Overview of Preparation Methods for High Performance Composite Materials

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Abstract. This article summarizes the advantages and disadvantages of the RTM process and its products. Based on a comprehensive analysis and comparison of common methods for improving the performance of RTM products, the principle and characteristics of the compression RTM (CRTM) process are highlighted and summarized. The research status at home and abroad analyzes the CRTM process parameters and their effects on the properties of composite materials, and points out the development trend of the CRTM process.

1. Introduction
The Resin Transfer Molding (RTM) process is a closed-mold molding process. The basic principles are: The homogeneous resin system is injected into a closed mold cavity in which a fiber reinforcement body is preliminarily laid, and a composite material product is obtained through curing, mold release, and post-treatment. [1-2] Because of the advantages of high efficiency, low cost, low pollution, and wide applicability, the RTM process is highly favored for surface quality, dimensional accuracy, and overall performance. [3] Its products are already in aerospace, Transportation, sporting goods, ships, construction and other fields have been widely used. [4-7]

Although RTM technology products have been widely used in many fields, their inherent limitations cannot be ignored: First, the products are prone to defects such as dry spots and bubbles, and the porosity is high, which seriously affects the performance and service life of the products; The fiber volume content of the product is relatively low (usually about 40%), not suitable for use as the main bearing structure; third, the process is not fully automated and is not suitable for mass production; the fourth is a mold with large area, thick section, and complicated structure. In the cavity, the resin is difficult to penetrate and the flow is not balanced. This dynamic process cannot be observed and it is more difficult to predict and control [8]. These problems make composite materials with high specific strength, specific stiffness, good fatigue resistance, and strong designability, which cannot be fully utilized [9]. Effective measures need to be taken to improve and improve the mechanical properties of composites formed by RTM technology. In order to enhance the carrying capacity of its products, it can promote the application of RTM composite products in various fields to a greater degree.
2. Common Ways to Improve RTM Product Performance

In order to solve the deficiencies of RTM process and its products, researchers at home and abroad have conducted a lot of research in raw material modification, molding process improvement, and optimization of the curing system.

2.1. Resin Modification

The resin matrix is an important part of the binder fiber in the composite material, structure shaping, transmission load, and protection fiber. The basic requirements of the RTM process for the resin system are: low viscosity (generally less than 0.8 Pa•S) and low viscosity at the injection temperature for a long time, moderate gel time and curing time, no precipitation of small molecules during curing, and low cure shrinkage. And with the reinforcing fiber compatibility is good. [10-13]

On this basis, the quality of RTM molded products and the level of performance are closely related to the resins used. The cured resin system must have good mechanical properties, especially when preparing load-bearing components, the mechanical properties are more important. A large number of studies have shown that, [14-16] through the modification of the resin, can not only reduce the shrinkage of some resins that meet the requirements of RTM process, but also reduce the precipitation of gas molecules and increase the mechanical properties, but also possible to make some resins with very good properties that are not suitable for the RTM process, under the premise of effectively maintaining their performance, and be suitable for the RTM process after modification. The modification methods used for different resins are different, and commonly used modification methods include blend modification, molecular structure modification, and inorganic element modification. [17, 18]

On the basis of satisfying the RTM process conditions, it is feasible to improve the mechanical properties of RTM composites by improving the mechanical properties of the resin matrix and its composites. However, the improvement of the mechanical properties of RTM molded composites by resin modification is rather limited and the effect is not very obvious.

2.2. Fiber Modification

The properties of composite materials mainly depend on the properties of the reinforcement material and the resin matrix, but are largely limited by the quality of the interface. A good interface bonding energy can effectively transfer the load and improve the mechanical properties of the composite material. The essence of the fiber and matrix composite process is to form a stress-transmitting interface on the fiber surface, and the role of the interface is to transfer the external force applied to the composite material through the interface to the reinforcement body through the interface. Fiber surface activity, surface energy, chemical functional group activity, and adhesion to the matrix determine the quality of the interface, which in turn has a significant influence on the mechanical properties of the composite. In view of these influencing factors, the current fiber modification is carried out in the following aspects: first, the introduction of reactive functional groups on the surface of the fiber, the improvement of resin wettability and chemical bonding on the fiber surface, [19] and the second is on the fiber surface. Etching, improve the fiber surface roughness, increase the specific surface area of the fiber, and then to form a mechanical interlocking structure with the resin matrix; [20] third is to remove the weak interface layer of the fiber surface.

Taking carbon fiber as an example, there are a large number of modifications methods currently studied [21]: First, the coating method. The principle is to form an intermediate layer on the fiber surface that can interact with the fiber and the resin matrix to achieve the purpose of interface enhancement. For example, Gong Ke [22] and other composite materials made of carbon fibers coated with a silane coupling agent can increase the tensile strength by 30% and the wear resistance by 3 times. The second is oxidation. Oxidation is a technique that is mainly used for the surface treatment of carbon fibers. The commonly used methods include liquid phase oxidation, gas phase oxidation, and anodic oxidation. The main principle is that through the oxidation treatment, the oxygen-containing functional groups are generated on the surface of the fiber, and the surface roughness and specific surface area of the fiber are also increased. Such as YU [23] and other K₂S₃O₈/AgNO₃ solution combination of PAN-based carbon
fiber surface oxidation treatment, can make CF/Epoxy composite interface shear strength increased by 62.5%. The third is high-energy radiation treatment. The principle is to use high-energy rays (such as electron beam, gamma ray, X-ray, ultraviolet, etc.) to bombard the surface of the fiber to make chemical reaction on the surface of the fiber to generate active radicals or graft reactive functional groups to increase the chemical bond between the fiber surface and the resin matrix. [24] In addition, high-energy ray etching carbon fiber surface can increase the surface roughness of the fiber, which is conducive to improve the binding force between the fiber and the substrate interface, and then improve the interface strength of the composite material. For example, when DILSIZ [25] uses plasma to treat the carbon fiber surface, the strength of the carbon fiber monofilament increases, and the elongation at break increases by 15%, the corresponding interlaminar shear strength and bending strength of the CF/Epoxy composite are both increased by 9%.

Although the mechanical properties of the composite materials prepared by using modified fibers can be improved, the fiber modification technology is still in the research and development stage, lacks a large-scale application foundation, and there are also problems such as high costs or complicated processes, [26, 27] not suitable for general promotion.

2.3. Curing system optimization

The curing of a polymer composite material is a cross-linking reaction of a polymer material, that is, the resin changes from a linear molecular structure to a network-like molecular structure. The cross-linking reaction is closely related to the final mechanical properties of the product and is an extremely critical step in the preparation of composites. The completeness of cross-linking reaction and whether the curing is uniform depends mainly on the curing time, temperature, and heating rate. For processes such as mold pressing that require pressure, the timing of pressurization and pressure are also important factors affecting the curing reaction. [28] Studying how to reasonably select and determine the curing parameters is an important part of the performance of raw materials as much as possible.

At present, the basic parameters such as the curing time and curing temperature of most common resin materials have become common knowledge. In actual use, only direct application or slight adjustment is needed, and the optimized space is minimal. The heating rate and the timing of the pressurization are different depending on the shape, size, thickness, and requirements of the components. However, most of the relevant research results can be used for reference, such as during the curing of large thickness composites. There will be temperature lag and temperature overshoot, resulting in uneven temperature and solidification in the thickness direction, resulting in non-uniform internal residual stress, fiber volume content and mechanical properties. [29, 31] Relevant scholars have done a lot of research, proposed a step-by-step curing process, [32] slow step heating [33] and other methods to better solve the problem of curing the large thickness of the wall plate.

In general, although optimizing the curing system is beneficial to improving the mechanical properties of composite materials, the space available for optimization is relatively limited, and some of the optimized processes require high equipment and technology, and their practicality is not strong.

2.4. Molding Process Improvement

Through the resin modification, fiber modification, and optimization of the curing system, the mechanical properties of RTM products can be improved to a certain extent, but they cannot break through the inherent limitations of traditional RTM processes. By improving the RTM process, the fiber volume content of products can be improved and products can be reduced. Porosity and other methods can completely break the performance limits of traditional RTM products, and are more efficient, more efficient, and more practical.

2.4.1. Soft Module-assisted RTM Process (Flexible RTM). The basic principle of the soft mold assisted RTM process is to place fiber reinforced preforms on unexpanded soft molds and place them in rigid negative molds. After injecting the resin, the soft molds are controlled to expand, and the normal extrusions are not yet gelled. Fiber/resin composites; heating at the same time to cure the fiber-reinforced
The method of controlling the expansion of the soft mold may be a method of heating the flexible mold to inflate the core mold or inflating the sealed flexible core mold. The former is called a thermal expansion soft mold assisted RTM process, and the latter is called an airbag assisted RTM process.

![Balloon Fiber preform](image)

**Fig. 1** Schematic diagram of the soft mode assisted RTM process principle (a) Laying of reinforcements and injection of resin; (b) Expansion of soft mold or bladder into place and curing

The advantages of the soft die expansion assisted RTM process is [35]: (1) The volumetric fiber content of the product is substantially increased (up to 60%) compared to the traditional RTM process through soft die extrusion of the preform. Has a certain role in reducing the porosity [36]; (2) It can integrally form large-size and complex-structured composite materials at a lower cost; (3) Since the product is fitted on a flexible mold with a certain elasticity, it has The internal complex structure of composite products is more convenient for demoulding.

However, there are some limitations in the soft-mold-assisted RTM process [37]: (1) The form of molded articles is limited. While the soft die method is expanding and squeezing, the outer surface of the product needs rigid die limitation, so this process is mainly used to form the hollow rotary structure; (2) The inner cavity size of the product is not easy to control precisely. The inner cavity size of the product is not formed by a rigid mold, and the dimensional accuracy is greatly affected by the expansion performance of the soft mold, and it is difficult to accurately control; (3) For the thermal expansion soft mold assisted RTM process, as the temperature rises, the expansion force of the core mold increases, but the resin viscosity gradually increases, which is not conducive to squeeze the rubber; for airbag assisted RTM process, the airbag thickness is limited, may be due to uneven expansion, resulting in uneven thickness of the product or rib structure displacement.

2.4.2. **Vacuum Assisted RTM Process (VARTM)**. The basic principle of the Vacuum Assisted RTM (VARTM) is to seal the cavity of the fiber reinforced material on the basis of the common RTM process, and connect the vacuum pump at the glue outlet; the resin is injected into the cavity. Vacuum is applied at the same time to complete the resin injection, curing and demoulding [38] (Figure 2). Cavity evacuation not only increases the pressure gradient of the resin filling, enhances the driving force of the resin flow, but also increases the apparent flow in the preform and the flow rate of the resin between the fiber bundles. More importantly, the cavity is discharged. The gas in the preform and especially the gas in the fiber bundle are discharged to facilitate the complete infiltration of the fiber, thereby reducing the defects of the product.
In addition, vacuum-assisted RTM derivatization process vacuum infusion molding process (abbreviated as VIMP) has a wide range of applications in the field of integral molding of large-size components. VIMP uses vacuum bags to seal fiber-reinforced bodies on single-sided hard molds. The pressure difference produced by the vacuum pumping presses the reinforcement body, and the resin flows from the rubber injection port to the rubber outlet by means of the vacuum negative pressure (Fig. 3).

Compared with the traditional RTM process, the VIMP process has the following advantages [39, 40]: (1) Only single-sided molds can be used, mold making is simpler; (2) Use of vacuum can increase fiber volume content and make product fiber volume content higher (up to 75% to 80%) [41]; (3) Vacuum assists the impregnation of the fibers by the resin and impregnates the fibers more fully; (4) The vacuum has the effect of evacuating the air in the fiber bundles to make the fibers impregnated more complete, thereby reducing the formation of microscopic pores, resulting in lower porosity products; (5) can be made of larger size composite components.

However, the VARTM process has a limited ability to increase the volume fraction of the fiber, and a longer molding cycle is required for the preparation of thick-sectioned and large-sized components, which to a certain extent limits its wide application in large load-bearing structures [42].

2.4.3. High Pressure RTM Process (HP-RTM). The principle of the HP-RTM process is to lay out a designed fiber preform in a rigid mold cavity, then inject the resin into the closed mold cavity with vacuum assisted by the injection equipment, and finally complete the high pressure provided by the hydraulic press. The impregnation and curing of the resin resulted in release of the product [43] (Figure 4). The process is an RTM process technology for high-efficiency, large-scale production of high-performance thermoset composite products. The advantages of the HP-RTM process mainly include [44]: (1) The resin filling time is short, rapid curing resin system can be used, the process cycle can be shortened, and the molding efficiency is greatly improved; (2) High-pressure injection and curing are conducive to the discharge of air and reduce the product porosity; (3) The product has good surface quality, high dimensional accuracy, and high process stability and repeatability.
However, the HP-RTM process has the following disadvantages: (1) High-pressure injection requires high injection equipment, high equipment costs (adhesive pressure up to 15 MPa), and (2) high-speed resin flow is not conducive to fiber infiltration, and can easily lead to displacement of fiber preforms, the use of a predetermined agent will affect the final performance of the product; (3) cannot effectively improve the fiber volume content of the product, the role of improving the mechanical properties of the product is not.

In addition to overcoming the disadvantages of the traditional RTM process through the above-mentioned improved RTM process, some researchers have also studied ways to improve the mechanics of RTM products by changing the way of injecting and dispensing rubber, optimizing the plastic injection, setting the rubber outlet and the flow channel, or optimizing the process parameters. Performance, these studies also have a certain effect on controlling and reducing the defects of RTM products and reducing the porosity [45-52].

From the above analysis, it can be found that these methods have a certain role in improving the mechanical properties of RTM products from different perspectives, and further expand the adaptability of RTM processes. However, no matter which method has its own shortcomings, especially in improving the mechanical properties of the products, they cannot play a significant role due to certain limitations.

3. Compression RTM Process (CRTM)

Compression resin transfer molding (CRTM), also known as injection-compression molding process (I/CM or I/C LCM), resin transfer-compression molding process (RT/CM). The thickness of the mold cavity is increased by the clearance reserved by the mold, so that there is a condition for depositing more reinforcing materials; The existence of a reserved gap makes the reinforcing material layer relatively loose, which can increase the permeability of the reinforcing material preform, so that the resin flows more smoothly in the cavity and the reinforcing body, and it is easier to realize the impregnation of the fiber-reinforced body with the resin; The existence of the compression process facilitates the discharge of air bubbles, compaction of the fibers and the resin, increases the volume content of the fibers of the product, and reduces the porosity of the product [53, 54].

CRTM process not only can greatly improve the mechanical properties of the product, but also can effectively shorten the process cycle and greatly expands its application range. In fact, the CRTM process appeared earlier, but due to limitations in equipment and demand, research and application are not widespread, and the development speed is slow. Until recently, the technology has gradually attracted the attention and favor of the industry.

3.1. Basic Principles and Features of the CRTM Process

The CRTM process is a combination of the molding process and the traditional RTM process. The specific process principle is to place the fiber reinforced material in a closed cavity in a reserved space. After the resin is injected, select the timing according to the need to compress until the mold is completely closed, gap disappeared in the cavity, solidified and molded to obtain the product (Figure 5).

![CRTM process schematic](image)

**Fig. 5** CRTM process schematic (a) Resin injection; (b) Compression.

The CRTM process greatly inherits the advantages of the RTM process and the molding process. The products produced by this process have the features of good appearance quality, high dimensional
accuracy, and fewer defects, as well as the advantages of high fiber volume content and low porosity. On this basis, it can effectively shorten the process cycle and reduce the RTM process restrictions on the thickness of the section. In addition, the process is also suitable for large-scale, automated production, and has a profound influence on large-scale composite material manufacturing companies with higher degree of automation [55]. However, the CRTM process requires high production equipment and the production cost is higher than that of the traditional RTM process, and the small-scale enterprise takes on difficulties. At the same time, due to the size and pressure of the press, the process cannot form large or over-complex components.

3.2. Research Status of CRTM Process at Home and Abroad

The research on CRTM and its related technologies abroad started earlier, and the research mainly focused on the following aspects:

(1) Experimental study of CRTM process. The main disadvantage of the RTM process lies in the fact that when a large-sized, high-fiber-volume component is prepared, the mold filling rate is slow, and defects such as dry spots and air bubbles tend to occur. Raising injection pressure can alleviate these problems to a certain degree, but it puts forward higher requirements for production equipment. Researchers using CRTM process CRTM to produce the same target requirements of the composite material required injection pressure will be substantially reduced, compressed composite material products prepared after the fiber volume content is much higher than the traditional RTM process [56]; process parameters The choice is closely related to the final performance of the product. The researchers also studied the relationship between CRTM process parameters and product performance and explored the optimal combination of parameters [57].

(2) Simulation of CRTM process. The focus and difficulty of the CRTM process is that its mold filling process is complex and difficult to predict and control. The researchers used a combination of simulation and experiment to study the resin filling process and pressure distribution in the CRTM process, which can optimize the placement of the preform, mold structure and injection parameters, greatly improving the test. Efficiency, reduced research costs [58-60]; numerical codes are also used to simulate the CRTM process, which simulates the entire process flow and the heat and force distribution during filling and curing processes, which is in good agreement with the actual situation and is more intuitive The image shows its advantages over RTM technology [61, 62]. In addition, there is also the use of finite element model or numerical code to establish the relationship between process parameters, study the interaction between parameters, and predict the process of resin flow, temperature, pressure, curing distribution and other conditions, determine the choice of parameters Principles [63-65].

(3) Applied research of CRTM process. The application research of CRTM technology aims to increase production efficiency and reduce production cost. It mainly studies the influence of process parameters on the production cycle in CRTM process and the requirements for production equipment, and proposes a method to balance efficiency and cost [66]. Through the establishment of a model to study the change of clamping force and compression pressure in the CRTM process, it can guide the mold design and reduce the cost of the mold [67]; The impact of CRTM process parameters on the mold filling process and the flow and heat transfer of different types of resin are carried out Th e study provides basis for selecting suitable parameters and resins to shorten the process cycle [68, 69].

China has conducted some research on the CRTM process, but there is still a large gap compared with foreign countries. In the CRTM process test study, Chang [70, 71] used the vacuum-assisted resin transfer molding-compression process (VRCRTM) to study the influence of process parameters on the mechanical properties of the products, and pointed out that the injection pressure is the most important influencing factor. The filling time and injection port pressure change under different compression timings. Luo Jun [72] proposed a compressive resin transfer molding method, which can significantly increase the fiber volume content of the product while ensuring the surface quality of the product and greatly improve the mechanical properties of the molded product. In the CRTM process simulation study, Luo Minjie [73] established a one-dimensional and two-dimensional model of the CRTM process, which
can simulate the flow front changes and filling processes under different parameters, and can provide reference for the preparation of composite products and mold design optimization. Yang [74] simulated the flow process and force field distribution in the CRTM process. Through experimental verification, it was confirmed that the simulation accuracy is extremely high, and the shape of the mold and the design of the injection hole can be well instructed.

3.3. Influencing Factors of CRTM Process

Although the CRTM process only adds a compression process based on the RTM process, many variables are introduced, such as reserved gaps, compression methods, compression pressure/compression speed, and compression timing. The influence of these variables on the process is more complex. There is a certain difficulty in studying the influence of these variables on the CRTM process. For this reason, most researchers use single-variable methods or purposefully select several variables to study their effect on the process.

(1) Reserve clearance. Reserving a gap can increase the cavity space, reduce the amount of compression of the fiber reinforced body after the mold clamping, thereby increasing its porosity, reducing the resistance to resin flow, and allowing the fiber to infiltrate more completely and rapidly. The filling time can be shortened; at the same time, the presence of gaps can also be used to lay more reinforcing fibers in the cavity, which can significantly increase the fiber volume content of the composite after compression.

There are two cases for the choice of reserved gap [75]: one is that there is no obvious gap between the fiber preform and the molding surface of the cavity, but the fiber coating does not receive the clamping pressure of the rigid mold, and the ply is loose. In this case, the value of the reserved gap is determined by the final thickness of the product, the volume content of the fiber and the apparent thickness of the reinforcement. It is also limited by the compression stroke of the mold part and the maximum pressure of the press; the other is fiber. There is a certain distance between the preform and the molding surface of the cavity. Young and Chiu [52] studied this situation and proposed a formula for estimating the reserved gap for a flat die. This study has a certain reference role in determining the reserved gap:

\[ h_0 = h_i + h_g - h = h_i + h\varphi - h = h_i - hV_f \]  

In the above formula, \( h_g \) is the distance between the surface of the dry fiber reinforcement and the molding surface of the cavity, \( h \) is the final thickness of the component, \( h_0 \) is the reserved gap of the die, \( h_i \) is the thickness of the uncompressed preform, and \( \varphi \) is the modulus The porosity of the reinforcement in the cavity, \( V_f \) is the volume fraction of the fiber.

(2) Compression method. Currently used compression methods are constant rate compression and constant pressure compression. When constant-rate compression is used, the mold closing speed is constant. With the relative movement of the yin and yang molds, the fiber layers in the mold cavity are gradually compacted, the porosity is gradually reduced, and the resin flow resistance and the resistance of the fiber reinforcement are increased. The pressure required for compression also increases, and decreases with the clearance of the mold. With constant pressure, due to the smaller initial resistance, the mold closing speed at the beginning is faster and constant. Similar rate compression, resistance will gradually increase, so the mold closing speed will gradually slow down until completely closed.

With constant-rate compression, too fast compression can shorten the filling time, but it is not conducive to the discharge of bubbles and resin impregnated fibers, and easily lead to fiber displacement, too slow not only does not improve the performance of the product will also make the filling time prolonged for no reason; When using constant pressure compression, the pressure selection is extremely important. When the pressure is less than the resistance when the mold is fully closed, the mold cannot be completely closed and products with thickness and fiber volume content cannot be produced. When the pressure is too high, the resin flows too fast and the fiber is difficult to infiltrate, it is not easy to discharge gas, and it easily leads to fiber displacement, resulting in decreased product performance.
(3) Compression timing. Compression timing can be broadly divided into two cases: First, the compression begins after the resin injection, and the second one begins before the resin injection ends. The different selection of the compression timing has a totally different effect on the process and product performance. Under normal circumstances, the beginning of compression before the resin injection is beneficial to shorten the process cycle, and the compression start time is different, and the length of the process cycle is also different; Compression begins after the injection, similar to the RTM process followed by the injection molding process, although it can also shorten the process cycle to a certain extent, but the greater role is to improve the product's fiber volume fraction and mechanical properties.

Chang [70] uses constant flow injection and constant pressure injection respectively for the injection pressure and charge of RTM process and CRTM process 3 compression timings (simultaneous start of injection and compression, end of injection and compression, and end of injection). Modal time was studied. Studies have shown that no matter which injection method is used, when the injection and compression start simultaneously, the time required for the resin to fill the mold is the shortest; when a constant pressure injection is used, the CRTM process chooses to compress at any time, and the time required for the resin to fill the mold is much shorter than that of the RTM.

4. Conclusion
The filling process of the CRTM process is intrinsically more complex than the RTM process. To realize all the advantages of the process, an effective process design is required. The current numerical model can simulate the filling and curing processes of shapes and simple products. Future research may focus more on the development of modeling and forming technology for complex-shaped components, which in a sense also determines the future development space for the CRTM process.

In general, the input cost and technical threshold of the CRTM process are high, but the CRTM process molded product has excellent performance. With the development of manufacturing technology, the CRTM process will be less and less restrictive, and the CRTM process will be even larger. The application space, especially in special fields such as aviation, aerospace, and warships, CRTM process will become the key to composite materials instead of metal materials, the process has outstanding advantages, such as increasing the product fiber volume content, reducing the porosity of the product, reducing The limitation of the thickness of components and the shortening of the molding process cycle will surely be further studied and applied more widely.

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