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EFFECT OF BALL-MILLING ON MECHANICAL PROPERTIES OF Mg – 3 % Al ALLOYS

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Abstract. The mechanical properties of any structural material are among the most influential factors in determining its potential application. The aim of this study is to look at how ball-milling affects the mechanical properties of Mg – 3 % Al alloy. Powder metallurgy approach integrating room temperature ball milling at different duration, cold powder compaction, sintering at inert atmosphere, and hot extrusion techniques were used to fabricate high quality ultra-fine grained and nanocrystalline Mg – 3 % Al alloy samples. X-ray diffraction (XRD) analysis revealed a quick reduction in grain size followed by saturation of the grain size at around 36 nm in 30 hrs of milling. In order to investigate the effect of grain refinement on the stress-strain response of the alloy, the extruded samples were then subjected to large compressive strains under uniaxial compressive loading. Micro-indentation studies were conducted with three separate peak loads of 3 N, 5 N, and 7 N to investigate the micromechanical behaviour of the alloy produced from different milling durations. The loading-unloading curve of the micro-indentation test was found to be strongly influenced by milling duration of the elemental powder. The grain refining affect was clearly seen in measurements of microhardness and indentation modulus.

Keywords: Mg – Al alloy, powder metallurgy, ball-milling, mechanical properties

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ВЛИЯНИЕ ШАРОВОГО ИЗМЕЛЬЧЕНИЯ НА МЕХАНИЧЕСКИЕ СВОЙСТВА СПЛАВОВ Mg – 3 % Al

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Аннотация. Механические свойства любого конструкционного материала являются одними из наиболее важных показателей, определяющих возможности его применения. Целью настоящей работы является изучение влияния шарового измельчения на механические свойства сплава Mg – 3 % Al. Для изготовления высококачественных ультратонких образцов из нанокристаллического сплава Mg – 3 % Al использовался метод порошковой металлургии, включающий шаровой помол при комнатной температуре различной продолжительности, холодное уплотнение порошка, спекание в инертной атмосфере, горячую экструзию. Рентгеноструктурный анализ (XRD) показал быстрое уменьшение размера зерен с последующим измельчением примерно до 36 нм за 30 ч. Экструдированные образцы были подвергнуты сильной деформации при одноосной сжимающей нагрузке с целью исследования влияния измельчения зерен на профиль напряжения и деформацию сплава. Для изучения микромеханического поведения сплава, полученного при различной длительности измельчения, были проведены исследования микродавления с тремя отдельными пиковыми нагрузками в 3, 5 и 7 Н. Было обнаружено, что на кривую загрузки – разгрузки сильно влияет продолжительность измельчения порошка. Влияние измельчения зерна было четко выражено при измерении микротвердости и значений вдавливания.

Ключевые слова: сплав Mg – Al, порошковая металлургия, измельние на шаровой мельнице, механические свойства

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Introduction

Magnesium-based alloys and their composites are gaining increased attention for a variety of applications, including those in the automotive, aviation, and defence industries [1 – 2]. One of the prime concerns for the automobile and aviation sectors is the reduction of fuel consumption and low tail-pipe emissions while improving safety. Thus, recent developments in lightweight and high-specific-strength materials, Mg – alloys and their composites are among the most viable options for addressing these issues due to their low density and enduring mechanical properties [3 – 4]. However, the HCP structure of magnesium limits it plasticity and corrosion resistance, which somewhat restricts its use at large scale in structural application. Despite the many obstacles encountered, there have been several studies on magnesium and its alloys and composite over the past few decades. Most of the studies include designing of alloy and composite and their optimization, microstructural refinement and the characterisation of the developed materials according to requirements. These studies were helpful in promoting the applications of magnesium and its alloys and composites. The open literature search suggests that the addition of alloying elements to magnesium has the potential to significantly enhance the basic properties of magnesium, and thus the trend in research is to develop magnesium alloys [5 – 6]. As an example, small weight percentage of Al, which primarily serves as a solid solution strengthening element, can be added to Mg to enhance its strength and ductility [7]. The addition of Al content is generally thought to be small (< 5 wt. %) in order to avoid the formation of second phase intermetallic particle $Mg_{17}Al_{12}$ that can reduce ductility. Grain refinement is another strategy that has been a focus of research over the last two decades, as it is widely regarded as one of the most successful technique for enhancing strength, ductility, and formability at the same time [8 – 9].

The main focus of this study is to design and produce Mg – 3 % Al alloys using powder metallurgy technique. As the distribution of the grain structure in the powder metallurgy process largely depends on the milling time, the powders of the alloying elements were milled for different duration to produce bulk extruded samples. The impact of ball milling on the macro and micro mechanical properties (TS, YS, and ductility) of the alloys was thoroughly investigated. The compressive behaviours of the alloys prepared with varying milling times were discussed. The study of micro-indentation tests under various loading conditions were reported.

1. Experimental procedures

Pure commercial Mg powder (particle size 60 – 300 μm, purity 98.5 %) and pure commercial Al powder (particle size 5 – 15 μm, purity 99.5 %) were obtained from Merck, Germany and Alfa Aesar, USA, respectively. Magnesium was selected as a base material because of its low structural density and high specific strength, while Al was chosen as an alloying element because of its capacity to improve Mg's strength and ductility. The powders were mixed at a weight ratio of 97:3 Mg to Al and then degassed and dried for 2 hrs at 200 °C under vacuum, and the subsequent manipulation was carried out in an argon filled stainless steel glove box. The oxygen and moisture levels were reduced to less than 1 ppm inside the glove box. The degassed powders were placed in a 500 ml hardened steel vial with 51 hardened steel balls (12.5 mm in diameter) and sealed within the glove box for conventional milling. To avoid excessive cold welding, small amounts of stearic acid were added to the powder. The ball milling of the powder mixture was carried out in an inert atmosphere for 0, 10, 20, and 30 hours using a Retsch planetary mill (Model: PM 400). The milling machine was set to 250 r.p.m. at room temperature. In order to avoid excessive temperature rise, 30 minutes of milling were alternated with 30 minutes of rest for cooling using the system's integrated fans. After milling for a stipulated period, the powders were opened inside the glove box, and then a hollow cylindrical compaction die was filled with the milled powder. The mechanically alloyed powders were cold-pressed with a 50-ton load through a solid cylindrical rod, and then they were sintered at 450 °C for 2 hours in an inert atmosphere. The cold compacted sintered billet (38 mm dia and 38 mm height) was then extruded at 350 °C with a 25:1 extrusion ratio. The billets were shocked at 350 °C for 30 minutes before extrusion to ensure uniform temperature. Fig. 1 is presented the flow chart of the fabrication process of the Mg – 3 % Al alloys prepared from the different milling durations.

X-ray diffraction linewidth analysis was used to determine the average grain size, constituent phases, and textural changes of milled powders and extruded samples. A Shimadzu Lab XRD-6000 X-ray diffractometer using Cu K_α (λ = 0.154 mm) radiation and operating in the $\theta - \theta$ geometry was used. The Hall-Williamson method was employed to estimate the grain size of the milled powder and their bulk extruded samples. The average grain size of the bulk extruded un-milled samples was estimated using a high-resolution optical microscope.

In order to investigate the effect of grain refinement via ball milling on the macro-mechanical properties of extruded Mg – 3 % Al alloy, quasi-static uniaxial compression tests were conducted on specimens prepared from as-received, 0, 10, 20 and 30 h milled powders. The test were performed at strain rate of $4 \cdot 10^{-4}$ s^{-1} on a fully automated Hounsfield mechanical testing machine (H50KS) as per ASTM E9-09 standard. Extruded cylindrical samples with a diameter of 6 mm and a length of 6 mm (aspect ratio (l/d) of unity) in accordance with the ASTM standard were subjected to a compressive load parallel to the extrusion direction. The fracture surface of compressive specimens was examined using a field emission scanning electron microscope (FESEM).

Microindentation tests were carried out using a load-controlled, completely automated MTR3/50-50/NI instrument (supplied by MICROTEST S.A., Spain) equipped with a Vickers indenter to investigate the micromechanical behaviour of the Mg – 3 % Al alloy fabricated from various milling hours. Load-displacement curves were recorded with three distinct peak loads of 3 N, 5 N, and 7 N, with a loading speed of 1 N per minute. Prior to the indentation test, the samples were metallographically polished to provide a flat surface. All indentation tests were performed at a constant temperature of 24 °C.

2. Result and discussion

The experimental results of the bulk Mg – 3 % Al alloys obtained from different milling hours are discussed in this section. The results of grain size measurements on dispersed milled powders and bulk extruded samples prepared from as-received, 0, 10, 20, and 30 h milled powders are presented in Table 1. It is clear evidence that the average grain size is reduced with the milling duration, whereas the average grain growth is around two to three fold in their bulk extruded samples. The observed grain growth was inevitable because of the application of high temperatures during sintering and hot extrussion that was required for high densities, the removal of adsorbates, and improved inter-particle bonding. The optically measured average grain size of the un-milled extruded sample was about 12 μm. The details pertaining to XRD analysis were reported in our previously published articles [9 – 10].

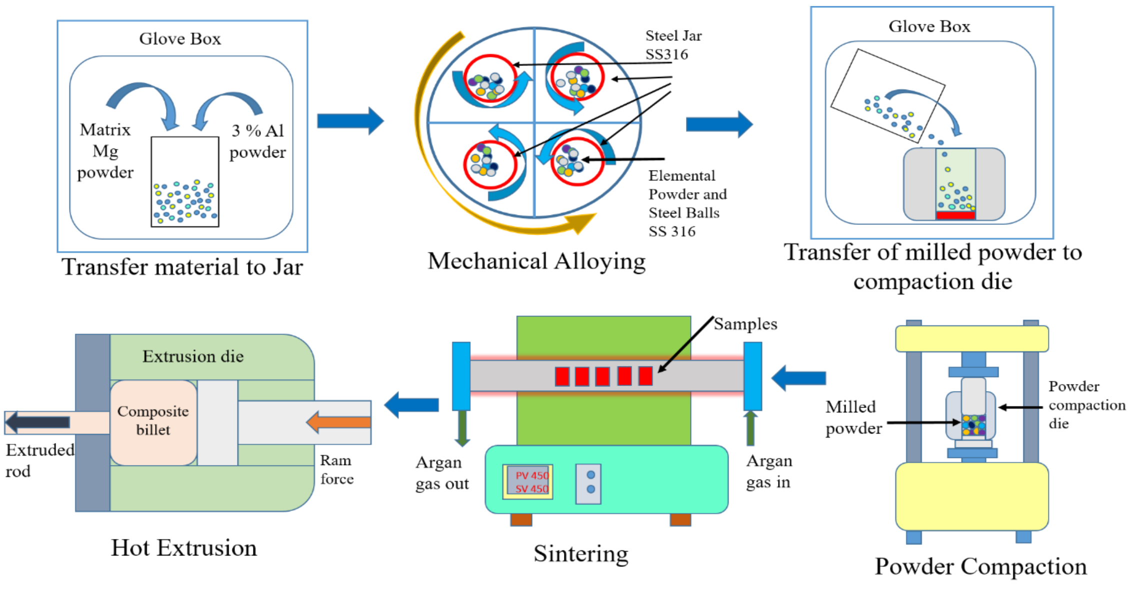


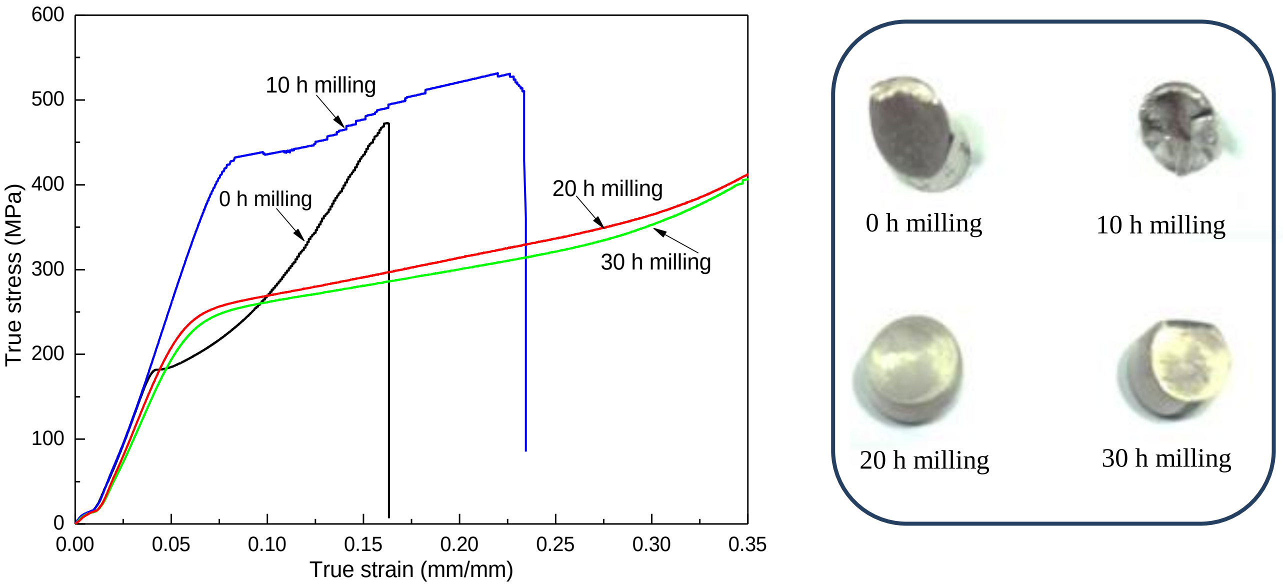
Fig. 1. Synthesis process of bulk Mg – 3 % Al alloys following powder metallurgy technique

Table 1

The average grain size of Mg – 3 % Al milled powders and their bulk extruded samples

Mg – 3 % Al alloy	Milling duration (in hrs.)			
	0	10	20	30
Powder	–	94 nm	40 nm	30 nm
Extruded samples	12 μm	148 nm	120 nm	91 nm

Fig. 2, *a* illustrates the true compressive stress-strain curves for the bulk Mg – 3 % Al alloys samples produced via the powder metallurgy and the corresponding deformed compressive specimens are presented in Fig. 2, *b*. The curves represent the compressive behaviour of samples prepared from as-received, 0, 10, 20 and 30 h milled powders, having 12 μm, 148 nm, 120 nm and 91 nm average grain sizes, respectively. The stress-strain curves of all four types of samples demonstrate that the milling duration has a significant effect on compressive deformation behaviours. The specimens produced from 20 h and 30 h milled powders withstood large strains without showing any indications of failure. The macroscopic appearance of the deformed samples shows that the samples were coined as a result of a softening or mushing effect. In contrast, the bulk extruded samples made from 0 h and 10 h milled powders showed relatively low strains to failure of about 16 and 23 %, respectively. The macroscopic appearance of their deformed samples reveals that failure occurred at 45 degrees to the compression loading axis. The 0 and 10 h milled samples exhibited work hardening behaviour. Conversely, the samples with 20 and 30 h milled sample showed elastic followed by large plastic behaviour. When the average grain size of the alloy samples reaches the nanoscale range due to longer milling durations (20 and 30 h), the work hardening and work hardening rate decline and almost transform into perfectly plastic-like behaviour. The reduction of work hardening rates in the nanocrystalline materials may be due to the incompetence to develop dislocation pileup and cell structures in the same manner as coarse-grained metals. A significant improvement in strength can be noticed in the samples made from 10 h milled powder when compared with the samples made from 0, 20 and 30 h milled powders. The substantial improvement in strength in the sample milled for 10 hours is a result of grain refinement in accordance with the Hall-Pitch relationship. While further milling reduces strength, this might be due to the activation of grain boundary sliding and diffusion that reduces work hardening. Mallick et al. [9 – 10] reported similar results obtained from the tensile behaviour of Mg – 3 % Al alloy. The alloys were prepared with the same procedure and milling duration.



a *b*

Fig. 2. Compression test on Mg – 3 % Al alloys (*a*) True stress-strain behaviour of the samples and (*b*) image of the deformed samples after test

Fig. 3, *a – d* presents the representative loading-unloading curves obtained from the microindentation test. The effect of indentation force on the indentation features of each batch of the samples was predicted by using three different maximum indentation forces (3, 5 and 7 N).

The indentation load (N) vs. penetration depth (μm) curves in all cases had a smooth appearance without signs of fracture or cracking. The response to induced elastic strain deformation is primarily represented by the loading curve, whereas the unloading curve represents the response to elastic recovery. As can be observed from Fig. 3, *a – d* the profile of the loading-unloading curves for various peak loads are in good agreement for every individual batch of Mg – 3 % Al alloy samples. As expected, the indentation depth increases with the increase of indentation load for each case. Fig. 4 depicts load-indentation depth curves for the samples prepared from 0, 10, 20 and 30 h milled powders. All the samples were subjected to maximum 5 N indentation load. The result revealed that the 10 h milled sample had a lower penetration depth than the un-milled sample. In addition, the nearly vertical unloading curve suggests a small recoverable (elastic) deformation in the 10 h milled sample. The higher hardness value in the alloy obtained from 10 h of milling was corroborated by the lower penetration depth and small elastic recovery. Smaller grains with a large grain boundary network accumulate strain energy and dislocation density, which may be responsible to enhance the hardness in 10 h milling sample [11]. In contrast, the samples prepared from 20 and 30 h milled powders exhibited higher indentation depths and consequently lower hardness values. The samples with longer milling times (20 and 30 h) may have had weaker solid solution strengthening, resulting in lower hardness values. The solution of Al in Mg grains may have decreased due to increase in the volume percentage of $MgAl_2O_4$ oxide particles during extended milling times. Based on TEM results, Li et al. [8] reported that the 30 h milled sample did not exhibit any dislocation activity after deformation. Table 2 lists the indentation depth, Vickers microhardness, and indentation modulus corresponding to various indentation loads for each category of alloy samples. The Oliver-Pharr method [12] was used to calculate the microhardness and indentation modulus at room temperature by using the indentation depth and the unloading portion of the indentation curve. In each batch of the synthesised alloy, the hardness diminishes steadily as the indentation depth increases, indicating indentation size effect (ISE) with decreasing hardness as the load is increased. This can be explained by the fact that the surrounding matrix of the indenter acts as a supple substrate at higher indentation depths, resulting a reduction in the strain hardening. In most cases, a similar trend was observed for indentation modulus. The indentation test results are consistent with the compressive parameters of the milled and un-milled samples.

The following main findings can be reached from the material development, mechanical property evaluation, and comparison:

Bulk Mg – 3 % Al alloy samples were successfully fabricated using a series of controlled processes, including room temperature ball milling at different time duration, cold compaction, conventional sintering under inert environment, and hot extussion.

The relationship between flow stress and average grain size of the bulk extruded Mg – 3 % Al alloy samples was demonstrated by the results of uniaxial compression tests at a strain rate of $4 \cdot 10^{-4}$ s^{-1} on mechanically milled bulk samples prepared from as-received, 0, 10, 20 and 30 h milled powders.

3. Conclusions

Milling time strongly affects yield strength, compressive strength, and strain to failure; as a result of grain refinement, these values increased nearly twofold, 12 and 44 %, respectively, in the 10 h milled sample when compared to the 0 h milled sample. However, when the milling period was raised to 20 and 30 h, the yield strength and compressive strength declined.

As milling time was increased, the work hardening and work hardening rate reduced, and the material tended towards perfectly plastic behaviour as grain size approached the nanoscale range.

Higher hardness values and lower indentation depth are associated with the sample made from 10 h milled powder when compared with the sample made from 0 h milled powder. In contrast, the samples made from 20 and 30 h showed high indentation depths and consequently lower hardness values when compared with 10 h milled sample.

The optimization of the milling parameters are crucial to develop Mg – 3 % Al alloy.

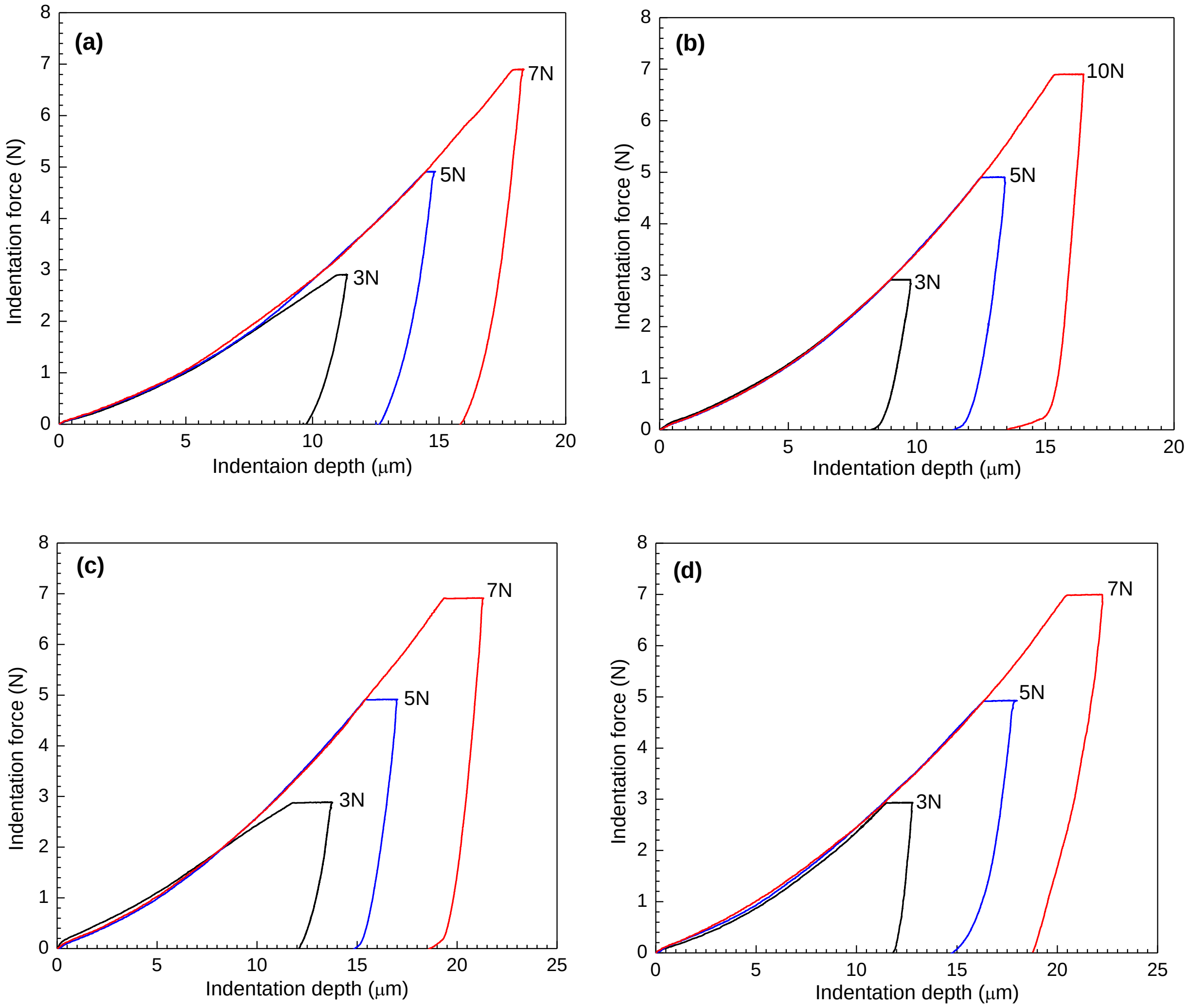


Fig. 3. Loading-unloading curve of the micro-indentation test for (*a*) 0 h milled sample, (*b*) 10 h milled sample, (*c*) 20 h milled sample and (*d*) 30 h milled sample

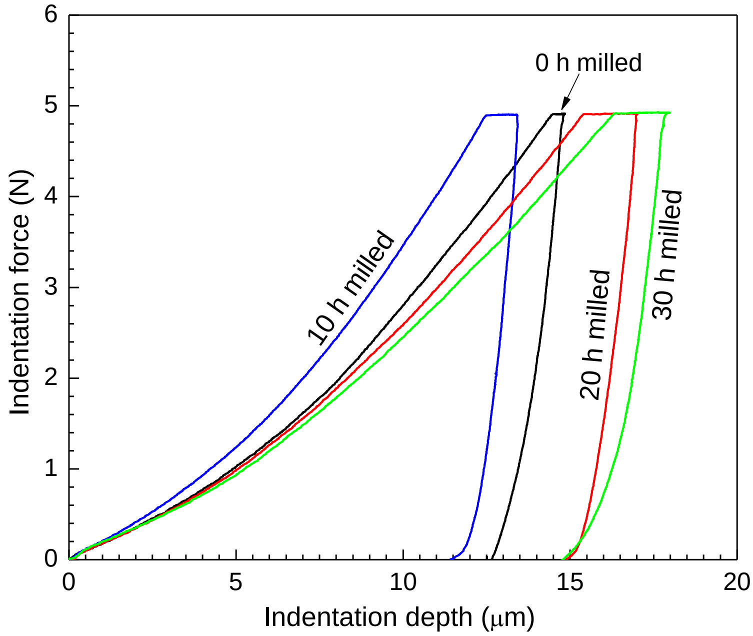


Fig. 4. Loading-unloading curves during micro-indentation under 5 N load for the Mg – 3 % Al alloys with varying milling duration

Table 2

Load dependent indentation

Milling time, hrs.	Load = 3 N			Load = 5 N			Load = 7 N		
	$h_{pen.}$, μm	Hv, kg/mm^2	$E_{ind.}$, GPa	$h_{pen.}$, μm	Hv, kg/mm^2	$E_{ind.}$, GPa	$h_{pen.}$, μm	Hv, kg/mm^2	$E_{ind.}$, GPa
0	10.8	111	62.3	14.4	102	57.1	18.3	89	53.5
10	9.4	156	66.7	12.7	140	82.2	16.5	92	59.2
20	10.8	110	67.7	17.0	71	68.6	20.1	63	67.9
30	13.2	78	67.6	17.4	72	58.4	22.8	61	35.0
* h_{pen} – penetration depth; H_v – micro-hardness; E_{ind} – indentation modulus									

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