be the stability of processability. This is indicated by the circularity framework developed for PCR-HDPE [88, 91], the techno-economic analyses of mechanical recycling [74], various recovery pathway comparisons [75], as well as standardized input–output analyses of LCA software [92, 93]. To place the *RSI*-based decision logic into a broader sustainability context, Figure 4 compares the typical ranges of CO_2 emissions and net energy demand associated with mechanical recycling, chemical recycling, and energy recovery routes, based on literature-reported life cycle assessment studies.

The common conclusion emerging from these frameworks is that a sustainable and economically viable material cycle cannot be ensured by purity or composition alone, but requires a preliminary, quantitative

Table 5. Comparison of HDPE and PP recycling routes from a decision-making perspective.

Recycling method	Main advantages	Main limitations
Mechanical recycling	Lowest energy demand and environmental impact; maintains material value through direct reuse in polymer applications; supports closed-loop strategies when material stability is sufficient; integrates well into existing processing infrastructure	Requires adequate chemical and rheological stability; performance is limited by degradation history, residual incompatibility, and variability in waste streams; increased risk of downcycling
Chemical recycling	Enables conversion of fractions unsuitable for mechanical recycling; allows recovery of feedstock-level intermediates; can complement mechanical routes by treating problematic side streams	High energy demand and process complexity; significant investment (CAPEX) and operational (OPEX) costs; limited industrial-scale deployment for polyolefins
Energy recycling	Robust treatment option for mixed or highly degraded waste; ensures controlled energy extraction when material recycling is no longer feasible	Irreversible loss of material value; high greenhouse gas emissions; not compatible with circular material flows

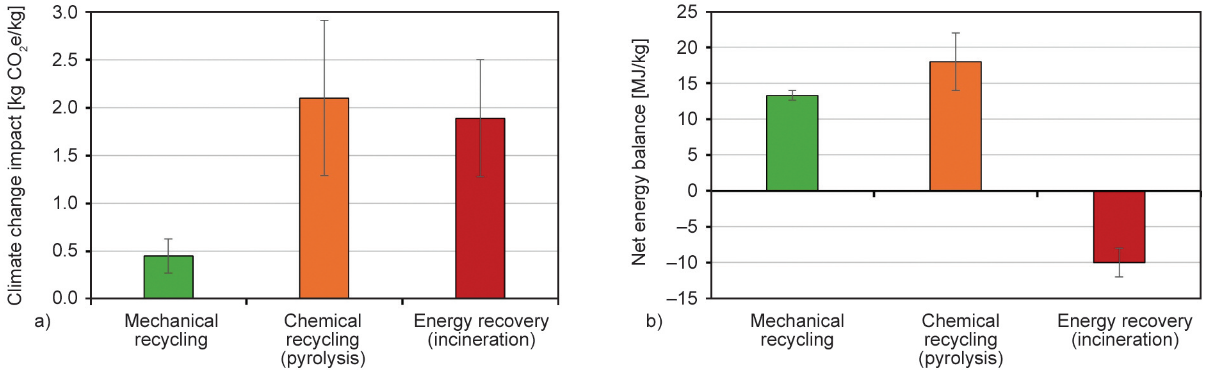

Figure 4. Comparative life cycle assessment indicators for mechanical recycling, chemical recycling (pyrolysis-based routes), and energy recovery of plastic waste. Data ranges and trends are synthesized from published LCA studies on HDPE and mixed plastic waste streams [8, 30, 31, 39, 46, 53, 61, 62, 65, 68, 71]. a) Climate change impact expressed as kg CO_2e per kg of treated plastic. b) Net energy balance [MJ/kg], where positive values indicate process energy demand and negative values represent net energy recovery.

assessment of rheological stability. The *RSI* provides this missing link: it offers an integrated, quantitative stability indicator that can be directly integrated into existing industrial assessment models, and which allows the recycling pathway of PCR-HDPE to be the subject of predictable, evidence-based and process stability–based decisions.

One of the novel features of *RSI* is that it does not rely solely on laboratory parameters, but can integrate all the rapid diagnostic tools that are already available in industrial sorting and quality control. NIR and HSI systems enable fast, in-line fraction identification [21–25, 27, 28], while expanding FTIR spectral libraries and standardized carbonyl stretching vibration analysis [48] provide a reliable basis for the rapid estimation of degradation history. These are complemented by machine learning–based identification and 'recyclate acceptance' systems [24–28, 49], as well as industry standards such as APR processing practices [73]. Separation solutions built into the process, such as steps that allow the removal of microplastics [72], also contribute to quality control early in the sorting process, resulting in more stable material flows. While these separation steps primarily serve process stability and quality control within the *RSI* framework, they may also concentrate microplastic-rich fine fractions into dedicated side streams rather than eliminating them from the system. Thanks to this diagnostic infrastructure, the *RSI* can be integrated into the industrial separation and quality control process virtually in real-time, thus becoming part of decision support. Table 6 clearly illustrates how quality assurance technologies, in particular NIR and FTIR, fit into the *RSI* framework as pre-screening diagnostic tools.

Based on the evidence presented, the *RSI* is an integrated decision support indicator that can bring together the main factors determining the degradation history, rheological behavior, and processability of PCR-HDPE. The advantage of the model is that it does not replace detailed analytical characterization, but organizes its results into a unified logical framework. As a result, *RSI* not only improves the predictability of the injection molding process but also contributes to the evidence-based selection of different recycling routes. The chapter concludes that the *RSI* fills a missing link, allowing for a true and stable return of PCR-HDPE in the circular system and providing decision-makers with information that traditional quality indicators cannot provide individually.

6. Industrial decision matrix based on the rheological stability index (*RSI*)

One of the greatest challenges in recycling PCR-HDPE is that processing plants must make decisions that simultaneously consider material quality, process stability, environmental impact, and economic risks. These decisions are based on large-volume feedstock streams where detailed analysis of each fraction is not possible. The *RSI* provides a framework that evaluates degradation history, melt viscosity change, incompatibility effects, and cycle stability in an integrated manner, thereby providing direct assistance in choosing a processing route.

The industrial application of *RSI* first addresses the question of whether a given PCR-HDPE fraction is

Table 6. Comparison of NIR/hyperspectral NIR and FTIR/ATR-FTIR technologies for identification and quality assurance of polyolefin-based wastes.

Technology	Advantages	Disadvantages/limitations
NIR/hyperspectral NIR [21–28, 48]	Fast, in-line polymer identification (HDPE, PP, LDPE, multilayer) >90% separation accuracy with high material throughput Manual MicroNIR: testing without sample preparation (PE, PP, PVC, PET, PS) Spectral recognition of individual layers in multilayer films Reduces incompatible contaminants, resulting in more stable regranulate	Black, soot-filled materials have poor reflectance, causing misidentification Surface sensitivity, lack of depth information Accuracy may be impaired with very heterogeneous, contaminated fractions
FTIR/ATR-FTIR [2, 6, 9, 14, 32–34, 38, 45, 48, 52]	Molecular fingerprinting, high type identification accuracy (PP, HDPE, PVC, PET, PS) Detects oxidation (1720 cm^{-1}), aging, chain scission (2960–2912 cm^{-1}) Nearly 100% identification accuracy with machine learning Suitable for regranulate grading and 'recyclate acceptance'	Not in-line technology Requires laboratory testing Provides only surface information (ATR), does not measure volumetric contamination Moisture, additives, or surface treatment can distort the spectrum

suitable for purely mechanical recycling. The key here is how well the melt can maintain a stable flow profile during the injection molding cycle and whether viscosity changes do not cause filling or pressure fluctuations [38, 39]. If the *RSI* is high, the mechanical path can be maintained stably, and the material can be processed predictably with adequate quality.

The second question is the optimal ratio of blended vHDPE/rHDPE systems. Table 3 clearly shows that blends can stabilize MFI, reduce rheological instability, and improve the consistency of mechanical properties [18]. This concept is further illustrated in Figure 5 by an illustrative distribution of *RSI* sub-indices for representative PCR-HDPE streams of different quality. High-*RSI* fractions can be processed without blending, while in the case of medium *RSI*, blending not only improves quality but also reduces risk. A low *RSI* indicates that viscosity fluctuations are so great that even the virgin material cannot fully stabilize it.

Stream A shows consistently high values for all four sub-indices, indicating stable chemical structure, rheological behavior and processability, and therefore does not require stabilization by blending. Stream B exhibits intermediate values, with reduced rheological and compatibility-related stability, supporting the need for blending with virgin material to mitigate viscosity fluctuations and reduce processing risk. In contrast, Stream C is characterized by uniformly low sub-index values, particularly for rheological and process stability, indicating severe degradation and instability. In this case, blending with virgin material is insufficient to restore stable processing, and mechanical recycling becomes technically unreliable.

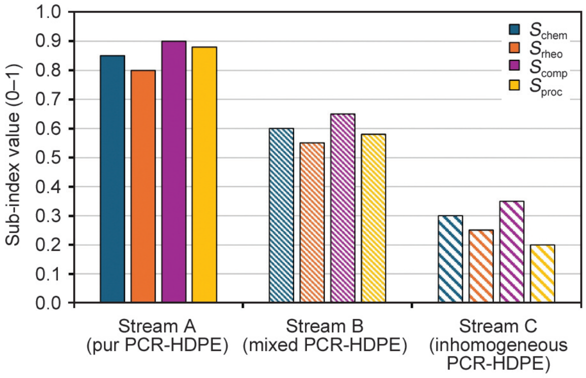

Figure 5. Illustrative distribution of *RSI* sub-indices (chemical stability, rheological stability, compatibility-related stability, and process stability) for representative PCR-HDPE streams of different quality. The relative trends shown are consistent with experimentally reported effects of degradation, blending, and rheological instability in mechanical recycling studies [18, 29, 38, 39, 54], and are intended to visualize how different stability components contribute to overall recycling suitability rather than to represent measured values for a specific material stream.

The third decision point is when it is appropriate to switch to the chemical route. The traditional approach makes this decision based on contamination or heterogeneity, but the literature suggests that these factors only partially explain mechanical unprocessability [7, 9, 13]. The *RSI* integrates parameters such as an increase in FTIR-based carbonyl index, a decrease in viscosity after multiple instances of melting, and fluctuations in injection molding pressure [29, 38, 39], which directly indicate rheological instability. When the *RSI* falls below a certain threshold, it indicates that the mechanical route can no longer be maintained with stable quality, and chemical depolymerization or pyrolysis becomes the rational route. This is in line with reviews on catalytic and thermochemical recycling of polyolefin-based wastes, which clearly indicate the relevance of the chemical route for high degradation [55–57, 94].

The *RSI* is also well-suited for rapid quality control methods. Table 6 showed that NIR and FTIR provide different, yet complementary information on the heterogeneity and degradation state of PCR fractions. The *RSI* organizes this information into a single decision structure, allowing manufacturers to quickly and reliably assess the rheological risk of incoming material. The industrial decision matrix based on the *RSI* can be broken down into three interrelated questions with qualitative rules:

(1) Is the material good enough for purely mechanical recycling?

If the *RSI* is in the ‘high stability’ range (high S_{rheo} and S_{proc}, moderate oxidation and limited incompatibility), a stable pressure and viscosity profile can be achieved during injection molding, with cycle fluctuation being small. The mechanical route is the primary option, suitable alone [38, 39, 67, 68, 89]. In such a case, the decision rule can be qualitatively stated as follows: *if the RSI is high, mechanical recycling is appropriate, there is no need for v/r mixing or an alternative route.* This approach is supported by the fact that cost calculations also indicate that excessive complexity can lead to hidden costs in mechanical recycling [95].

(2) Is vHDPE/rHDPE blending necessary for stabilization?
If the *RSI* is in the medium range (noticeable viscosity and cycle variance, but not critical, moderate incompatibility), the purely PCR-based mechanical route is still viable, but comes with process and quality risks. In such cases, blending with vHDPE can stabilize the MFI, rheological response, and mechanical properties [54, 68, 89]. The decision rule in qualitative form: *if the RSI is medium, the mechanical route is applicable in principle, but the use of a vHDPE/rHDPE blend is recommended to increase rheological and cycle stability.* Such decisions are also supported by large-scale models of circular supply chains that investigate the optimization of polyolefin flow [96].

(3) When does the chemical (or ultimately energetic) pathway become necessary?
If the *RSI* is in the low range (significant chain fragmentation, high carbonyl index, strong viscosity and pressure fluctuations, pronounced incompatibility), the mechanical route can no longer provide stable, reproducible quality, even with v/r mixing [7, 9, 12, 13, 17, 19, 29, 54]. In such cases, the qualitative rule is: *if the RSI is low, the purely mechanical route is unsafe, and chemical recycling (pyrolysis, oil recovery, olefin metathesis, etc.) becomes justified, while in extreme cases, energetic utilization remains the realistic option.* System assessments and life cycle analyses show that this switch can be justified not only technologically, but also economically and environmentally [55, 97].

These qualitative *RSI* levels can be directly linked to existing circularity and decision support frameworks. The circularity assessment framework for PCR-HDPE [91], techno-economic analyses of mechanical recycling [74], the comparisons of various recovery pathways [75] and standardized LCA input–output studies [92, 93] all indicate that it is not enough to decide based on contamination or MFI when choosing a technology, rheological stability must be explicitly included in the decision logic. These approaches are also consistent with scenario-based assessments that have examined the realistic yields and outcomes of different collection and sorting systems for post-consumer packaging waste [98]. The results of the scenario analysis indicate that decision points should not be determined along a single 'optimal' value, but should be considered in ranges related to rheological performance and material quality. The 'industrial decision matrix' based on *RSI* can therefore be interpreted as a rule-based decision support system rather than a descriptive overview:

- High *RSI*: direct mechanical recycling, with optional w/r mixing, low risk, favorable LCA and TEA indicators.
- Medium *RSI*: mechanical route is possible, but vHDPE mixing and closer process monitoring are recommended to reduce rheological risk.
- Low *RSI*: mechanical route can only be maintained with significant quality and economic risk, in which case the chemical (or ultimately energetic) route becomes a rational choice, justifiable from both LCA and TEA perspectives.

The industrial application of *RSI* results in a decision matrix that can be seen in Figure 6, which increases the efficiency of quality assurance, reduces processing instability, and identifies an optimal route for PCR fractions of different quality. Materials with high *RSI* values can be stably injection molded with low energy requirements and minimal cycle variation. For intermediate fractions, mixed systems can provide stabilization, while a low *RSI* is a clear indication that the mechanical route is no longer safe and chemical processing is more appropriate.

To illustrate the practical decision logic of the *RSI* framework under industrially relevant conditions, the following case study uses synthetic but realistic input data derived from established literature trends [29, 38, 39], rather than from a single specific recycling plant. Accordingly, Figure 7 presents a representative case study based on three PCR-HDPE streams: a clean, well-sorted fraction (Stream A), a mixed but still mechanically processable fraction (Stream B), and a highly heterogeneous waste stream (Stream C). The four *RSI* sub-indices (S_{chem}, S_{rheo}, S_{comp} and S_{proc}) were derived from experimentally accessible indicators and combined using three different weighting strategies reflecting typical industrial applications: equal weighting, packaging-oriented weighting, and engineering-oriented weighting.

Equal weighting represents a generic quality screening scenario, while packaging weights emphasize chemical stability and compatibility, and engineering weights prioritize rheological and process stability.

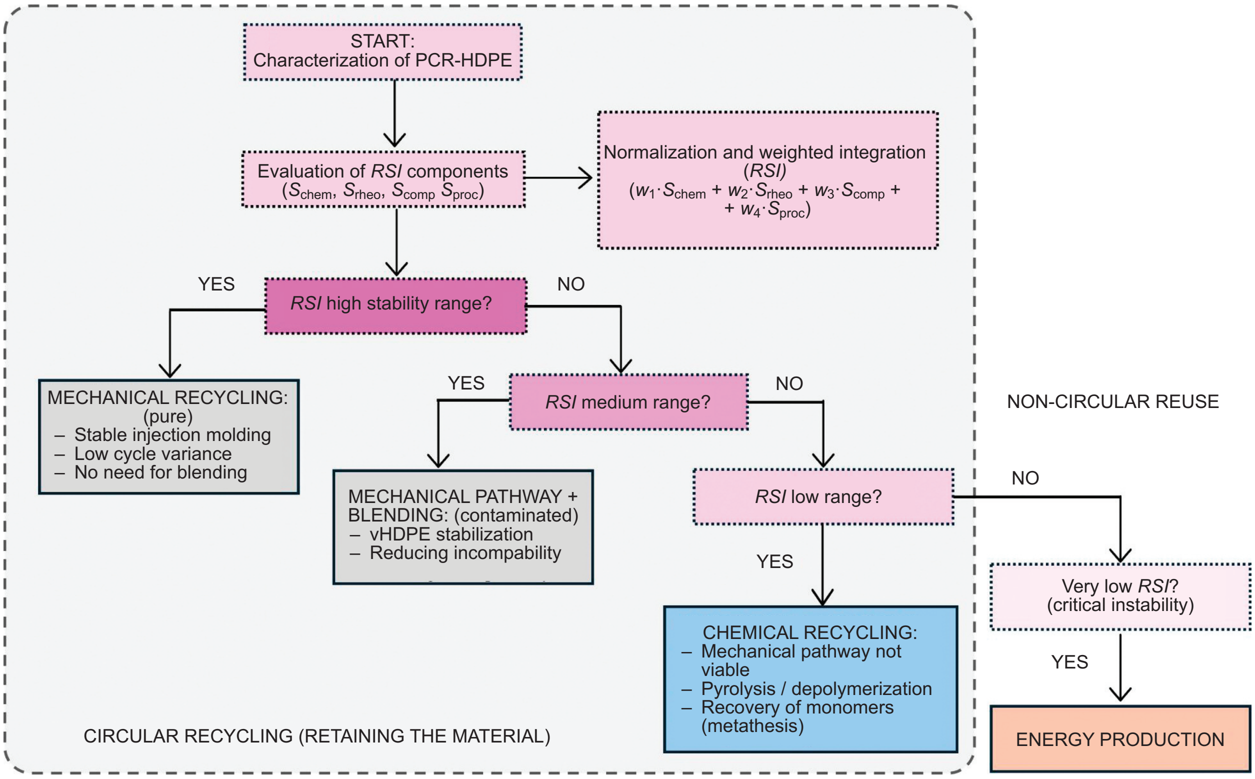

Figure 6. Industrial decision flow for PCR-HDPE based on the rheological stability index (*RSI*).

As illustrated in Figure 6, the resulting *RSI* values lead to consistent but application-dependent recycling decisions. Stream A exhibits high *RSI* values under all weighting schemes, confirming its suitability for direct mechanical recycling. Stream B falls into an intermediate *RSI* range, where mechanical recycling remains feasible but stabilization by blending or stricter process control is recommended. In contrast, Stream C consistently shows low *RSI* values, indicating that mechanical recycling would involve significant technical and economic risk and that chemical or energy recovery routes are more appropriate.

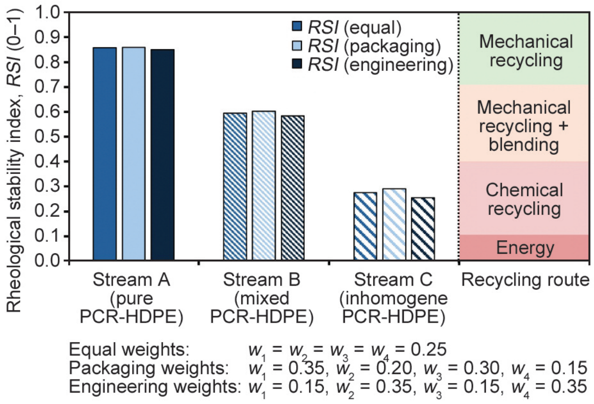

Figure 7. Application-weighted rheological stability index (*RSI*) values for representative PCR-HDPE streams. *RSI* values are calculated using equal, packaging-oriented, and engineering-oriented weighting schemes [18, 29, 38, 39]. Shaded regions indicate qualitative decision zones for direct mechanical recycling, mechanical recycling with blending, and chemical or energy recovery routes.

This case study illustrates how the *RSI* functions as a quantitative decision-support tool, translating multidimensional material quality information into clear, application-oriented recycling route recommendations. Overall, the *RSI* is a systemic decision support tool that brings together the most important factors of PCR-HDPE processability in a logical and predictable way. The integrated interpretation of degradation, viscosity change, incompatibility, and process stability allows for truly informed technological decisions in circular polyolefin treatment. Therefore, the *RSI* is a missing link that can become one of the most important tools for recycling PCR-HDPE fractions.

7. Conclusions

The assessment of post-consumer HDPE recycling has traditionally been based on heterogeneity, impurities, and polymer incompatibility, but available evidence suggests that these factors alone cannot explain the instability experienced during injection molding. The degradation history and the resulting viscosity change during collection, washing, and multiple processing cycles play a far more decisive role in the true processability of PCR-HDPE [9, 29, 32–34, 38, 39, 45] and are more directly related to cycle stability than any traditional quality parameter. It is clear from the literature that MFI, FTIR, and DSC provide valuable but fragmented information and are not suitable for predicting in-process behavior. In contrast, the pressure profile measured during injection molding, the evolution of fill viscosity and cycle-level fluctuation provide a stable indication of how much the material is able to maintain a uniform melt flow [38, 39]. The interrelated effects of degradation, incompatibility, and rheological instability, therefore, require an integrated approach.

This role is fulfilled by the Rheological Stability Index (*RSI*) introduced here, which uniformly characterizes the processability of PCR-HDPE based on polymer chemical stability, rheological behavior, incompatibility, and injection molding cycle stability. Although rheological instability is indirectly influenced by material composition, including the presence of different HDPE grades and residual impurities that may also promote additional degradation reactions, it represents an integrative response that captures their combined effects. The advantage of the model is that it forms a direct bridge between rapid diagnostic tools (NIR, HSI, FTIR) [21–28, 48, 72, 73, 79] and industrial decision-making, and can also be adapted to circular and techno-economic frameworks [91–93, 74, 75, 99].

The ‘so what?’ message is clear: the real limit to the circular recycling of PCR-HDPE is not determined by composition or purity, but by rheological stability. The *RSI* organizes this critical parameter into a quantitative and decision-supporting form, allowing industry players to predict processing risk, increase the reliability of mechanical recycling, and reduce cycle variance, energy requirements, and quality degradation. This integrated approach fills a missing link without which the circular industrial application of PCR-HDPE cannot become a stable and scalable system.

Future prospects and recommendations

the rheological stability index (*RSI*), introduced in this review, provides a structured framework for linking material degradation, rheological behavior, and process stability to recycling route selection for PCR-HDPE (Figure 6). While the concept is supported by extensive literature evidence, its broader impact depends on systematic validation, standardization, and industrial integration. The following perspectives outline short-, medium-, and long-term priorities for further development of the *RSI* framework.

In the short term, the primary priority is the experimental validation of *RSI* using well-defined PCR-HDPE datasets. Although individual *RSI* sub-indices are based on established characterization techniques, their combined interpretation as a unified stability indicator requires controlled studies across different waste streams, processing histories, and application domains. Repeated extrusion and injection molding experiments are crucial to quantify the relationship between viscosity evolution, degradation indicators, and process instability.

Closely linked to validation is the development of standardized measurement protocols. For *RSI* to become reproducible and transferable, harmonized guidelines are needed for sampling strategies, test conditions, and normalization procedures. These include definitions of viscosity drift, acceptable ranges of MFI variability, the selection of FTIR-based oxidation indicators, and the statistical treatment of process data, such as pressure and cycle-time fluctuations. The establishment of reference materials and benchmark datasets would further support interlaboratory comparison and calibration of *RSI* subindices.

In the medium term, the impact of *RSI* depends on its integration into existing industrial recycling infrastructures. Coupling *RSI* calculation with current sorting technologies, such as NIR or hyperspectral imaging, represents a promising direction. While these tools are primarily used for polymer identification, their data streams could support compatibility-related assessments and early prediction of rheological risk. In parallel, the creation of regional or European-scale *RSI* databases would enable the benchmarking of PCR-HDPE quality, the identification of recurring instability patterns, and data-driven optimization of recycling strategies across the value chain.

Another important medium-term development is the integration of *RSI* into techno-economic assessment (TEA) and life cycle assessment (LCA) frameworks. Although *RSI* is not an environmental indicator, it has a direct influence on processing efficiency, energy demand, and reject rates. Embedding *RSI*-informed decisions into LCA and TEA would improve the realism of sustainability assessments by explicitly accounting for material stability constraints.

Figure 8 presents an integrated decision-making framework in which rapid material diagnostics and *RSI*-based routing are used to guide the selection of mechanically, blended, chemically, or energetically viable recycling routes for PCR-HDPE, while systematically linking process choices to LCA, TEA, and life cycle cost (LCC) evaluations. Rather than promoting a single optimal pathway, the framework emphasizes context-dependent, industrially relevant decision-making across the recycling value chain.

An important practical implication of such integrated process frameworks is the handling of fine and microplastic-rich fractions. While early separation of fines can improve material flow stability and process reliability, it may also concentrate microplastics into dedicated side streams rather than eliminating them. From a system-level perspective, these fractions should therefore be treated as redirected material flows whose downstream handling and environmental implications must be explicitly considered within LCA-based sustainability assessments, rather than being treated as negligible losses.

In the long term, the *RSI* concept can be extended beyond PCR-HDPE to other polyolefin systems, including polypropylene, polyethylene copolymers, and multilayer structures. While the stability-based decision logic remains applicable, sub-indices and weighting strategies will need to be adapted to material-specific degradation mechanisms and processing sensitivities. A further long-term perspective is the development of AI-based predictive systems built on large *RSI* datasets. Such approaches could enable the proactive prediction of processability and the selection of recycling routes, provided that data quality, model transparency, and physical interpretability are carefully maintained.

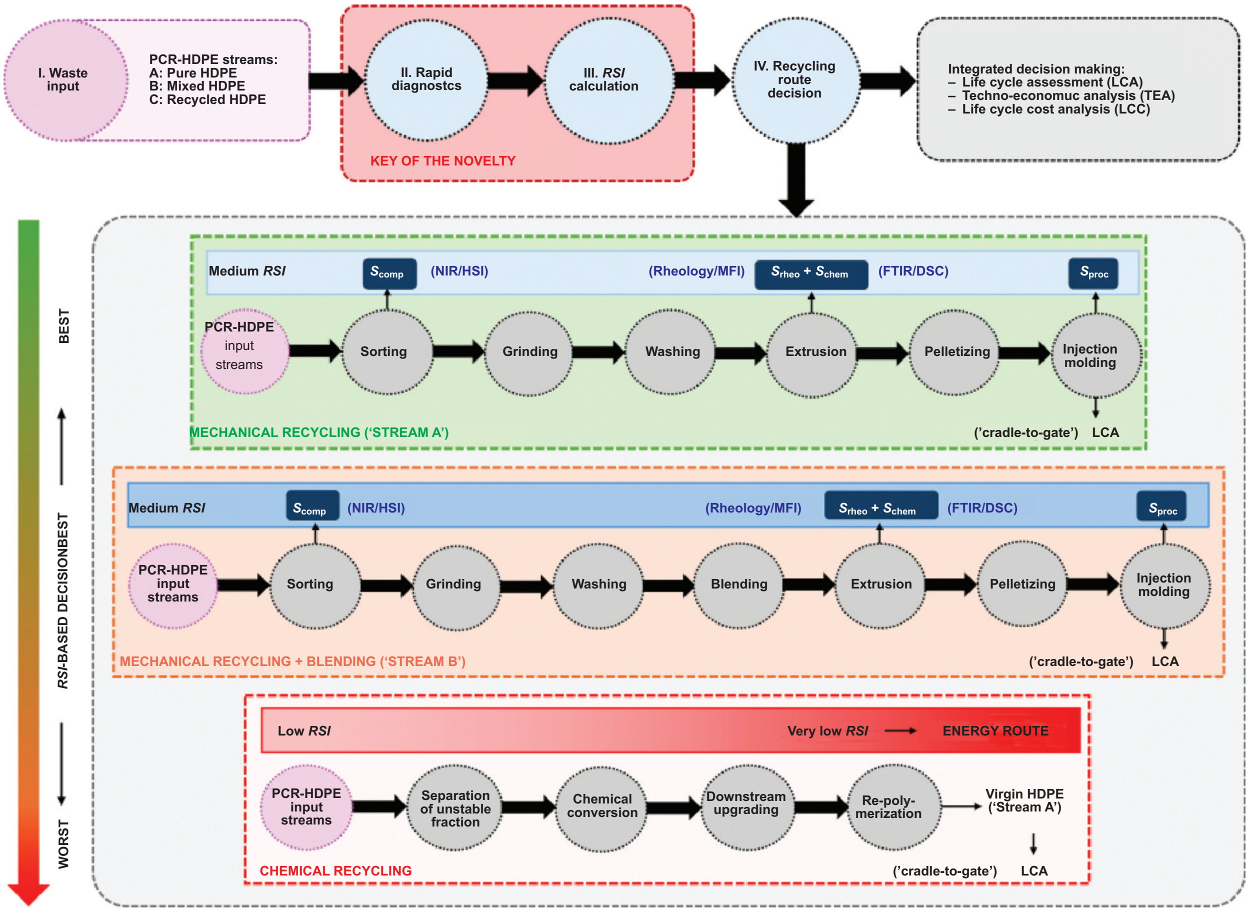

Figure 8. *RSI*-based integrated framework for recycling route selection and sustainability assessment of PCR-HDPE streams. Rapid diagnostics and *RSI* calculation classify input streams into high, medium, and low stability levels, guiding mechanical, blended, or chemical recycling pathways. The selected routes are subsequently evaluated using life cycle assessment (LCA) and techno-economic analysis (TEA), which link material stability, processability, and sustainability performance.

From an industrial perspective, *RSI* should initially be adopted as a qualitative risk classification tool rather than a strict numerical threshold. Early use can focus on identifying low-stability fractions associated with high processing risk, thereby reducing downtime, scrap rates, and energy consumption. With increasing experience and data availability, *RSI* can evolve into a quantitative, application-specific decision-support system. Overall, the *RSI* framework provides a clear pathway toward more reliable, transparent, and scalable recycling of PCR-HDPE, supporting the transition toward circular polyolefin systems.

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