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SOME ASPECTS REGARDING THE RECYCLING OF COMPOSITE MATERIALS USED IN AUTOMOTIVE

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Abstract: *The paper is presented a proper method for recycled composite materials used in the automotive industry. This process is complex and composed of many steps, where the recycled material is achieved from waste parts of PC+ABS plated with metal. The automated recycling installation has inserted a pickling chemical station for collecting the metal on wasted-injected composite parts. The main parts of recycling installation are plastics grinding mill, conveyor belt, filling conveyor, robot for transporting raw materials, pre-washing basin, acid pickling basins, washing basin, gas exhaust system and raw material dryer. The simulation is performed by MOLDEX 3D software to determine the basic parameter process of plastic injection moulding. The results of the experiments confirmed the method used for recycling the composite parts.*

Key words: *composite, pickling, plastic injection moulding, recycling, waste.*

1. INTRODUCTION

Recycling wastes are impetuous and necessary for reducing the raw materials which are not inexhaustible and to keep the green planet. The Geen Directives of EU and the 4R Plan present the basic principles regarding waste management such as *Reduce*, *Reuse*, *Recycle* and *Recover*, due to the final elimination of waste [1] Mwanza, B.G., [2] Ragossnig, A.M., Agamathu, M., [3-4].

The classical plastic recycling process is divided into many stages such as collection, sorting, washing, resizing, identification and separation, and compounding. In conformity with ISO 15270, the modern plastic recycling proceeding can be Energetical recycling, Mechanical recycling and Chemical recycling. Similar to ASTM 7209-06 these methods are named Quaternary recycling (energetic), Primary and Secondary recycling (mechanic), and Tertiary recycling (chemic) [5, 6].

The automotive industry produces a huge amount of industrial waste, of which plastics and composites are significant. Energy reduction must be closely related to the problem of location and environmental impact. One of the important sources of industrial plastics waste is resulting from the parts obtained by the plastics injection moulding process, which is the interest of this research.

Plastic Injection Moulding (PIM) is a well-known and great extension manufacturing process that is applied in the automotive manufacturing of plastic parts because of multiple and intricate details, minimizing the cycle time and lower product costs [7] Harper, C.A, [8], [9] Gillespie, L.K, [10] Hinojosa, R J.

Plastic injection moulding consists of some steps which are drying, mixing, melting, injection, packing, cooling and ejection, which are worked together. The quality and precision of injected moulded parts are major influenced by machine selection, design part and material properties [11] Jain, A.N., Tony, B., [12] Marwah. O.M.F. et al.

The main processing variables with a direct impact on PIM are speed, plastic temperature, plastic pressure, colling temperature and time. All of these parameters and optimised product designs and machining can be efficiently evaluated by the FEA of Moldex 3D software.

This research will present some aspects regarding the recycling composites used in

automotive which means the recycling process focusing on the pickling process, FEA of plastic injection moulding, recycled installation and mechanical tests of new recycled material.

2. RECYCLING PROCESS

The recycling process proposed in this paper is applied to the industrial wasted parts of ABS+PC composites plated with metals, Its goal is to recover the composite materials and plated metals that allow achieving new plastics and composites for the injection process of automotive parts. The recycling process is complex and has many stages such as:

- Collection and storage of industrial wastes resulted from the injection moulding process of automotive parts.
- Shredding and grinding process of industrial wasted parts.
- Pickling process of grinding wasted workpieces.
- Re-granulation of recycled material of ABS+PC composites.
- Characterization of recycled material and receiving new composites to be used in the plastic injection moulding process.
- Validation of the recycling process and automated recycled installation used for industrial wasted automotive parts.

The experimental processing is vast and includes multiple proceedings and tests, while in this paper the authors will present only some aspects regarding the recycling of composite parts such as the pickling process, FEA of plastic injection moulding, recycled installation and mechanical tests of recycled material.

2.1. Pickling process

After the industrial waste of plastics sorting and grinding the next step in the recycling process is pickling, which means a fast removal of the oxides from the plastic surface.

The chemical pickling process is performed inside of recycling installation in the basins of chlorohydrin acids (HCl) and nitric acids (HNO_3) that follow the installation course for the other process. Process and installation aim to recover the part of plastics and metal plated on the surfaces of waste composites.

The industrial wastes provided from ABS+PC named BAYBLEND T45PG were plated with different metals.
That material has been used for the fabrication of the samples.

The samples are divided into three types with different metals and layer thicknesses:

- **Sample A** has plated surfaces from 8-11 mm^2 obtained by shredding and grinding with Cr layers of between 0.5-1 µm, Ni between 40-70 µm and Cu between 50-70 µm.
- **Sample B** has plated surfaces from 50-60 mm^2 with layers of Cr between 0.5-1 µm, Ni between 40-70 µm and Cu between 50-70 µm.
- **Sample C** has plated surfaces from 60-350 mm^2 with layers of Cr between 0.5-1 µm, Ni between 40-70 µm and Cu between 50-70 µm.

Fig.1. Sample A used for pickling

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Fig.2. Sample B used for pickling

Fig.3. Sample C used for pickling

The pickling process has two main objectives, first is the determination of picking times for the removal of the metal layer plated, and second is the determination of the loading of

the acids used for tests. For the experiment, all samples used equal quantities of plastics, chemical substances and proceeding, such as 500 g of ABC+PC plated, 800 ml of HCl with 37% concentration, and 400 ml of HNO_3 with 55% concentration, diluting 250 ml of water. For the removal of the Cr layers, all the plastic-plated samples were immersed in HCl until the remaining only plastic surface was. The results of removal pickling times in Table 1 are presented.

Table 1

Removal pickling time of Cr for samples

	Sample A	Sample B	Sample C
Time (min)	2	3.2	4.5

After the removal of the Cr layers, all the samples were washed and went on to the next phase of the experiment to determine the removal pickling time of the Ni and Cu layers. The cleaned samples were immersed in HNO_3 to remove these two layers. It was recommended that during this immersion keep cool down the entire process experiment near 70-75°C for efficiency, which avoided the negative reaction of heating exhausted up to 93°C. The results of pickling time in Table 2 are presented.

Table 2

Removal pickling time of Ni and Cu for samples

	Sample A	Sample B	Sample C
Time (min)	26	41	49

Fig.4. Sample A after the pickling process

Fig.5. Sample B after the pickling process

Fig.6. Sample C after the pickling process

The next phase in the pickling process is the determination of the charge of the acids used in the metal experiment. Following the chemical pickling process, the acids used are loaded with metals pickled from the surfaces of plastics. To determine the degree of loading with both acids and metals some tests and results analysis were performed by applying the atomic absorbent proceeding with the AGILENT 200, model 240 FS AA. For the experiment, five equal samples of plated plastics were taken with a grain size between 8-10 mm^2 and a weight of 500 g. All the samples were immersed first in 800 ml of HCl with 37% concentration for 2 min for removal of the Cr layer. Next, all samples were immersed in 400 ml of HNO_3 with 55% concentration for approx. 28-30 min for each layer removal of Ni and Cu, due to a clean material. The test results are shown below.

Table 3

Sample of HCl for removal of Cr layer

	Metal (g/l)		
	Chrome	**Nickel**	**Copper**
Sample 1	0.2857	0.0835	1.4535
Sample 2	0.7585	0.1395	2.1195
Sample 3	1.1425	0.3504	3.5850
Sample 4	1.8408	0.4772	6.0450
Sample 5	1.8093	0.3134	6.3800

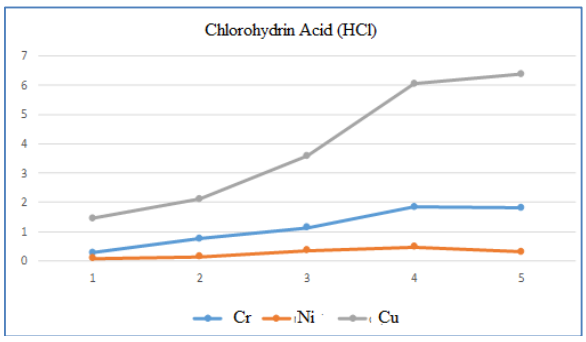

Fig.7. Diagram analysis of HCl for removal of Cr layer

Table 4

Sample of HNO_3 for removal of Ni and Cu layer

	Metal (g/l)		
	Chrome	**Nickel**	**Copper**
Sample 1	0.0061	32.7150	56.7500
Sample 2	0.0052	71.6500	126.9000
Sample 3	0.0079	58.1500	110.3000
Sample 4	0.0163	52.0750	90.1500
Sample 5	0.0171	71.6500	92.6500

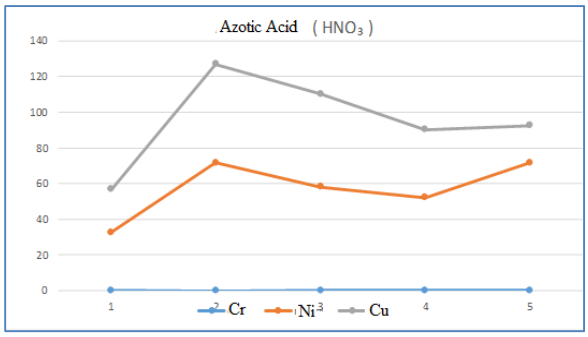

Fig.8. Diagram analysis of HNO_3 for removal of Ni and Cu layers

500 g of plated plastic is approx. 95-100 g of metal deposited divided by categories. The biggest amount of metal is Cu which is the support between plastic and other metals followed by Ni in a small amount and less of Cr because is applied in a thin film in conformity with the physical and mechanical properties of Cr. The acid density resulting after the pickling process obtained by lab measures in Table 5 is presented.

Table 5

Densities of acids used in the experiment

Sample	**Density (g/ml)**	**Temperature (°C)**	**Concentration (%)**
HCl	1.1285	26.0	37.57
HNO_3	1.4120	27.3	72.36

2.2 FEA of plastic injection moulding

The simulation was performed by **Moldex 3D** Viewer R16.0 software which is the world-leading CAE product for the plastic injection moulding industry with excellent class analysis and technology.

The program allows deep assessing of the simulation and FEA (Finite Element Analysis) for a large injection moulding process range and each to optimize product designs and machining.

The simulation can determine the injection mould parameters, the proper installation for the process and defects produced during the flow of material.

Previous research was focused on the FEA of the sample from ABS+PC BAYBLEND T65XF with a size of 165x192x80 mm, which results presented in the paper [13] Pop, PA, Ilea, S.

Now, the experiment is applied to different materials and geometry, such as the injected sample of ABS+PC BAYBLEND T45PG (Fig. 9) with the size of 165x192x2.5 mm, which is the material used for the recycling process.

The FEA results of the simulation injection process and the sample in the following Tables (Tabs.6-7) and Diagrams (Figs 9-15) are presented.

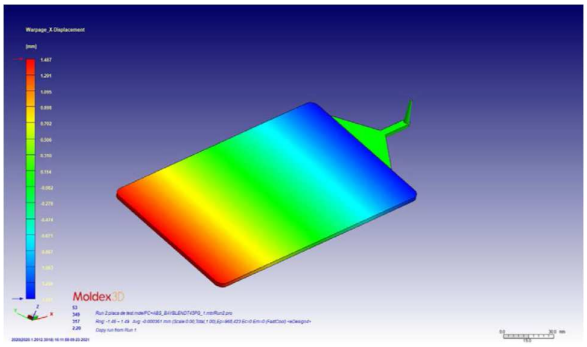

Fig.9. FEA of 3D injected sample used in the simulation analysis

The FEA diagrams show 4 steps of plastic injection moulding filling, packing, cooling and warpage, which determine the basic parameters process such as filling time, melt temperature, mould temperature, volumetric shrinkage, maximum injection pressure, injection volume, colling time, etc.

Table 6

Summary table-Mesh

Mesh Type	eDesign4
No. Cooling Channel	8
Material	BAYBLEND T45 PG
Part Dimension	165x95x2.5 (mm)
Mould Dimension	344x344x344 (mm)
MFI	12g/10min
Cavity (Part) Volume	39.131 (cc)
Cold Runner Volume	0.931688 (cc)
Element Number	968423
Part Elements	937426
NodeNnumber	1105924

Table 7

Summary table-Process Conditions

Filling Time	1.00 (sec)
Melt Temperature	250.0 (ºC)
Mould Temperature	70.0 (ºC)
Maximum Injection Pressure	221.00 (MPa)
Maximum Packing Pressure	221.00 (MPa)
Injected Volume	40.0627 (cc)
Packing Time	5.00 (sec)
Cooling Time	20.00 (sec)
Cycle Time	31.00 (sec)
VP Switch by Volume (%) Filled	95.00 (%)
Opening Mold Time	5.00 (sec)
Ejection Temperature	110.0 (ºC)
Air Temperature	25.0 (ºC)

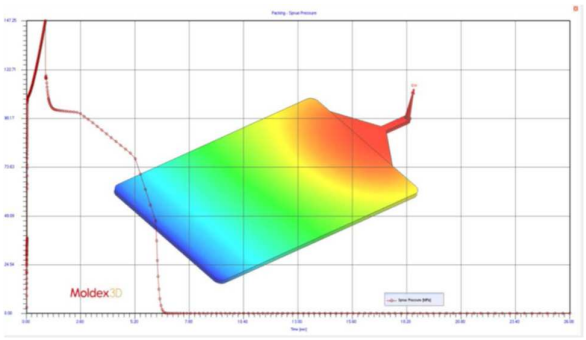

Fig.10. Diagram of packing pressure

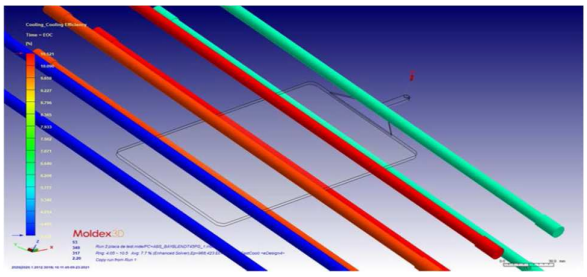

Fig.11. Diagram of cooling efficiency

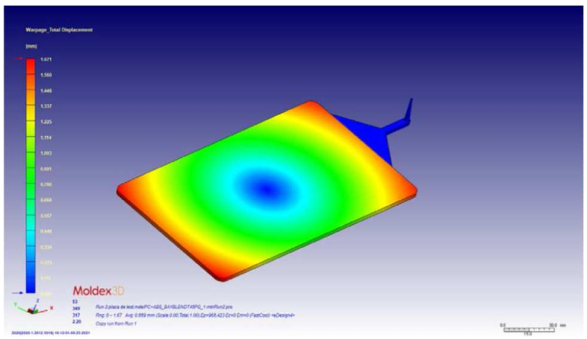

Fig.12. Diagram of warpage- Total displacement

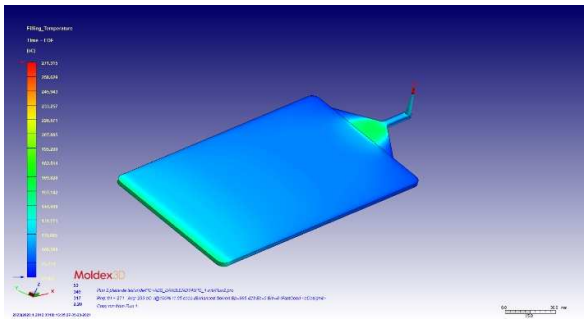

Fig.13. Diagram of filling temperature

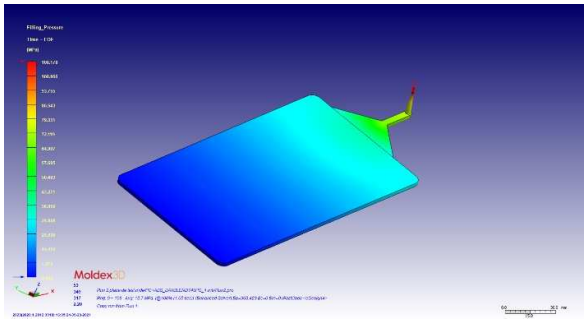

Fig.14. Diagram of filling pressure

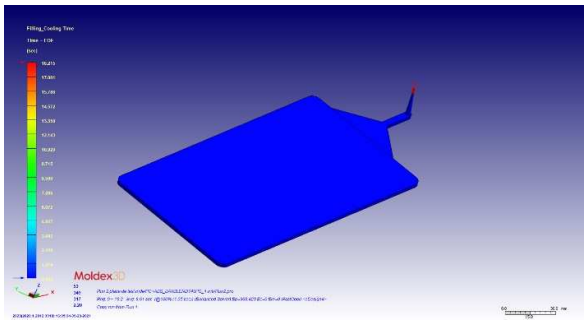

Fig.15. Diagram of filling -Cooling time

2.3. Recycled installation

The recycling installation is an automated concept station for recycling plastics that includes the complex system of pickling for industrial wasted plastics/composites with plated metals. The chemical pickling process is performed inside plated plastics and composites.

The pickling installation of industrial wasted composites is composed of the basins of chlorohydrin acids and nitric acids, recirculation pumps and filtration of acids from the two basins.

Through this process, the plated metal layers are removed from the plastic parts by successive immersion in these 2 acid basins, using a specific receipt to this process. Because during the process, gas emissions and temperature relief require more security criteria and labour

protection. The security systems are gas extraction hoods, gas washing towers, and the plant of wastewater and acid treatment. After the recovery of these materials, they will be processed to return to the processes for which they will be compatible.

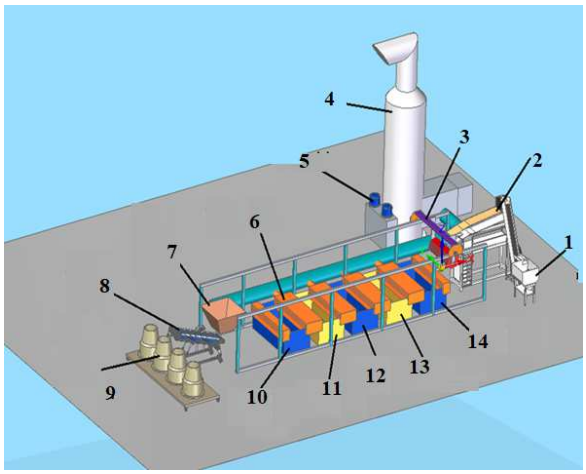

Fig. 16. Concept-station for recycling plastics
Where: **1**-Gringing mill, **2**-Filling conveyor, **3**-Robor for transporting of raw material, **4-**Gas washing tower, **5-**Acid filtration pumps, 6- Exhaust system, **7**- Raw material discharge tank, **8**-Conveyor belt, **9**-Drying bunkers, **10**-Wash basin, **11**-Nitric acid basin, **12**-Washing basin, **13**-Chlorohydric acids, **14**-Pre-washing basin.

This recycling station is composed of the following ensembles:

- The mill grinds plastic waste that is in the recycling process with a certain specific grain size.
- Conveyor belt, filling conveyor used to transport ground materials to predetermined process.
- Robot for transporting raw materials. It is a robot with 3-axis coordination used to transport the hopper to collect the materials that have been passed through the grinding process and transported to the pre-wash, pickling, washing and discharge process to the collection hopper.
- Raw material pre-washing basin, used for pre-washing the materials resulting from grinding.
- Acid pickling tanks, being used in the pickling process contain a significant amount of acids (HCl and HNO_3).
- Washing basins, are used to wash the waste that enters the pickling process.
- Exhaust gas installation, results from the pickling process where the chemical reaction takes place with the release of heat and gas emissions.
- Discharge tank of raw material used to evacuate the materials resulting from grinding, pre-washing and washing process,
- Dryers for drying the primary material at a temperature between 60-80°C, where the moisture accumulated by the material following previous processes is removed.
- Washing tower for the gases resulting from the pickling process to be able to remove air without loads of heavy metal particles into the atmosphere.
- Acid bath filtration pumps filter acids from impurities and metal particles that have precipitated and settled to the bottom of the basin.

The first recycling stage is the sorting of industrial plastic waste plated with metal, after which they are directed to a grinding mill. The waste has a granulation of 5x5 mm and the mill capacity is between 350-400 kg/hour. During the grinding process, a part of the plastic-plated waste is exfoliated contributing to metal layer removal.

The shredded material obtained is transported by a conveyor with a length of 6000 mm, and a width of 600 mm variable speed between 0-8 m/min. This conveyor supplies a hopper of 200 l with grinding material and has the aim to store material predestinated for the following processes. The hopper will be immersed in turn in the pre-washing basin, with a capacity of 2500 l of water mixed with a cleaning agent to remove certain impurities of dust resulting from grinding, and possible contamination with grease and paper.

The next stage represents the pickling process where the hopper with pre-washing material is immersed in an HCl basin of 2500 l to remove of Cr layer. At the end of this procedure, the hopper with the material is removed and immersed in the second pre-wash basin of 2500 l which contains the water to neutralize the hydrochloric acid and remove the acid vapours remaining on the material.

At the end of the pre-wash process, the hopper with the material will be immersed in a nitric acid basin of 2500 l to remove Ni and Cu

layers. Similarly, the neutralization procedure with acid and the removal of any acid vapours remaining on the material following the pickling process was achieved. After completion wash process the hopper is transported above a discharge tank the raw material. This tank was designed to prevent the material from spreading and to have the most precise control when handling the raw material in the process.

From the discharge tank, the raw material arrives on a conveyor where it is directed to the drying bunkers with a storage capacity of 500 kg each. Here, the raw material is kept at a temperature between 60-80°C, to remove the moisture accumulated by the material following the processes it went through.

The retention time in drying bunkers is 5-6 hours when it reaches the desired parameters, the new recycled material is removed outside and placed in bags and then stored in warehouses predestinated for this material.

Re-granulation, characterization of recycling samples and injection moulding part are the last proceedings to validate the recycling process and automated recycled installation.

2.4. Mechanical tests of recycled material

Tensile testing of plastic materials was made in conformity with EN ISO 527-2012 [14] and performed by INSTRON MF20 Instrument (Fig. 17).

Fig.17. INSTRON MF20 Instrument

The size of the test specimen is 150x10x5 mm, and the number of test specimens was 5. For testing, 3 types of test specimens were used, such as **A Specimen** is original ABS+PC BAYBLEND T45PG, **B Specimen** is ABS+PC BAYBLEND T45PG with 5 % recycled material, and **C Specimen** is 100 % recycled material (Fig. 18). The average results of tensile testing are presented in Table 8.

Fig.18. Test Specimens

Table 8

The tensile testing results of test specimens

	Test Specimen		
	A	**B**	**C**
Tensile load force(N)	1624.21	1592.17	1678.85
Tensile strain at break (%)	13.11	24.04	15.74
Tensile strength (MPa)	46.41	45.49	47.97
Modulus (MPa)	2550	2562	2631
Slop standard deviation at Modulus(MPa)	0.164	0.169	0.166

The difference results of tensile testing of the initial material and new recycled material are 3.17 % of Modulus and 3.29% of Tensile strength.

3. CONCLUSIONS

The chemical pickling process of plated plastics is more efficient for materials with small grains that allow the recovery of the metal and plastics.

The simulation with Moldex 3D for the injection moulding process of plastics can eliminate the possible mistakes in design and products and determine the main parameters process as a real base of PIM optimization.

The result of tensile testing applied on the initial material (sample A) and new recycled material (sample C) are appropriate results under

4% (e.g. the difference results between sample A and sample C for tensile strength is 3.22%, and modulus is 3.29%) that encourage to continue their characterization with more advanced tests to allow the validation of the recycling process and installation.

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Unele aspecte referitoare la reciclarea materialelor compozite utilizate la automobile

Lucrarea prezintă o metoda proprie de reciclare a materialelor compozite din industria auto, rezultate din componente industriale injectate. Procesul de reciclare este complex și cuprinde mai multe etape, unde materialul reciclat se obține din deșeuri industriale de PC+ABS placate cu metal. Instalația automatizata de reciclat are inserata o stație de decapare chimica pentru colectarea metalului din deșeuri industriale obținute din componente injectate. Părțile principale ale instalației de reciclare sunt o moara de măcinat deșeuri din plastic, banda transportoare, conveier de umplere, robot pentru transportarea materiei prime, bazin de pre-spălare, bazin de decapare cu acizi, bazine de spălare, instalație exhaustoare gaze și uscătoare pentru materia prima. Simularea se realizează prin intermediul software MOLDEX 3D pentru determinarea parametrilor de bază ai procesului de injectare prin matrițare a maselor plastice. Rezultatele experimentelor au confirmat metoda utilizata pentru reciclarea componentelor din materiale compozite..

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