Mixing of graphene nanoplatelets with magnesium alloy powders by electrostatic adsorption

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Abstract

Mg matrix composites reinforced by graphene nanoplatelets (GNPs) offers an efficient approach for improving the mechanical properties of Mg alloys. Unfortunately, the poor uniform dispersion of GNPs into Mg matrix vastly restricts their development. In addition, surface oxidation of Mg alloy powders is always serious. To alleviate these issues, pickling and surface modification technologies of ZK61 Mg alloy powders and mixing process with GNPs have been investigated. The results show that ZK61 alloy powders with smooth surface and low degree of oxidation can be obtained after being simultaneously mechanically stirred and ultrasonically treated for 30 min in a 0.2 vol% HF ethanol solution. They were then rinsed and dried, and modified by 0.3% wt% cetyl trimethyl ammonium bromide to carry a positive charge. Subsequently, GNPs ethanol suspension was poured into the modified ZK61 alloy powders solution and mechanically stirred for 10 min, and then a powder mixture that GNPs randomly attached on the Mg powders was obtained after drying. XPS analysis reveals that GNPs were adsorbed on the surfaces of the modified Mg powders by the mechanism of electrostatic adsorption. The achieved method for preparing GNPs/ZK61 alloy mixture powders provides a new strategy for fabricating Mg matrix composites reinforced by uniformly distributed GNPs.

1. Introduction

Mg alloys are the lightest commonly used metal structural materials with high stiffness/weight ratio, good castability and high damping capacity, and have a strong potential for applications in aerospace, automotive, motorcycle, electronic products and military fields [1–4]. Nevertheless, their actual applications are still very limited, far less than those of another kind of light metallic material, Al alloys, due to their relatively poor mechanical properties [5, 6]. Graphene (Gr), a two-dimensional material composed of carbon atoms with sp² hybrid orbitals, has not only excellent electrical, thermal and optical properties, but also excellent mechanical properties, being considered as an ideal nano-sized reinforcement for metal matrix composites [7, 8]. In contrast, graphene nanoplatelets (GNPs) that are composed of multi-layer Gr is more commonly used due to their more simple preparation method and resulted lower cost [8].

However, uniform dispersion of GNPs into metal matrix is more difficult than the other nano-reinforcements due to their large specific area [7, 9–12]. Similar to the other particles reinforced metal matrix composites, powder metallurgy (PM) is the most acceptable method for preparing GNPs reinforced composites [13]. For this method, high-energy ball-milling is always conducted in order to uniformly mix GNPs with matrix alloy powders, which not only damages the structure integrity of GNPs, but also results in anisotropic mechanical properties of the achieved composites [7, 14–18]. To overcome these shortcomings, two categories of mixing methods have been developed. One is that GNPs encapsulated by a layer of metal are first obtained by electrochemical deposition, and then mixed with alloy powders [19, 20]. The other is that GNPs are first dispersed by ultrasonic treatment in an organic solvent (i.e., ethanol) to achieve a GNP suspension liquid, and
then mixed with alloy powders by mechanical stirring to form a mixed suspension liquid, finally a mixture powder is obtained through filtering and drying [21], or the GNP suspension liquid is filtered and dried to gain dispersed GNP powders, and then ball-milled with alloy powders [22]. In fact, GNP may agglomerate again during filtering and subsequent drying due to their large specific area. In addition, it is essentially very difficult to uniformly mix these two kinds of powders through low-energy ball-milling due to their distinct morphologies: GNP have a 2D structure while alloy powders are always in a spherical 3D structure. It can be expected that if individual GNP are tightly attached on the surfaces of alloy powders through electrostatic attraction in a suspension liquid, and then a uniform mixture can be obtained after being filtered and dried. Based on this standpoint, a new technology named surface-modification was proposed [23]. For this technology, the surfaces of alloy powders and GNP were modified by adding a special reagent in their suspension liquid respectively to make them carry a positive or a negative charger before mixing. In this way, the dispersed GNP can be absorbed on the alloy powders through electrostatic force. But so far, the existing investigations on this aspect are very scarce and there is no special study on the detailed mixing process.

It is known that when cetyl trimethyl ammonium bromide (CTAB) dissolves into aqueous solutions, a kind of positive ions ([C16H33N(CH3)3]+) can be dissociated [24]. If Mg alloy powders are treated using this reagent in an aqueous solution and their surfaces may carry a positive charge. In addition, an electrostatically charged object can absorb small objects, especially with a thin slice shape, without static electricity since one end of the uncharged object near the electrostatically charged object can be induced an opposite electrical property to the electrostatically charged object [25]. Based on this theory, it is expected that GNP may be attached on the Mg alloy powders during mechanical stirring in a H2O-having solution in which a given amount of CTAB is added. So the effects of CTAB addition content and mechanical stirring time on the attaching status of GNP on ZK61 Mg alloy powders were investigated in this work. However, Mg alloys are very active and can easily react with O2/H2O [26], but the needed positive ions are not dissociated if there is no H2O in solution. To decrease the solution degree, an ethanol solution containing 10 vol% H2O was used as the solution for surface modification of Mg alloy powders. In addition, the long-stored raw Mg alloy powders often have a thick oxide layer on their surfaces, and the oxides should be removed prior to surface modification. Hydrofluoric acid (HF), a kind of weak acid that can lead MgO to dissolve into water or ethanol, i.e. being able to remove the oxides on Mg alloy powders, so it is widely used to pre-treat Mg alloy workpiece surface prior to electroplating [27, 28]. The concentration of HF in solution and corresponding treating time not only have large effect on oxide removal, but also on alloy loss resulted from corrosion [29, 30]. So the HF pickling was used to remove the oxide layers on ZK61 Mg alloy powders and the effects of main pickling parameters were studied.

2. Materials and methods

2.1. Raw materials

A kind of commercial ZK61 alloy powders with mean particle size of 34 μm was provided by Tangshan Weihao Magnesium Powder Co. Ltd, China, and their composition was Mg-5.2Zn-0.3Zr (in wt%). GNP with thickness of 2–4 nm and diameter of ~11 μm were purchased from Shenzhen Turing Evolution Technology Co. Ltd. CTAB used in this experiment was provided by Shanghai Zhongqin Chemical Reagent Co., Ltd.

2.2. Pickling of Mg alloy powders

60 g ZK61 alloy powders were first dispersed in 120 ml ethanol by ultrasonic treating at a frequency of 40 kHz and simultaneous mechanical stirring at a speed of 160 rpm for 5 min. Then 80 ml ethanol solution with a given concentration of HF was poured into the ZK61-ethanol solution (the volume fraction of HF in the total solution is 0.5 vol%, 1 vol%, 2 vol% and 3 vol% respectively) and continuously treated by ultrasonic and mechanical stirring for 10–90 min. Subsequently, the treated solutions were settled for 20 min and the liquid was dumped. The settled powders were then rinsed for different times (1–4 times) using 100 ml ethanol with simultaneous ultrasonic treatment and mechanical agitation for 5 min in order to verify the effect of rinsing time on the residual impurity (oxides and corrosion products) amount on Mg powders. Finally, the powders were dried in a vacuum drying oven at 323 K for 12 h.

2.3. Mixing of GNP and ZK61 alloy powders

50 g of ZK61 alloy powders obtained above were added into 100 ml ethanol solution with 10 vol% distilled water with a given amount of CTAB and ultrasonically treated for 20 min and then mechanically stirred for 1 h [31] to modify Mg powder surface. Subsequently, 200 ml 0.2 mg/ml GNP-ethanol suspension solution (that is obtained by ultrasonically treating 40 mg GNP in 200 ml ethanol for 2 h) was poured and mechanically stirred for different time. The ranges of used CTAB content and stirring time were 0.1–0.5 wt% of powders and
5–20 min, respectively. The achieved GNPs/ZK61 alloy powder suspension liquid was settled for 10 min, then the liquid was drained and the powders were dried in the vacuum drying oven at 323 K for 12 h.

2.4. Characterization

The morphologies of the raw GNPs and Mg powders, ultrasonically dispersed GNPs, pickled Mg powders and finally achieved GNPs-ZK61 alloy powders were characterized by a JSM-6700F scanning electron microscopy (SEM, JEOL ltd, Tokyo, Japan). The surfaces of the pickled Mg powders were analyzed by energy dispersive spectrometer (EDS, JEOL ltd, Tokyo, Japan) equipped on the SEM and their size were measured by a Mastersizer 2000 Laser particle size analyzer (Malvern Instruments Ltd, Malvern, UK). An AXIS SUPRA x-ray photoelectron spectrometer (XPS, Shimadzu Corporation, Kyoto, Japan) was adopted to characterize the surface conditions of the modified Mg alloy powders. A MFP-3D atomic force microscope (AFM, Oxford Instruments, Oxford, UK) was utilized to examine the thickness of GNPs after ultrasonic treatment.

3. Results and discussion

3.1. Pickling of ZK61 alloy powders

As shown by figure 1(a), there are a large number of non-spherical small particles between the raw ZK61 alloy powders and a lot of smaller white particles are attached on the surfaces of the powders. EDS spot analysis indicates that both these two kinds of particles are oxides (figure 1(b)), implying that the raw Mg powders have been seriously oxidized. So much oxide impurities should be removed prior to conducting the subsequent processes.

3.1.1. Effect of HF content on pickling

Figure 2 shows the morphologies of ZK61 alloy powders pickled using ethanol solutions with different HF concentrations under simultaneous ultrasonic treatment and mechanical stirring for 30 min. It is seen that all of the small-sized oxide impurity particles between the powders are disappeared after being pickled in 0.5 vol% HF solution, and only a small quantity of impurity agglomerate particles attach on the surfaces of part of powders (figure 2(a)), which are obviously different from those of the raw powders (comparing figures 1(a) and 2(a)). This means that HF is an effective agent for removing oxide impurities. As the HF content increases to 1 vol%, no obvious impurity can be seen on the powder surface (figure 2(b)) and only some small particles are faintly seen under large-magnification condition (see the insert in figure 2(b)). When the HF content further increases to and over 2 vol%, no impurities can be found (figures 2(c) and (d)) even under large-magnification condition (see the inserts in figures 2(c) and (d)). The whole surface is very clean and grain boundaries can be seen (the inserts in figures 2(c) and (d)). That is, only in view of the morphologies of alloy powders as shown in figure 2, 2 vol% HF solution is enough to remove the oxide impurities under the used treating conditions, i.e., simultaneous ultrasonic treating and mechanical stirring for 30 min.

It can be expected that the main constituent of oxide impurities should be MgO, and the following reaction occurs during pickling [32],

\[
\text{MgO} + 2\text{HF} = \text{MgF}_2 + \text{H}_2\text{O}
\]  

(1)

The generated MgF\(_2\) then dissolves into ethanol solutions. The present results indicate that the pickling solutions with less than 2 vol% HF are not enough for removing all of the oxides. Figure 2(a) shows that the...
morphology of the oxides attached on the powder surface is obvious different from that of the raw powders (comparing figure 1(a)). The oxides on the raw powders are in situ formed and uniformly distribute on the surfaces (insert in figure 2(a)), but those on the pickled powders by 0.5 vol% HF solution are in some small-sized particle agglomerates (figure 2(a)). It is suggested that during pickling, the oxide layers were peeled off from the powder surfaces and separated into small-sized particles due to reaction with HF and ultrasonic vibration. These oxide particles did not completely react and some of them were left in the solution due to absence of HF. During the subsequent settling, the residual oxide particles agglomerated and finally attached on the powders. As the HF concentration increases, the residual oxides gradually decrease, and completely disappear when the concentration is up to 2 vol% (figure 2(c)), and the further increase does not change the powder morphology (figure 2(d)). The EDS analysis indicates that oxygen content does not change as the HF concentration increases to 3 vol% (figure 3), which further demonstrates that 2 vol% HF is enough to remove the oxides in the raw powders. But figure 3 shows that the oxygen contents on the powders that are pickled by 2 vol% and 3 vol% HF solutions are not the desired zero, but about 1.5 wt%. This should be attributed to the slight oxidation of the pickled powders during their subsequent any chance to contact atmosphere such as during drying, storing and even EDS analyzing processes. That is, it is very difficult to absolutely avoid oxidation in practice. But it is true that the existing oxide impurities in the raw powders can be completely removed.

Figure 3 also indicates that the powder weight continuously decreases while the average size slightly increases as the HF concentration increases. It is expected that the weight reduction should be mainly attributed to the removal of oxides, including the individual oxide particles and oxide film on the powders. In addition, the corrosion of powders themselves also leads their weight decrease, especially the small-sized powders maybe completely dissolve into the solutions, which can be confirmed by comparing figures 2(a)–(d), the number of small powders gradually decreases as the HF concentration increases and there are almost no small powders after being pickled by 3 vol% HF solution. It is also just this reason that the average size of the pickled powders slightly increases as the concentration increases. So it is suggested that the HF content should be as low as possible under the condition of ensuring removal of existing oxides. The present results indicate that 2 vol% is a suitable concentration of HF.
3.1.2 Effect of ultrasonic treatment and mechanical stirring time on pickling

The results from the above section propose that 2 vol% HF solution is effective for removing the oxides in the original powders. So in this section, the effect of pickling time, i.e., ultrasonic treatment and mechanical stirring time on the pickling result was examined in 2 vol% HF solution in order to confirm the optimum pickling time.

As shown by figure 4, the change tendency of the powder morphologies with the pickling time is similar to that with the HF concentration discussed above. It can be seen that the attached oxides on powder surface gradually decrease as the pickling time is prolonged (figures 4(a) and (b)), and completely disappear when time is increased to 30 min (figure 3(c)), and further extension of pickling has no effect on the morphologies (figures 4(c) and (d)). Figure 5 shows that the oxygen content on powder surfaces also decreases as the time increases and does not change after 30 min These results imply that pickling for 30 min in 2 vol% HF solution can remove all of the oxides in the powders. Similarly, the further pickling only brings about the dissipation of powders due to corrosion, resulting in the decrease of powder weight (figure 5). So the pickling time should be as short as possible under the condition that all oxides are completely removed, and the present results indicate that pickling for 30 min in 2 vol% HF solution is appropriate. Figure 5 shows that the average powder size almost
does not vary with the pickling time. This should be attributed to the competition result between size decreases from partial corrosion of large-sized powders and size increases from dissipation of small-sized powders.

3.1.3. Effect of rinsing time on residual oxides
It must be noted that the results mentioned in the above sections are from examinations of the pickled powders after being rinsed for four times in ethanol under simultaneous ultrasonic treatment and mechanical stirring for 5 min. As shown by figure 6(a), there are still lots of oxide particles attach on the pickled powder surfaces after being settled and dried. This means that the pickling technique achieved from the above investigations, i.e., pickling for 30 min in 2 vol% HF ethanol solution under simultaneous ultrasonic treatment and mechanical stirring, cannot dissolve all of the oxides into solutions. To make all oxides to dissolve, the employed HF concentration or/and pickling time should be increased, but this must consume more powders due to enhanced corrosion or/and longer time corrosion. In addition, some HF must be left on powder surfaces in forms of $\text{H}^+$ and $\text{F}^-$ ions after drying, and their existence is harmful to the properties of the final materials. So the pickled powders should be rinsed to remove the residual oxides and HF. As shown by figures 6 and 2(c), the residual

Figure 5. Variations of weight, size and surface oxygen content of ZK61 alloy powders with pickling time.

Figure 6. SEM images of pickled ZK61 alloy powders before being rinsed (a) and after being rinsed for (b) one, (c) two and (d) three times in ethanol under simultaneous ultrasonic treatment and mechanical stirring for 5 min.
oxides gradually decrease with increasing rinsing time in ethanol, and completely disappear after being rinsed for four times. Together with the results of the above sections, it can be concluded that the appropriate pickling technique is that the raw powders are treated in 2 vol% HF ethanol solution by simultaneous ultrasonic treatment and mechanical stirring for 30 min, followed in turn by settling for 20 min, pouring out the liquid, rinsing four times in ethanol under ultrasonic vibrating and mechanical stirring for 5 min and drying in a vacuum furnace at 323 K for 12 h.

3.2. Mixing of ZK61 alloy powders with GNPs

The as-received GNPs always agglomerate in irregular particles as shown by figure 7(a) due to their large specific surface area. The inset in figure 7(a) indicates that the particle consists of lots of large-sized and closely-stacked GNPs. So they must be dispersed before they are incorporated with metal matrix, and the commonly used method is ultrasonic treatment in ethanol [33, 34]. According to a previous investigation from the authors [35], the raw GNPs were ultrasonically treated for 2 hours in ethanol and a suspension liquid with dispersed GNPs was then obtained. The dried GNPs seemly interconnect together in view of their general morphology (figure 7(b)), but in fact they are piled up in disordered small-sized platelets (see the inset in figure 7(b)). The result from AFM examination shows that the typical thickness of the resulting GNPs is about 3.4 nm, which is just equivalent to the size of 2–4 nm labeled by the merchant. All of these mean that the agglomerated GNPs are dispersed using the employed disperse process.

3.2.1. Effect of CTAB addition amount on mixing uniformity

As described in introduction, surface modification method is used to mix GNPs with Mg alloy powders and CTAB is acted as the surface modification agent of Mg powders. So the effect of addition amount of CTAB on mixing uniformity was first investigated in this section. Figure 8(a) visually shows the status of the mixture solution of GNPs and ZK61 alloy powders without CTAB via mechanically stirring for 10 min and subsequently settling for 10 min. It can be seen that Mg alloy powders all sink to the bottom of the beaker, forming a grey layer at the bottom (marked by A in figure 8(a)), and the upper liquid is in a dark color, which means that GNPs are still suspended in the liquid, and do not adsorb on the alloy powder surfaces. When 0.1 wt% CTAB is added, the color of the upper liquid obviously becomes light while the color of the alloy powders sunk to the bottom.

![Figure 7. SEM images of GNPs for (a) raw GNPs, (b) ultrasonically dispersed for 2 h. (c) AFM image of GNPs ultrasonically dispersed for 2 h. (d) Variation in thickness corresponding to the line in (c).](image-url)
becomes darker (marked by B in 8b). The former phenomenon means that the suspended GNPs in the liquid are obviously decreased, and the latter indicates that there are GNPs in the settled alloy powders, i.e., some GNPs are absorbed on the alloy powder surfaces. When the CTAB content is increased to 0.3% wt%, the color of the liquid becomes lighter and is in a transparent form (figure 8(c)), implying that the amount of GNPs suspended in the liquid is very small and the added GNPs are basically all attached on the alloy powders under this condition. As the CTAB content is further increased, there is no change compared with that treated by 0.5 wt% CTAB (figure 8(d)). So it can be concluded that 0.3 wt% is the smallest content of CTAB that the alloy powders can absorb the largest amount of GNPs. It is found that there is always a thin dark layer floated on the liquid surface (figures 8(b)–(d)). According to the authors’ previous investigation, there are some graphite particles in the raw GNPs, and they are not all turned into GNPs although exfoliation occurs during dispersing [36]. So the floated dark layer should be graphite particles and they will eventually be poured out together with the liquid.

3.2.2. Stirring time
The above section indicates that 0.3 wt% CTAB is adequate for Mg alloy powders to absorb GNPs. So the stirring time, another main parameter, on the mixing result is studied under maintaining the 0.3 wt% CTAB concentration and settling for 10 min. Figure 9(a) shows the status after being stirring for 5 min, which indicates that the upper liquid is in light gray and its color is deeper than that being stirred for 10 min (figure 8(c)). This difference should be attributed to that the employed time of 5 min is not enough for all GNPs to be adsorbed on the ZK61 alloy powder. But the liquid changes from light gray to dark gray again when the time is extended to 20 min, illustrating that the long-time mechanical force causes the GNPs adsorbed on the surface of ZK61 alloy powders to fall off again. That is, too short or too long stirring time is not conducive to the adsorption of GNPs on the alloy powders and 10 min is the suitable stirring time. So it can be summarized that GNPs can attach on the alloy powders after the pickled ZK61 alloy powders being modified in a 10 vol% H2O ethanol solution with addition of 0.3 wt% CTAB and then mechanically stirred with ultrasonically treated GNPs suspension liquid for 10 min.

3.3. Verification of mixing result and corresponding mechanism
In order to verify whether GNPs were actually attached on the ZK61 alloy powders, the suspension liquid achieved by adding 0.3 wt% CTAB and mechanically stirring for 10 min was settled for 10 min, the upper liquid was poured out, and the settled powders were dried in a vacuum furnace at 323 K for 12 h and then observed by SEM as shown by figures 10(a) and (b), as well as the powders achieved without addition of CTAB for
comparison (figures 10(c) and (d)). It can be found that some flocules are attached on the margin of each alloy powder (marked by A arrows in figure 10(a)), but almost nothing can be found on the powders treated without CTAB except rare particles (marked by box in figure 10(c)). As is known, GNPs are a kind of transparent or semi-transparent 2D material [37]. It is expected that only the GNPs in a warped or wrinkled form can be possibly seen under deep-color or dark background, and thus, the visible flocules on the margins of alloy powders should be GNPs. EDS analysis at point B in figure 10(a) indicates that these flocules are actually GNPs (figure 11(a)). In addition, as shown by figure 10(b), transparent GNPs can be dimly seen on the surface of alloy powder at the region marked by arrow C in figure 10(b), and EDS analysis at point D in figure 10(b) proves that GNPs indeed exist at this region (figure 11(b)). That is to say, GNPs may exist at the regions where no any foreign matters can be seen. EDS analysis at point E in figure 10(d) demonstrates that the particles are also GNPs (figure 11(d)), which may be accidentally attached to the powder during settling. Based on these results, it can be proposed that GNPs are randomly attached on the surfaces of alloy powders treated with 0.3 wt% CTAB. As a contrast, the results from EDS in any region (i.e., region F in figure 10(c)) on the surface of alloy powders treated without CTAB indicate that GNPs are rarely detected (figure 11(c)).

In addition, Mg alloys are usually easy to react with H₂O to form Mg(OH)$_2$ and H₂ [19], and the ethanol solution used in this section has 10 vol% H₂O. To verify the reaction degree, the regions at which no GNPs (marked by point F figure 10(c)) attached were examined by EDS. The results indicate that the oxygen content is relatively low (figure 11(c)), which is comparable to that of the powders that have been picked (figures 3 and 5).
This implies that the reaction of the alloy powders with H₂O is very limited. So CTAB is a proper surface modification reagent for mixing ZK61 alloy powders and GNPs in view of the mixing result and the oxidation degree of the alloy powders.

To confirm the expected electrostatic adsorption mechanism between GNPs and ZK61 alloy powders, the ZK61 alloy powders modified with 0.3 wt% CTAB were analyzed by XPS. From the resulting survey spectrum, Mg KLL peak, Zn 3p peak, Zr 3p peak, O 1 s peak and C 1 s peak can be detected (figure 12(a)). The former three peaks should be resulted from the Mg alloy powders themselves, the O 1 s peak is attributed to the slight oxidation after pickling as discussed above, and the C 1 s peak is originated from the CTAB. The high-resolution C 1 s spectra show that –CH₂CH₂– bonds exist at binding energies of 284.55 eV and 285.78 eV (figure 12(b)). CTAB, which contains a large amount of alkyl groups (–CH₂–), can be ionized in the H₂O-having ethanol solution [24], and thus leading the alloy powders to carry positive charge. In addition, GNPs will be induced to carry negative charge due to electrostatic induction when they are closed to the alloy powders with positive charge.
charge during mechanical stirring [25], and then are absorbed on the powder surfaces since they are light and have a slice shape. So the XPS result indirectly confirms that the attachment of GNPs on alloy powders is actually originated from electrostatic adsorption.

4. Conclusions

(1) The pickling in HF ethanol solution under simultaneous ultrasonic treatment and mechanical stirring is an effective method for removing the oxides in ZK61 alloy powders. Properly prolonging the pickling time (i.e., ultrasonic and mechanical stirring time) and increasing HF content are beneficial for reducing the oxidation degree of powder surface, but it will cause a certain amount of powder loss. Under the employed conditions in this work, the appropriate HF concentration and pickling time are 0.2 vol% and 30 min, respectively.

(2) CTAB is a suitable surface modification reagent for ZK61 alloy powders and the mixture powders that GNPs are randomly attached on the surfaces of ZK61 alloy powders were successfully prepared via mechanical blending the 10 vol% H2O ethanol solution of ZK61 alloy powders modified by 0.3 wt% CTAB and GNPs ethanol suspension achieved by ultrasonic treatment for 10 min. Too short or too long stirring time is not conducive to uniformly mix GNPs and ZK61 alloy powders.

(3) XPS analysis reveals that the attachment mechanism of GNPs on powder surface during the mixing process should be electrostatic absorption, in which ZK61 alloy powders may be positively charged after being modified with CTAB and GNPs may be negatively charged due to electrostatic induction when they are close to the alloy powders during stirring.

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