

Assessing the performance of blended byproduct caps for revegetation and closure of tailings storage facilities

G Taki *The University of Western Australia, Australia*

PF Grierson *The University of Western Australia, Australia*

N Saini *The University of Western Australia, Australia*

HEA Brand *Australian Synchrotron, Australia*

DV Murphy *Murdoch University, Australia*

TC Santini *The University of Western Australia, Australia*

Abstract

Establishment of a vegetative cover during closure of tailings storage facilities is a critical component of the development of an environmentally sustainable landscape after mining. However, establishing vegetation on fresh bauxite residue (alumina refining tailings) is constrained by the high alkalinity, salinity, sodicity, elevated concentration of trace elements, and low plant available nutrients in residues. Currently, design of store and release vegetative covers for closure of tailings storage facilities in southwest Australia requires excavation of local soils and importing nutrients and mulch to apply on top of the tailings storage facility. Where the residues are mostly benign, in situ remediation (application of amendments directly into tailings to remediate the chemical and physical conditions) techniques may be a viable approach to create a plant growth medium for closure and revegetation. Nevertheless, using imported soils and blending products is expensive. Neutralisation of bauxite residue disposal areas (BRDAs) for capping offers a potential alternative and substantial cost savings, especially when coupled with incorporation of materials to develop an improved substrate for plant growth.

*In this study, a new technique called ‘blended byproduct capping’ was developed for closure of the South32 Worsley Alumina BRDA in southwest Australia. The blended byproduct cap uses bauxite processing residues that are blended with available byproducts readily and cheaply available onsite at the refinery. Three types of bauxite processing residue (bauxite residue fines, bauxite residue fines plus 10% bauxite residue sand, and bauxite residue sand) were blended with three byproducts (fly ash from power generation, eucalypt mulch from site clearing, and gypsum from other operations nearby) either alone or in combination to create 15 potential capping materials. These capping materials were leached under glasshouse conditions for 18 weeks (three wetting and drying cycles, three weeks each) to assess changes in pH, EC, total elements and nutrients. The three best performing capping materials in terms of chemo-physical properties were then selected for germination and growth experiments. Germination rates of barley (*Hordeum vulgare*), ryegrass (*Lolium multiflorum*) and clover (*Trifolium spumosum*) were assessed and then surviving plants grown for four weeks after the first visible leaf was observed. Root and shoot biomass were harvested at the end of the experiment. More than 90% of barley and ryegrass seeds germinated. Clover germination was less than 60% both blended byproduct caps and potting mix. However, biomass and growth rates were significantly lower in blended byproduct caps compared to potting mix for all three species. Overall, we conclude that blended byproducts caps show significant promise as a cost-effective alternative for BRDA closure and revegetation but require further optimisation.*

Keywords: *bauxite residue deposit area, red mud, bauxite residue sand, germination success, plant growth, annual grass, clover*

1 Introduction

Bauxite refining residues (red muds) are highly alkaline, saline-sodic materials produced during the Bayer process for extraction of alumina from bauxite ore. Annually, 120–150 million tons of bauxite residue is produced and over 3 billion tons are in storage (Power et al. 2011; Santini & Banning 2016). Usually, these residues are placed into land-based storage areas, which may potentially contaminate the surrounding environment if natural hazards or containment failures occur (Kossoff et al. 2014). Consequently, appropriate management and remediation of residues is essential to reduce these risks. However, the management of bauxite residues is constrained by their harsh geochemical properties and low concentrations of plant available nutrients (Santini et al. 2021).

Currently, cap and store and/or in situ remediation are the two main approaches used to close and revegetate bauxite residues deposit areas (BRDAs). Cap and store involves importing capping materials (e.g. fresh top soil, fly ash etc.) from offsite to enclose the BRDA and as a substrate for plant growth. In contrast, in situ remediation involves applying organic and inorganic amendments directly into the residues, in order to improve properties sufficiently for a vegetation cover to be established on the tailings. The major drawbacks of cap and store are; the large quantities of material required, the challenge of providing sufficient nutrients over time for plant survival in the cap alone, and capillary rise of alkaline porewater from underlying tailings, which can then negatively impact the vegetation if the impermeable layer separating tailings from cap is compromised (Wehr et al. 2006). In situ remediation also presents challenges: decomposition and incorporation of organic or inorganic materials into, and their reaction with, the residues is a slow process and the majority of the underlying residue remains un-remediated for an extended period. In situ remediation approach generally does not incorporate an impermeable capping layer, meaning that the BRDA can generate large volumes of potentially toxic or damaging leachates during rainy seasons (Santini & Fey 2015). Consequently, a new approach called ‘blended byproduct capping’ has been developed for closure and revegetation of BRDAs to try and overcome some of these limitations. Blended byproduct capping involves creating a capping material by blending bauxite processing residues with other byproducts available onsite or from nearby operations that is both cost-effective and creates better conditions for plant survival.

In this study, we blended three types of bauxite processing residues [(i) bauxite residue fines (BRF); (ii) bauxite residue fines plus 10% bauxite residue sand (BRF + 10% BRS); and (iii) bauxite residue sand (BRS)] with three byproducts (fly ash, eucalypt mulch, and gypsum) to develop novel capping materials. Fly ash is commonly used to ameliorate sandy soil (Pathan et al. 2003), has been used as a capping material for BRDA closure (Santini & Fey 2015), and also as an amendment to promote bauxite residue neutralisation (Khaitan et al. 2008). Vegetation mulches can improve water holding capacity, aeration, drainage, organic C and microbial activity, and decrease pH, water loss and prevent wind and rainfall erosion (Ippolito et al. 2005; Chalker-Scott 2007; Mupangwa et al. 2013). Gypsum, generated as a byproduct in the phosphate fertiliser industry, has also been extensively used for bauxite residue remediation due to its ability to reduce pH and sodicity and increase aggregation (Courtney & Kirwan 2012; Bray et al. 2018). Although application of these byproducts may improve bauxite residue properties, any single byproduct is unlikely to ameliorate the bauxite residues properties to an extent suitable for plant growth. For example, a lack of nutrients and organic matter, as well as high pH, Na content and trace elements, inhibited plant growth in bauxite residue (Anderson et al. 2011; Fourrier et al. 2021). Therefore, combining multiple byproducts may enhance transformation of bauxite residue into a substrate that is more suitable for establishment and persistence of a vegetative cover.

The overall aim of this study was to evaluate the suitability of three blended byproduct caps of different textures (fines and sand content) to support plant growth and survival. Three annual crop species (*Hordeum vulgare*, *Lolium multiflorum* and *Trifolium spumosum*) were selected for a germination and growth trial. We expected ryegrass (*L. multiflorum*) to show the best germination and survival rate given its known high tolerance to alkalinity, salinity, and sodicity (Courtney & Mullen 2009; Jones et al. 2015). We expected all species to perform best on the byproduct cap that combined fines with 10% sand owing to a less alkaline pH, lower salinity, and overall improved substrate structure.

2 Materials and methods

All bauxite residues and capping material used in this study were provided from onsite by South32 Worsley Alumina in southwest Western Australia, except gypsum (obtained from commercial provider). First, blended byproduct capping materials were prepared with three types of residues: (i) bauxite residue fines (BRF); (ii) bauxite residue fines plus 10% bauxite residue sand (BRF + 10% BRS); and (iii) bauxite residue sand (BRS), amended with fly ash (10 wt%), eucalypt mulch (5 wt%), and gypsum (3 wt%). The blended mixtures were then leached/weathered for 18 weeks (three wetting cycles and three drying cycles, three weeks each; to mimic three wet-dry season cycles common in the Mediterranean-type climate of the region) before use in germination trials. Chemical properties of the capping materials were analysed at the CSBP Soil and Plant Analysis Laboratory in Western Australia (www.csbplab.com.au) (Table 1). Individual pots were prepared with 2.5 kg capping materials, using a transparent polyethylene bag lining inside of each pot before adding the materials to prevent significant drainage loss/leaching during the experiment. Given there have been no examples of successful revegetation of fresh bauxite refining residues to date (Di Carlo et al. 2020), we used a commercial potting mix as a positive control rather than negative control (i.e. unamended bauxite residues). Then 20 seeds of either barley, ryegrass or clover were sown per pot. *H. vulgare*, and *L. multiflorum* seeds were pre-treated with 1% sodium hypochloride (NaOCl) and 0.01 mM calcium chloride (CaCl₂). *T. spumosum* seeds were gently scoured with sandpaper to help break seed dormancy, and then treated with 1% NaOCl and 0.01 mM CaCl₂. Overall, 48 pots (four treatments × three crop species × four replicates) were established in a growth chamber at constant temperature of 20°C and a day length of 12 hours at The University of Western Australia in Perth, Western Australia.

Germination within each pot was recorded daily. Protruded visible cotyledon/s were considered as germinated. Growth was observed for four weeks, beginning after the first leaf appeared. Therefore, barley and ryegrass were harvested at 35 days after sowing while clover was harvested at 42 days after sowing (i.e. clover was much slower to germinate). Dead plants within each pot were recorded daily to determine the survival rate at the end of the experiment. To provide essential nutrients for plants, modified Hoagland solution (120.4 mL per pot) was applied in weekly doses to each pot containing K (235 mg/L), S (128.17 mg/L, calculated), Mg (48 mg/L), B (0.5 mg/L), Mn (0.5 mg/L), Zn (0.05 mg/L), Cu (0.02 mg/L), and Mo (0.01 mg/L) after dilution with deionised water. Di-ammonium phosphate was applied as source of N (280.38 mg) and P (310.12 mg) per pot. At harvest, plant height, number of leaves, and leaf-length were recorded. Leaves and stems were separated and then dried at 65°C for 72 hours to record shoot biomass. Roots were washed and preserved into 70% ethanol to measure the root length, then dried at 65°C for 72 hours to record root biomass. Leaves, shoots and roots are currently being analysed for nutrient and heavy metal content. Owing to timing and logistic limitations, only germination, survival, and shoot biomass data are discussed here.

Analysis of variance (ANOVA) and Post hoc Tukey Honestly Significant Difference tests were used to assess differences in germination, survival and shoot growth of the three species among different blends of capping materials. Data were analysed using GenStat 19th edition (www.vsnl.co.uk).

3 Results and discussion

3.1 Germination and survival rate

Germination in bauxite residues is commonly inhibited by the excessive salinity and sodicity (Courtney & Mullen 2009). In this study, salinity and exchangeable Na⁺ considerably decreased owing to the application of byproducts, along with pre-experiment preparation of bauxite residues by leaching/weathering (Table 1, EC, 4.37–4.64 dS/m to 0.18–0.33 dS/m and exchangeable Na⁺ 38.91–44.03 meq/100 g to 0.65–3.96 meq/100 g). Alkalinity of the capping materials decreased from very strongly alkaline (unamended capping materials pH, 10.5–10.6) to strongly or moderate alkaline level (amended capping materials pH, 8.3–8.8) after byproducts application and leaching/weathering. Addition of byproducts to the blended capping materials increased organic C and Ca content compared to initial bauxite residues (Table 1). Organic C was likely increased from residual unburned coal content within the fly ash (Santini & Fey 2015; Li et al. 2021).

Table 1 Chemical properties of byproducts capping materials

Chemical properties	Units	Unamended capping materials (before leaching)			Amended capping materials (after leaching)		
		*BRF	*BRF + 10% BRS	*BRS	*BRF	*BRF + 10% BRS	*BRS
pH		10.5	10.5	10.6	8.8	8.6	8.3
EC	dS/m	4.48	4.64	4.37	0.20	0.18	0.33
Ammonium Nitrogen	mg/kg	< 1	< 1	< 1	< 1	< 1	< 1
Nitrate Nitrogen	mg/kg	< 1	< 1	< 1	< 1	< 1	< 1
Colwell P	mg/kg	20	17	2	11	10	5
Colwell K	mg/kg	40	42	< 15	30	30	< 15
S	mg/kg	399.0	366.7	129.0	27.1	23.7	99.7
OC	%	0.14	0.18	0.11	0.89	1.02	0.87
DTPA Cu	mg/kg	1.76	1.47	0.49	0.25	0.23	0.21
DTPA Fe	mg/kg	11.10	12.00	7.00	6.00	4.00	5.40
DTPA Mn	mg/kg	0.39	0.64	0.07	0.08	0.09	0.07
DTPA Zn	mg/kg	0.27	0.24	0.35	0.10	0.18	0.19
Exc. Al	meq/100 g	0.030	0.040	0.020	0.010	0.010	0.020
Exc. Ca	meq/100 g	2.91	2.93	2.05	9.51	9.46	8.24
Exc. Mg	meq/100 g	0.23	0.17	0.01	0.22	0.20	0.03
Exc. K	meq/100 g	0.06	0.05	0.03	0.01	0.02	0.01
Exc. Na	meq/100 g	41.39	44.03	38.91	3.96	3.79	0.65
B	mg/kg	0.38	0.53	0.32	0.30	0.31	0.28

*BRF = bauxite residue fines; BRF + 10% BRS = bauxite residue fines + 10% bauxite residue sand; BRS = bauxite residue sand

Exchangeable Ca content increased due to gypsum application (Courtney et al. 2003; Ippolito et al. 2005). Barley (*H. vulgare*) and ryegrass (*L. multiflorum*) had 90–100% germination rates in both the potting mix and in the capping materials (Figure 1). By contrast, clover (*T. spumosum*) had significantly lower germination rates in both the potting mix and in the capping materials (maximum 53.8% germinated with BRS), regardless of residue type (Figure 1). This may be a result of overall lower seed viability but could also reflect differing conditions required to break seed dormancy of a dicotyledonous herb (*T. spumosum*) compared to annual grasses (Hudson et al. 2015).

All barley and ryegrass that germinated survived in all treatments; there were no significant differences among any combination of these two species and capping materials at the conclusion of the trial (Figures 2 and 3). Conversely, *T. spumosum* had significantly lower survival rates (Figures 2 and 3). The maximum survival rate of *T. spumosum* was observed in the potting mix (96.9%) and in the BRS (55.8%), which were significantly higher ($P \leq 0.05$) than within the BRF, and BRF + 10% BRS. These results suggested that the bauxite residues, particularly finer materials, are still hostile for *T. spumosum* and that this species is less tolerant of alkalinity compared to *H. vulgare* and *L. multiflorum*. Other clover species have also been found to be relatively intolerant to high alkalinity (e.g. *T. pratense*; Courtney & Mullen 2009).

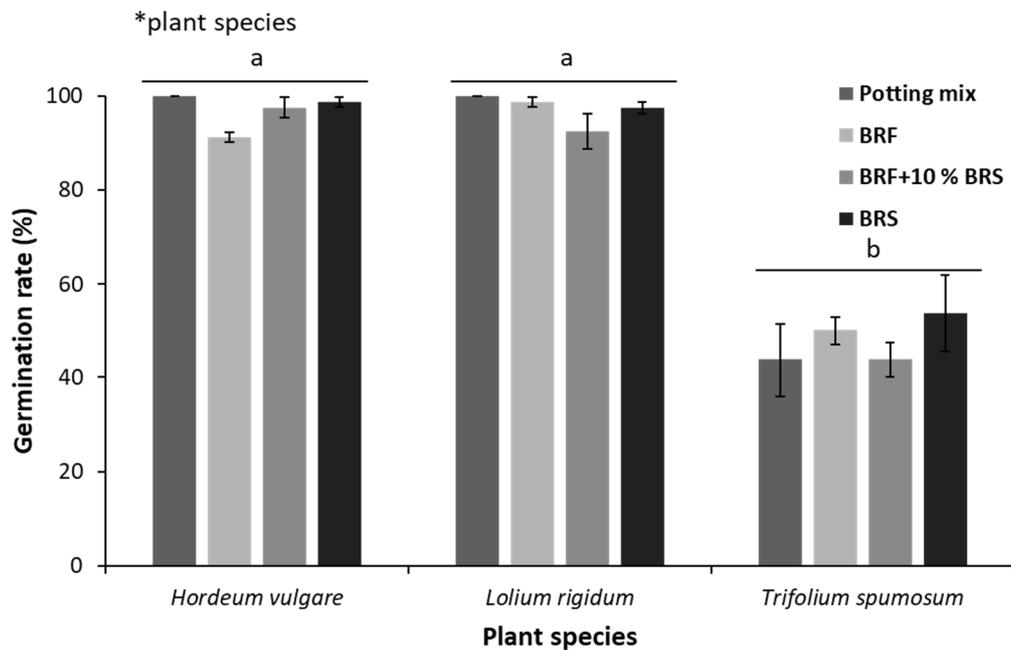


Figure 1 Germination rate of three plant species [(a) *H. vulgare*; (b) *L. multiflorum*; (c) *T. spumosum*] in the potting mix and different capping materials. Values are means of ± 1 standard error of four replicates. Means with different letters of each species are significantly different ($P \leq 0.05$) according to two-ways ANOVA. BRF = bauxite residue fines, BRF + 10% BRS = bauxite residue fines + 10% bauxite residue sand, BRS = bauxite residue sand

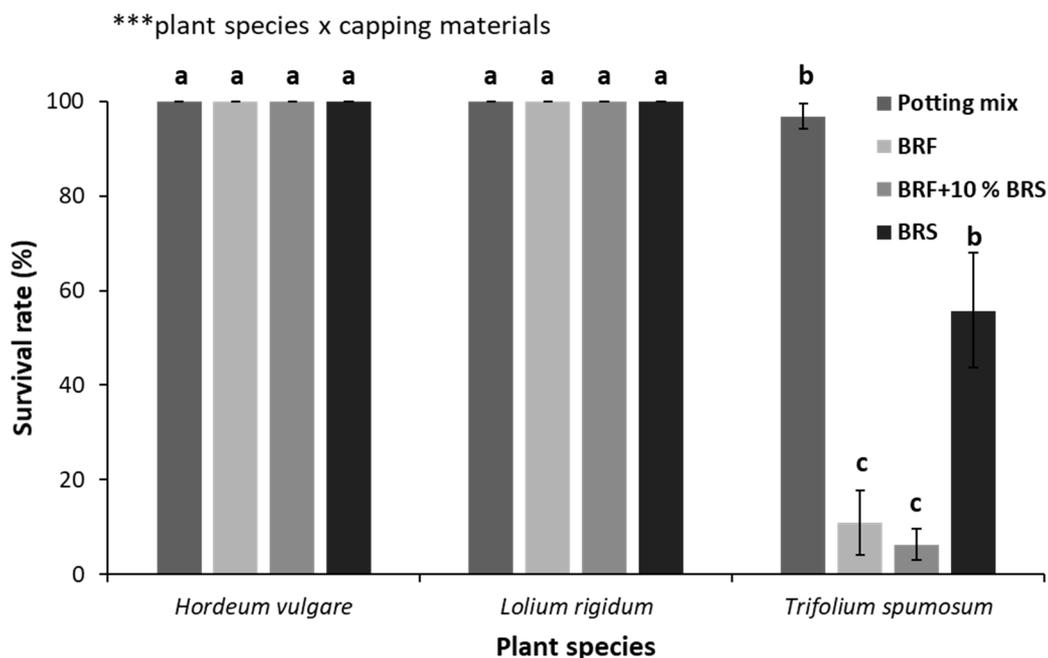


Figure 2 Survival rate of three plant species [(a) *H. vulgare*; (b) *L. multiflorum*; (c) *T. spumosum*] in the potting mix and different capping materials. Values are means of ± 1 standard error of four replicates. Means with different letters of each species are significantly different ($P \leq 0.05$) according to two-ways ANOVA. BRF = bauxite residue fines, BRF + 10% BRS = bauxite residue fines + 10% bauxite residue sand, and BRS = bauxite residue sand

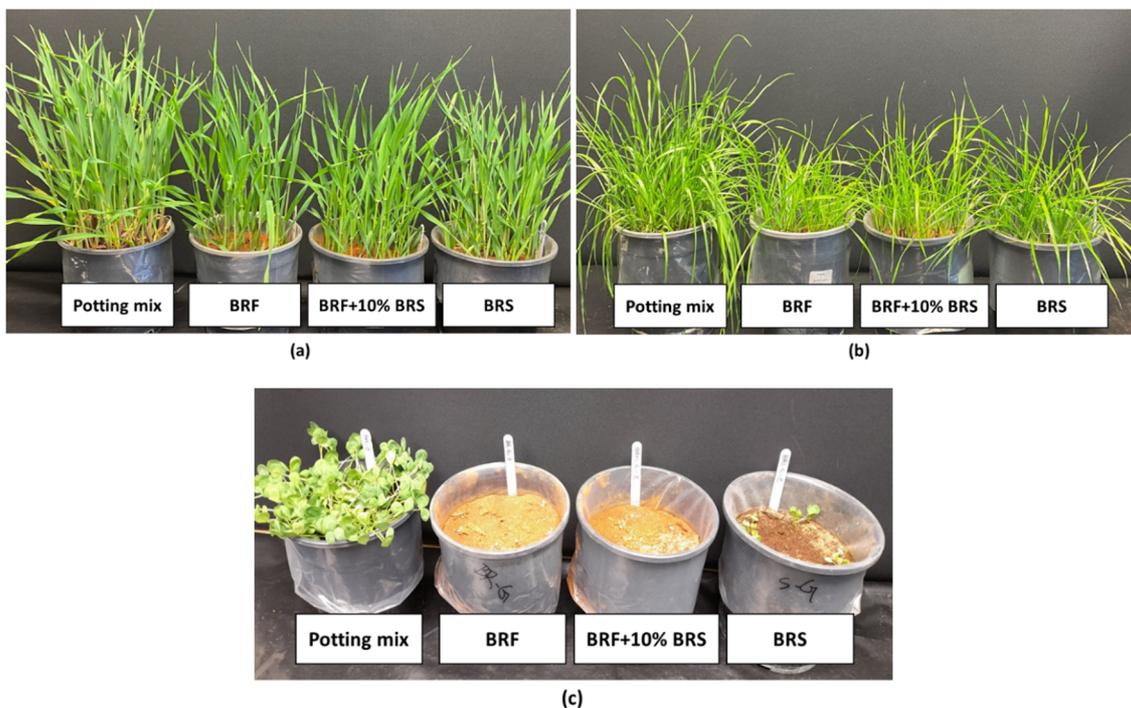


Figure 3 Plants at the end of harvesting at 35 days (a) *H. vulgare*; (b) *L. multiflorum*, and at 42 days (c) *T. spumosum* in the potting mix and different capping materials. BRF = bauxite residue fines, BRF + 10% BRS = bauxite residue fines + 10% bauxite residue sand, and BRS = bauxite residue sand

3.2 Shoot growth

Shoot dry weights of all three species were significantly lower in blended byproduct capping materials compared to the potting mix ($P \leq 0.05$) (Figures 3 and 4), despite applying nutrients to the capping materials throughout the experiment. The low germination and survival rate of *T. spumosum* (Figures 1 and 2) means that shoot dry weight data for this species should be interpreted with some caution owing to low replication (Figures 3 and 4).

Limited plant shoot biomass likely reflects the high alkalinity and potentially metal toxicity (especially Al and Na) in the blended byproduct caps. Salinity was below growth-restricting levels (0.18–0.33 dS/m, well below the plant growth restriction level of >2 dS/m; Hazelton & Murphy 2007). Nutrient deficiencies are very common in bauxite residues and generally leaching causes a further decrease of plant available nutrients; however, nutrients were applied throughout the growth phase to avoid nutrient limitation. Alkalinity is widely recognised to inhibit plant growth due to its buffering capacity of chemical reactions in the residues, which may limit nutrient availability (Di Carlo et al. 2020; Zhang et al. 2017). Alternatively, high pH also can enhance the availability of metals and trace elements in the residue, which can be toxic for plants (Di Carlo et al. 2019). Ongoing analysis of both shoot and root samples from this experiment should help elucidate how possible nutrient imbalances and/or toxicity may have impacted on plant growth.

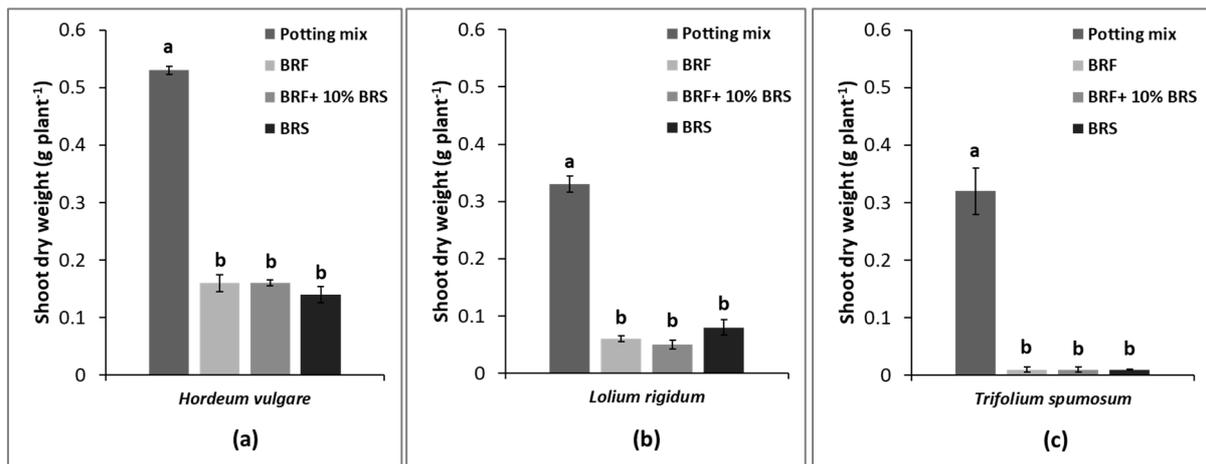


Figure 4 Shoot dry weight of (a) *H. vulgare*; (b) *L. multiflorum* (harvested at 35 days); (c) *T. spumosum* (harvested at 42 days) in the potting mix and different capping materials. Values are means of ± 1 standard error of four replicates. Means with different letters of each species are significantly different ($P \leq 0.05$) according to one-way ANOVA. BRF = bauxite residue fines, BRF + 10% BRS = bauxite residue fines + 10% bauxite residue sand, and BRS = bauxite residue sand

4 Conclusion

Overall, we conclude that blended byproducts caps show significant promise as a cost-effective alternative for BRDA closure and revegetation but require further improvement to reduce alkalinity and address possible nutrient imbalances. Our study showed application of byproducts along with a period of leaching/weathering improved geochemical properties of blended byproducts capping material, resulting in high seed germination, survival, and plant growth rates. Additional pre-treatment of both bauxite residues and organic amendments to improve biological functioning of blended byproducts may further enhance development of soil like properties. Utilisation of readily available onsite materials for capping is faster and with leaching is more cost-effective than using external material.

Blended byproduct caps have demonstrated potential to support the following strategic closure planning outcomes, once remaining alkalinity is removed and fertiliser is added:

- Reduced capital expenditure (CAPEX) costs (compared with cap and store) due to decreased reliance on import of soil capping materials.
- Decreased time to closure due to faster vegetation establishment.
- Potential for reduced ongoing maintenance due to decreased erosion and capillary rise.
- Improved confidence in meeting closure success criteria and progressive certification of this land for future custodial transfer.

Acknowledgement

The authors acknowledge South32 Worsley Alumina Pty Ltd and Australian Research Council (ARC) for their funding to conduct this research. The first author thanks Australian Government for Research Training Program (RTP) Fees Offset scholarship, The University of Western Australia for University Postgraduate Award (UPA), and the Australian Institute of Nuclear Science and Engineering (AINSE) for Postgraduate Research Award (AINSE-PGRA) to support his study.

References

Anderson, JD, Bell, RW & Phillips, IR 2011, 'Bauxite residue fines as an amendment to residue sands to enhance plant growth potential—a glasshouse study', *Journal of Soils and Sediments*, vol. 11, pp. 889–902.

- Bray, AW, Stewart, DI, Courtney, R, Rout, SP, Humphreys, PN, Mayes, WM & Burke, IT 2018, 'Sustained bauxite residue rehabilitation with gypsum and organic matter 16 years after initial treatment', *Environmental Science & Technology*, vol. 52, no. 1, pp. 152–161.
- Chalker-Scott, L 2007, 'Impact of mulches on landscape plants and the environment — a review', *Journal of Environmental Horticulture*, vol. 25, no. 4, pp. 239–249.
- Courtney, R, Timpson, JP & Grennan, E 2003, 'Growth of *Trifolium pratense* in red mud amended with process sand, gypsum and thermally dried sewage sludge', *International Journal of Mining, Reclamation and Environment*, vol. 17, no. 4, pp. 227–233.
- Courtney, R & Mullen, G 2009, 'Use of Germination and Seedling Performance Bioassays for Assessing Revegetation Strategies on Bauxite Residue', *Water, Air, & Soil Pollution*, vol. 197, pp. 15–22.
- Courtney, R & Kirwan, L 2012, 'Gypsum amendment of alkaline bauxite residue – plant available aluminium and implications for grassland restoration', *Ecological Engineering*, vol. 42, pp. 279–282.
- Di Carlo, E, Boullemant, A & Courtney, R 2019, 'A field assessment of bauxite residue rehabilitation strategies', *Science of The Total Environment*, vol. 663, pp. 915–926.
- Di Carlo, E, Boullemant, A & Courtney, R 2020, 'Ecotoxicological risk assessment of revegetated bauxite residue: Implications for future rehabilitation programmes', *Science of The Total Environment*, vol. 698.
- Fourrier, C, Luglia, M, Keller, C, Hennebert, P, Foulon, J, Ambrosi, JP, ... Criquet, S 2021, 'How Raw and Gypsum Modified Bauxite Residues Affect Seed Germination, Enzyme Activities, and Root Development of *Sinapis alba*', *Water, Air, & Soil Pollution*, vol. 232, p. 309.
- Hazelton, P & Murphy, B 2007, *Interpreting Soil Test Results: What do all the Numbers mean?*, CSIRO Publishing, Melbourne.
- Hudson, AR, Ayre, DJ & Ooi, MK 2015, 'Physical dormancy in a changing climate', *Seed Science Research*, vol. 25, no. 2, pp. 66–81.
- Ippolito, JA, Redente, EF & Barbarick, KA 2005, 'Amendment effects on pH and salt content of bauxite residue', *Soil Science*, vol. 170, no. 10, pp. 832–841.
- Jones, BEH, Haynes, RJ & Phillips, IR 2015, 'Addition of an organic amendment and/or residue mud to bauxite residue sand in order to improve its properties as a growth medium', *Journal of Environmental Management*, vol. 95, no. 1, pp. 29–38.
- Kossoff, D, Dubbin, WE, Alfredsson, M, Edwards, SJ, Macklin, MG & Hudson-Edwards, KA 2014, 'Mine tailings dams: characteristics, failure, environmental impacts, and remediation', *Applied Geochemistry*, vol. 51, pp. 229–245.
- Khaitan, S, Dzombak, DA & Lowry, GV 2008, 'Neutralization of bauxite residue with acidic fly ash', *Environmental Engineering Science*, vol. 26, no. 2, pp. 431–440.
- Li, GK, Fischer, WW, Lamb, MP, West, AJ, Zhang, T, Galy, V, ... Ji, J 2021, 'Coal fly ash is a major carbon flux in the chang jiang (yangtze river) basin', *Proceedings of the National Academy of Sciences*, vol. 118, no. 21.
- Mupangwa, W, Twomlow, S & Walker, S 2013, 'Cumulative effects of reduced tillage and mulching on soil properties under semi-arid conditions', *Journal of Arid Environment*, vol. 91, pp. 45–52.
- Pathan, SM, Aylmore, LAG & Colmer, TD 2003, 'Properties of several fly ash materials in relation to use as soil amendments', *Journal of Environmental Quality*, vol. 32 no. 2, pp. 687–693.
- Power, G, Gräfe, M & Klauber, C 2011, 'Bauxite residue issues: I. Current management, disposal, and storage practices', *Hydrometallurgy*, vol. 108, no. 1–2, pp. 33–45.
- Santini, TC & Fey, MV 2015, 'Fly ash as a permeable cap for tailings management: Pedogenesis in bauxite residue tailings', *Journal of Soils and Sediments*, vol. 15, no. 3, pp. 552–564.
- Santini, TC & Banning, NC 2016, 'Alkaline tailings as novel soil forming substrates: reframing perspectives on mining and refining wastes', *Hydrometallurgy*, vol. 164, pp. 38–47.
- Santini, TC, Wang, JC, Warren, KL, Pickering, G & Raudsepp, MJ 2021, 'Simple organic carbon sources and high diversity inocula enhance microbial bionutralization of alkaline bauxite residues', *Environmental Science & Technology*, vol. 55, no. 6, pp. 3929–3939.
- Wehr, JB, Fulton, I & Menzies, NW 2006, 'Revegetation strategies for bauxite refinery residue: A case study of Alcan Gove in northern territory, Australia', *Journal of Environmental Management*, vol. 37, no. 3, pp. 297–306.
- Zhang, H, Li, X, Nan, X, Sun, G, Sun, M, Cai, D & Gu, S 2017, 'Alkalinity and salinity tolerance during seed germination and early seedling stages of three alfalfa (*Medicago sativa* L.) cultivars', *Legume Research*, vol. 40, no. 5, pp. 853–858.