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(54) **Magnesium alloys**

(57) A casting heat-resistance Mg alloy improved in heat resistance without relying upon expensive RE contains Cu. More specifically, it contains Al (8.0 weight %), Cu (1.0-5.0 weight %), Zn (2.0 weight %), Be (0.01 weight %) and Mg (the rest). The alloy can be prevented from

deteriorating in corrosion resistance by adjusting the added amount of Cu to 1.0-1.5 weight %. The corrosion resistance of the alloy can be improved more by adding 0.5 to 1.0 weight % Mn as well.

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DescriptionBACKGROUND OF THE INVENTION

5 Field of the Invention

[0001] The present invention relates to magnesium alloys, and more particularly to magnesium alloys for use in casting.

Background Art

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[0002] Magnesium (Mg) has the specific gravity of 1.74, which is approximately two thirds of the specific gravity of aluminum (Al) and approximate one fourth of the specific gravity of iron (Fe). Magnesium is most lightweight among practical metals, and has various favorable properties such as excellent specific tensile strength, specific rigidity, shielding of electromagnetic waves, heat conduction, size invariance and cutting facility, as well as availability for recycle. Because of these properties, magnesium is used in bodies of cell phones, note-type personal computers and mobile electronic devices that require weight saving. Also for weight saving of vehicles, which is the most effective way for improving the fuel consumption rate, the car industry is promoting development and practical use of automotive parts made of magnesium alloys (hereafter called Mg alloys as well).

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[0003] Mg-Al-Zn based AZ91D alloys and Mg-Al based AM60B alloys are known as typical magnesium alloys for use in casting. Names of the magnesium alloys are determined in accordance with the ASTM standards. The JIS standards (Japanese Industrial Standards) also follow them. The notation system according to these standards assigns specific alphabetical letters to respective elements contained in alloys. More specifically, the notation system uses A for aluminum (Al), Z for zinc (Zn), M for manganese (Mn), K for zirconium (Zr), E for rare earth elements, S for silicon (Si), Q for silver (Ag), L for lithium (Li), C for copper (Cu), W for yttrium (Y), H for terbium (Tb), for example. Numerals following the atomic symbols represent quantities of the components, and alphabetical letters suffixed at the ends indicate the sequential order of establishment of the alloys.

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[0004] The AZ91D alloy is not only excellent in balance between its castability, such as flow, hot cracking resistance and shrinkage property, and its mechanical properties, but it is also economical. Therefore, applications of the AZ91D alloy widely range from vehicle parts to electronic devices. On the other hand, the AM60B alloy is used mostly in vehicle parts because of its better ductility than the AZ91 D alloy.

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[0005] Exemplary applications of the AZ91D alloy are key lock housings, sunroofs, retractable roofs, glove boxes, ashtrays, and so on. Exemplary applications of the AM60B alloy are beams of instrument panels, pedal brackets, sheet frames, airbag housings, steering columns, pedal brackets, ABS mount brackets, road wheels, and so on.

[0006] For weight saving of vehicles, the use of magnesium alloys in transmissions, engine-peripherals and other parts, which are subject to severe environments, is under research. However, existing magnesium alloys including AZ91D and AM60B involve the issue of creep strain or deformation under the temperature of 100°C or more, and the creep deformation degrades the dimensional accuracy of the bolt-tightening portions and degrades the tightening strength.

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[0007] To cope with this problem, development of heat-resistant magnesium alloys usable under severe thermal environments has been started. Some alloys of the Mg-Al-Si and Mg-RE systems are already known as heat-resistant Mg alloys. In addition, engine blocks and transmission cases as actual products of Mg-Al-Si based alloys and Mg-RE based alloys have been made commercially available.

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[0008] However, Mg-Al-Si based alloys such as AS21 and AS41 are liable to seize up to the mold in the course of casting, and their heat resistance is still insufficient for use in engine parts. On the other hand, Mg-RE based alloys, WE54, ZE41 and QE22 are good in heat resistance, but they are expensive and will increase the cost of the products.

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[0009] More specifically, rare earth elements RE are too expensive to use as additive elements. Furthermore, alloys containing RE involve the problems that their melts degrade in fluidity and are liable to seize up to the mold, which lead to degrading castability and strength at room temperature.

[0010] In existing approaches for development of heat-resistant Mg alloys, there are roughly two tendencies: development of Mg-RE based alloys based upon WE54 and QE22 alloys and development based upon Mg-Al based alloys such as AZ91D and AM60B. In review of the prehistory of Mg-RE based alloys, they were developed as substitutes for Mg-Th based alloys. Therefore, these alloys demonstrate sufficient heat resistance at 200 through 300 °C. However, since the target of their development was military and car-racing purposes, these alloys have not been prepared for commercial production required for use in public vehicles. Development of Mg-RE based alloys tends to incline toward alloys adjusted in amount of additive Zn or Ag to improve the castability and alloys added with Ca or Si to enhance the heat resistance. On the other hand, development of alloys based on the Mg-Al system has become active in recent years and, for enhancing heat resistance, various trials have been made to add combinations of RE, Ca, Si and Sr to Mg-Al based alloys such as AZ91 D and AM60B, for example, as basic alloys. However, these additive elements, if

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added excessively, not only degrade the castability, but also deteriorate the room temperature strength and the corrosion resistance.

SUMMARY OF THE INVENTION

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[0011] It is therefore an object of the present invention to provide a heat-resistant casting Mg alloy enhanced in heat resistance without relying on expensive RE.
[0012] A further object of the invention is to provide a heat-resistant casting Mg alloy well balanced in heat-resistance and room temperature strength.
 10 **[0013]** A still further object of the invention is to provide a heat-resistant casting Mg alloy that can be inhibited from deterioration of corrosion resistance while keeping well balanced in heat resistance and room temperature strength.
[0014] According to the present invention, those objects are accomplished by providing a casting Mg alloy comprising an Mg-Al based alloy containing approximately 5.0-9.0 weight % Al and added with Cu.
 15 **[0015]** The Mg-Al based alloy preferably contains approximately 8.0 weight % aluminum (Al). If the amount of Cu added to the alloy is in the range approximately from 1.0 to 5.0 weight %, heat resistance and room temperature strength will be well balanced. However, it is preferable to limit the amount of Cu to the range approximately from 1.0 to 1.5 weight % and add manganese (Mn) to prevent deterioration of the corrosion resistance. Preferable amount of Mn added here is approximately 0.5-1.0 weight %.

BRIEF DESCRIPTION OF THE DRAWINGS

- [0016]** FIG. 1 shows creep curves of sample Mg alloys added with 1.0-5.0 weight % Cu and comparative alloys.
[0017] FIG. 2 is a diagram plotting creep rate of sample Mg alloys added with 1.0-5.0 weight % Cu together with that of comparative alloys.
 25 **[0018]** FIG. 3 is a graph showing 0.2% proof strength of sample Mg alloys added with 1.0-5.0 weight % Cu at 23°C (room temperature) and 150°C (high temperature), together with that of comparative alloys.
[0019] FIG. 4 is a graph showing maximum tensile stress of sample Mg alloys added with 1.0-5.0 weight % Cu at 23°C (room temperature) and 150°C (high temperature), together with that of comparative alloys.
 30 **[0020]** FIG. 5 is a graph showing tensile elongation of sample Mg alloys added with 1.0-5.0 weight % Cu at 23°C (room temperature) and 150°C (high temperature), together with that of comparative alloys.
[0021] FIG. 6 is a graph showing corrosion rate of sample Mg alloys added with 1.0-5.0 weight % Cu, together with that of comparative alloys.
[0022] FIG. 7 is a graph showing corrosion rate of sample Mg alloys containing 1.5 weight % Cu and 0.25-1.0 weight % Mn, together with that of comparative alloys.
 35 **[0023]** FIG. 8 shows creep curves of sample Mg alloys containing 1.5 weight % Cu and 0.25-1.0 weight % Mn, together with those of comparative alloys.
[0024] FIG. 9 is a diagram plotting creep rate of sample Mg alloys containing 1.5 weight % Cu and 0.25-1.0 weight % Mn, together with that of comparative alloys.
[0025] FIG. 10 is a graph showing 0.2% proof strength of sample Mg alloys containing 1.5 weight % Cu and 0.25-1.0 weight % Mn at 23°C (room temperature) and 150°C (high temperature), together with that of comparative alloys.
 40 **[0026]** FIG. 11 is a graph showing maximum tensile stress of sample Mg alloys containing 1.5 weight % Cu and 0.25-1.0 weight % Mn at 23°C (room temperature) and 150°C (high temperature), together with that of comparative alloys.
[0027] FIG. 12 is a graph showing maximum tensile elongation of sample Mg alloys containing 1.5 weight % Cu and 0.25-1.0 weight % Mn at 23°C (room temperature) and 150°C (high temperature), together with that of comparative alloys.

DETAILED DESCRIPTION OF THE INVENTION

- [0028]** For the benefit of a balance among heat resistance, cost, castability and other factors, the Inventors considered it advantageous to use Mg-Al based alloys as a master and add one or more elements that contribute to enhancement of heat resistance. Consequently, the Inventors selected copper (Cu) as an element that can enhance the heat resistance. Cu not only enhances heat resistance without degrading room temperature strength and castability, but also is available at a lower cost than rare earth elements (RE). Cu enhances heat resistance by accelerating solid solution and generating compounds in the system. Solid-soluble limit of Cu was examined on a Mg-Al-Cu ternary alloy, and it has been confirmed that Cu can be solid-soluble to the maxim, 3.0 weight %, when Al is added to Mg by 8.0 weight %. Mg-Al based alloys are added with 5.0-9.0 weight % Al to improve castability and room temperature strength. Therefore, from the viewpoint of heat resistance and other properties, appropriate amount of Al to be added will be 5.0-9.0 weight %, and preferable amount of Al is 8.0 weight %.
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[0029] Since Zn has a function to improve the mechanical strength of Mg and the fluidity of the melt during casting,
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room temperature strength and castability similar to those of the most demanded AZ91D alloy by adding Zn. However, if the amount of added Zn reaches 2.0% or more, it disturbs alloying. Therefore, appropriate amount of Zn is 2.0 %. Additionally, since the use of SF₆, etc. for preventing combustion of magnesium will be prohibited for protection of the environment, beryllium (Be) having a combustion-preventing effect was added by 0.1 %. Beryllium prevents the surface of the Mg melt from contact with and oxidation by air, and prevents combustion of the melt. Therefore, AZ91D alloys are added with a slight amount of Be as well.

[0030] Mg alloys significantly deteriorate in creep deformation resistance under high temperature environment, and enhancement of its heat resistance typified by creep deformation resistance is required. Creep deformation may occur in a material and will result in changing the material in size from the designed one also when a tightening force determined by a machine design is applied by a fastening element such as a bolt. Therefore, creep deformation resistance is one of the most important properties of materials. Regarding the term "creep deformation" used here, if a metal is exposed to continuous application of a stress of a level even lower than the maximum tensile stress, the metal deforms and breaks down in the long run. This phenomenon of deformation with time under a certain stress is called "creep deformation" or simply "creep".

[0031] Mechanism of creep deformation is different between that occurring at low temperatures and that occurring at high temperatures for the following reason. That is, heat softening occurs at or over the temperature of 0.5 T_m, and brings about dislocations and hole behaviors different from those at lower temperatures. The symbol "T_m" is the melting point in absolute temperature, which is 461.5 K for pure Mg.

[0032] Based upon the above review, creep deformation resistance was examined as an index of heat resistance with the following five Mg alloys prepared as samples different in amount of added Cu.

[0033] (a) First sample: 1.0 % Cu-Mg alloy, containing 8.0 weight % Al, 1.0 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be and the remainder weight % Mg

[0034] (b) Second sample: 1.5 % Cu-Mg alloy, containing 8.0 weight % Al, 1.5 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be and the remainder weight % Mg

[0035] (c) Third sample: 3.0 % Cu-Mg alloy, containing 8.0 weight % Al, 3.0 weight % Cu, .20 weight % Zn, 0.01 weight % Be and the remainder weight % Mg

[0036] (d) Fourth sample: 4.0 % Cu-Mg alloy, containing 8.0 weight % Al, 4.0 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be and the remainder weight % Mg

[0037] (e) Fifth sample: 0.1 % Cu-Mg alloy, containing 8.0 weight % Al, 5.0 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be and the remainder weight % Mg

[0038] Melts for the first to fifth sample alloys were prepared in the following manner. First prepared is a crucible plated with molten alumina (SUS430 stainless steel). The crucible was heated to 750°C in an electric furnace, and weighed amounts of pure Mg, pure Al, pure Cu, Al-2.98%Be master alloy and Zn of Norsk-Hydro were molten. The melts of the alloys were next poured into a mold, and cooled at room temperature. Thus, the first to fifth sample alloys were obtained.

As comparative alloys, AZ91D and AS21B were used. For preparing a melt of AS21B, high-purity Si (6N) was used.

[0039] To examine heat resistance of the alloys, creep test and hot tensile test were conducted. Composition details of the comparative alloys AZ91 D and AS21 B were as follows.

Composition of AZ91D used as the first comparative alloy: Al (8.7 weight %), Zn (0.7 weight %), Be (0.0013 weight %), and Mg (the rest).

Composition of AS21 B used as the second comparative alloy: Al (2.5 weight %), Zn (0.2 weight %), Si (1.2 weight %) and Mg (the rest).

[0040] Creep test is the commonest test for examining heat resistance. It reveals deformation conditions of materials at high temperatures. The examination was conducted by using a single type creep tester. Under the test conditions of test temperature: 150°C, test load: 50 MPa, warm-up time length: 24 hours, and test time length: 100 hours, creep elongation and creep rate were measured.

[0041] When developing heat resistant materials, it is important that the materials exhibit their properties at room temperature even at high temperatures. Therefore, the tests were conducted at different temperature conditions, namely at a high temperature (150°C) and at room temperature (23°C).

[0042] FIG. 1 shows creep curves of the first to fifth sample alloys (S1 through S5) and the first and second comparative alloys (C1 and C2). It is acknowledged from the creep curves of FIG. 1 that the first to fifth sample alloys exhibit creep strains lower than those of the AS21B and AZ91D alloys as the first and second comparative alloys. In addition, while the sample alloys added with 3.0-5.0 weight % Cu (third to fifth sample alloys) decreased in creep stress as the amount of added Cu increased, the sample alloys added with 1.0-1.5 weight % Cu (first and second alloys) exhibited equivalent values of creep strain. From these results, it has been confirmed that creep deformation resistance is enhanced by addition of 1.0 weight % or more Cu.

[0043] FIG. 2 shows the relation between creep rates and time of the first to fifth sample alloys (S1 to S5) as well as the first and second comparative alloys (C1 and C2) on logarithmic scales. Since any of the first to fifth sample alloys as well as the first and second comparative alloys exhibits a creep rate in form of a straight line having a negative

inclination, they are assumed to be in the stage of transient creep.

[0044] 0.2% proof strength, maximum tensile stress and tensile elongation of respective alloys are shown in FIGS. 3, 4 and 5. With reference to FIG. 4, the comparative alloy, AZ91D, exhibited the highest maximum tensile stress both at room temperature and at the high temperature. In contrast, tensile strength of the first to fifth sample Mg alloys (S1 to S5) added with Cu was slightly lower than that of the comparative alloy AZ91D at room temperature, but it became approximately equal at the high temperature. As to the 0.2% proof strength of FIG. 3, although the sample alloys exhibited similar tendency at room temperature, the alloys containing 3.0 weight % or more Cu exhibited higher 0.2% proof strength than AZ91D alloy at the high temperature. Regarding the elongation shown in FIG. 5, the AZ91D alloy largely varied in tensile elongation from 8.0% at room temperature to 24.3% at the high temperature. On the other hand, in the first to fifth sample Mg alloys containing Cu, variance in tensile elongation tended to decrease with increase of the amount of Cu added. From this result, Cu is appreciated to increase its effect of suppressing tensile deformation at high temperatures as its amount of addition increases. Thus, it has been confirmed that 1.0-5.0 weight % Cu added to Mg-8.0% Al alloys enhances heat resistance properties of the alloys, such as creep resistance and hot tensile resistance.

[0045] Although the heat resistance can be improved by adding 1.0-5.0 weight % Cu, electrode potential of Cu is as high as 0.153 V as compared to electrode potential -2.363 V of Mg. Because of this large potential difference, Cu may degrade the corrosion resistance.

[0046] With the first to fifth sample alloys as well as the first and second comparative alloys, salt water immersion test was also conducted. As the salt water for the test, 5.0 weight % NaCl water solution was prepared by dissolving special grade NaCl (sodium chloride) into distilled water. The first to fifth sample alloys as well as the first and second comparative alloys were immersed into the test salt water for 12 hours, and they were washed with distilled water. After that, the alloys were immersed into 10% chromium oxide water solution heated to 100 °C, and corrosive products (magnesium hydroxide adhering on the alloys were removed. The, the corrosion rate of each alloy was calculated by introducing its mass loss, pre-test surface area and test time into the following equation to evaluate the corrosion resistance.

[0047]

$$\text{Corrosion rate (mm/year)} = \{ \text{corrosion amount (mg)} \times 365 \\ \text{(day/year)} \times 10 \text{ (mm/cm)} \} / \{ \text{density (1810 mg/cm}^3\text{)} \times \text{surface area (cm}^2\text{)} \\ \times \text{lapsed time (day)} \}$$

[0048] FIG. 6 shows corrosion rates of the first to fifth sample alloys and the first and second comparative alloys. The Mg alloys added with Cu (first to fifth sample alloys) exhibited higher corrosion rates than those of the comparative alloys AZ91D and AS21B, and were inferior in corrosion resistance. The first to fifth sample alloys tend to deteriorate in corrosion resistance as the amount of Cu added increases. However, there is a large difference in corrosion rate between the alloys containing Cu by 1.5% or less (first and second sample alloys) and the alloys containing Cu by 3.0% or more (third to fifth sample alloys). The Mg alloys containing 1.5% or less Cu (first and second sample alloys) have lower corrosion rates.

[0049] To improve the corrosion resistance, Mn was added as a trial. Since Mn was considered to separate out as simple substance without forming compounds when added by 1.0% or more, corrosion resistance test and heat resistance test were conducted with alloys added with 0.25-1.0 weight % Mn.

[0050] For this study, three samples (sixth to eighth sample alloys) were prepared by adding different amounts of Mn to the aforementioned second sample alloy (1.5% Cu-Mg alloy containing 8.0 weight % Al, 1.5 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be and the rest amount of Mg as follows.

[0051]

(a) Sixth sample: 0.25% Mn-Mg alloy containing 8.0 weight % Al, 1.5 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be, 0.25 weight % Mn and the rest amount of Mg.

(b) Seventh sample: 0.5% Mn-Mg alloy containing 8.0 weight % Al, 1.5 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be, 0.5 weight % Mn and the rest amount of Mg.

(c) Eighth sample: 1.0% Mn-Mg alloy containing 8.0 weight % Al, 1.5 weight % Cu, 2.0 weight % Zn, 0.01 weight % Be, 1.0 weight % Mn and the rest amount of Mg.

[0052] Melts of the sixth to eighth sample alloys were prepared in the same manner as the first to fifth sample alloys. To add Mn, an Al-Mn master alloy and high-purity aluminum were molten in a high-frequency melting furnace, and Al-Mn master alloys adjusted in components and having the target compounding ratios were used. Further, Mg melt was heated to 800°C in an electric furnace and, after adding the Al-Mn master alloy, the melt was stirred for 60 seconds approximately.

[0053] Fig. 7 shows corrosion rates of the sixth to eighth Mn-added sample alloys (S6 to S8), first and second comparative alloys (AZ91D and AS21 B) and the second sample alloy (1.5% Cu-Mg alloy) as another comparative alloy. With reference to Fig. 7, when the amount of Mn is 0.25 weight % (sixth sample alloy), the corrosion rate is 40.96 mm/year, which is not so different from 46.23 mm/year of the alloy not containing Mn (second sample alloy). In contrast, in case of the alloys containing Mn by 0.5 weight % or more (seventh and eighth sample alloys), the corrosion rate decreased to 22.80-26.18 mm/year. This demonstrates that addition of 0.5-1.0 weight % Mn contributes to improvement of corrosion resistance.

[0054] Figs. 8 and 9 show creep curves and creep rates of the sixth to eighth sample alloys and the first, second comparative examples and the second sample alloy. All of the alloys added with Mn exhibited creep strain equivalent to those of alloys not containing Mn. Creep rates appeared in form of similar straight lines as well. It is assumed from this result that addition of Mn does not adversely affect the creep deformation resistance.

[0055] Figs. 10 through 12 show 0.2% proof strength, maximum tensile stress and tensile elongation of the sixth to eighth sample alloys, first and second comparative alloys, and the second sample alloy. With reference to Figs. 10 through 12, alloys containing 0.5% and 1.0% Mn (seventh and eighth samples) exhibited equivalent tensile properties to those of the alloy not containing Mn (second sample) both at the high temperature (150°C) and at room temperature (23°C). In the alloy containing 0.25% Mn (sixth sample), maximum tensile stress and tensile elongation were significantly low at the high temperature (150°C). In general, increase of temperature activates dislocations and increases tensile deformation of a material. However, the 0.25% Mn alloy (sixth sample) exhibited the low tensile property probably because some defects such as inclusions existed in the sample piece and functioned to start the fracture.

[0056] From the result of the test and review, it has been confirmed that corrosion resistance can be enhanced by adding 0.5-1.0 weight % Mn to Mg-8.0% Al-1.5% Cu alloy and that a slight amount of AlMn compound existing among crystal grains of the Mg-8.0% Al-1.5% Cu alloy contributes to enhancement of corrosion resistance without degrading the creep deformation resistance and hot tensile properties.

Claims

1. A casting Mg alloy comprising:

a Mg-Al based alloy containing approximately 5.0 through 9.0 weight % Al; and
Cu added to the Mg-Al based alloy.

2. The casting Mg alloy according to claim 1 wherein the Mg-Al based alloy contains approximately 8.0 weight % Al.

3. The casting Mg alloy according to claim 1 or 2 wherein the added amount of Cu is approximately 1.0 through 5.0 weight %.

4. The casting Mg alloy according to claim 1 or 2 wherein the added amount of Cu is approximately 1.0 through 1.5 weight %.

5. The casting Mg alloy according to claim 4 wherein Mn is added.

6. The casting Mg alloy according to claim 5 wherein the added amount of Mn is approximately 0.5 through 1.0 weight %.

7. A casting Mg alloy comprising:

a Mg-Al based alloy; and
approximately 1.0 through 1.5 weight % Cu, approximately 2.0 weight % Zn and approximately 0.01 weight % Be that are added to the Mg-Al based alloy.

8. The casting Mg alloy according to claim 7 wherein the Mg-Al alloy contains approximately 5.0 through 9.0 weight % Al.

9. The casting Mg alloy according to claim 7 wherein the Mg-Al alloy contains approximately 8.0 weight % Al.

10. A casting Mg alloy comprising:

a Mg-Al based alloy; and
approximately 1.0 through 1.5 weight % Cu, approximately 2.0 weight % Zn, approximately 0.01 weight % Be

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and approximately 0.25 through 1.0 weight % Mn that are added to the Mg-Al based alloy.

11. The casting Mg alloy according to claim 10 wherein the Mg-Al alloy contains approximately 5.0 through 9.0 weight % Al.

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12. The casting Mg alloy according to claim 10 wherein the Mg-Al alloy contains approximately 8.0 weight % Al.

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FIG. 1

(Creep Curves of Cu-added Mg Alloys, AZ91D and AS21B Alloys)

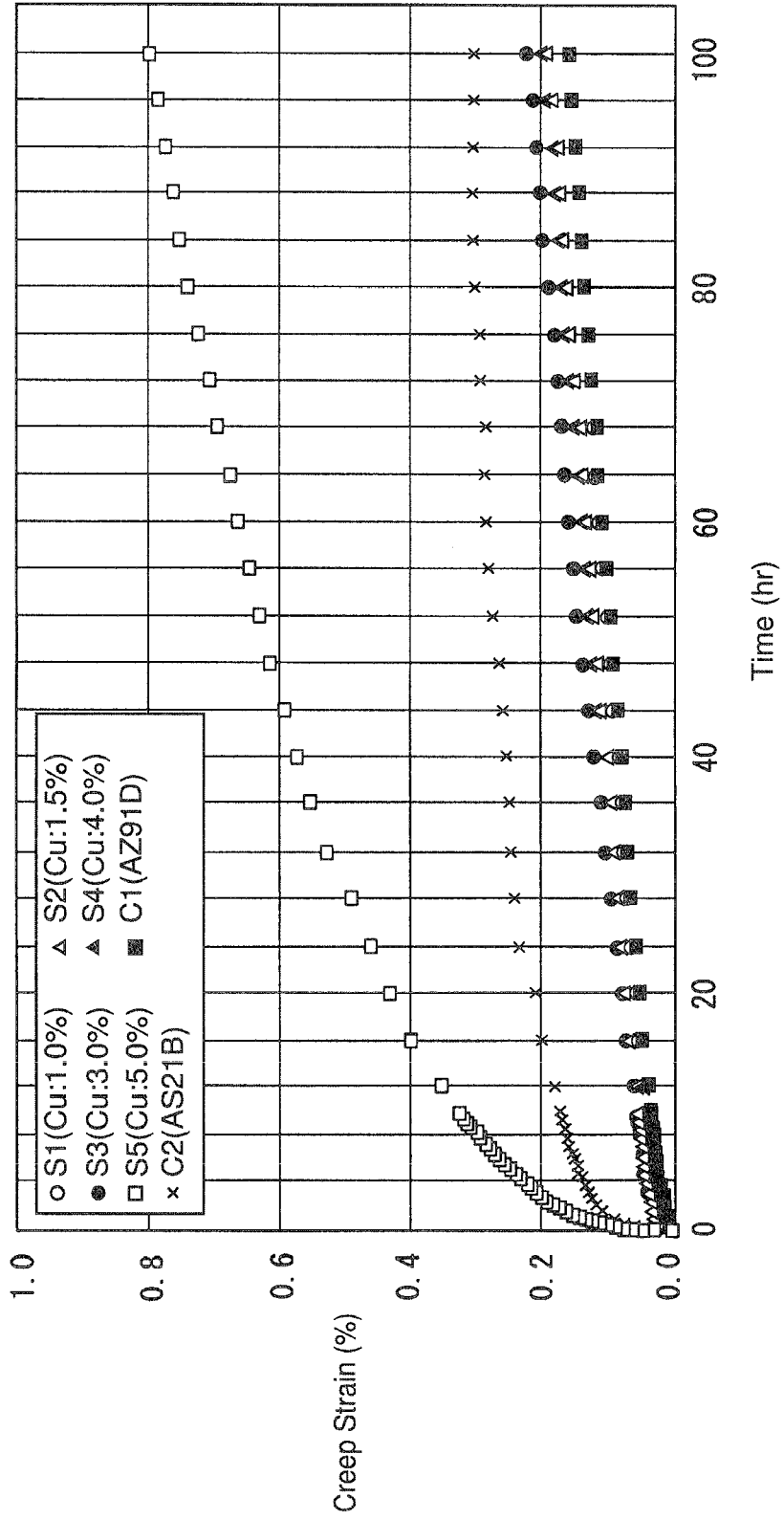


FIG. 2

(Creep Rate of Cu-added Mg Alloys, AZ91D and AS21B Alloys)

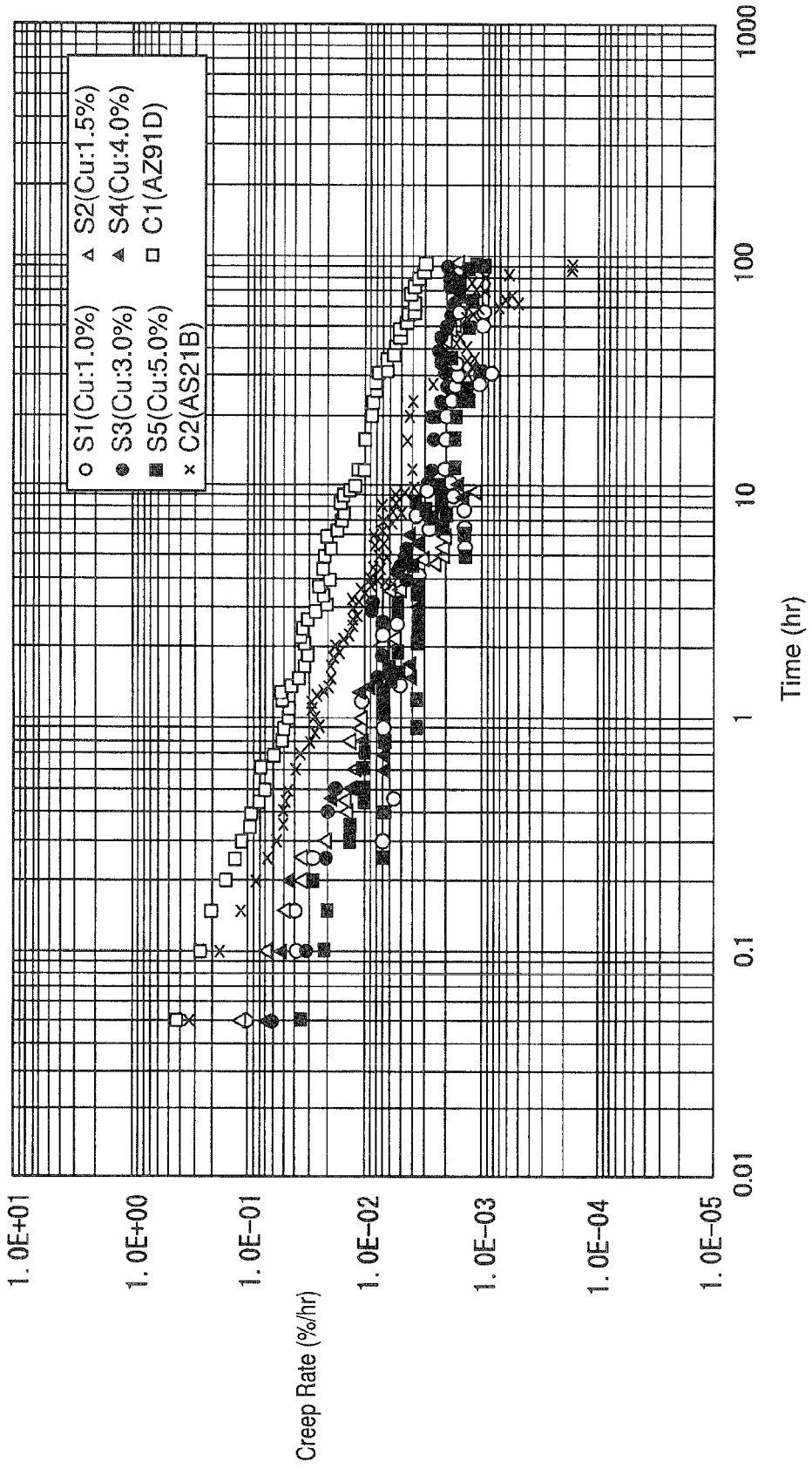


FIG. 3

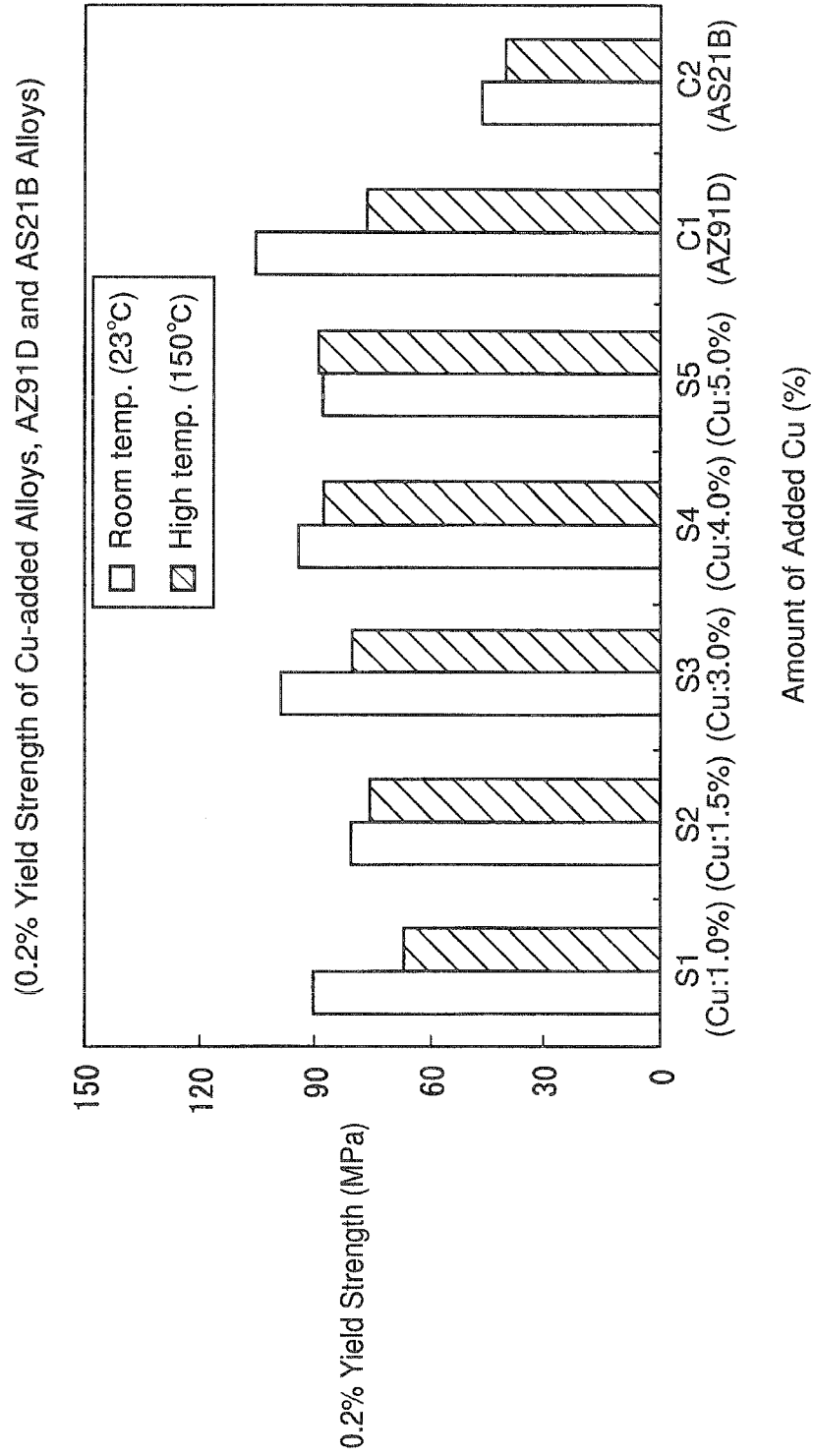


FIG. 4

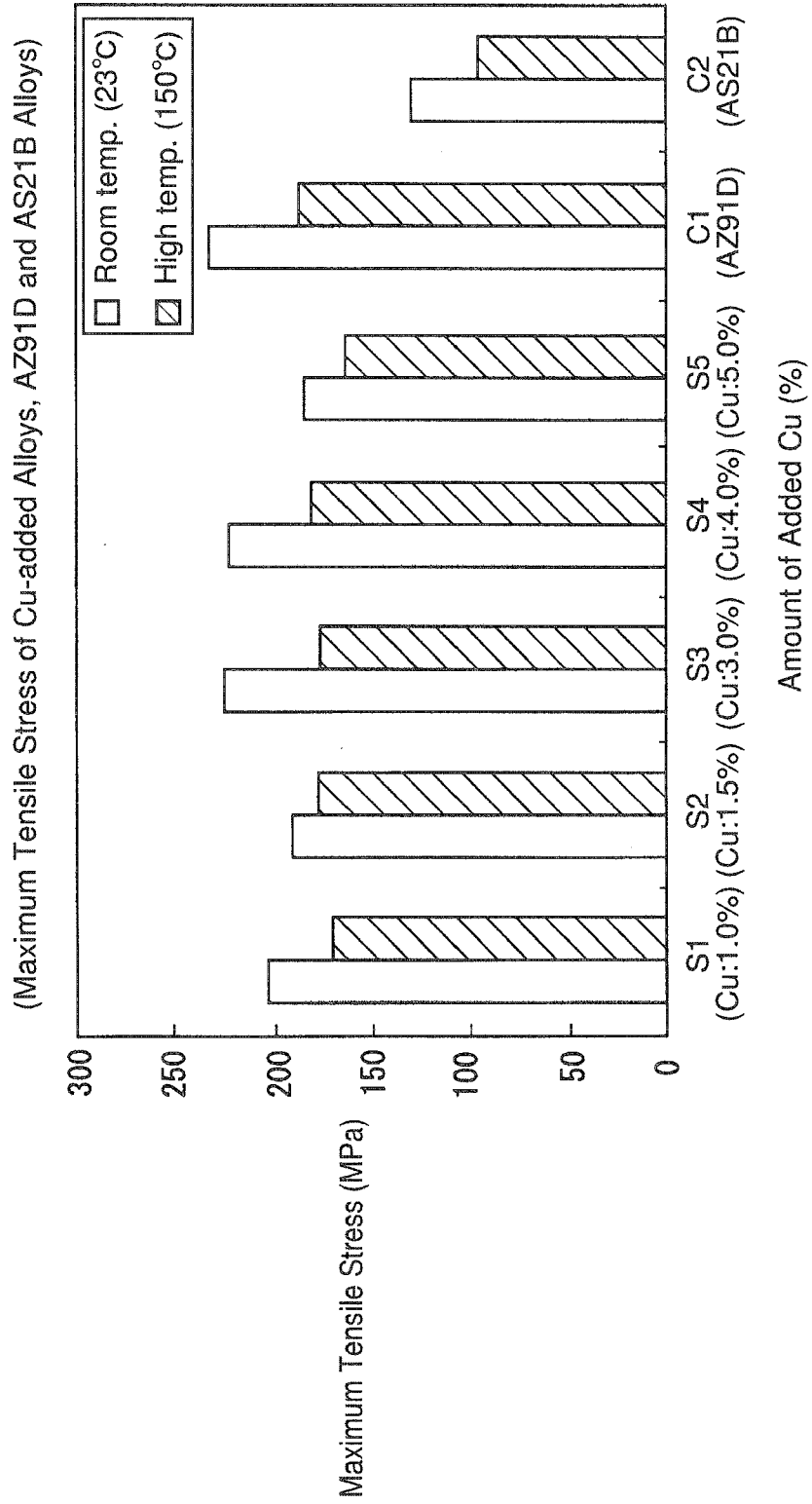


FIG. 5

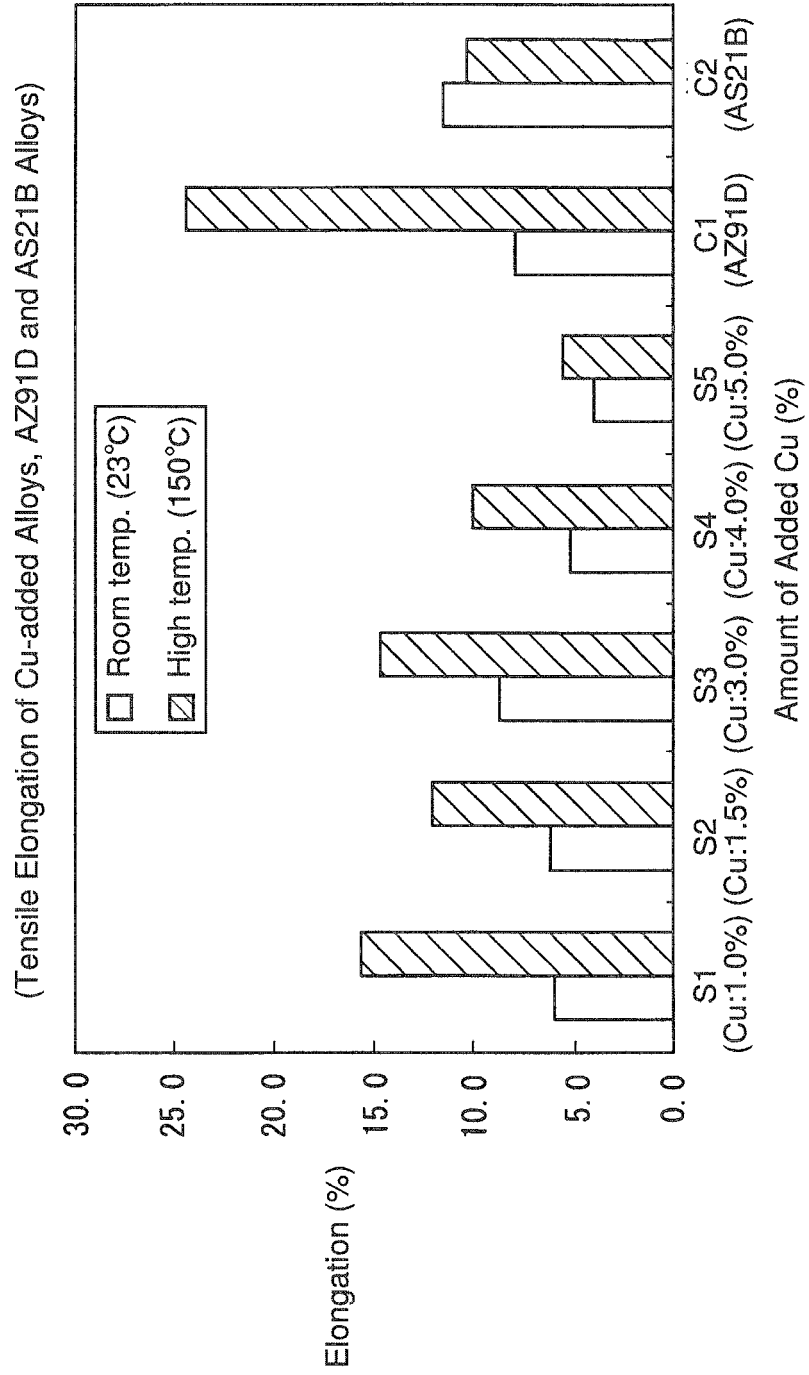


FIG. 6

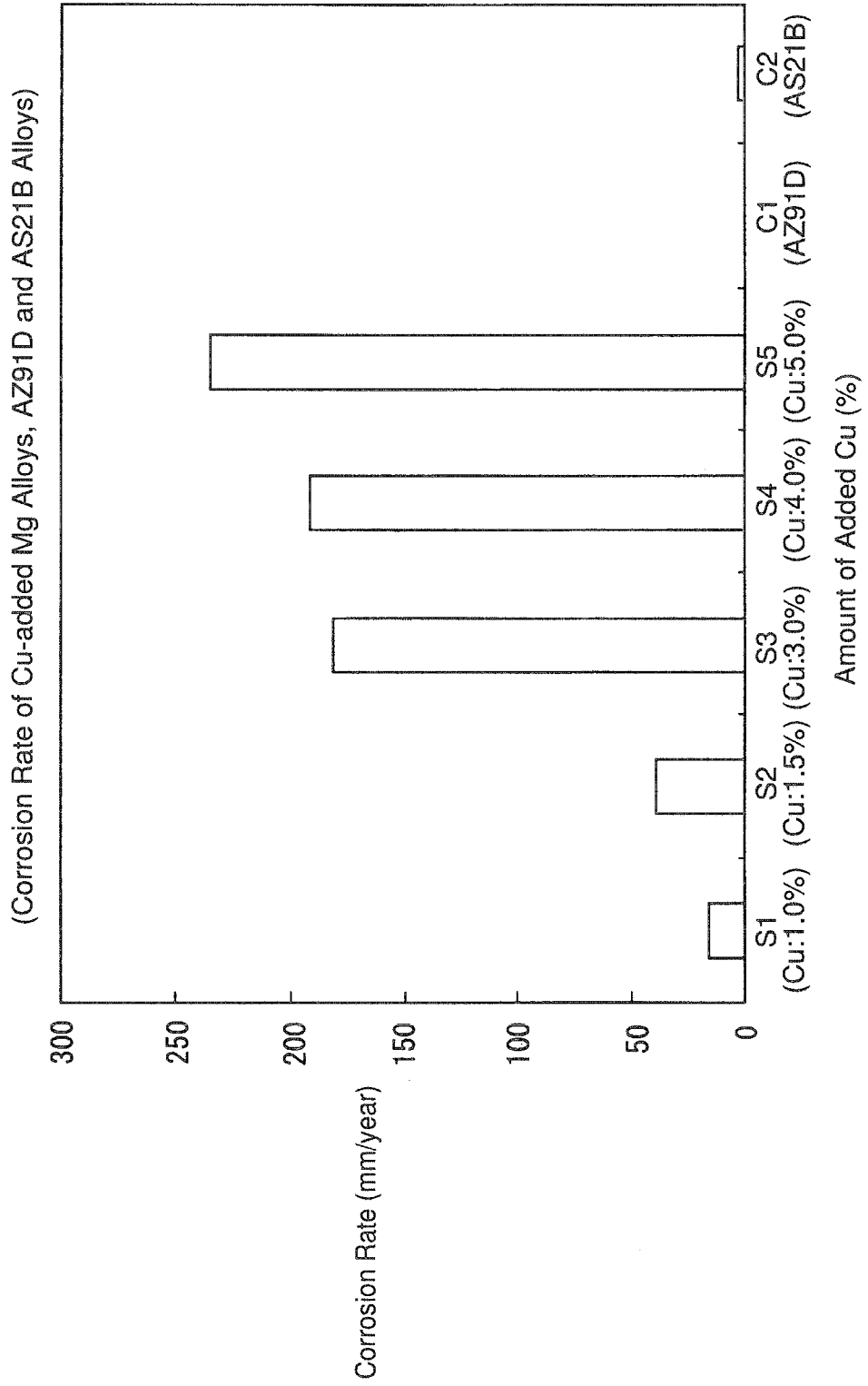


FIG. 7

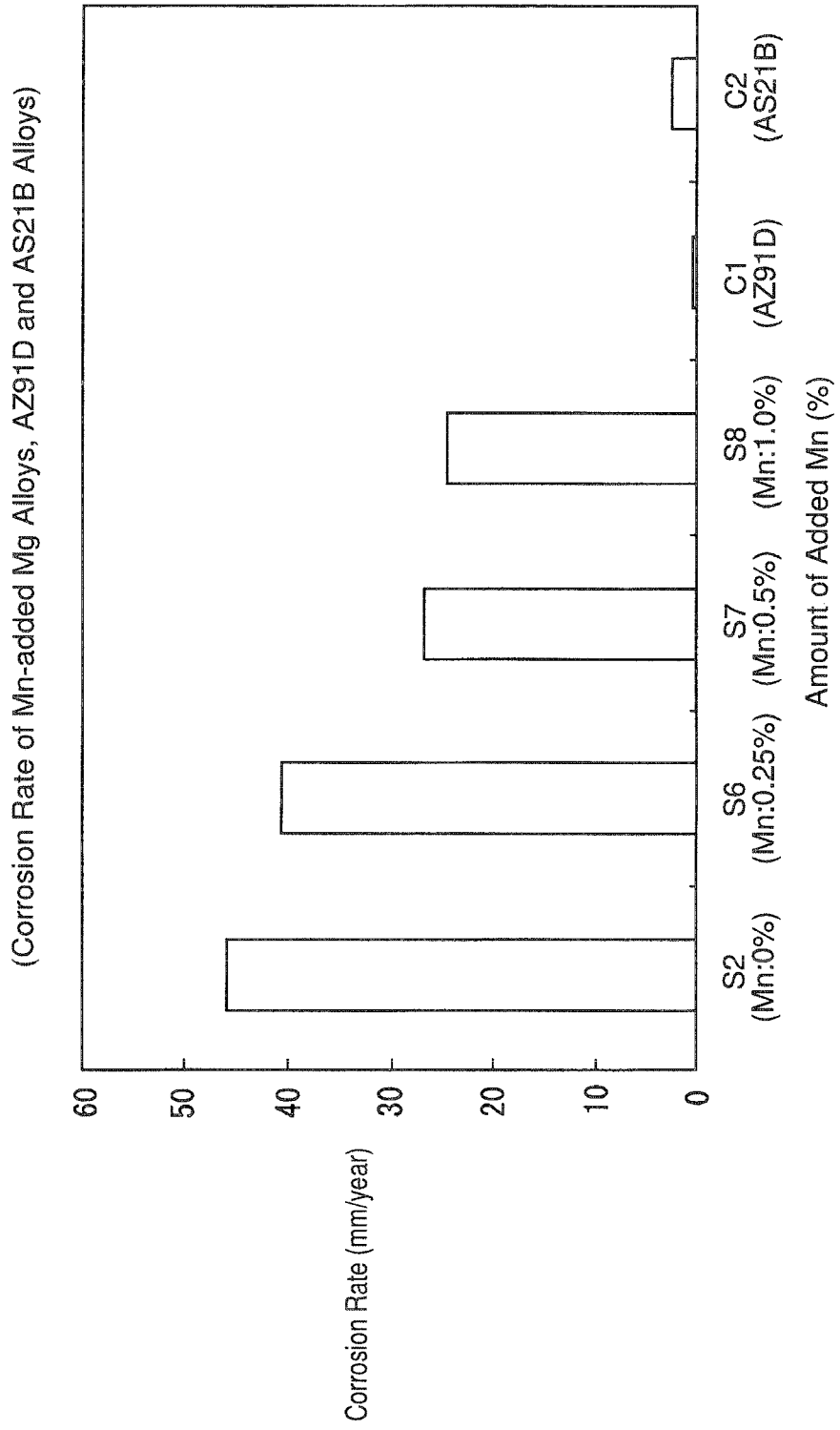


FIG. 8

(Creep Curves of Mn-added Mg Alloys, AZ91D and AS21B Alloys)

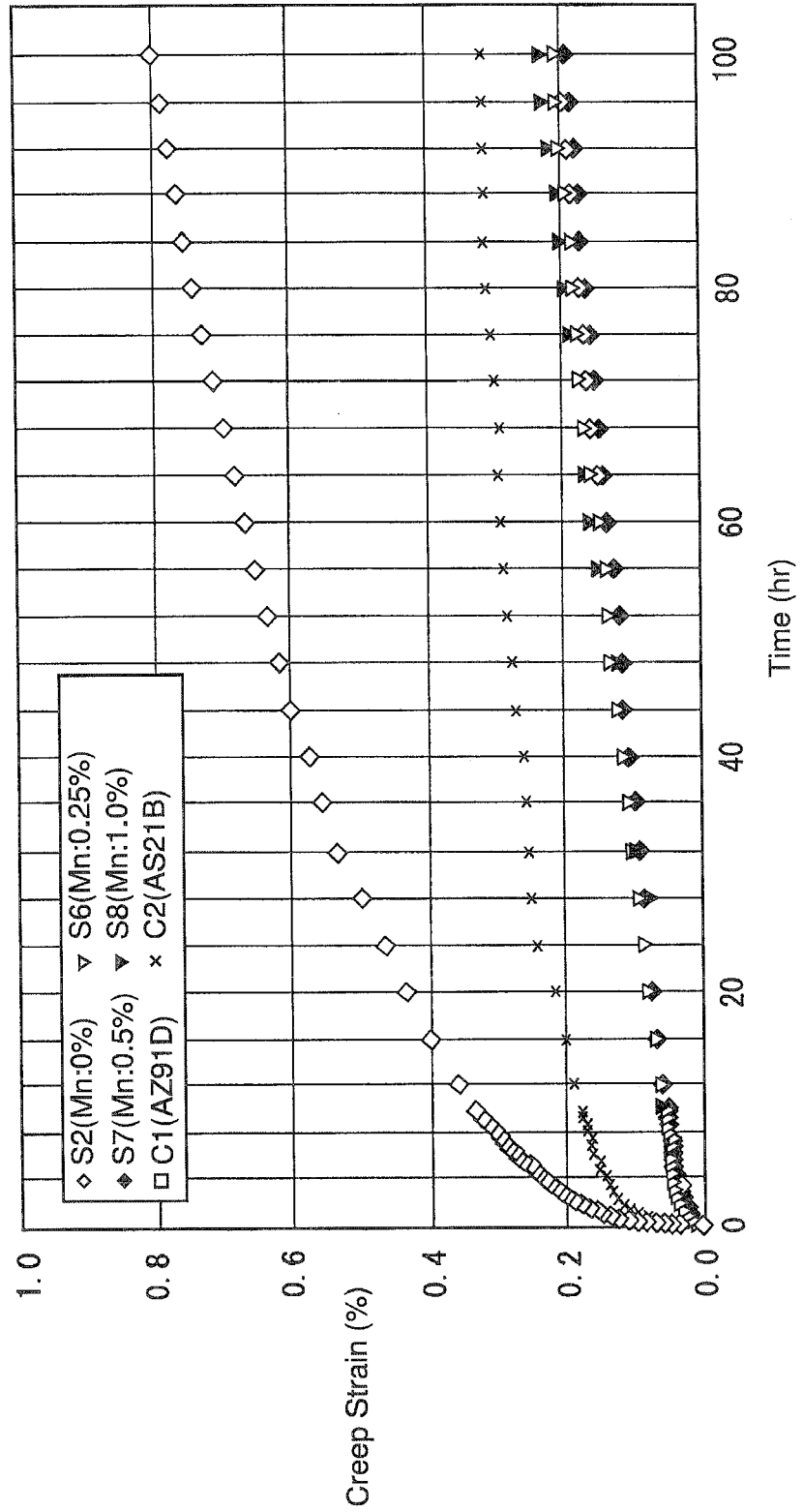


FIG. 9

(Creep Rate of Mn-added Mg Alloys, AZ91D and AS21B Alloys)

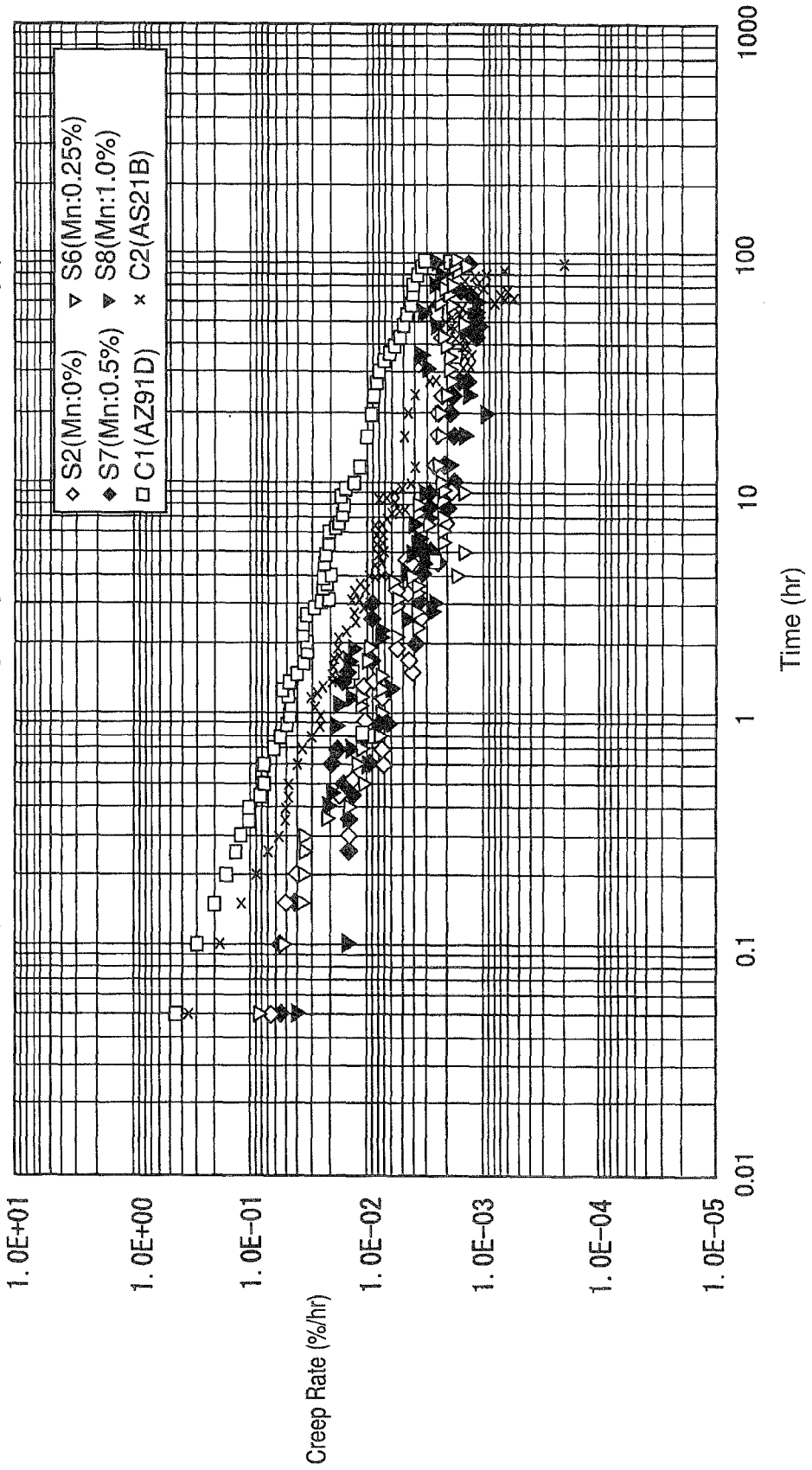


FIG. 10

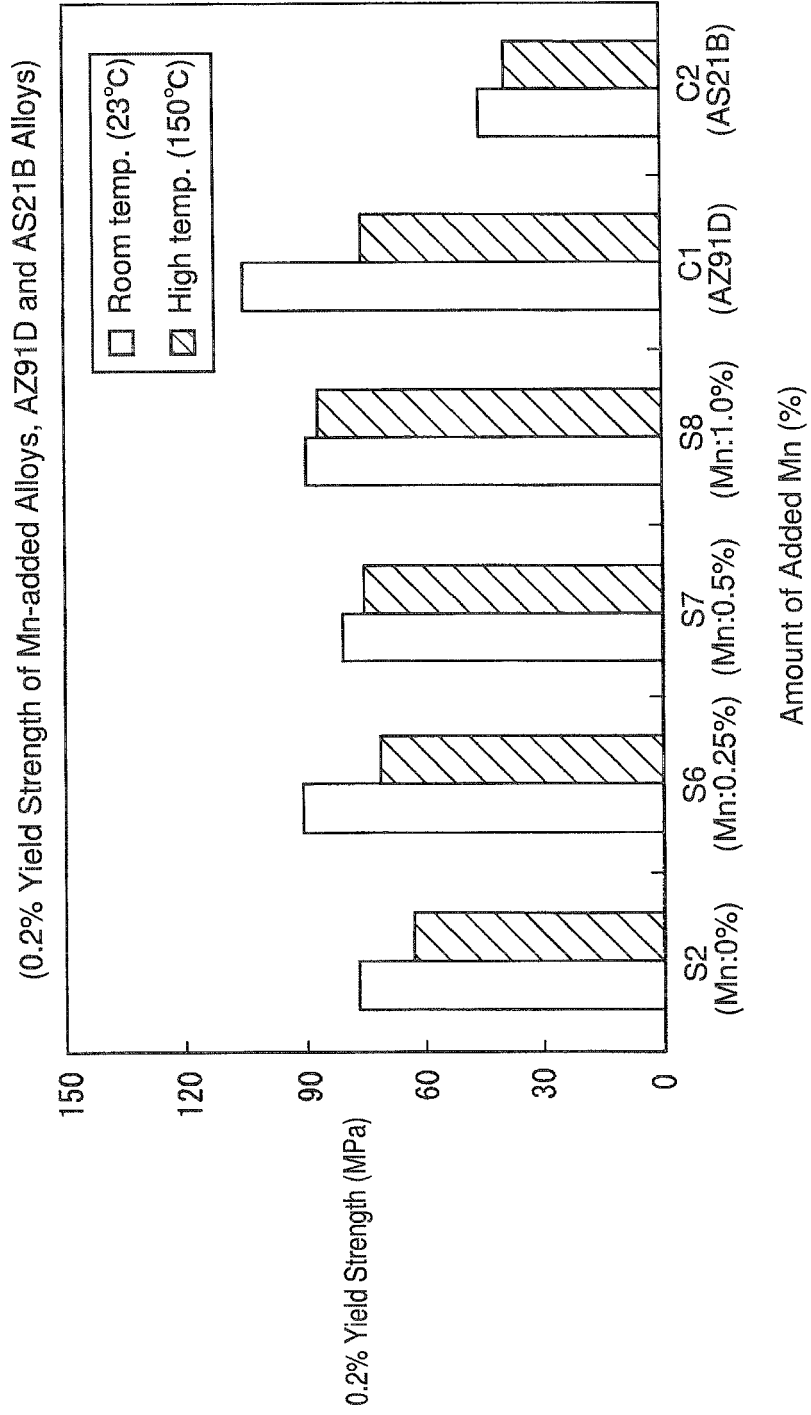


FIG. 11

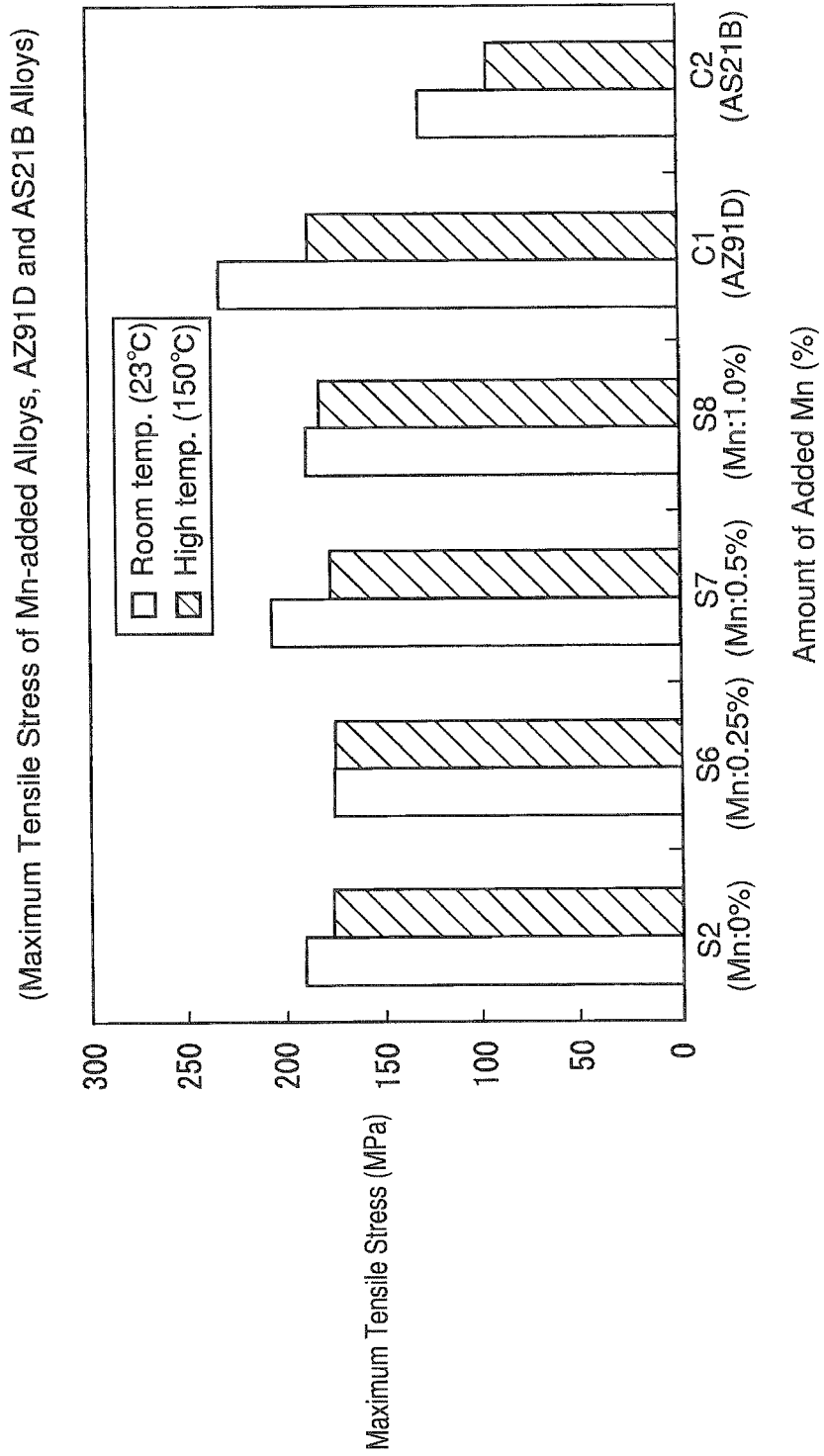


FIG. 12

