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Green evaluation of microwave-assisted leaching process of high titanium slag on life cycle assessment

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Abstract: A greenness evaluation index and system of microwave-assisted leaching method were established. The effects of the life cycle assessment variables, such as the resource consumption, environment impact, cost, time and quality, were investigated, and the concept of green degree was applied in the production of synthetic rutile. An analytic hierarchy process was utilized to assess matrix of greenness evaluation. The Gauss-Seidel iterative matrix method was employed to solve the assessment matrix and obtain the weights and membership functions of all evaluation indexes. A fuzzy decision-making method was applied to build the greenness evaluation model, and then the scores of green degree in microwave-assisted leaching process was obtained. The greenness evaluation model was applied to the life cycle assessment of the microwave-assisted leaching process. The results show that the microwave-assisted leaching process has advantages over the conventional ones, with respect to energy-consumption, processing time and environmental protection.

Key words: life cycle assessment; greenness evaluation; microwave-assisted leaching; high titanium slag

1 Introduction

Titanium dioxide pigment accounts for more than 92% consumption of titanium minerals[1]. Rutile is the major raw material for the production of TiO_2 pigment and its main applications include manufacture of paint, paper and plastics. However, the supply of high grade natural rutile is limited[2−4]. Thus, producing synthetic rutile from abundantly available high titanium slag becomes a major alternative. How to produce high grade synthetic rutile with low energy consumption and less environment pollution becomes more and more emergent[5−6]. A new process eliminates the environmental pollution from production source, maintains titanium resources sustainable development and prepares synthetic rutile from high titanium slag by microwave-assisted leaching, which removes partial iron content and produces high grade TiO_2[7−9].

Compared with conventional heating techniques, the main advantage of microwave heating is that microwave heating is both internal and volumetric heating. Since there are a high efficiency to convert electricity energy to electromagnetic energy and no thermal conductivity mechanism involved, the heating is very rapid, uniform and highly energy efficient, resulting in energy saving and shortening the processing time[10−13]. In addition, additional advantages include greater control of the microwave heating process, no direct contact between the heating source and heated materials and reduced equipment size and waste[14−15].

The life cycle assessment (LCA) is one of the most widely used and internationally accepted methods for the evaluation of the environmental impacts of products and systems[16]. The LCA calculates the environmental burden of products extraction from raw materials through manufacturing, transporting, utilizing and disposing and identifies the particular stage of the life cycle, which causes the maximum environmental damage. Recently, many statistical and mathematical methods have been developed for LCA process[17−18]. The analytic hierarchy process (AHP) is a general problem-solving method that is useful in making complex decisions, for example, the multi-criteria decisions, based on the variables that do not have exact numerical consequences. The AHP approach determines the weight qualitatively

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by constructing multi-level decision structures and forms pair wise comparison matrices[19].

The focus of this research is to compare the preparation conditions of synthetic rutile from high titanium slag of high grade synthetic rutile content by microwave-assisted leaching and conventional leaching. The effects of the fuzzy comprehensive assessment variables, in which five decision-making aspect factors, such as resource consumption, environmental impact, cost, time and quality, are considered simultaneously.

2 Experimental

2.1 Materials

High titanium slag was prepared from ilmenite ore by carbothermal reduction in an electric arc furnace. The chemical composition of the high titanium slag is listed in Table 1. From Table 1, the slag contains 64.33% TiO_2, 25.79% Ti_2O_3 and 5.26% FeO. This particular slag is considered as a high grade slag since it contains a relatively great amount of titanium. The slag also contains 1.04% MnO, 2.75% Al_2O_3, 2.30% MgO, 2.57% SiO_2 and minor elements, such as S, P and C.

Table 1 Chemical composition of high titanium slag (mass fraction, %)

TiO_2	Ti_2O_3	FeO	Al_2O_3	SiO_2
64.33	25.79	5.26	2.75	2.57
MnO	MgO	S	P	C
1.04	2.30	0.049	0.014	0.049

2.2 Instruments

The high titanium slag was treated using the self-made microwave-assisted leaching equipment. The microwave system consists of a magnetron, a power controller, a matched load, a wave guide and a multi-mode cavity. The schematic diagram of the microwave heating equipment is shown in Fig.1. The microwave power supply for the microwave heating equipment consists of two magnetrons with frequency of 2.45 GHz and power of 1.5 kW, which are cooled by water circulation. A polytetrafluoroethene column container with inner diameter of 300 mm and length of 200 mm was positioned at the center of the microwave stainless steel oven. An attached infrared pyrometer (Marathon Series, Raytek, USA) used to monitor the temperature of the sample has the circular crosswire focusing on the sample cross-section.

2.3 Procedure

Prior to the use, high titanium slag was crushed and sieved to obtain particles with size less than 0.2 mm.

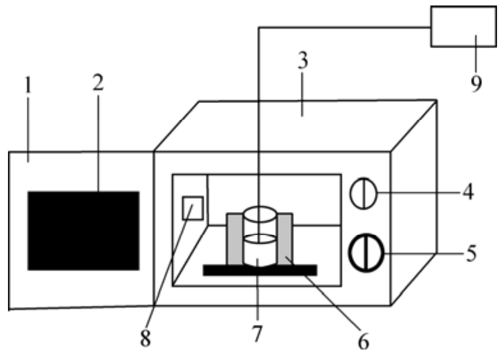

Fig.1 Schematic diagram of self-made microwave-assisted leaching equipment: 1—Oven door; 2—Observation door; 3—Microwave multi-mode cavity; 4—Timer; 5—Power controller; 6—Fireproof materials; 7—Raw materials; 8—Ventilation hole; 9—Temperature measurement system

Subsequently, high titanium slag was loaded on a ceramics boat which was placed inside a stainless steel tubular reactor, whose internal diameter is 38 mm. The samples were heated to 120 ℃ at a heating rate of 5 ℃/min in the drying oven and held at a this temperature for 2 h. After drying, the samples were cooled to room temperature. The total mass of sample was 100 g. The sample was placed in the muffle furnace and heated to 950 ℃ for 60 min, and then it was naturally cooled in the furnace to room temperature. After reaction, the sample with mass of about 80 g was then subjected to microwave-assisted acid leaching. The microwave-assisted conditions were as follows: hydrochloric acid concentration 20%, reaction temperature 80 ℃, solid-liquid ratio 1:10 and reaction time 60 min. Thus, synthetic rutile was prepared from high titanium slag by microwave-assisted leaching process. Simultaneously microwave-assisted leaching conditions were compared with the conventional acid leaching conditions.

3 Analytic hierarchy process

3.1 Assessment of hierarchical structure

Like many other methods, AHP allows decision makers to create a model for a complex problem with a hierarchical structure[20−21]. In this study, the top level of the hierarchy structure on life cycle assessment is the overall goal of the leaching process of high titanium slag. The following levels describe the tangible criterion and sub-criterion that contribute to the goal. The bottom level is formed by the alternatives to make evaluations in terms of criterion. The hierarchical structure on life cycle assessment of leaching process of high titanium slag is shown in Fig.2.

3.2 Pairwise comparison

As soon as the AHP logic hierarchical structure is

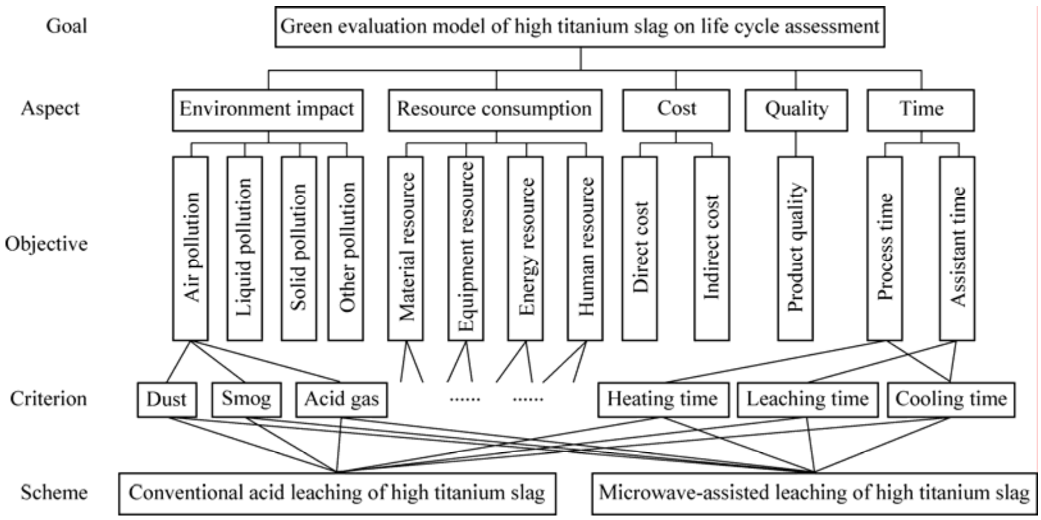

Fig.2 Hierarchical structure on life cycle assessment of leaching process of high titanium slag

formulated, one should first yield the judgment matrices based on pairwise comparison of all elements in each hierarchy level with respect to the higher hierarchy level according to certain criteria of comparison within set scales[22]. The Saaty's scales of pairwise comparisons are listed in Table 2[23].

Table 2 Scale of pairwise comparisons

Importance	Definition	Explanation
1	Of equal value	Two activities contribute equally to objective
3	Slightly more value	One is slightly in favor over another
5	Essential or strong value	One is strongly in favor over another
7	Very strong value	Dominance of one element proved in practice
9	Extreme value	Highest order dominance of one element over another
2, 4, 6, 8	Intermediate values	When compromise is needed between two adjacent judgments

For each criterion and weight of the hierarchy structure, all the relevant elements are compared in pairwise comparison matrices as follows

$$P=\begin{bmatrix} 1 & \frac{w_1}{w_2} & \Lambda & \frac{w_1}{w_n} \\ \frac{w_2}{w_1} & 1 & \Lambda & \frac{w_2}{w_n} \\ \mathrm{M} & \mathrm{M} & \mathrm{O} & \mathrm{M} \\ \frac{w_n}{w_1} & \frac{w_n}{w_2} & \Lambda & 1 \end{bmatrix} \tag{1}$$

where P is comparison pairwise matrix; w_1 is the relative importance of criterion 1; w_2 is the relative importance of criterion 2; w_n is the relative importance of criterion n.

An eigenvalue method and the Gauss-Seidel iterative matrix are used to calculate the relative weight of elements in each pairwise comparison matrix. The biggest eigenvalue is obtained from the following equation.

$$\det[\boldsymbol{P}-\lambda_{\max}\boldsymbol{I}]=0 \tag{2}$$

where $\lambda_{\max}$ is the biggest eigenvalue of matrix $\boldsymbol{A}$; $\boldsymbol{I}$ is unit matrix. Then, the relative weight ($\boldsymbol{A}$) of matrix ($\boldsymbol{P}$) is obtained from the following equation:

$$(\boldsymbol{P}-\lambda_{\max}\boldsymbol{I})\times\boldsymbol{A}=0 \tag{3}$$

The biggest eigenvalue is obtained by solving the eigenvalue Eq.(2) and shown in Eq.(4). The relative weight was obtained by solving and normalizing the eigenvalue Eq.(3).

$$\lambda_{\max}=\frac{1}{n}\sum_{i=1}^{n}\left\{\frac{\sum_{j=1}^{n}a_{ij}\times w_j}{w_i}\right\} \tag{4}$$

3.3 Definition of assessment matrices

Assessment aspects matrix is expressed as

$$\boldsymbol{U}=\{U_1,\ U_2,\ \cdots,\ U_i\} \tag{5}$$

where i is the number of assessment aspects.

Assessment objective matrix is expressed as

$$\boldsymbol{U}_i=\{U_{i1},\ U_{i2},\ \cdots,\ U_{ij}\} \tag{6}$$

where j is the number of assessment aspects of $\boldsymbol{U}_i$.

Assessment criterion matrix is expressed as

$$\boldsymbol{U}_{ij}=\{U_{ij1},\ U_{ij2},\ \cdots,\ U_{ijk}\} \tag{7}$$

where k is the number of assessment criterion of $\boldsymbol{U}_{ij}$.

3.4 Definition of weight matrices

The weight of assessment aspects is expressed as

$$\boldsymbol{A}=\{a_1, a_2, \cdots, a_i\} \tag{8}$$

where a_i is the ith weight of assessment aspects and the boundary conditions can be given as follows.

$$\begin{cases} 0 < a_i \le 1 \\ \sum_{i=1}^{i} a_i = 1 \end{cases} \tag{9}$$

The weight of assessment objective is expressed as

$$\boldsymbol{A}_i=\{a_{i1}, a_{i2}, \cdots, a_{ij}\} \tag{10}$$

where a_{ij} is the jth weight of assessment objective of a_i and the boundary conditions can be given as follows:

$$\begin{cases} 0 < a_{ij} \le 1 \\ \sum_{j=1}^{j} a_{ij} = 1 \end{cases} \tag{11}$$

The weight of assessment criterion is expressed as

$$\boldsymbol{A}_{ij}=\{a_{ij1}, a_{ij2}, \cdots, a_{ijk}\} \tag{12}$$

where a_{ijk} is the kth weight of assessment criterion of a_{ij} and the boundary conditions can be given as follows.

$$\begin{cases} 0 < a_{ijk} \le 1 \\ \sum_{k=1}^{k} a_{ijk} = 1 \end{cases} \tag{13}$$

3.5 Definition of assessment matrices

The fourth level of fuzzy assessment matrices is expressed as

$$\boldsymbol{R}_{\mathrm{ij}} = \begin{bmatrix} r_{i11} & r_{i12} & \Lambda & r_{i1n} \\ r_{i21} & r_{i22} & \Lambda & r_{i2n} \\ \mathrm{M} & \mathrm{M} & \mathrm{M} & \mathrm{M} \\ r_{im1} & r_{im2} & \Lambda & r_{imn} \end{bmatrix} \tag{14}$$

where r_{imn} is the value of the fuzzy subset membership function between mth assessment criterion and nth assessment criterion.

The third level of fuzzy assessment matrices is expressed as

$$\boldsymbol{R}_{ij} = \boldsymbol{A}_{ij} \circ \boldsymbol{R}_{ijk} \tag{15}$$

where $\boldsymbol{R}_{ijk}$ is the fourth level fuzzy assessment matrices; $\boldsymbol{A}_{ij}$ is the weight matrices of assessment criterion; o gives the fuzzy relation comprehensive algorithms.

The second level of fuzzy assessment matrices is expressed as

$$\boldsymbol{R}_i = \boldsymbol{A}_i \circ \boldsymbol{R}_{ij} \tag{16}$$

where $\boldsymbol{R}_{ij}$ is the third level fuzzy assessment matrices; $\boldsymbol{A}_i$ is the weight matrices of assessment objective.

The first level of fuzzy assessment matrices is expressed as

$$\boldsymbol{R} = \boldsymbol{A} \circ \boldsymbol{R}_i \tag{17}$$

where $\boldsymbol{R}_i$ is the second level fuzzy assessment matrices; $\boldsymbol{A}$ is the weight matrices of assessment aspects.

3.6 Definition of green grade matrix

In order to obtain a final result, the following expression is used to represent the green grade matrix set of each assessment aspects.

$$\boldsymbol{V}=\{V_1, V_2, \cdots, V_i\} \tag{18}$$

where V_i is the ith assessment grade of assessment aspects.

In order to obtain sensible comparisons, the expert judging and multi-fuzzy assessment theory are applied in the life cycle assessment process. Accordingly, the fuzzy assessment set $\boldsymbol{V}$ can be expressed by the green grade of the assessment aspects as follows:

$$\boldsymbol{V}=\{1.0, 0.8, 0.7, 0.6, 0.5\} \tag{19}$$

3.7 Green evaluation results

The green evaluation results of high titanium slag under microwave-assisted leaching and conventional acid leaching are obtained from the following equation, which can be derived from Eq.(17) and Eq.(19).

$$Y=\boldsymbol{V}\boldsymbol{R}^{\mathrm{T}}\times 100 \tag{20}$$

where Y is the green evolution results.

4 Results and discussion

In order to demonstrate the universal application of the greenness evaluation index and assessment system of leaching process of high titanium slag for life cycle assessment, the microwave-assisted leaching process and conventional acid leaching process are compared with each other under same evaluation system. The weight values are used and the assessment matrices of both conventional acid leaching and microwave-assisted leaching are shown in Tables 3 and 4, respectively. The comparison of these two leaching process can be considered as a decision making model since it guides decision makers to choose best alternative for the experiment.

From the above theoretical work, the detailed calculation processes can be carried out according to fuzzy comprehensive assessment method and analytic hierarchy process. The fuzzy comprehensive assessment results are shown in Table 5. The results show that microwave-assisted leaching process have advantages

Table 3 Life cycle assessment of conventional acid leaching of high titanium slag

Assessment aspect	Weight	Assessment objective	Mass/g	Assessment criterion	Weight	Assessment matrice				
Environment impact	0.25	Air pollution	0.30	Dust	0.30	0.2	0.3	0.2	0.2	0.1
				Smog	0.30	0.1	0.3	0.2	0.2	0.2
				Acid gas	0.40	0.2	0.3	0.3	0.1	0.1
		Liquid pollution	0.25	Leaching liquid	0.45	0.2	0.3	0.1	0.3	0.1
				Washing liquid	0.25	0.2	0.3	0.2	0.2	0.1
				Absorbing liquid	0.30	0.3	0.2	0.3	0.1	0.1
		Solid pollution	0.25	Waste slag	0.40	0.1	0.4	0.2	0.2	0.1
				Additive	0.35	0.2	0.2	0.2	0.3	0.1
				Adhesive	0.25	0.1	0.3	0.4	0.2	0
		Other pollution	0.20	Noise	0.25	0.4	0.1	0.1	0.3	0.1
				Thermal radiation	0.30	0.5	0.2	0.1	0.1	0.1
				Other	0.45	0.2	0.3	0.1	0.3	0.1
Resource consumption	0.25	Material resource	0.35	Materials consume	0.35	0.1	0.3	0.2	0.4	0
				Materials utilization	0.40	0.1	0.3	0.2	0.2	0.2
				Materials recovery rate	0.25	0.2	0.5	0.2	0.1	0
		equipment resource	0.20	Equipment utilization	0.70	0.3	0.3	0.2	0.1	0.1
				Automation	0.30	0.4	0.2	0.2	0.1	0.1
		Energy resource	0.30	Energy utilization	0.55	0.2	0.2	0.4	0.1	0.1
				Renewable resource	0.35	0.1	0.4	0.3	0.1	0.1
				Surplus energy	0.10	0.2	0.2	0.4	0.1	0.1
		Human resource	0.15	Professional	0.55	0.1	0.3	0.4	0.1	0.1
				Administrator	0.25	0.2	0.4	0.1	0.2	0.1
				Knowledge	0.20	0.1	0.5	0.3	0.1	0
Cost	0.2	Direct cost	0.60	Design cost	0.45	0.2	0.3	0.3	0.1	0.1
				Materials cost	0.30	0.5	0.2	0.1	0.1	0.1
				Processing cost	0.25	0.2	0.4	0.1	0.2	0.1
		Indirect cost	0.40	Maintenance fee	0.55	0.2	0.3	0.2	0.2	0.1
				governance fee	0.30	0.3	0.1	0.4	0.1	0.1
				Waste disposal fee	0.15	0.3	0.2	0.2	0.2	0.1
Quality	0.15	Product quality	1.0	Product content	0.40	0.1	0.3	0.4	0.1	0.1
				Microstructure	0.40	0.2	0.5	0.1	0.2	0
				Surface area	0.20	0.3	0.1	0.1	0.3	0.2
Time	0.15	Process time	0.65	Heating time	0.35	0.3	0.1	0.3	0.2	0.1
				Cooling time	0.65	0.1	0.2	0.4	0.1	0.2
		Assistant time	0.35	Leaching time	0.35	0.3	0.1	0.3	0.1	0.2
				Cooling time	0.65	0.3	0.4	0.1	0.2	0

Table 4 Life cycle assessment of microwave-assisted leaching of high titanium slag

Assessment aspects	Weight	Assessment objective	Mass/g	Assessment criterion	Weight	Assessment matrice				
Environment impact	0.25	Air pollution	0.30	Dust	0.30	0.4	0.3	0.2	0.1	0
				Smog	0.30	0.5	0.3	0.1	0.1	0
				Acid gas	0.40	0.3	0.3	0.2	0.1	0.1
		Liquid pollution	0.25	Leaching liquid	0.45	0.4	0.2	0.2	0.1	0.1
				Washing liquid	0.25	0.5	0.2	0.2	0.1	0
				Absorbing liquid	0.30	0.3	0.3	0.2	0.2	0
		Solid pollution	0.25	Waste slag	0.40	0.2	0.4	0.2	0.1	0.1
				Additive	0.35	0.4	0.1	0.3	0.1	0.1
				Adhesive	0.25	0.4	0.2	0.1	0.2	0.1
		Other pollution	0.20	Noise	0.25	0.3	0.3	0.1	0.2	0.1
				Thermal radiation	0.30	0.4	0.3	0.2	0.1	0
				Other	0.45	0.3	0.1	0.4	0.1	0.1
Resource consumption	0.25	Material resource	0.35	Materials consume	0.35	0.5	0.1	0.2	0.1	0.1
				Materials utilization	0.40	0.4	0.1	0.3	0.1	0.1
				Materials recovery rate	0.25	0.5	0.2	0.1	0.1	0.1
		Equipment resource	0.20	Equipment utilization	0.70	0.3	0.2	0.3	0.1	0.1
				Automation	0.30	0.4	0.1	0.3	0.1	0.1
		Energy resource	0.30	Energy utilization	0.55	0.4	0.1	0.2	0.3	0
				Renewable resource	0.35	0.3	0.4	0.1	0.1	0.1
				Surplus energy	0.10	0.3	0.3	0.3	0.1	0
		Human resource	0.15	Professional	0.55	0.3	0.2	0.3	0.1	0.1
				Administrator	0.25	0.2	0.5	0.1	0.1	0.1
				Knowledge	0.20	0.3	0.4	0.2	0.1	0
Cost	0.2	Direct cost	0.60	Design cost	0.45	0.6	0.1	0.1	0.1	0.1
				Materials cost	0.30	0.5	0.2	0.1	0.1	0.1
				Processing cost	0.25	0.4	0.2	0.3	0.1	0
		Indirect cost	0.40	Maintenance fee	0.55	0.2	0.4	0.2	0.1	0.1
				governance fee	0.30	0.4	0.1	0.3	0.2	0
				Waste disposal fee	0.15	0.5	0.1	0.2	0.1	0.1
Quality	0.15	Product quality	1.0	Product content	0.40	0.3	0.2	0.3	0.1	0.1
				Microstructure	0.40	0.3	0.3	0.3	0.1	0
				Surface area	0.20	0.3	0.1	0.4	0.1	0.1
Time	0.15	Process time	0.65	Heating time	0.35	0.4	0.3	0.1	0.1	0.1
				Cooling time	0.65	0.3	0.3	0.2	0.1	0.1
		Assistant time	0.35	Leaching time	0.35	0.4	0.1	0.3	0.1	0.1
				Cooling time	0.65	0.3	0.3	0.1	0.2	0.1

Table 5 Fuzzy comprehensive assessment results for microwave-assisted leaching and conventional acid leaching

Leaching condition	Green degree
Microwave-assisted leaching	86.51
Conventional acid leaching	81.28

over the conventional one, with respect to energy-consumption, processing time and environmental protection, since the microwave-assisted leaching is very rapid, uniform and highly energy efficient. Thus, the microwave-assisted leaching process is better than conventional acid leaching process, which fulfill with the aim of green manufacture.

4 Conclusions

1) The green degree of microwave-assisted leaching is 86.51, a relatively high value, indicating that microwave-assisted leaching is better than conventional acid leaching, especially in terms of environment impact and resource consumption, which is consistent with the aim of life cycle assessment.

2) Microwave-assisted leaching process of high titanium slag has a potential to provide a new method to prepare synthetic rutile with high efficiency and low energy consumption.

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