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- **ICHIKAWA Ryu**
Naka-shi,
Ibaraki 311-0102 (JP)
- **IGARASHI Makoto**
Naka-shi,
Ibaraki 311-0102 (JP)
- **OKADA Kazuki**
Naka-shi,
Ibaraki 311-0102 (JP)

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(71) Applicant: **Mitsubishi Materials Corporation**
Tokyo 100-8117 (JP)

(74) Representative: **Gille Hrabal**
Partnerschaftsgesellschaft mbB
Patentanwälte
Brucknerstraße 20
40593 Düsseldorf (DE)

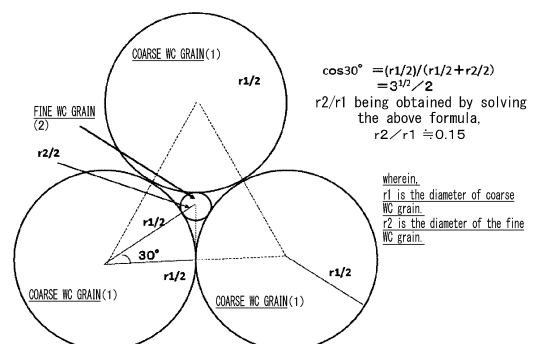
(72) Inventors:
• **KAWAHARA Keisuke**
Naka-shi,
Ibaraki 311-0102 (JP)

(54) **WC-BASED CEMENTED CARBIDE CUTTING TOOL HAVING EXCELLENT DEFECT RESISTANCE AND CHIPPING RESISTANCE, AND SURFACE-COATED WC-BASED CEMENTED CARBIDE CUTTING TOOL**

(57) A WC-based cemented carbide cutting tool and a surface-coated WC-based cemented carbide tool, in which in the WC-based cemented carbide cutting tool, the WC-based cemented carbide has a component composition containing Co: 5 to 14 mass%, Cr₃C₂: 0.1 to 1.4 mass%, and a the WC balance containing inevitable impurities, in a case where a grain size distribution is obtained by measuring grain sizes of WC grains in a cross-section of the WC-based cemented carbide, a plurality of local maximums are present in the grain size distribution, and in a case where a grain size corresponding to a most frequent value formed on a coarse grain side is represented by r₁, and a grain size corresponding to a most frequent value formed on a fine grain side is represented by r₂, a grain size ratio r₂/r₁ is 0.15 or more and 0.60 or less, and an area ratio A₂×100/(A₁+A₂) is 5 to 35 area%, A₁ being a total area of WC grains having a grain size of 0.75 to 1.20 times of r₁ and A₂ being a

total area of WC grains having a grain size of 0.50 to 1.20 times of r₂.

FIG. 1



Description

[Technical Field]

[0001] The present invention relates to a WC-based cemented carbide cutting tool (also referred to as "WC-based cemented carbide tool") having excellent plastic deformation resistance and exhibiting excellent chipping resistance in the cutting of a hard-to-cut material such as stainless steel, and a surface-coated WC-based cemented carbide cutting tool.

[0002] Priority is claimed on Japanese Patent Application No. 2019-057278, filed March 25, 2019, the content of which is incorporated herein by reference.

[Background Art]

[0003] Since WC-based cemented carbide has high hardness and toughness, a WC-based cemented carbide tool having a cutting tool body made of WC-based cemented carbide is known as a cutting tool which exhibits excellent wear resistance and has a long life over long term use.

[0004] However, in recent years, various proposals have been made to further improve the cutting performance and tool life of the WC-based cemented carbide tool according to the type of a work material, cutting conditions, and the like.

[0005] For example, Patent Document 1 proposes cemented carbide which has a hard phase mainly containing tungsten carbide and a binder phase mainly containing an iron group element (including cobalt, the cobalt content is preferably 8 mass% or more in the cemented carbide), and satisfies $B/A \leq 0.05$, where A is the number of grains of the tungsten carbide and B is the number of tungsten carbide grains having one or less point of contact with other tungsten carbide grains, so that the plastic deformation resistance of the cemented carbide is improved, and as a result, the life of a WC-based cemented carbide tool is increased in the wet continuous cutting of carbon steel and stainless steel.

[0006] Patent Document 2 proposes a WC-based cemented carbide tool which has: a Co amount of 10 to 13 mass%; a ratio of a Cr amount to the Co amount of 2% to 8%; at least one of TaC and NbC contained so that a total amount of TaC and NbC is in a range of 0.2 to 0.5 mass%; a balance consisting of WC; and hardness of 88.6 HRA to 89.5 HRA, in which a ratio D80/D20 of a diameter D80 corresponding to 80% in terms of WC integrated grain size to a diameter D20 corresponding to 20% in terms of integrated grain size in area ratio on a polished surface is in a range of $2.0 \leq D80/D20 \leq 4.0$, D80 is in a range of 4.0 to 7.0 μm , and a WC adhesion degree c is $0.36 \leq c \leq 0.43$ so that the adhesion of a work material is prevented and the fracture resistance is improved in the cutting of a hard-to-cut material typified by stainless steel.

[0007] Patent Document 3 proposes a WC-based cemented carbide drill in which in a case where a component composition of WC-based cemented carbide is represented by WC-x mass% Co-y mass% Cr_3C_2 -z mass% VC, $6 \leq x \leq 14$, $0.4 \leq y \leq 0.8$, $0 \leq z \leq 0.6$, and $(y+z) \leq 0.1x$ are satisfied, the WC-based cemented carbide has a WC adhesion degree C so that in a case where the WC adhesion degree C of the WC-based cemented carbide is represented by $C = 1 - V_b^{\alpha} \cdot \exp(0.391 \cdot L)$, a value V_b of a binder phase volume fraction of the WC-based cemented carbide in the above formula is $0.11 \leq V_b \leq 0.25$, a value L of (standard deviation of grain size distribution of WC grains)/(average WC grain size) is in a range of $0.3 < L < 0.7$, and a coefficient α satisfies $0.3 \leq \alpha \leq 0.55$, and thus toughness is improved with no reduction in hardness and rigidity and fracture resistance is thereby increased in the cutting of an A1 alloy, carbon steel, and the like.

[0008] Patent Document 4 proposes a WC-based cemented carbide tool in which in a case where a WC-WC adhesion interface length is represented by L1 and a WC-Co adhesion interface length is represented by L2,

a formula of $R > (0.82 - 0.086 \times D) \times (10/V)$

is satisfied to improve the thermal plastic deformation resistance and toughness of the WC-based cemented carbide tool in the cutting of an Ni-based heat resistant alloy.

$$R = (L1) / ((L1) + (L2))$$

D: WC area average grain size (μm), which is in a range of $0.6 \leq D \leq 1.5$

Here, D is a grain size of WC when the area ratio of WC is 50%.

V: binder phase volume (vol%), which is in a range of $9 \leq V \leq 14$.

[0009] Patent Document 5 proposes a method of manufacturing a WC-based cemented carbide tool having a bimodal grain size distribution, including: wet-mixing WC powders having various grain size distributions together with a binder metal and a pressure forming agent without kneading; drying; pressure-forming; and sintering, in which grains of the WC powder are classified into two groups of a small grain group having a maximum grain size a_{max} and a large grain group having a minimum grain size b_{min} , each group containing at least 10% of the total amount of the WC grains,

$b_{\min} - a_{\max} > 0.5 \mu\text{m}$ is satisfied, and the grain size variation in each group is $>1 \mu\text{m}$.

[0010] It is also proposed that the grains of the small grain group are previously coated with a grain growth inhibitor, the grains of the large grain group are previously coated with a binder metal, the WC-based cemented carbide tool includes WC, 4 to 20 wt% of Co, and <30 wt% of a cubic carbide as a mixture or solid solution containing TiC, TaC, NbC, or WC, the WC grains are classified into two groups, and a weight ratio of coarse WC grains of 2.5 to $6.0 \mu\text{m}$ to fine WC grains of $1.5 \mu\text{m}$ or less is in a range of 0.25 to 4.0 .

[0011] In addition, the WC-based cemented carbide tool produced by the above manufacturing method exhibits excellent wear resistance in the turning of stainless steel.

[0012] Patent Document 6 proposes a WC-based cemented carbide tool for an end mill in which WC-based cemented carbide containing chromium carbide has two types of tungsten carbide phases having different average grain sizes dispersed in a binder phase, a ratio of the average grain sizes of the two types of tungsten carbide phases satisfies $3 \leq \text{average grain size (large)} / \text{average grain size (small)} \leq 7$, a volume ratio therebetween satisfies $1.5 \leq \text{volume of average grain size (large)} / \text{volume of average grain size (small)} \leq 3$, the binder phase contains 0.5 to 2.0 wt% of chromium carbide and 5 to 15 wt% of Co, and a maximum grain size of the tungsten carbide phase is $3.0 \mu\text{m}$ or less.

[0013] In addition, for example, the WC-based cemented carbide tool for an end mill exhibits excellent strength, toughness, and crack propagation resistance in the end mill machining of alloy steel by dispersing two types of tungsten carbide grains having different grain sizes in the binder phase and performing solid-solution strengthening of the chromium carbide in the binder phase.

[Citation List]

[Patent Documents]

[0014]

[Patent Document 1]

Japanese Patent No. 6256415

[Patent Document 2]

Japanese Unexamined Patent Application, First Publication No. 2017-88999

[Patent Document 3]

Japanese Unexamined Patent Application, First Publication No. 2017-148895

[Patent Document 4]

Japanese Unexamined Patent Application, First Publication No. 2017-179433

[Patent Document 5]

Japanese Patent No. 4970638

[Patent Document 6]

Japanese Patent No. 3605740

[Summary of Invention]

[Technical Problem]

[0015] According to the conventional WC-based cemented carbide tools proposed in Patent Documents 1 to 6, the cutting performance and tool characteristics of the WC-based cemented carbide tool are improved by controlling the point of contact between the WC-WC grains, the grain size of the WC grains, the grain size distribution of the WC grains, the WC adhesion degree, and the like.

[0016] However, in the conventional tools, the plastic deformation resistance is not sufficient in the cutting of a hard-to-cut material such as stainless steel, and it is difficult to suppress the advance of fractures due to insufficient toughness. Accordingly, the tool life was short due to the occurrence of abnormal damage such as deformation or chipping of an edge tip.

[Solution to Problem]

[0017] In order to provide a WC-based cemented carbide tool exhibiting excellent plastic deformation resistance and chipping resistance in the cutting of a hard-to-cut material such as stainless steel, the present inventors have pursued intensive studies focusing on the form of WC grains of WC-based cemented carbide, and found the following findings.

[0018] That is, the inventors have found that in a case where WC grains form a texture state approximately close to a close-packed structure in a WC-based cemented carbide tool, the WC grains form a strong skeleton structure, and as

a result, a contact length between the WC-WC grains is increased, the occurrence of grain boundary sliding at the interface of the WC-WC grains is reduced, and thus the plastic deformation resistance is improved.

[0019] It has also been found that since coarse WC grains and fine WC grains are mixed in the alloy texture, even in a case where fractures occur in the WC-based cemented carbide, the advance of linear fractures is suppressed, and as a result, the WC-based cemented carbide has improved toughness.

[0020] Accordingly, in a case where a WC-based cemented carbide tool in which WC grains in WC-based cemented carbide have a texture close to a close-packed structure is provided for cutting of a hard-to-cut material such as stainless steel, deformation of an edge tip of the tool is suppressed due to an improvement in the plastic deformation resistance, and the occurrence of abnormal damage such as chipping is also suppressed due to the suppression of the advance of fractures, so that the life of the tool can be increased.

[0021] The present invention is contrived based on the above findings, and has the following aspects.

(1) A WC-based cemented carbide cutting tool including: a cutting tool body made of WC-based cemented carbide, in which the WC-based cemented carbide has a component composition containing Co: 5 to 14 mass%, Cr_3C_2 : 0.1 to 1.4 mass%, and a the WC balance containing inevitable impurities, in a case where a grain size distribution is obtained based on area by measuring grain sizes of WC grains in a cross-section of the WC-based cemented carbide, a plurality of local maximums are present in the grain size distribution, and in a case where a grain size corresponding to a most frequent value formed on a coarse grain side is represented by r_1 , and a grain size corresponding to a most frequent value formed on a fine grain side is represented by r_2 , a grain size ratio r_2/r_1 is 0.15 or more and 0.60 or less, and an area ratio $A_2 \times 100 / (A_1 + A_2)$ is 5 to 35 area%, A_1 being a total area of WC grains having a grain size of 0.75 to 1.20 times of r_1 and A_2 being a total area of WC grains having a grain size of 0.50 to 1.20 times of r_2 .

(2) The WC-based cemented carbide cutting tool according to (1), in which the WC-based cemented carbide further contains at least one selected from TaC, NbC, TiC, and ZrC in a total amount of 4 mass% or less.

(3) A surface-coated WC-based cemented carbide cutting tool including the WC-based cemented carbide cutting tool according to (1) or (2) and a hard coating layer formed on at least a cutting edge of the WC-based cemented carbide cutting tool. The contents of Cr_3C_2 , TaC, NbC, TiC, and ZrC in (1) and (2) are values obtained by expressing a Cr amount, a Ta amount, an Nb amount, a Ti amount, and a Zr amount measured in a cross-section of the WC-based cemented carbide in terms of carbide, respectively.

[Advantageous Effects of Invention]

[0022] In a WC-based cemented carbide tool and a surface-coated WC-based cemented carbide cutting tool according to an aspect of the present invention (hereinafter, referred to as "WC-based cemented carbide tool of the present invention" and "surface-coated WC-based cemented carbide cutting tool of the present invention"), Co and Cr_3C_2 , which are components of WC-based cemented carbide constituting a body of the tool, and optional TaC, NbC, TiC, and ZrC have a specific composition range. In addition, since WC grains in the WC-based cemented carbide have a texture state approximately close to a close-packed structure, a contact length between the WC-WC grains is increased, the occurrence of grain boundary sliding at a WC-WC interface is suppressed, the cutting tool has excellent plastic deformation resistance, and deformation of an edge tip is suppressed. In addition, since coarse WC grains and fine WC grains are mixed in the alloy texture, even in a case where fractures occur in the WC-based cemented carbide, the advance of linear fractures is suppressed, and resistance to abnormal damage such as chipping is improved.

[Brief Description of Drawings]

[0023]

Fig. 1 shows a schematic view of a case where a close-packed structure formed of three coarse WC grains and one fine WC grain is formed in WC-based cemented carbide. r_2/r_1 obtained by solving a formula of $\cos 30^\circ = (r_1/2) / (r_1/2 + r_2/2) = 3^{1/2}/2$ satisfies $r_2/r_1 \geq 0.15$ (where r_1 is a diameter of the coarse WC grain. r_2 is a diameter of the fine WC grain).

Fig. 2 shows a schematic view of a case where a close-packed structure formed of four coarse WC grains and one fine WC grain is formed in WC-based cemented carbide. r_2/r_1 obtained by solving a formula of $\cos 45^\circ = (r_1 + r_2) / (2 \times r_1) = 1/\sqrt{2}$ satisfies $r_2/r_1 \geq 0.414$ (where r_1 is a diameter of the coarse WC grain. r_2 is a diameter of the fine WC grain).

Fig. 3 shows a schematic view of the measurement of a flank face plastic deformation amount of a cutting edge. The upper view (rake face) is a plan view, and the lower view (flank face) is a side view. As the flank face plastic

deformation amount of the cutting edge, the amount of deformation of a ridge of the cutting edge pushed by cutting is measured after cutting with reference to the ridge of the cutting edge not undergoing deformation before cutting. As a specific measuring method, a line segment is drawn with respect to a main cutting edge-side flank face of the tool on a ridge where the main cutting edge-side flank face and the rake face intersect at a position sufficiently away from the cutting edge, the line segment is extended in a cutting edge portion direction, and a part where a distance between the extended line segment and the ridge of the cutting edge portion (in a direction vertical to the extended line segment) is the longest is measured and defined as the flank face plastic deformation amount of the cutting edge.

[Description of Embodiments]

[0024] Hereinafter, the present invention will be described in detail.

Co:

[0025] Co is contained as a main binder phase forming component of WC-based cemented carbide. In a case where the Co content is less than 5 mass%, sufficient toughness cannot be maintained. In contrast, in a case where the Co content is greater than 14 mass%, the alloy rapidly softens, desired hardness required for a cutting tool cannot be obtained, and deformation and wear remarkably progress. Therefore, the Co content in the WC-based cemented carbide was set to 5 to 14 mass%.

Cr_3C_2 :

[0026] In Cr_3C_2 , Cr forms solid solutions in Co which forms the main binder phase, and causes solid solution strengthening of Co, thereby increasing the strength of the WC-based cemented carbide. However, in a case where the Cr_3C_2 content is less than 0.1 mass%, the above action does not sufficiently occur. In contrast, in a case where the Cr_3C_2 content is greater than 10% of the Co content, a composite carbide of Cr and W precipitates, the toughness is reduced, and it becomes a starting point of the occurrence of fractures.

[0027] Since the upper limit of the Co content is 14 mass% in the present invention, the upper limit of the Cr_3C_2 content is 1.4 mass%, that is 10% of the upper limit of the Co content.

[0028] Therefore, the Cr_3C_2 content in the WC-based cemented carbide was set to 0.1 to 1.4 mass%.

TaC, NbC, TiC, ZrC:

[0029] The WC-based cemented carbide of the present invention may further contain at least one selected from TaC, NbC, TiC, and ZrC as a component of the WC-based cemented carbide in a total amount of 4 mass% or less.

[0030] TaC, NbC, TiC, and ZrC all have an effect of increasing oxidation resistance and crater wear resistance. However, in a case where the total content in terms of carbide is greater than 4 mass%, the wear resistance is insufficient. In addition, agglomerates are likely to be formed, which become a starting point of the occurrence of fractures.

[0031] Therefore, in a case where at least one selected from TaC, NbC, TiC, and ZrC is contained as a component in the WC-based cemented carbide, the total content thereof is preferably 4 mass% or less.

[0032] The contents of Cr_3C_2 , TaC, NbC, TiC, and ZrC described above are values obtained by expressing a Cr amount, a Ta amount, an Nb amount, a Ti amount, and a Zr amount measured by EPMA for the WC-based cemented carbide in terms of carbide, respectively.

Close-Packed Structure of WC Grains:

[0033] WC grains in the WC-based cemented carbide in a WC-based cemented carbide tool of the present invention have a close-packed structure or a texture close to the close-packed structure, and this will be described with the schematic views of Figs. 1 and 2.

[0034] First, Fig. 1 will be described.

[0035] Fig. 1 shows a schematic view in which one fine WC grain 2 having a grain size r_2 invades into a region surrounded by three coarse WC grains 1 having a grain size r_1 , and a close-packed structure formed of three coarse WC grains 1 and one fine WC grain 2 is formed.

$$\text{Here, } \cos 30^\circ = (r_1/2)/(r_1/2 + r_2/2) = 3^{1/2}/2$$

is satisfied. Therefore, $r_2/r_1 \leq 0.156$ is obtained in a case where the above formula is solved.

[0036] Therefore, in order to form the close-packed structure as shown in the schematic view of Fig. 1, the value of r_2/r_1 is required to be approximately 0.15.

[0037] Here, it is assumed that the grain sizes of the coarse WC grains 1 are all r_1 , and the grain sizes of the fine WC grains 2 are all r_2 .

[0038] Next, Fig. 2 will be described.

[0039] Fig. 2 shows a schematic view in which one fine WC grain 2 having a grain size r_2 invades into a region surrounded by four coarse WC grains 1 having a grain size r_1 , and a structure close to a close-packed structure formed of four coarse WC grains 1 and one fine WC grain 2 is formed.

[0040] Here, assuming a virtual lattice as shown by the dotted line in Fig. 2, which has a lattice length a , $2 \times r_1 = \sqrt{2} \times a$ and

$$r_1 + r_2 = a$$

are satisfied. Therefore, $r_2/r_1 \leq 0.414$ is obtained in a case where the above formula is solved.

[0041] Therefore, in order to form the approximately close-packed structure as shown in the schematic view of Fig. 2, the value of r_2/r_1 is required to be approximately 0.42. The value obtained here is a calculated value. Accordingly, the inventors performed an experiment for confirmation with reference to the value, and found that an effect of improving the plastic deformation resistance is obtained up to $r_2/r_1 = 0.60$. Thus, the upper limit of the grain size ratio was set to $r_2/r_1 = 0.60$.

[0042] Here, it is also assumed that the grain sizes of the coarse WC grains 1 are all r_1 , and the grain sizes of the fine WC grains 2 are all r_2 .

[0043] The value of r_2/r_1 obtained above is calculated on the assumption of the coarse WC grains 1 having the same size and the fine WC grains 2 having the same size. However, since WC grains in actual WC-based cemented carbide have different sizes, the above idea cannot be directly applied as a method for forming a close-packed structure of the WC grains.

[0044] However, by applying the above idea to WC grains as a raw material powder for producing WC-based cemented carbide, it can be useful for forming a close-packed structure of the WC grains in WC-based cemented carbide after sintering.

[0045] For example, in a case where a mixed powder consisting of a coarse grain WC powder P1 having a grain size d_1 and a fine grain WC powder P2 having a grain size d_2 is prepared, mixed with another raw material powder containing a Co powder, pulverized, mixed and stirred while suppressing the degree of pulverization as much as possible, and then sintered to produce a WC-based cemented carbide sintered body, a sintered body texture is formed in which WC grains having a grain size r_1 and WC grains having a grain size r_2 coexist in the WC-based cemented carbide sintered body.

[0046] Here, r_1 or r_2 of the WC-based cemented carbide exhibits little change in grain size since the degree of pulverization is suppressed to a low level in the step of pulverizing, mixing, and stirring the raw material powder. Accordingly, r_1 or r_2 of the WC grains of the sintered body can be said to be approximately the same as d_1 or d_2 .

[0047] In this case, in a case where the value of d_2/d_1 in the WC raw material powder is in a range of 0.15 to 0.60, the ratio r_2/r_1 of r_2 to r_1 of the WC grains in the WC-based cemented carbide is in a range of approximately 0.15 to 0.60, and a close-packed structure or a texture close thereto is prepared.

[0048] However, since an actual WC powder does not have a single grain size such as d_1 or d_2 but has a grain size distribution, it is required to consider a grain size distribution of the WC powder as a raw material powder or a grain size distribution of the WC grains in the WC-based cemented carbide in order to form the close-packed structure of the WC grains in the WC-based cemented carbide.

[0049] Accordingly, the inventors focused on a most frequent value, which is a factor that characterizes the grain size distribution, and performed an experiment by associating a grain size r_1 corresponding to the most frequent value formed on the coarse grain side of $3 \mu\text{m}$ or more with a grain size r_2 corresponding to the most frequent value formed on the fine grain side of less than $3 \mu\text{m}$ in the grain size distribution having a plurality of local maximums. As is clear from examples to be described later, it was experimentally confirmed that WC-based cemented carbide in which WC grains have a structure close to a close-packed structure can be produced in a case where a sintered body is produced by blending a coarse grain WC powder and a fine grain WC powder so that a grain size ratio r_2/r_1 is 0.15 or more and 0.60 or less and a total area ratio of WC grains having a grain size of 0.50 to 1.20 times r_2 in the total of a total area A_1 of WC grains having a grain size of 0.75 to 1.20 times r_1 and a total area A_2 of WC grains having a grain size of 0.50 to 1.20 times r_2 , that is, $A_2 \times 100 / (A_1 + A_2)$ is 5 to 35 area%.

[0050] That is, according to the examples to be described later, in a case of $r_2/r_1 > 0.60$, the grain size difference between the coarse WC grains and the fine WC grains is too small, which disturbs an ideal packed structure. In addition, in a case of $r_2/r_1 < 0.15$, even in a case where the fine WC grains enter the gap between the coarse WC grains, the

grains cannot come into contact with each other, and an ideal packed structure cannot be provided. Thus, it is not possible to provide an effect of improving the plastic deformation resistance.

[0051] Furthermore, in a case where the blending ratio of the fine grain WC powder in the total of the coarse grain WC powder and the fine grain WC powder is less than 5 vol%, the fine WC grains cannot exhibit an effect of sufficiently increasing the contact length between the WC-WC grains, and in a case where the above blending ratio is greater than 35 vol%, the coarse WC grains in the WC-based cemented carbide cannot form a close-packed structure which forms a base skeleton.

[0052] As described above, in the present invention, in order to form a close-packed structure or a structure close thereto of the WC grains in the WC-based cemented carbide, it is important to adjust the grain size distribution of each of the coarse grain WC powder and the fine grain WC powder as a WC raw material powder to be used, and to adjust the blending amounts of the coarse grain WC powder and the fine grain WC powder, and a WC-based cemented carbide tool formed of WC-based cemented carbide obtained by appropriately adjusting the grain size distributions and the blending amounts exhibits excellent plastic deformation resistance and chipping resistance in the cutting of a hard-to-cut material such as stainless steel.

[0053] A characteristic of the manufacturing steps in the present invention is to use a method of selecting, mixing, and sintering such a raw material WC that a grain size ratio d_2/d_1 and a grain amount ratio of grains having a grain size d_1 and grains having a grain size d_2 can be made to be equal to a grain size ratio r_2/r_1 and a grain amount ratio of grains having a grain size r_1 and grains having a grain size r_2 , respectively, and thus a target alloy texture is achieved. Specifically, for the raw material WC, polycrystalline WC was not employed since there was a possibility of a deviation from the original grain size due to the grain crushing and pulverization in the mixing process. Only monocrystalline WC was used. As the monocrystalline raw material WC used, a crushed product without agglomeration was used in order to prevent the grains from being crushed during the mixing process and the size thereof from deviating from the original grain size for the same reason as described above. The WC grain size distribution can be approximated by a Gaussian function where a horizontal axis represents the WC grain size and a vertical axis represents the WC frequency in the measurement by a laser diffraction and scattering type grain size distribution measuring method, and a raw material WC was selected in which a value of 3σ of the coarse WC grains was equal to or less than a value of $0.20 \times d_1$ and a value of 3σ of the fine WC grains was equal to or less than a value of $0.20 \times d_2$, where σ is a standard deviation calculated from the Gaussian function approximated. In addition, in the mixing method, in order to prevent that the WC grains are pulverized and their sizes are thus changed from the original grain size of the raw material WC, mixing with low pulverization energy, preferably medialess mixing was introduced. Furthermore, in the sintering method, in a case where the sintering is performed at a high temperature for a long period of time, the WC grains grow and the growth causes a deviation from the original grain size. Accordingly, the sintering was performed under such optimum sintering conditions of a low temperature and a short period of time that defects (pores) were not generated in the alloy.

[0054] Based on the above thought, the WC-based cemented carbide tool of the present invention can be produced by, for example, the following steps.

[0055] First, two types of monocrystalline WC powders having different grain size distributions are blended so as to have a predetermined blending ratio, a raw material powder formed of a Co powder and a Cr_3C_2 powder or a raw material powder further containing one or more of a TaC powder, an NbC powder, a TiC powder, and a ZrC powder as necessary is further added, and the powders are blended and mixed without the application of a large crushing force by, for example, medialess mixing by an attritor with a reduced amount of media, preferably an ultrasonic homogenizer, a cyclone mixer, or the like to produce a mixed powder.

[0056] Then, a powder compact is produced by molding the mixed powder. The powder compact is sintered under conditions of a low temperature and a short period of time under a vacuum atmosphere: heating temperature: $1,300^\circ\text{C}$ or higher and $1,500^\circ\text{C}$ or lower, preferably $1,300^\circ\text{C}$ or higher and $1,400^\circ\text{C}$ or lower; and heating holding time: 15 to 120 minutes, preferably 15 to 60 minutes to produce WC-based cemented carbide so that changes in the WC shape and the grain size distribution due to the growth of the grains are suppressed.

[0057] Then, the WC-based cemented carbide can be machined and ground to produce a WC-based cemented carbide tool having a desired size and a desired shape.

[0058] In the WC-based cemented carbide tool produced in the above steps, the contact length between the WC-WC grains is increased, the occurrence of grain boundary sliding at the interface of the WC-WC grains is reduced, and thus the plastic deformation resistance is improved. In addition, since the coarse WC grains and the fine WC grains are mixed in the alloy texture, even in a case where fractures occur in the WC-based cemented carbide, the advance of linear fractures is suppressed and the toughness is improved.

[0059] Furthermore, by forming a hard film such as a Ti-Al or Al-Cr-based carbide, nitride, or carbonitride, or Al_2O_3 on at least the cutting edge of the WC-based cemented carbide cutting tool by coating using a film forming method such as PVD or CVD, a surface-coated WC-based cemented carbide cutting tool can be produced.

[0060] In the production of the surface-coated WC-based cemented carbide cutting tool, the type of the hard film and the film forming method employed may be a film type and a film forming method which have already been known to

those skilled in the art, and are not particularly limited.

[0061] The WC-based cemented carbide tool and the surface-coated WC-based cemented carbide cutting tool of the present invention will be described in greater detail with reference to examples.

[Examples]

[0062]

(a) First, as powders for sintering, two types of monocrystalline WC powders (a coarse grain WC powder having a most frequent value of d_1 (μm) in its grain size distribution and a fine grain WC powder having a most frequent value of d_2 (μm) in its grain size distribution) having different grain size distributions, a Co powder, a Cr_3C_2 powder, a TaC powder, an NbC powder, a TiC powder, and a ZrC powder are prepared.

These powders were blended so as to have a blending composition shown in Table 1, that is, blended so as to have a WC grain size ratio and a blending ratio of coarse grains and fine grains in which d_2/d_1 was 0.15 or more and 0.60 or less and (blending amount of fine WC grains having the most frequent value of d_2)/(blending amount of coarse WC grains having the most frequent value of d_1 +blending amount of fine WC grains having the most frequent value of d_2) satisfied 5% to 35%. Thus, powders for sintering were produced.

Table 1 shows the blending compositions (mass%) of various powders, the values of the grain sizes d_1 and d_2 corresponding to the most frequent values of the two types of WC powders in the grain size distributions, and the blending ratios of the fine WC grains and the coarse WC grains.

The average grain sizes (D50) of the Co powder, the Cr_3C_2 powder, the TaC powder, the NbC powder, the TiC powder, and the ZrC powder are in a range of 1.0 to 3.0 μm .

(b) The powders for sintering prepared by blending so as to have a blending composition shown in Table 1 were wet-mixed by medialess attritor mixing at a rotation speed of 50 rpm for 8 hours, dried, and then pressure-formed at a pressure of 100 MPa to produce powder compacts.

(c) Then, these powder compacts were sintered under conditions of heating temperature: 1,300°C or higher and 1,500°C or lower and heating holding time: 15 to 120 minutes under a vacuum atmosphere, and WC-based cemented carbides were produced.

(d) Then, the WC-based cemented carbides were machined and ground to produce WC-based cemented carbide tools 1 to 11 (hereinafter, referred to as the present invention tools 1 to 11) of an insert shape of CNMG120408-GM shown in Table 3.

[0063] WC-based cemented carbide tools 1 to 9 of comparative examples (hereinafter, referred to as the comparative example tools 1 to 9) were manufactured for comparison.

[0064] In steps of manufacturing the tools, powders for sintering having a blending composition shown in Table 2, that is, a WC grain size ratio and a blending ratio of coarse grains and fine grains in which d_2/d_1 was 0.15 or more and 0.60 or less or (blending amount of fine WC grains having the most frequent value of d_2)/(blending amount of coarse WC grains having the most frequent value of d_1 +blending amount of fine WC grains having the most frequent value of d_2) was out of a range of 5% to 35% were wet-mixed by medialess attritor mixing at a rotation speed of 50 rpm for 8 hours, dried, and then press-formed at a pressure of 100 MPa to produce powder compacts.

[0065] Then, the comparative example tools 1 to 9 shown in Table 4 were produced by performing the same steps as (c) and (d) in the manufacturing steps of the present invention tools 1 to 11.

[0066] In a cross-section of the WC-based cemented carbide of each of the present invention tools 1 to 11 and the comparative example tools 1 to 9, the contents of Co, Cr, Ta, Nb, Ti, and Zr, which were the components of the cemented carbide, were measured at 10 points by EPMA. An average value of the measured values was defined as a content of each component.

[0067] Each of the contents of Cr, Ta, Nb, Ti, and Zr was calculated by expressing in terms of carbide.

[0068] Tables 3 and 4 show the average contents of the components.

[0069] Next, the cross-section of the WC-based cemented carbide of each of the present invention tools 1 to 11 and the comparative example tools 1 to 9 was observed by an electron scanning microscope (SEM) using an electron backscatter diffraction method (hereinafter, referred to as EBSD). The area of each WC grain was measured by analyzing a crystal orientation mapping image of the WC grains, a diameter when the WC grain was approximated to a circle of the same area was calculated, and a grain size distribution graph was made with a vertical axis representing a grain area ratio (a ratio of a total area of WC grains having the corresponding grain size to a total area of all WC grains in the measurement range) and a horizontal axis representing the grain size. In this case, the horizontal axis is divided by 0.2 μm width, and the minimum value is 0.1 μm . The WC grain size is a value in the middle of the dividing width.

[0070] The cross-section used for observation by EBSD and SEM is an optional cross-section, and in this example, a cross-section of the alloy processed by ion milling by 100 μm or more from the outermost surface of the flank face was observed. The observation range in EBSD was a field of view of $24 \times 72 \mu\text{m}$, and the number of WC grains was set to 4,000 or more in the measurement range. The observation conditions in EBSD included a pixel size of $0.1 \mu\text{m} \times 0.1 \mu\text{m}$ and a capturing time of 15 ms/point. The observation range in SEM was a field of view of $24 \times 72 \mu\text{m}$, and the number of WC grains was set to 4,000 or more in the measurement range. The observation conditions in SEM included an acceleration voltage of 15 kV.

[0071] In this case, in a grain size distribution having a plurality of local maximums, a grain size corresponding to the most frequent value formed on the coarse grain side of 3 μm or more was represented by r_1 , and a grain size corresponding to the most frequent value formed on the fine grain side of less than 3 μm was represented by r_2 .

[0072] Regarding the WC grains extracted by the EBSD method, a total area A_1 (μm^2) of WC grains having a grain size of 0.75 to 1.20 times r_1 and a total area A_2 (μm^2) of WC grains having a grain size of 0.50 to 1.20 times r_2 were obtained, and a total area ratio ($A_2 \times 100 / (A_1 + A_2)$ area%) of the WC grains having a grain size of 0.50 to 1.20 times r_2 to a total of the total area A_1 of the WC grains having a grain size of 0.75 to 1.20 times r_1 and the total area A_2 of the WC grains having a grain size of 0.50 to 1.20 times r_2 was obtained.

[0073] Tables 3 and 4 show the measurement results. Since crushed monocrystalline WC grains which are difficult to pulverize and crush are used, and a medialess mixing method avoiding the pulverization of the grains and sintering conditions of a low temperature and a short period of time for suppressing the growth of the grains are introduced in the manufacturing steps, it has been found that " r_2/r_1 " approximately reflects " d_2/d_1 " of the raw material and " $A_2 \times 100 / (A_1 + A_2)$ " approximately reflects the blending ratio of the coarse WC grains and the fine WC grains "(blending amount of fine WC grains having the most frequent value of d_2) / (blending amount of coarse WC grains having the most frequent value of d_1 + blending amount of fine WC grains having the most frequent value of d_2)".

[Table 1]

Raw Material Powder Type	Blending Composition (mass%)										
	Co	Cr ₃ C ₂	TaC	NbC	TiC	ZrC	WC				
							Coarse WC Grains		Fine WC Grains		(reference blending ratio) Fine WC Grains/ (Fine WC Grains+Coarse WC Grains)
							Blending Amount (mass%)	d1 (μm)	Blending Amount (mass%)	d2 (μm)	
A	5.0	0.3	1.5	0.3	1.1	-	65.0	7.6	26.8	2.8	29.2
B	9.1	0.5	1.8	-	-	-	69.6	6.4	19.0	1.5	21.4
C	7.1	0.7	1.2	0.3	0.5	0.2	67.6	6.7	22.4	1.0	24.9
D	10.0	0.5	-	-	-	-	74.0	7.0	15.5	2.5	17.3
E	8.5	0.7	2.0	0.5	0.5	-	79.0	4.8	8.8	2.9	10.0
F	11.1	1.0	1.0	1.0	1.0	1.0	71.2	4.0	12.7	2.0	15.1
G	9.6	0.6	0.1	0.1	-	0.1	58.2	4.2	31.3	1.4	35.0
H	7.5	0.1	-	0.5	0.5	-	67.6	5.9	23.8	2.6	26.0
I	6.3	0.5	-	2.1	1.0	0.5	85.1	6.2	4.5	1.8	5.0
J	12.0	0.8	-	-	1.8	0.6	78.0	3.2	6.8	0.9	8.0
K	14.0	1.4	0.7	-	0.9	2.1	55.7	3.5	25.2	0.7	31.1
											(reference value) d2/d1
											0.37
											0.24
											0.15
											0.36
											0.60
											0.50
											0.33
											0.44
											0.29
											0.28
											0.20

[0074] In the column of "WC", inevitable impurities are also contained.

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Raw Material Powder Type	Blending Composition (mass%)											
	Co	Cr ₃ C ₂	TaC	NbC	TiC	ZrC	WC				(reference blending ratio) Fine WC Grains/ (Fine WC Grains+Coarse WC Grains)	(reference value) d2/d1
							Coarse WC Grains		Fine WC Grains			
							Blending Amount (mass%)	d1 (μm)	Blending Amount (mass%)	d2 (μm)		
a	15.0	0.8	2.4	-	1.0	0.5	65.8	3.5	14.5	0.3	18.1	0.09
b	7.5	0.3	0.6	2.0	-	0.5	85.1	3.3	4.0	0.9	4.5	0.27
c	10.5	1.0	-	0.3	1.1	-	52.3	4.1	34.8	0.9	40.0	0.22
d	7.9	0.1	0.1	1.5	1.4	0.4	75.3	4.2	13.3	2.8	15.0	0.67
e	4.7	0.1	-	0.3	1.6	1.5	87.8	4.3	4.0	0.9	4.4	0.21
f	9.5	0.6	1.7	0.2	-	0.2	70.2	6.3	17.6	0.8	20.0	0.13
g	13.2	1.5	1.4	0.5	-	-	53.7	3.9	29.7	0.8	35.6	0.21
h	8.6	-	-	-	1.0	0.2	85.8	4.6	4.4	2.9	4.9	0.63
i	6.5	0.4	1.5	2.3	-	0.6	61.3	4.0	27.4	0.5	30.9	0.13

[0075] In the column of "WC", inevitable impurities are also contained.

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[Table 3]

Present Invention Tool Type	Raw Material Powder Type	Com position (mass%)							r1 (μm)	r2 (μm)	r2/r1	A2/(A1+A2) (area%)
		Co	Cr ₃ C ₂	TaC	NbC	TiC	ZrC	WC				
1	A	5.0	0.3	1.5	0.3	1.1	-	balance	7.6	2.8	0.37	29.2
2	B	9.1	0.5	1.8	-	-	-	balance	6.4	1.5	0.24	21.4
3	C	7.1	0.7	1.2	0.3	0.5	0.2	balance	6.7	1.0	0.15	24.9
4	D	10.0	0.5	-	-	-	-	balance	7.0	2.5	0.36	17.3
5	E	8.5	0.7	2.0	0.5	0.5	-	balance	4.8	2.9	0.60	10.0
6	F	11.1	1.0	1.0	1.0	1.0	1.0	balance	4.0	2.0	0.50	15.1
7	G	9.6	0.6	0.1	0.1	-	0.1	balance	4.2	1.4	0.33	35.0
8	H	7.5	0.1	-	0.5	0.5	-	balance	5.9	2.6	0.44	26.0
9	I	6.3	0.5	-	2.1	1.0	0.5	balance	6.2	1.8	0.29	5.0
10	J	12.0	0.8	-	-	1.8	0.6	balance	3.2	0.9	0.28	8.0
11	K	14.0	1.4	0.7	-	0.9	2.1	balance	3.5	0.7	0.20	31.1

[0076] In the column of "WC", inevitable impurities are also contained.

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[Table 4]

Comparative Example Tool Type	Raw Material Powder Type	Com position (mass%)							r1 (μm)	r2 (μm)	r2/r1	A2/(A1+A2) (area%)
		Co	Cr ₃ C ₂	TaC	NbC	TiC	ZrC	WC				
1	a	15.0	0.8	2.4	-	1.0	0.5	balance	3.5	0.3	0.09	18.1
2	b	7.5	0.3	0.6	2.0	-	0.5	balance	3.3	0.9	0.27	4.5
3	c	10.5	1.0	-	0.3	1.1	-	balance	4.1	0.9	0.22	40.0
4	d	7.9	0.1	0.1	1.5	1.4	0.4	balance	4.2	2.8	0.67	15.0
5	e	4.7	0.1	-	0.3	1.6	1.5	balance	4.3	0.9	0.21	4.4
6	f	9.5	0.6	1.7	0.2	-	0.2	balance	6.3	0.8	0.13	20.0
7	g	13.2	1.5	1.4	0.5	-	-	balance	3.9	0.8	0.21	35.6
8	h	8.6	-	-	-	1.0	0.2	balance	4.6	2.9	0.63	4.9
9	i	6.5	0.4	1.5	2.3	-	0.6	balance	4.0	0.5	0.13	30.9

[0077] In the column of "WC", inevitable impurities are also contained.

[0078] In addition, the following wet continuous cutting test was performed on the present invention tools 1 to 11 and the comparative example tools 1 to 9, each of which was in a state of being screwed to a tip portion of an insert holder of tool steel by a fixing jig.

Work Material: round bar of JIS-SUS304 (HB170),
Cutting Speed: 100 m/min,
Cutting Depth: 2.0 mm,
Feeding: 0.5 mm/rev,
Cutting Time: 5 minutes, and
Using wet water-soluble coolant

[0079] After the wet continuous cutting test, a flank face plastic deformation amount of the cutting edge was measured, and the wear state of the cutting edge was observed. Regarding the flank face plastic deformation amount of the cutting edge, a line segment was drawn with respect to a main cutting edge-side flank face of the tool on a ridge where the main cutting edge-side flank face and the rake face intersected at a position sufficiently away from the cutting edge, the line segment was extended in a cutting edge portion direction, and a part where a distance between the extended line segment and the ridge of the cutting edge portion (in a direction vertical to the extended line segment) was the longest was measured and defined as the flank face plastic deformation amount of the cutting edge. A wear state when the flank face plastic deformation amount was 0.04 mm or more was defined as edge tip deformed. Fig. 3 shows a schematic view of the measurement of the flank face plastic deformation amount.

[0080] Table 5 shows the measurement results.

[Table 5]

Present Invention Tool	Flank Face Plastic Deformation Amount (mm)	Wear state	Comparative Example Tool	Flank Face Plastic Deformation Amount (mm)	Wear state
1	0.012	normal wear	1	0.076	edge tip deformed
2	0.026	normal wear	2	0.046	edge tip deformed
3	0.029	normal wear	3	0.081	edge tip deformed
4	0.024	※ Medium- level crater wear occurred	4	0.050	edge tip deformed
5	0.029	normal wear	5	-	※ unevaluated due to occurrence of edge tip fracture
6	0.017	normal wear	6	0.045	edge tip deformed
7	0.023	normal wear	7	0.044	edge tip deformed ※ Edge tip chipping occurred
8	0.025	normal wear	8	0.048	edge tip deformed
9	0.017	normal wear	9	0.079	edge tip deformed
10	0.022	normal wear			
11	0.028	normal wear			

[0081] A hard coating layer having an average layer thickness shown in Table 6 was formed by coating on the cutting edge surfaces of the present invention tools 1 to 4 and the comparative example tools 1 to 4 by a PVD method or a CVD method to produce the present invention surface-coated WC-based cemented carbide cutting tools (hereinafter, referred to as "the present invention coated tools") 1 to 4 and comparative example surface-coated WC-based cemented carbide cutting tools (hereinafter, referred to as "comparative example coated tools") 1 to 4.

[0082] A wet continuous cutting test to be shown below was performed on each of the coated tools, a flank face plastic deformation amount of the cutting edge was measured, and the wear state of the cutting edge was observed.

Cutting Conditions:

Work Material: round bar of JIS-SUS304 (HB170),

Cutting Speed: 150 m/min,

Cutting Depth: 2.0 mm,

Feeding: 0.5 mm/rev,

Cutting Time: 5 minutes, and

Using wet water-soluble coolant

Table 7 shows the results of the cutting test.

[Table 6]

Tool Type			Type of Hard Coating Layer (components, composition)	Average Layer Thickness of Coating Layer
Present Invention Coated Tool	Comparative Example Coated Tool	1	(Ti _{0.4} Al _{0.6})N	3.0
		2	(Ti _{0.5} Al _{0.5})N	1.5
		3	TiN (1 μm)/TiCN (5 μm)/Al ₂ O ₃ (2 μm)	8.0
		4	TiN (0.5 μm)/TiCN (4 μm)/Al ₂ O ₃ (1 μm)	5.5
(Note) The composition shows an atomic ratio.				

[Table 7]

Type		Flank Face Plastic Deformation Amount (mm)	Wear state of Cutting Edge	Type		Flank Face Plastic Deformation Amount (mm)	Wear state of Cutting Edge
Present Invention Coated Tool	1	0.008	normal wear	Comparative Example Coated Tool	1	0.065	edge tip deformed
	2	0.018	normal wear		2	0.041	edge tip deformed
	3	0.020	normal wear		3	0.072	edge tip deformed
	4	0.012	normal wear		4	0.043	edge tip deformed

[0083] According to the test results shown in Tables 5 and 7, it has been found that the present invention tools and the present invention coated tools exhibit excellent plastic deformation resistance without the occurrence of chipping.

[0084] In contrast, the comparative example tools and the comparative example coated tools had poor chipping resistance and plastic deformation resistance, and had a short life.

[Industrial Applicability]

[0085] As described above, a WC-based cemented carbide tool and a coated tool of the present invention have excellent plastic deformation resistance and chipping resistance in a case where the tools are provided for cutting of a hard-to-cut material such as stainless steel. Even in a case where the tools are applied to other work materials and

cutting conditions, the tools exhibit excellent cutting performance over a long period of use, and the life of the tools are expected to be increased.

[Reference Signs List]

[0086]

- 1: Coarse WC grain
- 2: Fine WC grain
- 3: Rake face
- 4: Flank face
- 5: Cutting edge
- 6: Flank face plastic deformation amount
- 7: Line segment drawn by extending ridge where flank face and rake face intersect
- L: Lattice length a

Claims

1. A WC-based cemented carbide cutting tool comprising:
 - a cutting tool body made of WC-based cemented carbide,
 - wherein the WC-based cemented carbide has a component composition containing Co: 5 to 14 mass%, Cr_3C_2 : 0.1 to 1.4 mass%, and a the WC balance containing inevitable impurities,
 - in a case where a grain size distribution is obtained by measuring grain sizes of WC grains in a cross-section of the WC-based cemented carbide, a plurality of local maximums are present in the grain size distribution, and
 - in a case where a grain size corresponding to a most frequent value formed on a coarse grain side of $3\text{ }\mu\text{m}$ or more is represented by r_1 , and a grain size corresponding to a most frequent value formed on a fine grain side of less than $3\text{ }\mu\text{m}$ is represented by r_2 , a grain size ratio r_2/r_1 is 0.15 or more and 0.60 or less, and
 - a total area ratio $A_2 \times 100 / (A_1 + A_2)$ is 5 to 35 area%, A_1 being a total area of WC grains having a grain size of 0.75 to 1.20 times of r_1 and A_2 being a total area of WC grains having a grain size of 0.50 to 1.20 times of r_2 .
2. The WC-based cemented carbide cutting tool according to Claim 1,
 - wherein the WC-based cemented carbide further contains at least one selected from TaC, NbC, TiC, and ZrC in a total amount of 4 mass% or less.
3. A surface-coated WC-based cemented carbide cutting tool comprising:
 - the WC-based cemented carbide cutting tool according to Claim 1 or 2; and
 - a hard coating layer formed on at least a cutting edge of the WC-based cemented carbide cutting tool.

FIG. 1

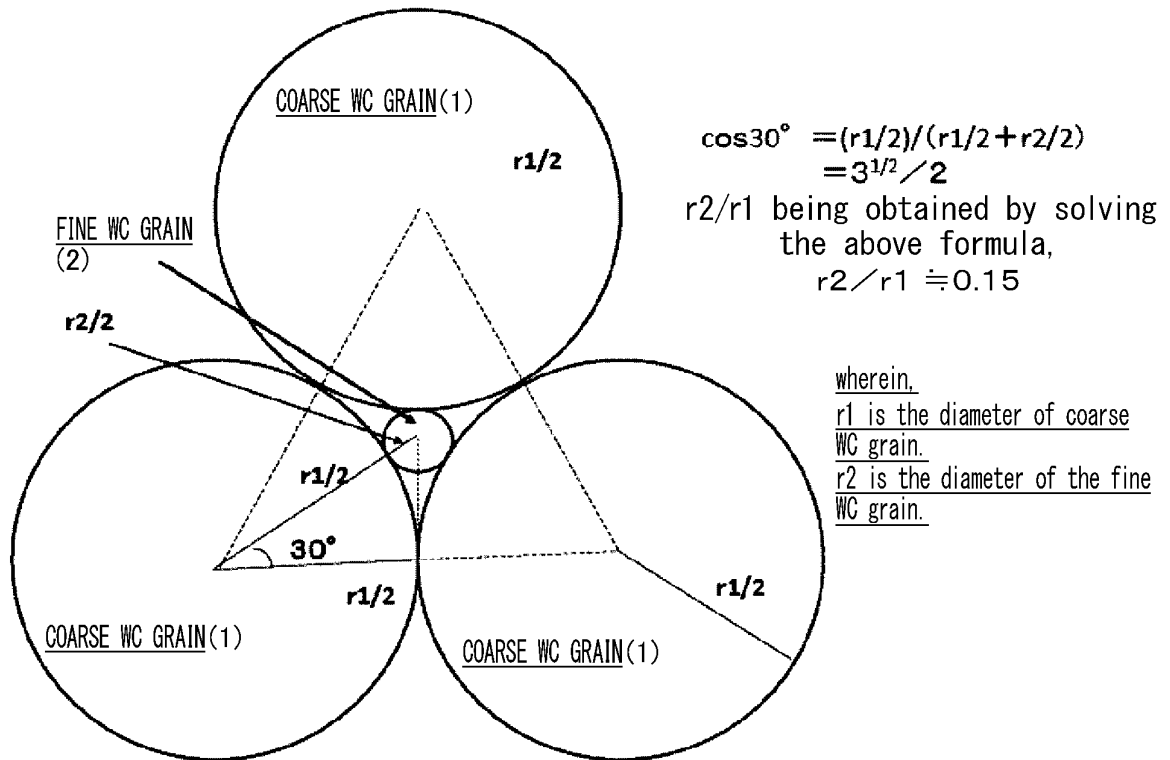


FIG. 2

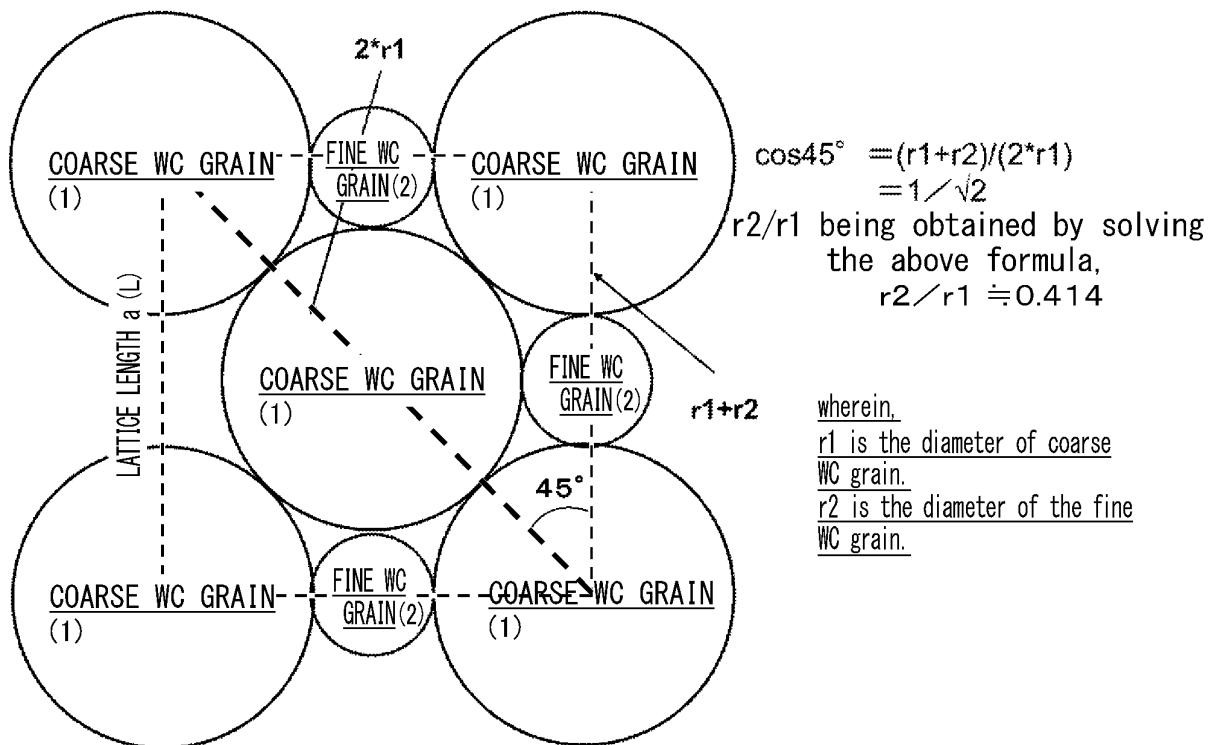
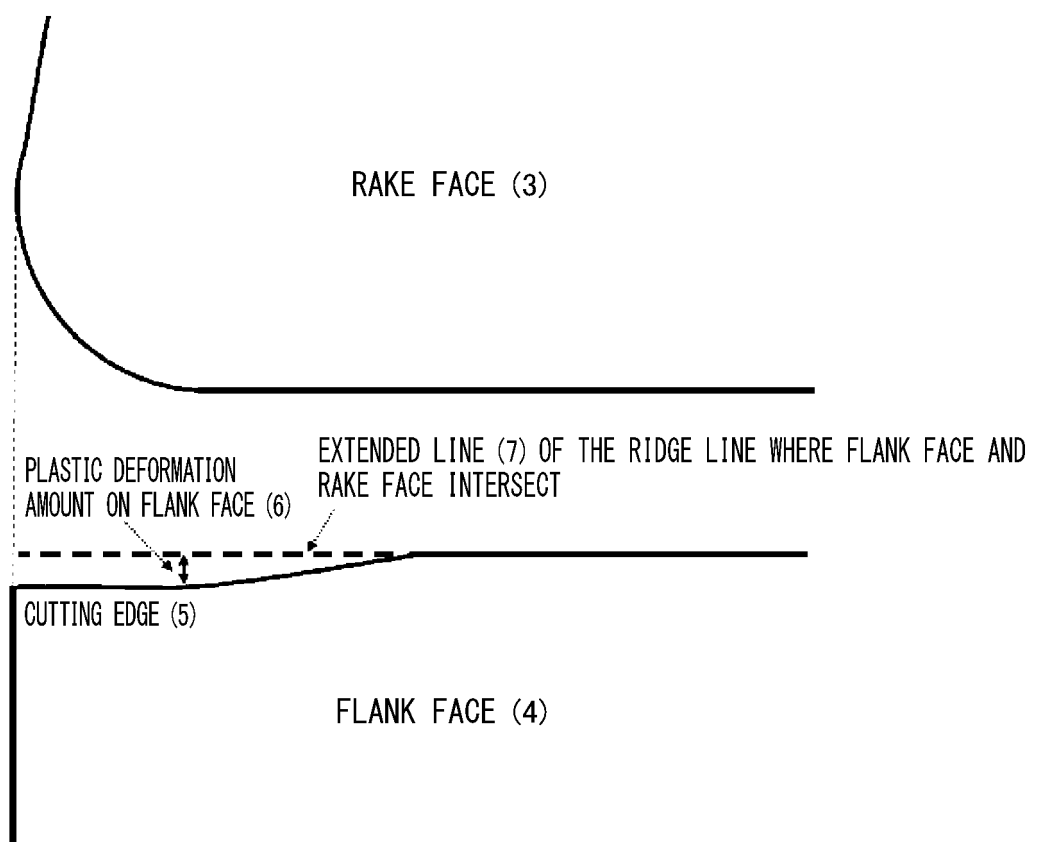


FIG. 3



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INTERNATIONAL SEARCH REPORT

International application No.

PCT/JP2020/013232

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A. CLASSIFICATION OF SUBJECT MATTER

Int.Cl. B22F3/24(2006.01)i, B23B27/14(2006.01)i, C22C29/08(2006.01)i,
C22C1/05(2006.01)i

FI: C22C29/08, B23B27/14B, C22C1/05H, B22F3/24102A

According to International Patent Classification (IPC) or to both national classification and IPC

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B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

Int.Cl. B22F3/24, B23B27/14, C22C29/00-29/18, C22C1/05

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Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Published examined utility model applications of Japan 1922-1996

Published unexamined utility model applications of Japan 1971-2020

Registered utility model specifications of Japan 1996-2020

Published registered utility model applications of Japan 1994-2020

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

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C. DOCUMENTS CONSIDERED TO BE RELEVANT

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Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	JP 8-302441 A (SUMITOMO ELECTRIC INDUSTRIES, LTD.) 19.11.1996 (1996-11-19), entire text	1-3
A	JP 2005-256076 A (TUNGALOY CORP.) 22.09.2005 (2005-09-22), entire text	1-3
A	JP 9-227981 A (TOKYO TUNGSTEN CO., LTD.) 02.09.1997 (1997-09-02), entire text	1-3
A	US 2011/0315051 A1 (OLSEN, G. T.) 29.12.2011 (2011-12-29), entire text	1-3

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☐	Further documents are listed in the continuation of Box C.	☒	See patent family annex.
*	Special categories of cited documents:	"T"	later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"A"	document defining the general state of the art which is not considered to be of particular relevance	"X"	document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
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"L"	document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)	"&"	document member of the same patent family
"O"	document referring to an oral disclosure, use, exhibition or other means		
"P"	document published prior to the international filing date but later than the priority date claimed		

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Date of the actual completion of the international search
26.05.2020Date of mailing of the international search report
09.06.2020

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Name and mailing address of the ISA/
Japan Patent Office
3-4-3, Kasumigaseki, Chiyoda-ku,
Tokyo 100-8915, Japan

Authorized officer

Telephone No.

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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No.

PCT/JP2020/013232

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JP 8-302441 A 19.11.1996 (Family: none)
 JP 2005-256076 A 22.09.2005 (Family: none)
 JP 9-227981 A 02.09.1997 (Family: none)
 US 2011/0315051 A1 29.12.2011 WO 2011/162775 A1
 entire text
 EP 2607511 A2
 CA 2802854 A
 AU 2012261560 A

REFERENCES CITED IN THE DESCRIPTION

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