# Effect of Coupling Agent on Interfacial Bonding Properties of Viscoelastic Magnetic Abrasive Tools and Finishing Performance

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***Abstract* — In magnetic abrasive finishing processes, abrasive tools play a major role on finishing effects. Finishing of complex shaped components needs advanced abrasive tools to produce nano level surface finish. As new abrasive tools, viscoelastic magnetic abrasive tools, made from a mixture of polymer matrix (Polymethyl-vinyl siloxane rubber), magnetic medium, abrasives, and other additives, have excellent viscoelasticity and fluidity. But easy debonding between the magnetic medium, the abrasives and the matrix leads to the uncertain property of the tools. Therefore, those tools have a short life and further disadvantages such as the instability of the physical and chemical properties in the finishing processing and low bonding strength between the filler particles and the matrix. This paper establishes a mathematical model of interface debonding, proposes a surface modification method for the filler particles using silane coupling agents or titanate coupling agents, analyzes the mechanism of the bonding between the surface-modified filler particles and the matrix, and determines the finishing effects of coupling agent on the change in average surface roughness and the surface defects by finishing exepriments on H62 brass tube. It is concluded that the effects of surface modification of the Fe and SiC particles by the coupling agent are favorable and the finishing effect of coupling agent KH-550 is the best. The surface treatment can improve the dispersion and compatibility of the Fe and SiC particles in the matrix, increase the interface bonding strength, and improve the processing capability of viscoelastic magnetic abrasive tools.**

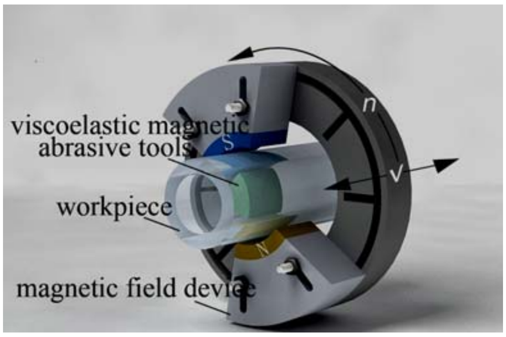
***Keywords — Viscoelastic magnetic abrasive tools；Finishing；Coupling agent；Surface treatment；Interface bonding***

## I. INTRODUCTION

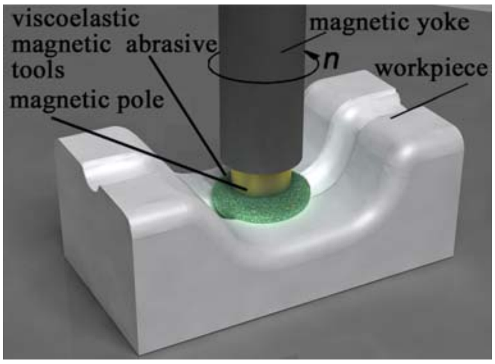
Magnetic abrasive finishing is a new kind of high-precision surface finishing technology based on magnetic abrasives employed in a magnetic field. Magnetic abrasives commonly used at present are tiny particles composed of abrasive powder, ferromagnetic materials, and binders. The technique has good flexibility and adaptability and can be used for the finishing of complex parts, internal and cylindrical surfaces, planes, and for chamfering and deburring. However, magnetic abrasives are difficult to be bought into full contact with the surface to be processed because of narrow grooves, complex cavities, and other surface irregularities, resulting in unfavorable processing effects. Meanwhile, the abrasives are easily dispersed with corresponding low ratio of repeated usage and the cost is high[1,2,3,4,5,6].

Researchers at Taiyuan University of Technology propose a new kind of viscoelastic magnetic abrasive tool, in which at a certain viscosity and elasticity of the polymer matrix, ferromagnetic particles and abrasives are filled with appropriate proportions. Then, the components are fully mixed, thereby producing magnetic abrasive tools with tailored viscoelasticity and fluidity (Fig. (1).). These tools can be well adapted to various complex curved parts by the formation of a "flexible polishing film" on the machined surfaces. The technique has the advantages of simple preparation, low processing temperature, and wide range of applications [7,8]. The principle of viscoelastic magnetic abrasive finishing of internal and complex surfaces is shown in Fig. (2), namely, a certain amount of viscoelastic magnetic abrasives is placed on the machined surface and, because of its own liquidity, being in full contact with the machined surface, forming a "flexible polishing film." Under the action of a magnetic field, the viscoelastic abrasive tools exert magnetic forces on the work-piece surface and fulfill a complex relative movement by the magnetic pole's rotation and axially reciprocating linear motion, thus achieving the finishing of the whole machined surface.

Fig.1 Viscoelastic Magnetic Abrasive Tools




(a) Internal surface




(b) Complex surface

Fig.2. Viscoelastic Magnetic Abrasives Finishing Schematic Diagram

X. H. Li and W. H. Li of Taiyuan University of Technology have conducted experimental studies on the finishing of flat, cavity, and complex surfaces using viscoelastic magnetic abrasives tools. After 15 min of finishing, the surface roughness value decreased by 2–3 levels, and the surface quality was greatly improved[9,10]. However, the abrasive tools were heated, adhered to the work-piece, and the filler particles could debond from matrix in the finishing process. Therefore, the service life of the abrasive tools was shortened and the surface quality of the work-piece deteriorated. This paper analyzes the causes of interface debonding of viscoelastic magnetic abrasive tools. Moreover, surface treatment is applied to the tools and the resulting properties are determined by finishing experiments.

## II. MATHEMATICAL MODEL OF INTERFACE DEBONDING

Viscoelastic magnetic abrasive tools are composed of a polymer matrix (Polymethyl-vinyl siloxane rubber) with a relatively low modulus, ferromagnetic particles, and abrasives with a relatively high modulus. The polymer matrix is very different from the abrasive and ferromagnetic particles in terms of structure, and their chemical and physical properties are incompatible, resulting in fully developed interfaces between them. The finishing performance and service life of viscoelastic magnetic abrasive tools are largely dependent on the interfacial bonding strength between matrix and abrasives or ferromagnetic particles. In the case of a uniform distribution throughout the interface zone, stress is evenly transferred and the abrasive tools show excellent comprehensive performance. However, during the finishing process, a large number of ferromagnetic particles debonds from the matrix and aggregates at the surface of the abrasive tools due to the action of the magnetic force, leading to loss of the finishing effect[11,12].

W. Peng of the National University of Defense Technology developed the two-phase sphere-analysis model of viscoelastic de-wetting of the composite solid propellants based on the elasticity–viscoelasticity correspondence principle. He studied the mechanisms of interfacial de-wetting and derived a simple formula for the local critical de-wetting stress at the interface between particle and matrix [13].

The model uses a spherical cell as representative volume element (RVE) of the material, which contains two model phases in form of concentric spheres. If the radius of the spherical particles is $a$ and the outer diameter of the spherical matrix is $b$, the volume fraction of the particles is $f=\left(\frac{a}{b}\right)^3$. If a spherically symmetric load $F(t)$ is applied to the outer surface of the spherical cell, in the moment of de-wetting ($t_c$), the critical de-wetting stress is as follows:

$$\sigma_{rr}^{p}(t_c)=\frac{2G_c(1-f)}{3F(t_c)a(1-v_m)} \tag{1}$$

where $G_c$ is the interface bonding energy per unit of surface area, $p$ and $m$ represent the corresponding amounts of filler particles and matrix, respectively, and $v_m$ is the loading rate.

The filler particles of viscoelastic magnetic abrasives are composed of two components, namely ferromagnetic particles and abrasives. In the finishing process, compared with the abrasives, the ferromagnetic particles are not only affected by the interaction with the work-piece, but also by the magnetic force. Therefore, the interface between ferromagnetic particles and matrix is easier debonded, implying that the radius and volume fraction of the particles prominently affect the performance of the viscoelastic magnetic abrasive tools. Therefore, Eqn. (1) of the critical de-wetting stress at the interface between particles and matrix is modified, resulting in Eqn. (2).

$$\sigma_{rr}^{p}(t_c)=\alpha\times\frac{2G_1(1-f)}{3F(t_c)a(1-v_m)}+\beta\times\frac{2G_2(1-f)}{3F(t_c)a(1-v_m)} \tag{2}$$

where $R_T$ is the radius of the ferromagnetic particles, $R_M$ is the radius of the abrasives, $f_T$ is the volume fraction of ferromagnetic particles, $f_M$ is the volume fraction of abrasives, $G_1$ is the interface bonding energy between ferromagnetic particles and matrix, $G_2$ is the interface bonding energy between abrasives and matrix, $\alpha$ is the impact factor of the ferromagnetic particles on the critical debonding stress, and $\beta$ is the impact factor of the abrasives on the critical debonding stress.

As can be seen from Eqn. (2), the larger the size and volume fraction of the filler particles, the easier debonding. However, in the preparation of the viscoelastic magnetic abrasive tools, in order to ensure the finishing effect, the size and volume fraction of the particles have to be within specified ranges, the usual particle size is 100–300 mesh and the volume fraction varies from 30% to 50%. Because of the effect of the magnetic field, the ferromagnetic particles easily drop out in the finishing process. To ensure effective cementation of the filler particles in the matrix, active functional groups were introduced into the matrix, making the interface of the two phases additionally linked by chemical bonds.

## III. COUPLING-AGENT INDUCED SURFACE MODIFICATION AND ACTION MECHANISM

The ferromagnetic particles and abrasives of viscoelastic magnetic abrasive tools are inorganic materials, and the matrix is a polymer. If the ferromagnetic particles and abrasives are directly filled into the matrix, it is difficult to establish chemical bonds at the interfaces of the respective two phases to ensure their effective cementation in the matrix because of their poor physic-chemical compatibility. Moreover, the stress concentration in the interface zone will become more and more serious with the increase in size and volume fraction of the incorporated particles, thereby easily causing interface debonding. It is feasible to modify the surface of the inorganic particles by chemical reaction or physical action using a surface modifier. The dispersion, stability, compatibility, and interfacial bonding performance of the filler particles with the matrix can be significantly improved by their surface modification, which increases the performance and service life of the viscoelastic magnetic abrasive tools.

The coupling agent is a substance having amphoteric structure. One end of its molecules can react with the filler particles by the formation of chemical bonds and the other end can form chemical bonding and physical entanglement with the polymer matrix, setting up a "molecular bridge" between the filler particles and the polymer matrix, thereby enhancing their interaction and enhancing the interfacial binding force[14,15].

Considering that the matrix is Polymethyl-vinyl siloxane rubber(MVQ) and the filler particles are Fe and SiC particles, silane and titanate coupling agents were selected for the surface modification of the filler particles to improve the performance of the viscoelastic magnetic abrasives.

### A. Silane Coupling Agent

The basic structure of the silane coupling agent is shown in Eqn. (3):

$$\mathrm{R_n-SiX_{(4\text{-}n)}} \tag{3}$$

R represents organic hydrophobic groups with strong affinity and reaction ability with polymer molecules, such as methyl, vinyl, amino, and naphthenic groups, X represents hydrolyzable groups that can decompose if exposed to aqueous solutions, water in air, or react if adsorbed at the surfaces of inorganic substances. Typical X groups include alkoxy, aryloxy, and acyl.

Before the preparation of the viscoelastic magnetic abrasives tools, the surfaces of the filler particles are treated by the silane coupling agent. The X groups of the coupling agent generate dehydration reaction, thereby forming low-molecular polymer with hydroxy groups that in turn can react with hydroxy of the inorganic surface via hydrogen bonds. The filler material is then heated and dried at a certain temperature, forming covalent bonds by partly dehydration, thus resulting in a cover layer of the silane coupling agent on the surface of the filler particles. Once adding the modified filler particles into the matrix, the R groups of the silane coupling agent would react with the groups of the polymer matrix, thus forming covalent bonds.

### B. Titanate Coupling Agent

The structure of the titanate coupling agent is shown in Eqn. (4):

$$\overleftarrow{(R_1-O)_m}^{\text{React with inorganics}} \rightarrow T_i-\overrightarrow{(O-X-R_2-Y)_n}^{\text{React with organic}} \tag{4}$$

under the constraint of $1\le m\le 4$, $1\le m+n\le 6$. $R_1$ is the short chain that can react with the alkyl group at the surface of the inorganic filler particles, forming a monolayer. $R_2$ is the long alkane chain showing entanglement and miscibility with the polymer molecule. $X$ represents groups containing C/N/S/P and other elements. $Y$ indicates functional groups crosslinking with the titanate coupling agent, such as hydroxy and epoxy groups.

In the preparation of viscoelastic magnetic abrasive tools, isopropoxide at one end of the titanate agent would generate coupling reaction with hydroxyl at the surface of the filler particles, thus forming an organic active monolayer on the surfaces of the ferromagnetic and abrasive particles. This process changes the hydrophilicity of the particles into lipophilicity, thereby increasing their compatibility with the polymer matrix and resulting in unsaturated long carbon chains entangled in the polymer matrix. The reaction mechanism between the titanate coupling agent and filler particles is shown in Fig. (3).

## IV. SURFACE-MODIFICATION PROCESS OF THE FILLER PARTICLES

Viscoelastic magnetic abrasive tools generally use iron powder as magnetic medium and silicon carbide as abrasive. Therefore, the surface-modifying methods regarding the filler particles should be determined according to the chemical properties of SiC and Fe for improving the interface bonding strength.

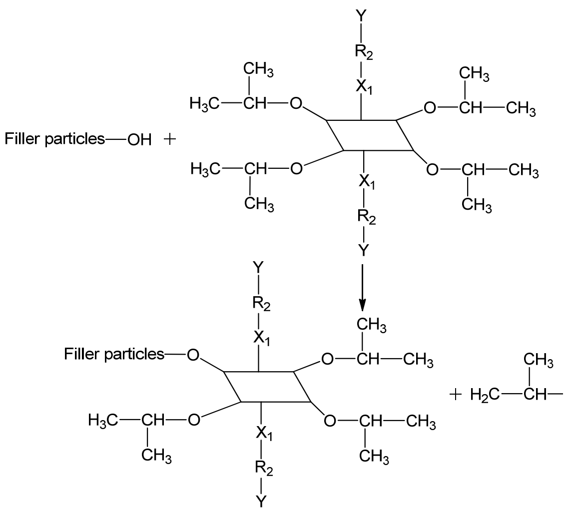


Fig.3 Reaction Mechanism Between Titanate Coupling and Filler Particles

### *A. Surface-Modification Process of SiC Particles*

If exposed to air, SiC is easily oxidized, forming an oxide film on its surface. The main component of the oxide film is SiO2, which can absorb the moisture contained in air, forming silanol (Si–OH), as shown in Fig. (4). The larger the mesh number used for the sample preparation, the smaller the size of the SiC particles and the greater their surface energy. Therefore, small SiC particles are easily oxidized.

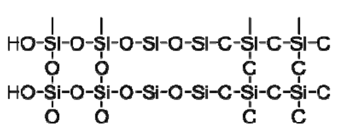


Fig.4 The Structure Diagram of The SiC Oxide Film

The hydroxy groups (–OH) at the surface of the SiC oxide film can react with alkoxy very easily. One end of the silane coupling agent contains alkoxy groups and the other end contains groups that can react with the polymer. Therefore, the silane coupling agent KH-550 (chemical name: 3-aminopropyl tri-ethoxysilane (H2N(CH2)3Si(OCH2CH3)3) with an organic hydrophobic group is selected as surface modifier. The surface-modification process is shown in Fig.(5).

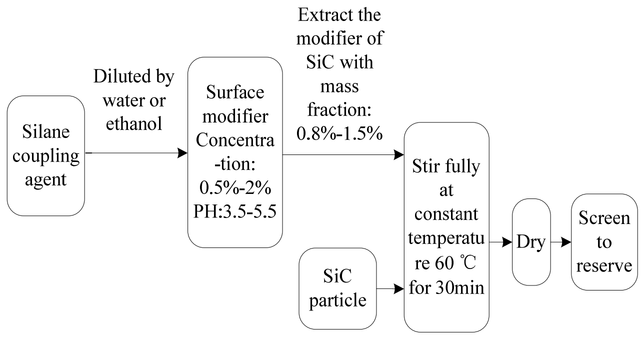



Fig.5 SiC Surface Modification Treatment Process

After the surface modification, the silane coupling agent will react with the alkoxy groups at the surface of the SiC particles, thereby forming a coating layer. The R groups of the silane coupling agent point outward, resulting in strong affinity to the polymer matrix, as shown in Fig. (6).

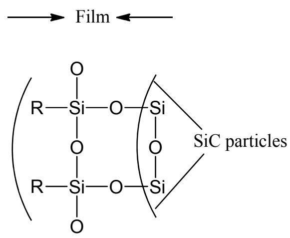



Fig.6 The Surface Chemical Bonding Model of SiC Particles After The Surface Modification Using Silane Coupling Agent

### *B. Fe Surface-Modification Process*

Fe does not easily react with oxygen in dry air, but readily forms iron oxide at the surface in moist air and, more difficultly, hydroxyl. Since only the surface of Fe contains hydroxyl, the silane coupling agent can contribute to the bonding strength of the interface between Fe paritcles and matrix. Therefore, iron powder should first be poured into alkaline solution, inducing hydroxylation and removing grease, dust, and other impurities from its surface. Then, the iron powder is immersed into water for cleaning. Thus, the surface of the iron powder would be coated with a continuous film of water molecules. Finally, after filtering, the iron powder is completely immersed into the prepared silane coupling agent, thereby enhancing the number of alkaline hydroxyl groups at the surface of Fe, forming a continuous, stable, and tight silane film. The surface-modification process is shown in Fig.( 7).

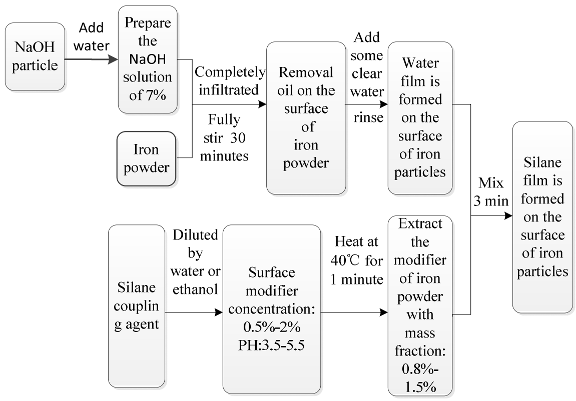



Fig.7 Fe Surface Modification Process

The abrasive and the modified magnetic phase are mixed with the polymer matrix by efficient stirring, preparing the viscoelastic magnetic abrasive tools.

## V. EXPERIMENTAL STUDY

### *A. Experimental Device And Conditions*

The experiment device is shown in Fig. (8). The device includes four motors, the work-piece fixing device, belt drive, the magnetic field generator, magnetically axial feed device, and magnetic pole vibration device. Motor 1 rotates the work-piece. The magnetic field generator is composed of two permanent magnets and the yoke. The poles are arranged perpendicular to the inner yoke. Motor 2 can rotate the magnetic field generator by the belt drive. Motor 3 can drive the magnetic field generator along the axial direction. Finally, Motor 4 can drive the poles, thereby realizing axial vibrations. When the viscoelastic magnetic abrasive tools were placed in the cavity of the work-piece, they would cling to the inner surface of the work-piece under the action of the magnetic force. When the magnet and/or the work-piece moved, the viscoelastic magnetic abrasive tools would achieve finishing of the inner surface. The experimental parameters are shown in Table I.

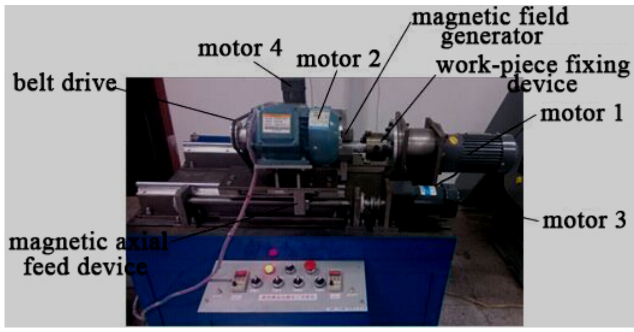



Fig.8 Experimental Device

TABLE I. THE EXPERIMENTAL PARAMETERS

| Items | Parameters |
|---|---|
| Sample(external diameter×inner diameter×length) | H62 brass tube（$\varphi$35mm×$\varphi$28mm×60mm） |
| The magnetic field generator | Two poles arranged in 90° |
| The magnetic induction intensity in the machining region（T） | 0.6～0.8 |
| The distance between sample and the pole（mm） | 3 |
| The rotating speed/(r/min) | 460 |
| The axial vibration | Amplitude 15mm，Frequency 2.3Hz |
| The finishing time(min) | 15 |
| The mass of viscoelastic magnetic abrasive (g) | 20 |

### *B. Influence of The Coupling Agent on The Finishing Effect*

The viscoelastic abrasive tools used in the experiments are shown in Table II.

Finishing experiments of three work pieces were performed to determine the effect of the surface modification of the viscoelastic abrasive tools. The surface roughness curve in dependence of time is shown in Fig. 9. As evident from Fig. (9), the surface-roughness value decreased from 0.944 µm to 0.226 µm (reduced by 0.718 µm) without the coupling agent. However, using Fe and SiC particles surface modified by the silane coupling agent (whose amount is 1% of the mass of the modified particles), the surface roughness value of the three work pieces was reduced by 0.723 µm, 0.751 µm, and 0.744 µm, to 0.198 μm, 0.201 μm, and 0.187 μm, respectively. Obviously, the finishing effect of the surface-modified viscoelastic magnetic abrasive tools is better than that of the non-modified ones.

TABLE II. VISCOELASTIC MAGNETIC ABRASIVE TOOLS

| parameters / The viscoelastic magnetic abrasive tools | | The mass ratio of Fe：SiC | Mesh number of Fe | Mesh number of SiC | Percentage of the filler particles |
|---|---|---|---|---|---|
| Filler particles modified by coupling agent | The silane coupling agent KH-550 (1%) | 3：1 | 600 | 200 | 50% |
| Filled particles non-modified by coupling agent | None | | | | |

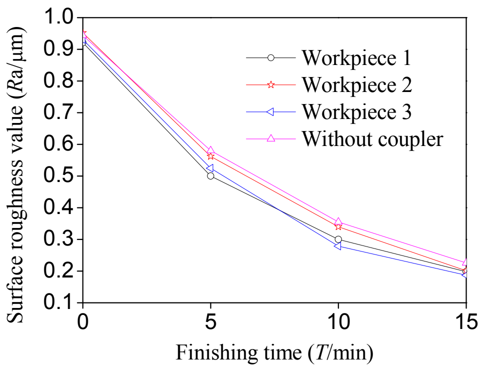



Fig.9 Influence of The Coupling Agent on Finishing Effect

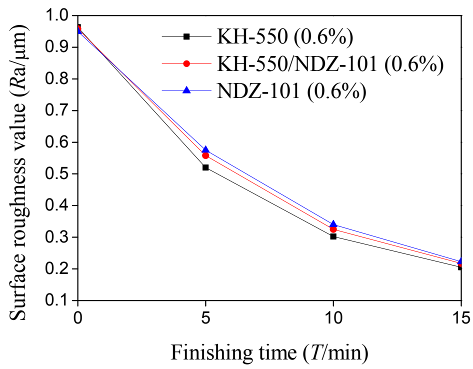
### *C. Influence of The Species and Amount of The Coupling Agent on The Finishing Effect*

After the surface modifications of the Fe and SiC particles using the silane, titanate, and mixed coupling agent (with mass ratio of the silane to the titanate coupling agent of 1:1), we prepared nine kinds of viscoelastical magnetic abrasive tools and performed corresponding finishing tests. The mass percentage of the coupling agent to the modified filler particles is (a) 0.6%, (b) 1.3%, and (c) 2.0%. The parameters are listed in Table III. The surface roughness curves in dependence of time are shown in Fig. (10). As can be seen from Fig. 10, at the amount of 0.6% of the coupling agent, the surface roughness value slightly decreased but was almost the same as that without coupling agent. Apparently, the amount of the coupling agent was too small and the surface modification of the Fe and SiC particles not sufficient to effectively improve their bonding strength with the MVQ matrix. When 2.0% of the coupling agent was used, the improvement of the surface-roughness value slightly decreased in comparison to that for the finishing without the coupling agent as a result of the excessive amount of the coupling agent reducing the viscosity and shear stress of the viscoelastic magnetic abrasive tools. In addition, impurities at the interface formed that further reduced the interface bonding strength and finishing effect. When 1.3% of the coupling agent was used, the surface-roughness value most strongly improved. Using the silane coupling agent KH-550, the surface-roughness value decreased from 0.968 μm to 0.157 μm, thus reduced by 0.811 μm. According to practical experience, the ideal amount of KH-550 is 0.8%–1.5%.

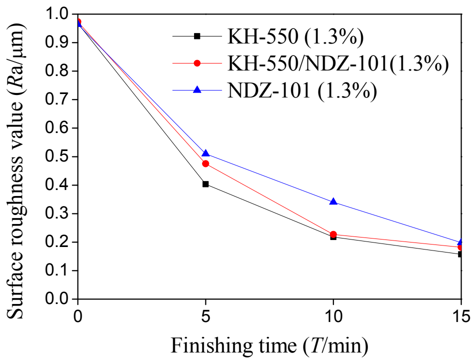
As can be seen from Fig. (10). (a)–(c), the effect of the surface modification of the silane coupling agent KH-550 is the best and in the case of the titanate coupling agent NDZ-101, it is the worst. In addition, the effect of the mixed coupling agent decreases between them. The mechanisms of the two coupling agents on the surface-modified particles are different, i.e., the silane coupling agent can form covalent bonds with Fe or SiC particles, whereas the titanate coupling agent can form organic monolayers on the surfaces of the particles. Therefore, the silane coupling agent can improve the interface bonding strength and finishing effect of the viscoelastic magnetic abrasive tools.

TABLE III. THE PARAMETERS OF VISCOELASTICAL MAGNETIC ABRASIVE TOOLS

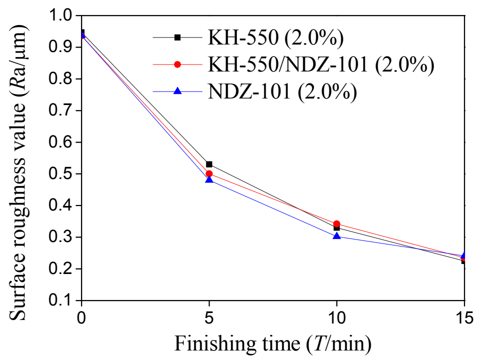
| Parameters | The mass percentage of coupling agent to filler particles | | | The mass ratio of Fe: SiC | Mesh number of Fe | Mesh number of SiC | Percentage of the filler particles |
|---|---|---|---|---|---|---|---|
| | Silane coupling agent KH-550 | Titanate coupling agent NDZ-101 | Mixed Coupling agent KH-550 /NDZ-101 | | | | |
| Abrasive tools 1 | 0.6% | 0.6% | 0.6% | 3：1 | 600 | 200 | 50% |
| Abrasive tools 2 | 1.3% | 1.3% | 1.3% | | | | |
| Abrasive tools 3 | 2.0% | 2.0% | 2.0% | | | | |




(a) the amount of 0.6% coupling agent




(b) the amount of 1.3% coupling agent



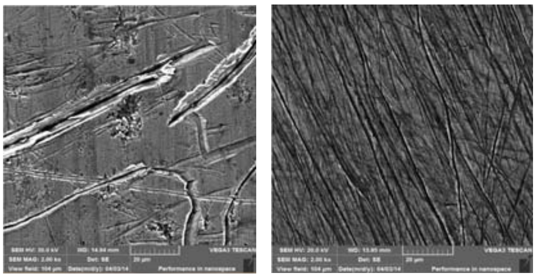

(c) the amount of 2.0% coupling agent

Fig.10 The Influence of The Types and Amounts of Coupling Agent on Finishing Effect

### *D. Finishing Effect*

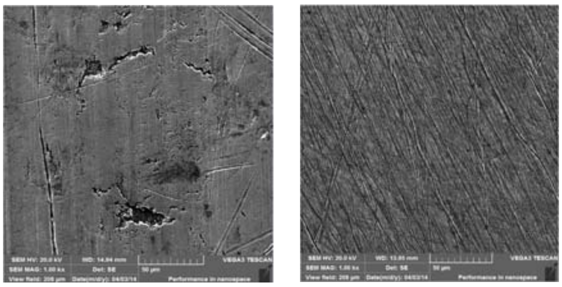
For 1.3% of the coupling agent, the contrast photos of the surface microtopography before and after finishing using viscoelastic magnetic abrasive tools (finishing H62 tube) are shown in Fig. (11). and Fig. (12).

As can be seen from Figs. (11).(a) and (12).(a), there are surface defects, such as inclusions, scratches, and grooves of different depth on the H62 work-piece. However, after finishing for 15 min using the modified viscoelastic magnetic abrasive tools, the surface defects disappeared. The H62 tube had smoother surface, denser structure, and strong corrosion resistance. Its surface quality improved greatly.



(a) Before Finishing (b) After Finishing

Fig.11 The Contrast Photos of The Surface Micro-Topography Before and After Viscoelastic Magnetic Abrasive Tools Finishing H62（2000X）



(a) before finishing (b) after finishing

Fig. (12). The Contrast Photos of The Surface Micro-Topography Before and After Viscoelastic Magnetic Abrasive Tools Finishing H62（1000X）

## VI. CONCLUSION

In this paper, the effect of coupling agent on interfacial bonding properties and finishing performance have been reported. Based on this study, following conclusions can be concluded:

(1) The effects of coupling agent on interfacial bonding properties between the Fe and SiC particles are favorable. It can improve the dispersion and compatibility of the Fe and SiC particles in the matrix, increase the interface bonding strength, and improve the processing capability of viscoelastic magnetic abrasive tools.

(2) The finishing effect of coupling agent KH-550 is the best. The surface-roughness value decreased from 0.968 μm to 0.157 μm, thus reduced by 0.811 μm.

(3) Finishing 15 min using the modified viscoelastic magnetic abrasive tools, the surface defects of the H62 tube disappeared. The surface became smooth, and the surface quality improved greatly.

## CONFLICT OF INTEREST

The authors confirm that this article content has no conflicts of interest.

## ACKNOWLEDGMENT

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